THE RUSSIAN FEDERATION MINISTRY
ON SCIENCE AND EDUCATION

NATIONAL RESEARCH NUCLEAR UNIVERSITY MEPhI
(MOSCOW ENGINEERING PHYSICS INSTITUTE)

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ENVIRONMENTAL PROTECTION IN CLOSED NUCLEAR FUEL CYCLE AND NUCLEAR WEAPONS NON-PROLIFERATION PROBLEM

This book is recommended
by the Training and Methodological Association of higher schools
in the educational direction
140300 "Nuclear Physics and Technologies"
to be used as a textbook by the higher school students who are taught in
the educational direction "Nuclear Physics and Technologies"

Moscow 2014

УДК 621.039.516.4:502.1(075)
ББК 31.46я7
Е58

Environmental protection in closed nuclear fuel cycle and nuclear weapons non-proliferation problem: Textbook / *V.A. Apse, A.N. Shmelev, E.G. Kulikov, G.G. Kulikov.* M.: NRNU MEPhI, 2014. 220 p.

The problems related with closing nuclear fuel cycle (NFC) are considered from the following two points of view, namely negative ecological effects of the open and closed NFC on the environment, and increased threats to proliferation of fissile materials which could be used to manufacture nuclear explosive devices (NED). Main stages of the closed NFC, starting from mining and primary treatment of uranium ores and ending by ultimate disposal of radioactive wastes (RAW) from radiochemical reprocessing of spent nuclear fuel (SNF), are briefly characterized. The textbook presents some results obtained in numerical evaluations of the effects produced at various stages of the closed NFC on human health, on the environment, and on a possibility to divert fissile materials from peaceful energy applications to terrorist purposes. The most promising strategies for energy utilization of reactor-grade plutonium and weapon-grade plutonium in nuclear power reactors at NPP are analyzed with accounting for potential applicability of reactor-grade plutonium as a NED charge. The textbook underlines a reasonability to use isotopic denaturing of uranium and plutonium for reliable non-proliferation of nuclear weapons.

The textbook is intended for training of the specialists in nuclear technologies, operation of NFC facilities and nuclear non-proliferation.

The textbook was prepared and published at the expense of the International Science and Technology Center (ISTC) within the frames of the Responsible Science Program of Sub-Program SB159 “Culture of Nuclear Non-Proliferation”.

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ISBN 978-5-7262-1976-9

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INTRODUCTION

Before addressing main topics of the present textbook, i.e. ecological and nuclear non-proliferation problems of nuclear fuel cycles, it is necessary for readers to get some acquaintance with basic conceptions of nuclear power industry.

Peaceful as well as military applications of nuclear energy are based on various nuclear technologies dealing with nuclear materials (NM). Nuclear materials are those substances without which it is impossible to actuate the following two self-sustaining nuclear reactions accompanied by release of huge energy amounts:

1. Chain fission reaction of heavy nuclei.

For example, neutron-induced fission of isotope ^{235}U results in production of two (in very rare cases, three) fission products (FP), in emission of 2.5 fission neutrons (in average) which can continue the chain fission reaction, and in intense generation of thermal energy (about 200 MeV per one fission).

$$^{235}U + n \rightarrow FP_1 + FP_2 + (2\text{-}3)n + 200\ MeV.$$

That is why nuclear materials include all uranium and thorium isotopes (natural NM) and isotopes of artificial transuranium isotopes (mainly isotopes of plutonium, neptunium, americium and curium). Also, nuclear materials include highly radioactive artificial uranium isotope ^{233}U (half-life $T_{1/2} = 1{,}6 \cdot 10^5$ years), which can be produced by neutron irradiation of natural thorium.

2. Thermonuclear fusion reaction of light nuclei.

For example, fusion reaction of light hydrogen isotopes, namely reaction of deuterium with tritium is able to produce stable helium, high-energy neutrons and about 21 MeV of thermal energy:

$$D + T \rightarrow {}^4He + n + 21\ MeV.$$

That is why nuclear materials include two hydrogen isotopes: deuterium and tritium. Abundance of stable deuterium in natural hydrogen is

about 0,015%. Natural hydrogen does not contain its heavier isotope (tritium) because of its rapid radioactive decay ($T_{1/2}$ = 12,3 years). Lithium is also regarded as a nuclear material because its light isotope ^{6}Li can be used for intense production of tritium through ^{6}Li(n,α)T reaction. Micro cross-section of ^{6}Li(n,α)T reaction in thermal point (E_n = 0,025 eV) is sufficiently large (about 940 barns). Natural lithium contains 7,5% ^{6}Li.

Thus, the following NM categories are under consideration now:

1. Initial NM – natural uranium and natural thorium, depleted uranium, i.e. uranium with reduced content of ^{235}U.
2. Special NM – enriched uranium, i.e. uranium with increased content of ^{235}U, plutonium with any isotope composition and artificial uranium isotope ^{233}U.
3. Transuranium elements (Np, Am, Cm, Bk, Cf).
4. Deuterium, tritium, lithium and heavy water.

The first three NM categories are related with nuclear power based on fission reactions of heavy isotopes while the fourth NM category is related with fusion reactions of light isotopes. As thermonuclear power facilities are not built and put in operation yet, main attention in the textbook is given to nuclear technologies dealing with the first three NM categories.

Nuclear technologies include the procedures intended for NM production, storing, applications, transportation, reprocessing for repeat usage of secondary NM or ultimate disposal of technological wastes.

For a long time the world public expresses deep concerns about links between nuclear technologies and safe vital activity of the humankind. Therefore, the textbook gives the largest attention to analysis of these links. The term “safety” should be interpreted here in a wide sense including radiation safety, nuclear safety, non-proliferation safety (or security) and ecological safety.

The term **”radiation safety”** means a sufficient protection against the striking effects caused by direct exposure to any type of ionizing radiations.

The term **”nuclear safety”** means an inadmissibility for the self-sustaining uncontrolled chain fission reaction to initiate and propagate. Serious violations of the nuclear safety requirements can lead to a nu-

clear explosion, thermal explosion or, at least, to the flash of ionizing radiation and over-exposure of operation staff members.

The term **"non-proliferation safety (or security)"** means a sufficient NM protection against their thefts or diversion for manufacturing of nuclear explosive devices or radiological weapons. Presently, the IAEA experts propose to use the term "nuclear security" for designation of this nuclear non-proliferation aspect that differs in principle from the aforementioned term "nuclear safety".

The term **"ecological safety"** means an inadmissibility of unacceptably strong negative impact of nuclear technologies on the environment including radiation, chemical and thermal effects. One else negative effect is related with a necessity to alienate large territories for safe operation of some nuclear facilities.

Nuclear fuel is a nuclear material containing nuclides which can be split (fissioned) by neutrons. The following NM can be regarded as fissionable nuclides:

1. Natural uranium and thorium isotopes.
2. Artificial plutonium isotopes (products of consecutive neutron captures beginning from ^{238}U).
3. Isotopes of artificial transuranium elements (Np, Am, Cm and so on).
4. Artificial uranium isotope ^{233}U (product of neutron capture by ^{232}Th).

As a rule, uranium, thorium and plutonium isotopes with even mass numbers ("even" nuclides ^{238}U, ^{232}Th, ^{240}Pu, ^{242}Pu) can be fissioned only by high-energy neutrons (energy thresholds for neutron-induced fission reactions of these nuclides cover the range from 1 MeV to 1.5 MeV). On the contrary, uranium and plutonium isotopes with odd mass numbers ("odd" nuclides ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu) can be fissioned by neutrons with any energy values including thermal neutrons. Moreover, the lower neutron energy, the more intense fission reaction can occur.

Energy spectrum of fission neutrons is a fast neutron spectrum with mean energy about 2.1 MeV. Besides, these fast neutrons undergo intense slowing down, and their energies sharply drop down below the threshold levels for fission reactions of even nuclides. This means that it is very difficult to maintain the chain fission reaction by even nuclides only because a small fraction of fission neutrons has the energies high enough to overcome the threshold levels. At the same time, it is desirable and quite possible to slow down fission neutrons to thermal

energies and, thus, provide the best conditions for initiation and propagation of the chain fission reaction on odd uranium and plutonium nuclides.

Nuclear fuel containing only natural fissionable nuclides (^{235}U, ^{238}U, ^{232}Th) is named as a primary nuclear fuel. Nuclear fuel containing artificial fissionable nuclides (^{233}U, ^{239}Pu, ^{241}Pu) is named as a secondary nuclear fuel.

Natural fissionable nuclides ^{238}U and ^{232}Th are of little use as a nuclear fuel because they can be fissioned by fast neutrons only. However, these nuclides can be used to produce artificial well-fissionable (or fissile) nuclides ^{239}Pu and ^{233}U, respectively, i.e. for reproduction (or breeding) of secondary nuclear fuel. That is why these nuclides are often named fertile nuclides.

The present nuclear energy systems are based upon the use of natural uranium containing the following three isotopes:

1. ^{238}U; natural abundance – 99,28%; half-life $T_{1/2} = 4{,}5 \cdot 10^9$ years;
2. ^{235}U; natural abundance – 0,71%; half-life $T_{1/2} = 7{,}1 \cdot 10^8$ years;
3. ^{234}U; natural abundance – 0,0054%; half-life $T_{1/2} = 2{,}5 \cdot 10^5$ years.

By the way, the Earth's age (approximately 10 billion years) is comparable with ^{238}U half-life.

It is interesting to note here that ^{234}U is a member of ^{238}U decay family: ^{234}U is produced by α-decay of ^{238}U and two consecutive, relatively rapid β-decays of intermediate nuclides:

$$^{238}\text{U}(\alpha, T_{1/2}=4{,}5 \cdot 10^9 \text{ years})^{234}\text{Th}(\beta, T_{1/2}=24 \text{ d})^{234}\text{Pa}(\beta, T_{1/2}=6{,}7 \text{ h})^{234}\text{U}$$

All uranium isotopes are radioactive materials. They can emit α-particles whose energies cover the range 4,5÷4,8 MeV and undergo spontaneous fission followed by neutron emission: for example, ^{238}U emits ~13 n/(s·kg).

Uranium isotope ^{235}U is the only natural nuclear material which can be fissioned by neutrons of any energy including thermal neutrons (the lower neutron energy, the better fissionability of ^{235}U) with emission of excessive fast neutrons. Just thanks to these fission neutrons it becomes possible for the chain fission reaction to initiate. Unfortunately, natural uranium contains a rather small fraction of ^{235}U (~0,71%). The overwhelming majority of nuclear power reactors in operation now applies

enriched uranium, i.e. uranium containing 2-5% ^{235}U instead of 0,71% ^{235}U in natural uranium. Some research reactors still use uranium enriched with ^{235}U up to 90% and above. Currently, the IAEA insistently recommends the states-participants to arrange gradual transfer of their research reactors on the use of uranium fuel containing below 20% ^{235}U. Critical mass of 20%-uranium is equal to ~830 kg. Successful theft of so large uranium mass and manufacturing of a primitive but transportable nuclear explosive device is quite unlikely feasible.

Enriched uranium contains relatively larger ^{235}U quantity than ^{235}U abundance in natural uranium. There are the following categories of enriched uranium depending on ^{235}U content (X_5):

1. Low-enriched uranium with X_5 below 5%.
2. Middle-enriched uranium with X_5 from 5% to 20%.
3. Highly-enriched uranium with X_5 from 20% to 90%.
4. Weapon-grade uranium with X_5 above 90%.

Depleted uranium (X_5 < 0,71%) is a by-product of the uranium enriching process. Contemporary technologies of uranium isotope enrichment can produce depleted uranium with ^{235}U content at the level of 0,2-0,3%.

When capturing neutron, main uranium isotope ^{238}U transforms into secondary nuclear fuel, namely fissile plutonium isotope ^{239}Pu, after two consecutive β-decays of intermediate nuclides:

$$^{238}U(n,\gamma)^{239}U(\beta,T_{1/2}=23,5´)^{239}Np(\beta,T_{1/2}=2,3\ d)^{239}Pu.$$

Similarly, fissile uranium isotope ^{233}U can be produced by neutron irradiation of natural thorium. When capturing neutron, the only long-lived thorium isotope ^{232}Th transforms into secondary nuclear fuel, namely fissile uranium isotope ^{233}U, after two consecutive β-decays of intermediate nuclides:

$$^{232}Th(n,\gamma)^{233}Th(\beta,T_{1/2}=23,3´)^{233}Pa(\beta,T_{1/2}=27,4\ d)^{233}U.$$

However, these conversions of natural fertile isotopes (^{238}U, ^{232}Th) into secondary nuclear fuel isotopes (^{239}Pu, ^{233}U) require that primary nuclear fuel, i.e. fissile uranium isotope ^{235}U, must be placed into the reactor core in such a quantity which makes it possible to initiate the

self-sustaining chain fission reaction. The chain fission reaction can generate a large enough amount of fission neutrons to produce secondary nuclear fuel through radiative neutron captures by fertile isotopes. Large fraction of fertile uranium isotope ^{238}U in primary fuel of nuclear power reactors (95-98%) can realize a partial reproduction of nuclear fuel.

Fuel element (fuel rod or fuel pin) is a main constructive form of nuclear fuel in the reactor core. Cylindrical fuel rod consists of a central active part (fuel meat) containing fissile and fertile isotopes and hermetical cladding around. Usually, the claddings are made of metals (Zr-based alloys and stainless steels). In spherical fuel elements of high-temperature gas-cooled reactors (HTGR) micro fuel particles are clad by thin layers of silicon carbide and pyrolytic carbon, and then these fuel particles are uniformly dispersed in graphite matrix.

Fuel rods are united into fuel assemblies (FA) containing from several fuel rods up to several hundred fuel rods. Inside FA fuel rods are stiffly fastened by the spacing grids (spacers). Also, certain conditions must be guaranteed to provide a reliable heat removal by coolant from fuel rods and compensate temperature-induced expansion of structural and fuel materials.

Complete set of all fuel assemblies disposed in a nuclear reactor constitutes the reactor core where the controlled chain fission reaction can be initiated to convert nuclear energy into thermal energy and then into electrical energy. It appears the reactor core plays a similar role with a traditional thermal pile, or boiler, where fossil organic fuel (charcoal, oil or natural gas) is burnt to produce heat. This analogy allows us to use such habitual terms as “fuel”, “incineration” or “burn-up” although no any burning or incinerating processes, in their traditional sense, occur in the nuclear reactor core.

Indeed, there are many substantial distinctions between nuclear and organic fuel. Main distinctions are briefly described below.

1. Significantly higher calorie content in nuclear fuel.

Incineration of one carbon atom in chemical reaction with oxygen can produce thermal energy at the level of 4 eV only:

$$C + O_2 \rightarrow CO_2 + 4\ eV.$$

Fission of one ^{235}U nucleus by neutrons can produce thermal energy at the level of 200 MeV:

$$^{235}U + n \rightarrow FP_1 + FP_2 + (2\text{-}3)n + 2\cdot10^8 \text{ eV}.$$

Taking into account different atomic weights of uranium and carbon isotopes (235:12), calorie content of ^{235}U fission reaction exceeds calorie content of ^{12}C oxidation reaction (per one atomic mass unit) by a factor of $2{,}5\cdot10^6$.

$$E(1\,\text{kg } ^{235}U) = \frac{1000}{235}\times 6\cdot10^{23}\times 2\cdot10^8 \text{ eV};$$

$$E(1\,\text{kg } ^{12}C) = \frac{1000}{12}\times 6\cdot10^{23}\times 4 \text{ eV};$$

$$\frac{E(1\,\text{kg } ^{235}U)}{E(1\,\text{kg } ^{12}C)} = \frac{12}{235}\times\frac{2\cdot10^8}{4} \approx \frac{5\cdot10^7}{20} = 2{,}5\cdot10^6.$$

Such a large ratio reflects the fact that intra-nuclear energy is much higher than energy of chemical (inter-atomic or inter-molecular) reactions. Large calorie content of nuclear fuel can substantially reduce mass and volume of fuel needed to produce the same energy. Thus, expenses for fuel transportation and storage can be considerably decreased. Moreover, nuclear fuel creates a new important factor, namely geographical independency of NPP site placement on placement of uranium mines and nuclear fuel fabrication plants. This property of nuclear fuel allows the humankind to correct unfairness of the nature consisting in extremely non-uniform geographical distribution of organic and nuclear energy resources.

2. Impossibility to reach complete incineration of all fissile nuclides for one irradiation cycle.

During time period of full-power reactor operation the reactor core must contain nuclear fuel in the quantity larger than its critical mass. That is why for one irradiation cycle it is possible to incinerate only so

fraction of nuclear fuel quantity that exceeds its critical mass and provides super-criticality, or reactivity margin needed to make up the negative effects caused by fuel burn-up and build-up of fission products.

Usually, nuclear fuel burn-up is evaluated either by FP quantity per total fuel mass (for example, 10% fuel burn-up means that 10% of fuel mass was burnt-up and converted into FP) or by quantity of produced thermal energy per total fuel mass, MWd/t. It may be shown that 1% of fuel burn-up is approximately equal to specific energy yield of 10 GWd/t.

Typical, currently achievable values of fuel burn-up are as follows:

- Heavy-water CANDU-type reactors - 10÷12 GWd/t, or 1÷1,2% of heavy metals (HM).
- Light-water reactors (LWR) of VVER, PWR and BWR type - 40÷50 GWd/t (or 4-5% HM).
- Fast LMFBR-type reactors – up to 100 GWd/t (or ~10% HM).

Upon exhaustion of the reactivity margin, spent fuel assemblies have to be replaced by fresh ones completely or partially. Spent nuclear fuel (SNF) contains large amounts of fertile and fissile nuclides. One ton of SNF discharged from VVER-440 contains, in addition to 30 kg FP, about 950 kg ^{238}U, 12,5 kg ^{235}U and 6,5 kg of plutonium isotopes (mainly, ^{239}Pu and ^{240}Pu).

3. Possibility to organize repeat usage (recycle) of fertile and fissile nuclides.

The recycle can reduce significantly the demands for natural uranium mining and for its isotope enrichment with ^{235}U.

4. Possibility to organize reproduction of fissile nuclides.

Fissile nuclides can be reproduced in any nuclear reactor which, in addition to fissile nuclides, contains fertile nuclides ^{238}U or ^{232}Th. When capturing neutron, fertile nuclide ^{238}U converts into fissile nuclide ^{239}Pu. Similarly, ^{232}Th converts into fissile nuclide ^{233}U. The reproduction process is conventionally characterized by the breeding ratio (BR), i.e. by ratio of secondary fuel generation rate to primary fuel incineration rate. Depending on the BR value, the following options of nuclear fuel reproduction are marked out: partial reproduction (BR < 1); full reproduction (BR = 1) and extended reproduction (breeding), if BR > 1.

The reproduced secondary fuel can slow down the reactivity slump, prolong the reactor lifetime and generate some additional amount of thermal energy. In stationary operation mode of typical LWR, when the reactor core contains both fresh and partially incinerated FA, total generation rate of thermal energy includes a considerable contribution (up to 40% and above) from fissions of secondary fissile nuclide ^{239}Pu.

Thanks to some neutron-physical peculiarities, the best conditions for extended reproduction (breeding) of nuclear fuel can be formed in fast breeder reactors loaded with mixed uranium and plutonium dioxides, i.e. with mixed oxide (MOX) fuel. Fast breeder reactors are able to produce such plutonium quantity that is sufficient to meet fuel demands of the reactor-producer and create an initial fuel loading for a new reactor-consumer. If large stockpiles of natural uranium are available or if there are no incentives for intense NPP deployment, then fast reactors can operate in fuel self-sustainability regime with the BR value about unity.

Similar situations can be formed for mixed thorium-uranium fuel. Good neutron-multiplying properties of ^{233}U and huge abundance of natural thorium keep a customary interest to nuclear power reactors loaded with thorium-uranium fuel. However, nuclear technologies related with fabrication of fresh and reprocessing of spent (Th-U) fuel assemblies encountered some specific difficulties, and till now these technologies are not developed yet up to an industrial scale.

5. “Incineration” of nuclear fuel does not require oxidizer.

Incineration of conventional organic fuel in traditional thermal power plants requires roughly three-fold mass of oxygen taken from the Earth’s atmosphere. Moreover, the incineration process is followed by release of toxic wastes (smoke, ashes, sulphur and nitrogen oxides).

“Incineration” of nuclear fuel does not require an oxidizer at all. Radioactive fission products and spent nuclear fuel, which may be regarded as nuclear wastes, are retained within fuel rods for a rather long time period and, then, after appropriate treatment, transported into well-protected geological repositories.

Data on fuel consumption and waste production by two electrical power plants (TPP and NPP) of the same power (1000 MWe) are presented in Table I.1.

The well-known Kyoto protocol concerning global warming-up of the Earth's climate proposes to introduce some ecological constraints on release of CO_2 into the atmosphere by coal-fired TPP and establish certain economic sanctions for the exceeding of these constraints at the level of 60 US dollars (about 40 euros) per one ton of carbon dioxide. Following from the scale of global coal-fired power system, it is easy to evaluate total scope of these economical sanctions.

Table I.1

Fuel consumption and waste production by electrical power plants

TPP-1000	VVER-1000
Coal consumption – $2{,}3 \cdot 10^6$ t/year Oxygen consumption – $6{,}2 \cdot 10^6$ t/year	^{235}U consumption – 1,0 t/year
Wastes	
CO_2 – $8{,}5 \cdot 10^6$ t/year Ashes – $2{,}3 \cdot 10^5$ t/year The wastes are released into the atmosphere	Radioactive wastes – 1,0 t/year Spent nuclear fuel – 35÷40 t/year The wastes are retained in spent fuel rods for a long time

6. Accumulation of radioactive FP. Residual heat generation after reactor shutdown. Induced radioactivity of structural materials and coolant.

Fission reactions of heavy nuclides produce fission products, i.e. relatively lighter nuclides whose mass numbers cover the range from ~70 a.m.u. up to ~160 a.m.u. As a rule, fission reaction is an asymmetrical act, i.e. instead of splitting a heavy nucleus into a couple of fission products with approximately equal masses, fission reaction produces two nuclides with mass ratio about 2:3 (95 a.m.u. and 140 a.m.u., for example). Common form of FP yields dependency on their mass is roughly the same for all fissile nuclides and for all neutron energies. (a symmetrical two-peak curve, as is presented in Fig. I.1). FP accumulation rate in nuclear power reactors is about 1000 kg per one GWe·year.

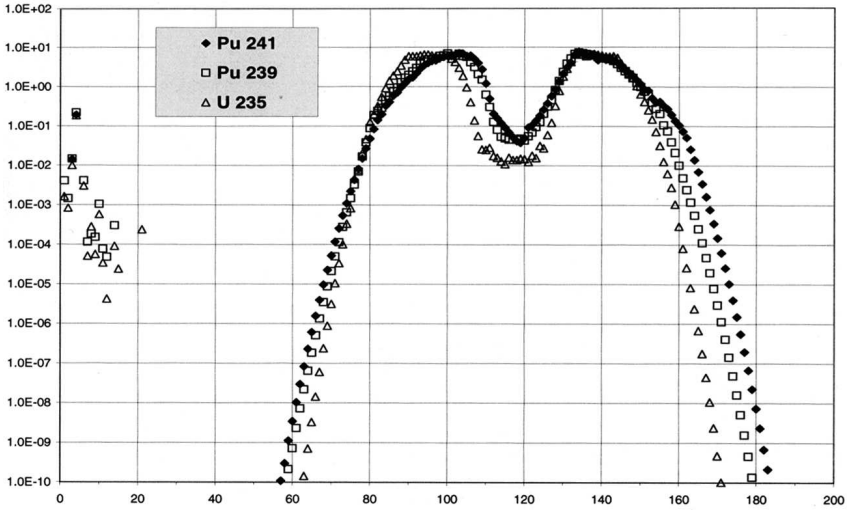


Fig. I.1. Dependency of FP yields on their atomic mass

Fission products consist of about 200 radioactive isotopes belonging to 36 chemical elements including daughter products of their radioactive decays. Half-lives of these radionuclides cover the very broad time range: from several milliseconds up to several million years. Depending on half-lives, the following FP categories can be marked out: short-lived, middle-lived and long-lived nuclides. Main type of FP radioactivity is a β-decay. Each radioactive FP is a starting isotope for decay chain consisting of 4-5 consecutive decays and ending by a stable nuclide.

Besides fission products, spent nuclear fuel contains also transuranium isotopes, intense emitters of α- and β-radiation. A particular attention should be given to minor actinides (MA) consisting of ^{237}Np (neptunium fraction), ^{241}Am and ^{243}Am (americium fraction), ^{244}Cm and ^{245}Cm (curium fraction). Chemical properties of minor actinides are very close to those of rare-earth fission products. Therefore, at the stage of SNF reprocessing and FP extraction, minor actinides and rare-earth FP are removed together. As a consequence, a special RAW category is being formed, namely MA-containing RAW. All minor actinides are fissile or fertile nuclides. That is why minor actinides must put under

strict control in order to prevent proliferation of weapon-suitable nuclear materials.

Main channels for MA generation in nuclear reactors are as follows:
1) $^{235}U(n,\gamma)^{236}U(n,\gamma)^{237}Np$;
2) $^{241}Pu(\beta^-, 14\ лет)^{241}Am(n,\gamma)^{242m}Am(n,\gamma)^{243}Am(n,\gamma)^{244}Cm(n,\gamma)^{245}Cm$;
3) $^{242}Pu(n,\gamma)^{243}Am(n,\gamma)^{244}Cm(n,\gamma)^{245}Cm$.

MA generation rates are presented in Table I.2 for LWR loaded with traditional uranium oxide (UOX) fuel and with advanced mixed uranium-plutonium (MOX) fuel.

Table I.2

Generation rate of minor actinides in LWR

Nuclide	$T_{1/2}$, years	MA generation rate, kg/GWe·year	
		UOX	MOX
^{237}Np	$2{,}1\cdot10^6$	20,4	15,1
^{241}Am	432	1,3	6,0
^{243}Am	7380	2,5	21,8
^{244}Cm	18,1	0,9	15,6
^{245}Cm	8500	0,1	1,7
Total	-	25,2	60,2

Significant fraction of SNF β- and γ-activity is caused by short-lived FP. Therefore, SNF radioactivity rapidly decreases with time after SNF withdrawal from the reactor core. Residual heat generated by spent FA in the cooling pool is mainly caused by FP and MA radioactive decays. Time dependency of residual heat generation rate is quite similar to the aforementioned time dependency of SNF radioactivity - rapid exponential slump just after withdrawal followed by gradual approach to a plateau level constituting several percents of nominal reactor power.

The induced radioactivity of steel in-vessel structures is mainly caused by the following radionuclides: ^{63}Ni ($T_{1/2}$ = 100 years), ^{60}Co ($T_{1/2}$ = 5,3 years) и ^{55}Fe ($T_{1/2}$ = 2,7 years). These radionuclides are produced by neutron irradiation of stable chemical elements, components of stainless steels. Total radioactivity of steel LWR structures is equal to

~50 MCi (BWR) and ~5 MCi (PWR) at the reactor shutdown. Afterwards, the total radioactivity rapidly decreases and gradually (in the process of 20-30–year staying in the cooling pool) approaches to the plateau (1 MCi for BWR-type reactors and 0,1 MCi for PWR-type reactors), approximately 2% of initial radioactivity.

The induced radioactivity of metal NPP structures becomes more and more urgent problem as NPP lifetime expires.

Nuclear fuel, being involved into the processes of its fabrication, usage and reprocessing, passes a series of consecutive stages which can be united into a general concept of nuclear fuel cycle (NFC).

Main NFC stages

1. Mining of uranium ores and uranium extraction.
2. Nuclear fuel fabrication:
   - 2a. Production of uranium concentrate in the form of uranium octaoxide U_3O_8.
   - 2b. Conversion of uranium concentrate into uranium hexafluoride UF_6.
   - 2c. Uranium enrichment with ^{235}U.
   - 2d. Manufacturing of fuel rods and fuel assemblies.
3. The use of nuclear fuel in nuclear reactors of various types (plutonium-producing, power or research reactors).
4. Interim storage of spent fuel assemblies (SFA) in the cooling pools at NPP.

The following two options may be chosen for the next NFC stages, namely once-through, or open NFC and closed NFC.

If the open NFC option was chosen, then:

5. Transportation and ultimate disposal of SFA in deep geological formations. This stage is a final step of the open NFC.

If the closed NFC option was chosen, then:

6. Transportation of SFA to a spent fuel reprocessing plant.
7. Extraction of radioactive wastes, their treatment and ultimate disposal in deep geological formations.
8. Extraction of primary and secondary nuclear fuel for multiple uses (recycles) in re-fabricated fresh fuel rods and fuel assemblies. In reality, this is a return to point 2.

The following three NFC variants can be marked out:

A. The open NFC

Main stages of the open NFC:

1. Mining of uranium ore.
2. Production of uranium concentrate U_3O_8.
3. Conversion of uranium concentrate U_3O_8 into uranium hexafluoride UF_6.
4. Isotope uranium enrichment.
5. Fabrication of nuclear fuel in form of fuel rods and fuel assemblies.
6. Use of nuclear fuel in nuclear reactors.
7. Interim SNF storing in the cooling pools at NPP.
8. Ultimate disposal of SNF in deep geological repositories.

B. The closed NFC with uranium recycle

Main stages of the closed NFC:

1. Mining of uranium ore.
2. Production of uranium concentrate U_3O_8.
3. Conversion of uranium concentrate U_3O_8 into uranium hexafluoride UF_6.
4. Isotope uranium enrichment.
5. Fabrication of nuclear fuel in form of fuel rods and fuel assemblies.
6. Use of nuclear fuel in nuclear reactors.
7. Interim SNF storing in the cooling pools at NPP.
8. SNF reprocessing: separation of uranium, plutonium and radioactive wastes.
9. Recycle of extracted uranium to the stage 4, i.e. to the isotope uranium re-enrichment.
10. Plutonium storing in the dedicated warehouses.
11. Ultimate disposal of RAW in deep geological repositories.

C. The closed NFC with uranium and plutonium recycle

Main stages of the closed NFC:

1. Mining of uranium ore.
2. Production of uranium concentrate U_3O_8.
3. Conversion of uranium concentrate U_3O_8 into uranium hexafluoride UF_6.

4. Isotope uranium enrichment.
5. Fabrication of nuclear fuel in form of fuel rods and fuel assemblies.
6. Use of nuclear fuel in nuclear reactors.
7. Interim SNF storing in the cooling pools at NPP.
8. SNF reprocessing: separation of uranium, plutonium and RAW.
9. Recycle of extracted uranium and plutonium to the stage 5, i.e. to the fabrication of mixed oxide fuel.
10. Ultimate disposal of RAW in deep geological repositories.

These variants of NFC schemes are sown in Fig. I.2.

Presently, only seven states are able to reprocess spent nuclear fuel: the USA, Great Britain, France, Russian, China (nuclear powers), India and Japan. But the US administrations decided to stop reprocessing of spent fuel assemblies discharged from commercial NPP till effective and proliferation-proof SNF reprocessing technology is developed.

Currently in the world there are the following two opposite and controversial viewpoints on reasonability of the NFC closure:

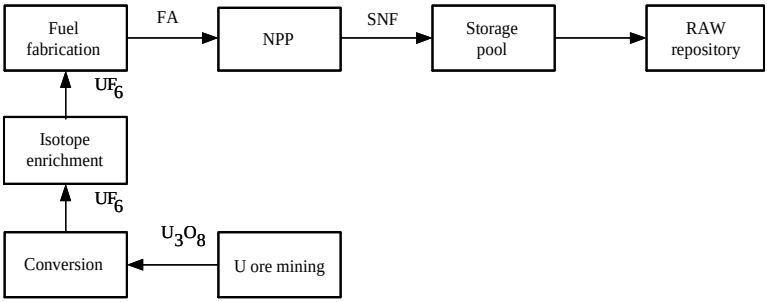
1. The NFC closure is an unreasonable action because it assumes radiochemical SNF reprocessing, extraction, transportation and application of primary fuel (mainly, regenerated uranium) and secondary fuel (mainly, plutonium) for re-fabrication of fresh nuclear fuel. Thus, the NFC closure creates a series of complicated technological and political problems, including:

a. Possibility for terrorist groups to steal fissile materials for manufacturing of nuclear explosive devices.

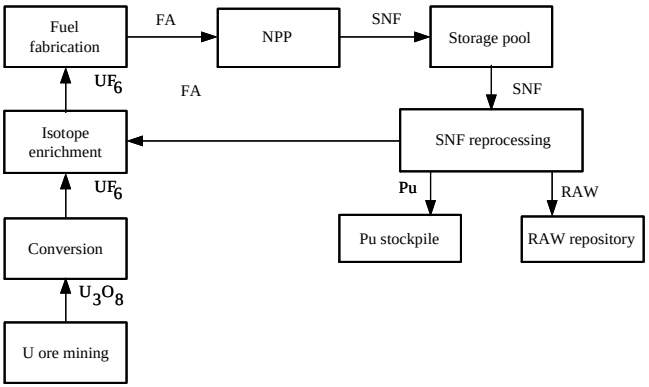
b. Complicacy and jeopardy of SNF reprocessing technologies.

c. Complicacy and jeopardy of RAW treatment and ultimate disposal in geological repositories.

This viewpoint is held by the US Government. The US Presidents Ford and Carter, in the late 1970s, prohibited radiochemical reprocessing of SFA discharged from commercial nuclear power reactors. However, scientific investigations of the problems related with SNF reprocessing and recycle were continued but within a reduced scope. Spent fuel assemblies are considered as a RAW form suitable for ultimate disposal in deep geological repositories. The RAW repositories must be equipped with some technical tools capable to retrieve SFA containers for further reprocessing, if target priorities in the US nuclear policy would be changed.



A)



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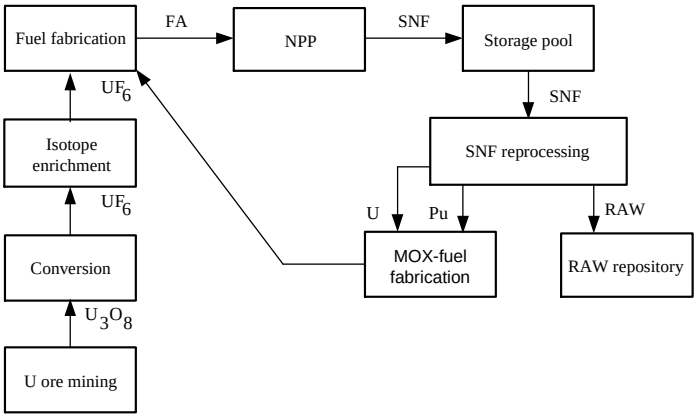


C)

Fig. I.2. Layouts of the open NFC and two options of the closed NFC

2. The opposite viewpoint does not regard SNF as wastes suitable only for ultimate disposal. The viewpoint regards SNF as a valuable nuclear material that contains both primary and secondary fuel which can be extracted and multiply used for energy production. The NFC closure is considered as a main strategic pathway towards national energy independency.

Technological difficulties of SNF reprocessing, RAW treatment and ultimate disposal are estimated as very complicated and radiation-dangerous but all the difficulties can be successfully overcome by currently available methods and technical tools.

Potential jeopardy of NM thefts and unauthorized use in the closed NFC is recognized too but the NM non-proliferation problems are considered as completely resolvable by means of already available domestic and international safeguard systems.

This viewpoint is supported by the Governments of France, Japan and Russia. The brightest example is a position of Japan. Practically, Japan has no available its own resources of fossil organic and nuclear fuel. So, Japan is not able to form a self-dependent power system based on coal, gas or oil incineration. Moreover, Japan has a series of substantial reasons to reject nuclear power option at all. Firstly, Japan is the only country in the world that was subject to the well-known nuclear bombardment in 1945. Secondly, Japan is a densely populated country placed on relatively small territory with intense seismic activity. Chernobyl-like nuclear accident is able to envelop all the country. Nevertheless, development of nuclear energy system based on fast breeder reactors with extended reproduction of secondary fuel in the closed NFC opens an opportunity for Japan to reach energy independency with very limited import of natural uranium.

The USA never encountered a problem of national energy independency. There are large deposits of fossil organic fuel and natural uranium in the USA. The US nuclear power system includes 104 units with total electrical power about 100 GWe; NPP share in total energy production is equal to 20%). In 2011 global nuclear power system consisted of ~450 units with total capacity of 375 GWe, i.e. above one-fourth fraction of global nuclear power is produced by American NPP. No new NPP were built in the USA for the last 30 years. Besides, typical American NPP is a privately owned commercial enterprise. Thus, from

the USA standpoint, there are no any economical, political and non-proliferation incentives to arrange the closed NFC.
In 2011 total power of nuclear energy system in Japan was equal to 45 GWe (30% of total energy production), several new nuclear power units are under construction (15 GWe, in total). Japan is vitally interested in the NFC closure to reach self-sustainability of energy resources.

Control questions to Introduction

1. What is nuclear fuel? Call main components of nuclear fuel.
2. What are the primary nuclear fuel and the secondary nuclear fuel?
3. Call main distinctions between nuclear fuel and fossil organic fuel.
4. Call main stages of the open and the closed NFC.
5. What are main difficulties for the NFC closure?

CHAPTER 1. MAIN STAGES OF THE CLOSED NUCLEAR FUEL CYCLE

1.1. Mining and primary treatment of uranium ore

For the beginning, the following information can be presented about discoveries of natural nuclear materials – uranium and thorium.

German chemist M. Klaproth is considered as a scientist who performed uranium discovery in 1789. Klaproth precipitated a yellow compound by dissolving pitchblende extracted from silver mines in Jachymov (Czech Republic now) in nitric acid. Klaproth erroneously assumed the yellow substance was the oxide of a new yet undiscovered chemical element. He named the newly discovered element after the planet Uranus. In 1841 the French chemist E. Peligot isolated the first sample of metal uranium.

Thorium was discovered in 1828 by the Norwegian mineralogist M. Esmark, identified by the Swedish chemist J. Berzelius and named after Thor, the Norse god of thunder. Despite such a terrible name, thorium was never used in any military purposes. In pure thorium ores thorium consists of isotope ^{232}Th only. Thorium decays with emission of α-particles, its half-life ($T_{1/2}$ = 1,4·10^{10} years) is about three-fold longer than half-life of main uranium isotope ^{238}U ($T_{1/2}$ = 4,5·10^{9} years). Geological evaluations showed that natural thorium resources exceeded natural uranium resources up to the same degree (three-fold exceeding). Average uranium abundance in the Earth's crust is estimated as 2-4 ppm while thorium abundance is three times larger, i.e. 12-15 ppm.

Because of their strong chemical activity, uranium and thorium are not found in the nature as pure metals but only in form of complex chemical compounds. In total, nearly 200 uranium and thorium-containing minerals are known today.

Because of strong chemical activity of uranium, because of high solubility of uranium compounds in water that leads to active uranium transport in the Earth's crust, there are relatively few regions in the world with rich deposits of uranium ores. According to some geological evaluations, sea and ocean water contains about 4·10^{9} t natural uranium (~3,3 mg/m^3, or 0,003 ppm, as an average content). For comparison:

total natural uranium resources in the Earth's crust are evaluated in 10^{14} tons (2-4 ppm, as an average content).

The following categories of uranium ores can be marked out depending on uranium content:

1. Very rich ores contain above 1% U.
2. Rich ores contain 0,5-1% U.
3. Medium ores contain 0,25-0,5% U.
4. Ordinary ores contain 0,09-0,25% U.
5. Poor ores contain below 0,09% U.

In average, the mined ores contain about 0,1% U, i.e. these are ordinary and poor uranium ores.

Natural uranium resources are evaluated on the following two cost categories:

1. Cheap uranium costs below 80 US dollars per 1 kg U_3O_8.
2. Expensive uranium costs above 80 US dollars per 1 kg U_3O_8.

The threshold cost (80 US dollars/kg U_3O_8) differentiates the competitiveness areas of NPP and coal-fired TPP. If natural uranium costs below 80 US dollars/kg U_3O_8, then NPP produces the cheaper electrical energy than TPP does, and vice versa.

The following four categories of natural uranium resources can be marked out depending on the completeness of geological information:

1. Reasonably assured uranium resources (RAR).
2. Inferred uranium resources (IR), i.e. uranium deposits at peripheral wings of reasonably assured resources.
3. Prognosticated uranium resources refer to those expected to exist in well-known uranium provinces.
4. Speculative uranium resources refer to those expected to exist in geological provinces that may host uranium deposits.

The first and second categories are the most trustworthy ones. Information on global uranium resources (as of January 1, 2009) and uranium production rate in 2006-2008 is presented in Table 1.1 and Table 1.2. As is seen, the reasonably assured uranium resources are evaluated as $3{,}52 \cdot 10^6$ t, the inferred uranium resources - $1{,}88 \cdot 10^6$ t, i.e. about $5{,}4 \cdot 10^6$ t in total, including $3{,}7 \cdot 10^6$ t of cheap uranium and $1{,}7 \cdot 10^6$ t of expensive uranium. As of January 1, 2009, the world nuclear power (373 GWe) required 59 thousand tons of natural uranium a year. Under such a consumption rate, the cheap uranium resources will be sufficient

for 63 years, the expensive uranium resources can prolong this time interval on 28 years, i.e. total cheap and expensive uranium resources will be able to meet the demands of the world nuclear power for natural uranium during 91 years.

Table 1.1

Uranium resources, thousand tons (2009)

No.	Country	RAR		IR	
		< 80 $/kg	< 130 $/kg	< 80 $/kg	< 130 $/kg
1	Australia	1163	1176	449	497
2	Canada	337	361	111	124
3	Kazakhstan	234	336	242	316
4	Brazil	158	158	74	121
5	SAR	142	195	91	100
6	China	101	116	49	56
7	Russia	100	181	58	299
Σ(1-7)		2235 of 2516 (89%)	2523 of 3525 (72%)	1074 of 1226 (88%)	1513 of 1879 (81%)

Table 1.2

Uranium production rate, thousand tons

No.	Country	2006	2007	2008
1	Canada	9,86	9,48	9,00
2	Australia	7,59	8,60	8,43
3	Kazakhstan	5,28	6,63	8,51
4	Niger	3,44	3,19	3,03
5	Russia	3,19	3,41	3,52
6	Namibia	3,08	2,83	4,40
7	USA	1,80	1,75	1,49
Σ(1-7)		34,24 of 39,62 (86%)	35,89 of 41,24 (87%)	38,38 of 43,88 (87%)

Nearly 85% of the reasonably assured and inferred uranium resources are placed in seven countries: in America (Canada, Brazil), Africa (SAR), Eurasia (Kazakhstan, China, Russia) and in Australia. The same situation takes place with uranium production. Annual rate of natural uranium production (40-44 thousand tons) does not meet the

demands of the world nuclear power (59 thousand tons). The deficit of natural uranium is covered by the previously mined uranium ores.

Some data on total capacities of national nuclear energy systems in 2011 and on NPP shares in gross electricity generation are presented in Table 1.3.

Table 1.3

The world nuclear power in 2011

No.	Country	Total nuclear power, GWe	NPP share, %
1	USA	100,4	19
2	France	63,1	78
3	Japan	44,7	30
4	Russia	23,7	18
5	South Korea	19,7	35
6	Ukraine	13,1	48
7	Canada	12,7	15
8	Germany	12.1	18
9	China	11,8	2
10	Great Britain	10,1	15
	Total	312,0 of 373 (84%)	16

As is seen, the countries possessing main deposits of uranium ores are the main producers of natural uranium too but they are not obligatory the possessors of the well-developed nuclear power system. For example, all African countries and Australia have no NPP in operation at all. Quite the contrary, Asian countries Japan and South Korea have no any available resources of natural uranium, both these countries are the main uranium importers in the world, but there are the well-developed nuclear power systems in these states.

Of 312 GWe produced by the countries with the well-developed nuclear power industry, about 113 GWe are generated by North American countries, 123 GWe – by European countries and 76 GWe – by Asian countries. The remaining 61 GWe are generated by the countries, non-members of the top-ten list.

The following four methods are mainly used for recovery of natural uranium:

1. Underground extraction from uranium mines.
2. Uranium extraction from open-cast mines.
3. Underground leaching, or in-situ leaching of uranium deposits.
4. Uranium extraction from seawater.

When uranium-containing minerals are already recovered from the Earth's crust with application of the first two methods, uranium ore undergoes the hydro-metallurgical (HM) treatment. The hydro-metallurgical technologies are based on good solubility of the uranium-bearing minerals by acidic and alkaline solutions.

Natural uranium can be recovered from uranium ores by means of the following consecutive procedures:

1. Crashing and physical concentration of uranium ore by removal of the barren (dead) rocks.
2. Leaching (dissolution) of uranium ore in acidic or carbonate solutions.
3. Selective separation of uranium from the solutions or pulps by technologies of sorption, extraction and chemical precipitation.
4. Production of dry uranium concentrate (~95% U_3O_8).
5. Production of pure (refined) uranium compounds with application of the affinage technologies.

The following methods can be used to concentrate uranium ore by separation of the uranium-bearing rocks from the barren rocks:

1. Radiometrical separation. The radiometrical method is based on the higher radioactivity of the uranium-bearing rocks. Uranium ore is milled into pieces with typical sizes about 20-30 cm. The ore pieces are examined by monitoring the natural gamma-radioactivity of each ore piece and removing the barren pieces. This method is able to remove up to 50% of the barren rocks.

2. Gravitational separation. The gravitational method is based on different densities of the uranium-bearing minerals (6, 5-10,5 g/cm^3) and the barren rocks (2,5-2,7 g/cm^3). Uranium ore is milled into pieces with typical sizes about 1 mm, and the ore pieces are put into a water-filled vessel. The heavier pieces sink onto bottom and can be next collected. The gravitational method is often combined with a floatation separation.

3. Floatation separation. The floatation method is based on different densities and different abilities to be moistened by water of the uranium-bearing minerals and the barren rocks. Uranium ore is milled into pieces with typical sizes about 0,1 mm, and the ore pieces are put into a water-filled vessel. Air flow is pumped from the bottom. The lighter pieces of the barren rocks are sticking to the air bubbles and going to the water surface while the heavier pieces of the uranium-bearing minerals gradually sink onto the vessel bottom where they can be then collected. The separation process can be quickened by introducing some floatation reagents into the water-filled vessel to change purposefully natural ability of the uranium-bearing minerals to be moistened by water.

The next step in the HM-treatment is a leaching (removal by dissolving) of uranium compounds from uranium ore. Depending on chemical composition of uranium ore, one of two leaching materials can be used, namely acidic or carbonate solutions.

The acid leaching is a more widely used technology. Sulphuric acid H_2SO_4, nitric acid HNO_3 and hydrochloric acid HCl may be used as a leaching reagent. The carbonate leaching is applied at large content of impurities which can actively interact with acidic solutions. Soda $NaHCO_3$, sodium bi-carbonate Na_2CO_3 and ammonium carbonate $(NH_4)_2CO_3$ may be also used as a leaching reagent.

After uranium compounds were leached from uranium ore, these uranium compounds can be selectively derived from liquid acidic or carbonate solutions by using the following three methods:

1. Sorption on organic ion-exchange resins.
2. Extraction by organic liquid (extractant).
3. Chemical precipitation from solutions.

Uranium can be sorbed from pulps and clarified solutions. Extraction and chemical precipitation of uranium compounds can be performed from the well-clarified solutions only. Uranium-bearing solutions can be clarified by:

1. Settling of solid particles in large vessels.
2. Filtration of the solutions obtained after removal of solid particles by settling through thick layers of sand, silica gel and activated charcoal.

The sorption method is based on the selective ability of some organic ion-exchange resins to sorb primarily uranium compounds on

their surface. Small spherical granules of an ion-exchange resin are mixed with uranium-bearing solution, and the granules sorb selectively uranium compounds. Since ion-exchange resins are lighter than liquid solutions, the granules can be easily collected and removed for further de-sorption of uranium compounds from their surface. The uranium washing off the granules is named as a de-sorption, or elution process with an eluate as a final product. Neutral or alkaline soda solutions are widely used as eluents.

Other method is also used to derive uranium compounds from acidic or carbonate aqueous solutions. This is a method of uranium extraction by organic substances. From viewpoint of a general chemistry, the extraction process is based on a solvation reaction that can unite molecules of quite different materials into a single stable compound (solvate). The simplest example of the solvation reaction is a hydration of salts that leads to formation of stable hydrates with the following chemical formula "salt·nH_2O". The uranium extraction process is based on the property of some organic dissolvents (extractants), immiscible with water, to form complex chemical compounds with uranium salts. The extraction process must be followed by the re-extraction process, i.e. dissolution of uranium-bearing solvates by excess quantity of a neutral dissolvent and, thus, formation of a highly concentrated uranium solution.

When the clarified acidic or carbonate solution contacts with organic extractant, uranium is distributed between aqueous and organic phases. The most uranium quantity goes into organic phase. Then, these phases are separated, and uranium re-extracted from the organic phase. Light water or low-concentrated nitric acid HNO_3 can be used as re-extractants. Several consecutive applications of the extraction-re-extraction process can derive up to 99,7% U contained in the mined natural uranium ore.

One else method of uranium derivation from acidic or carbonate solutions is a chemical precipitation. The precipitation process can deal with the clarified acidic or carbonate solutions produced by leaching of uranium ore and with large volumes of low-concentrated uranium-bearing solutions produced by the de-sorption or re-extraction processes.

Uranium compounds can be precipitated by introducing some appropriate reagents (precipitants) into the uranium-bearing solutions. The following substances can be used as precipitants: hydrogen peroxide H_2O_2, ammonium hydrate NH_4OH, caustic soda NaOH, magnesium oxide MgO, etc. The precipitation process produces insoluble hydrates of uranium oxides $UO_X \cdot nH_2O$ which fall as a sediment onto a bottom. Then, the uranium-bearing precipitates can be picked up and dried.

The precipitated, picked up and dried uranium concentrate is a final material produced by HM-treatment of the mined uranium ore (solid form).

Thus, HM-treatment of solid uranium ores includes the following main steps:

1. Transportation of the mined uranium ore to HM-plants.
2. Crashing of the uranium ore and physical concentration of uranium compounds.
3. Leaching of uranium compounds from the uranium ore.
4. Application of sorption, extraction and chemical precipitation processes.
5. Application of de-sorption, re-extraction and chemical precipitation processes.

However, the in-situ leaching (ISL) process makes it possible to work without transportation, crashing, physical concentration and leaching of the uranium ore. The ISL method consists of the following steps:

1. Drilling of the injection and output wells into the uranium ore body.
2. Injection of liquid dissolvents into the uranium ore body for leaching of uranium compounds.
3. Pumping out of the produced solutions through the output wells after a certain time interval.

Then, these solutions undergo the aforementioned procedures of HM-treatment (sorption-de-sorption, extraction-re-extraction and chemical precipitation).

As a rule, the ISL wells are drilled not deeper than 100 m. Main disposition scheme of the ISL wells represents a square five-well cluster with four injection wells in four apices of the square and one output well in the square center. Pitch of the cluster is not longer than 30 m. Acidic (at low content of carbonates in the uranium ore body) or alkaline (at high content of carbonates in the uranium ore body) are used as

uranium dissolvents. Uranium concentrations in the output solutions can reach ~200 mg/l, i.e. about 0,02%, or 200 ppm.

The ISL process can be applied only if the uranium ore body is characterized by the following properties:

1. The uranium ore body is placed between two water-tight strata so that the uranium-bearing solutions could not leak from the deposit region.
2. The uranium ore body is porous enough for the leaching dissolvent to penetrate easily and deeply into the ore body.

Seawater can be also regarded as a very low concentrated uranium-bearing solution. In a global scale, seawater of the world seas and oceans contains about $4 \cdot 10^9$ t U but with as low concentration as ~0,003 mg/l, or 0,003 ppm. So, total uranium resources in seawater are larger by three orders of magnitude than total reasonably assured and inferred uranium resources in the Earth's crust (~$5{,}4 \cdot 10^6$ t U). Unfortunately, derivation of natural uranium from seawater encountered the following difficulties:

1. Large volumes of seawater to be pumped through a uranium extraction installation.
2. Flow of fresh seawater can not be mixed with spent water effluents produced by a uranium extraction installation.
3. Large volumes of chemical reagents and wastes.

Thus, HM-treatment of uranium ore can produce dry uranium concentrate as a mixture of uranium oxides (mostly, U_3O_8). Uranium concentrate consists of practically all uranium quantity that previously contained in uranium ore. But uranium concentrate derived from the ore contains also some accompanying impurities. Really, uranium concentrates contains about 94-95% of uranium oxides and 4-5% of undesirable impurities. That is why it is necessary to clean uranium concentrate by removing all impurities, and a particular attention should be given to elements which could play a role of parasitic neutron absorbers in nuclear reactors. Some isotopes of boron, cadmium, hafnium, of rare-earth elements (europium, gadolinium and samarium) are strong neutron absorbers and so they must be removed from uranium concentrate.

Therefore, the next NFC stage is a fine purification of uranium concentrate from undesirable impurities (especially, from neutron-absorbing elements) with application of affinage processes. The most

developed and mastered affinage technology is based on the aqueous extraction process with application of tri-butyl-phosphate (TBP) as an extractant. TBP is a complex organic ether $(C_4H_9)_3PO_4$ with the following scheme of chemical links:

$$\begin{array}{l} C_4H_9 - O \diagdown \\ C_4H_9 - O - P = O \\ C_4H_9 - O \diagup \end{array}$$

TBP density (0,973 g/cm^3) is close and slightly lower than density of light water. TBP is a very viscous liquid and, to reduce its viscosity, TBP is usually diluted with neutral organic liquids (kerosene, for instance). From the standpoint of nuclear technologies, the most significant TBP property consists in its excellent ability to extract selectively uranium compounds from any uranium-bearing solutions. TBP can extract uranyl-nitrate $UO_2(NO_3)_2$ from its aqueous solutions by four orders of magnitude more effectively than impurities. One TBP liter can retain up to 440 g U.

The aqueous extraction affinage process includes the following procedures:

1. Dissolution of uranium concentrate by nitric acid with production of uranyl-nitrate:

$$U_3O_8 + 8\ HNO_3 = 3\ UO_2(NO_3)_2 + 2\ NO_2 + 4\ H_2O.$$

2. Mixing of the uranyl-nitrate solution with TBP. Main fraction of uranyl-nitrate goes into organic phase.

$$UO_2(NO_3)_2 + 2\ TBP \rightarrow UO_2(NO_3)_2 \cdot 2\ TBP.$$

3. Separation of aqueous and organic phases.
4. Derivation of pure uranyl-nitrate from organic phase by the chemical precipitation process. Here the following two ways can be applied:

a. Hydrogen peroxide H_2O_2 is used as a precipitant. Hydrate of uranium peroxide $UO_4 \cdot 2H_2O$ falls into deposit.

b. Ammonium bicarbonate NH_4HCO_3 is used as a precipitant. Ammonium-uranyl-carbonate $(NH_4)_4UO_2(CO_3)_3$ falls into deposit.

Calcination of both these products (hydrate of uranium peroxide and ammonium-uranyl-carbonate) can produce, depending on the calcination temperature range, the following impurity-free uranium oxides: UO_3 at 240-350^{0}C, U_3O_8 at 580-620^{0}C and UO_2 at 750-800^{0}C.

1.2. Conversion and isotopic enrichment of natural uranium

All contemporary nuclear power reactors are fueled with enriched uranium compounds, i.e. uranium containing the larger ^{235}U fraction than that in natural uranium (~0,71%). Thermal light-water reactors (LWR) constitute a basis for the global nuclear power industry, and they are fueled with uranium dioxide enriched up to 2-5% ^{235}U.

A whole series of nuclear technologies have been developed for uranium isotope enrichment. All the technologies are based on the mass difference of main uranium nuclides ^{235}U and ^{238}U (3 a.m.u.). This difference of nuclear masses causes different deviations of ionized atoms traveling in a magnetic field (electromagnetic technology), different probabilities for light and heavy atoms to penetrate through a porous wall (gas diffusion technology), different spatial distributions of light and heavy atoms in a centrifugal field (gas centrifuge technology and separation nozzle process).

Advanced and very promising laser technologies are under intense development now. There are two varieties of laser technology for uranium isotope enrichment, namely atomic and molecular options.

Atomic vapor laser isotope separation (AVLIS) technology is based on selective excitation of ^{235}U atoms under laser irradiation. Then, the excited ^{235}U atoms can be ionized by additional laser radiation, and ^{235}U ions can be easily separated from electrically neutral ^{238}U atoms.

Molecular laser isotope separation (MLIS) technology is based on selective excitation of gaseous $^{235}UF_6$ molecules under laser irradiation. Then, the excited $^{235}UF_6$ molecules can be chemically dissociated by additional laser radiation with production of solid $^{235}UF_5$ powder.

Some chemical methodologies of isotope separation are based on different isotope stabilities in different chemical compounds of the same chemical element. The chemical methodologies use reactions of isotope exchange between two different compounds of one chemical element. Individual isotopes are accumulated in that chemical element

where they are more stable. For example, natural boron (20% ^{10}B and 80% ^{11}B) can be enriched with its light isotope ^{10}B, strong absorber of thermal neutrons, to fabricate highly efficient control rods needed to provide safe LWR operation. When mixing BF_3 with $BF_3O(CH_3)_2$, the following isotope exchange reaction can occur with gradual accumulation of ^{10}B in organic phase:

$$BF_3 + BF_3O(CH_3)_2 \rightarrow {}^{11}BF_3 + {}^{10}BF_3O(CH_3)_2$$

A similar effect can be achieved in the isotope exchange reactions of different uranium compounds, desirably, with different uranium valences. As is known, ^{235}U is more chemically stable in six-valence uranium compounds while, on the contrary, ^{238}U – in four-valence compounds. Such a fine distinction in chemical properties of uranium isotopes can be used for their separation.

Plasma isotope separation technology is based on the effect of ion cyclotron resonance. Any charged particles (ions, for example), when coming into a constant magnetic field, begin rotating around the force lines of the magnetic field with a certain frequency (ion cyclotron frequency –ICF) and with a certain orbital radius. The ICF value depends on the ion mass, so ^{235}U and ^{238}U ions are characterized by their own ICF values. Orbital radii of ^{235}U and ^{238}U ions depend on their energy. If the alternating electrical field with the frequency equaled to the ICF value of ^{235}U, for instance, is applied, then energy of the electrical field is selectively absorbed by ^{235}U ions only. As a consequence, energy of ^{235}U ions increases, and orbit of their rotation extends. Thus, an opportunity arises to separate spatially ^{235}U ions from ^{238}U ions.

Quality of isotope separation (enriching) technologies can be evaluated by the following two parameters: efficiency and energy consumption. Efficiency of the enriching technology is defined by its ability to upgrade relative content (abundance) of necessary isotope after one step of the enriching process. Energy consumption of the enriching technology is defined in the terms of energy expenses per a separative work unit (SWU). The concept of separative works and their measuring units is described below.

Schematically, the process of uranium enrichment can be characterized by such a way. Initial uranium mass F (feed) and relative ^{235}U con-

tent X_F are main input parameters of the process. Main output parameters of the process are mass of enriched uranium P (product), relative ^{235}U content in the product X_P, mass of depleted uranium W (waste or tails) and relative ^{235}U content in the waste X_W.

F (feed), $X_F(^{235}U)$
↓
System of separative steps
↓
Depleted uranium - W (waste), $X_W(^{235}U)$
+
Enriched uranium - P (product), $X_P(^{235}U)$

Mathematical definition of material balance in the uranium enrichment process can be written as a system of the following two equations:

1. Balance of uranium mass: $F = P + W$.
2. Balance of ^{235}U mass: $X_F \cdot F = X_P \cdot P + X_W \cdot W$.

This is a system of two equations with three unknown variables (F, P and W). Fortunately, by dividing both equations by P, the system can be transformed into the resolvable system of two equations with two unknown variables F/P and W/P:

$$\frac{F}{P} = 1 + \frac{W}{P};$$

$$X_F \cdot \frac{F}{P} = X_P + X_W \cdot \frac{W}{P}.$$

By solving the system, the following characteristics of the isotope separation process can be determined:

a. Factor of natural uranium consumption per the product mass unit:

$$\frac{F}{P} = \frac{X_P - X_W}{X_F - X_W};$$

b. Factor of the waste production per the product mass unit:

$$\frac{W}{P} = \frac{X_P - X_F}{X_F - X_W};$$

c. Division factor of the feed flow θ:

$$F = P + W = \theta \cdot F + (1 - \theta) \cdot F;$$

$$\Theta = \frac{P}{F} = \frac{X_F - X_W}{X_P - X_W}.$$

Some numerical examples:

a. Production of weapon-grade uranium from natural uranium:

$$X_F = 0{,}71\%;\ X_P = 90\%;\ X_W = 0{,}25\%.$$

Then

$$\frac{F}{P} = \frac{X_P - X_W}{X_F - X_W} = \frac{89{,}75}{0{,}46} \approx 195.$$

This means that production of 25 kg (one Significant Quantity for weapon-grade uranium) requires about 5000 kg of natural uranium contained, in average, in about 5000 t of natural uranium ore.

b. Production of reactor-grade uranium from natural uranium:

$$X_F = 0{,}71\%;\ X_P = 4\%;\ X_W = 0{,}25\%.$$

Then

$$\theta = \frac{P}{F} = \frac{X_F - X_W}{X_P - X_W} = \frac{0{,}46}{3{,}75} \approx 0{,}12.$$

This means that about 120 kg of enriched reactor-grade uranium (4% ^{235}U) and 880 kg of depleted uranium (0,25% ^{235}U) can be obtained from 1000 kg of natural uranium.

The following parameters can be introduced as they can be helpful for characterization of the uranium enrichment process:

1. Relative concentrations of ^{235}U in the feed, product and waste:

$$R = \frac{X_F}{1 - X_F}; \quad R' = \frac{X_P}{1 - X_P}; \quad R'' = \frac{X_W}{1 - X_W}.$$

2. The single-stage separation factor:

$$\alpha = \frac{R'}{R} = \frac{X_P / (1 - X_P)}{X_F / (1 - X_F)}.$$

3. The single-stage depletion factor:

$$\beta = \frac{R}{R''} = \frac{X_F / (1 - X_F)}{X_W / (1 - X_W)}.$$

4. The single-stage enrichment gain: $\varepsilon' = \alpha - 1$.

5. The single-stage depletion gain: $\varepsilon'' = \beta - 1$.

1.2.1. Separation work

The methodology for quantitative evaluation of the efforts expended to separate ^{235}U and ^{238}U from each other has been developed by English physicists R. Peierls and P. Dirac. They proposed to use a certain function U that can characterize a total value of any uranium isotope composition. For example, total value of the feed material is defined by multiplying the feed mass F by a certain dimensionless function $V(X_F)$ that depends only on a specific concentration of the desired isotope ^{235}U, i.e.

$$U_F = F \cdot V(X_F).$$

The $V(X)$ function is called the separation potential function. Before the uranium enrichment process started, total value of the feed material $U_F = F \cdot V(X_F)$. After the uranium enrichment process ended, total value of the obtained materials is a sum of the product value $U_P = P \cdot V(X_P)$ and the waste value $U_W = W \cdot V(X_W)$, i.e. total value of isotopic composition increased on:

$$\Delta U = (U_P + U_W) - U_F = P \cdot V(X_P) + W \cdot V(X_W) - F \cdot V(X_F). \qquad (1)$$

The value gain ΔU is chosen as a main characteristic of the separative work scope needed to divide the initial binary isotope composition into two new materials, namely enriched uranium and depleted uranium.

The separation potential function $V(X)$ is dimensionless, and so the separative works are measured on the feed, product and waste mass units (kilograms, for instance). Also, as it follows from the definition, the separative work scope is independent on the applied isotope separation technology.

If the following mathematical operations are performed, then the exact formula for the separation potential function $V(X)$ can be derived:

1. Equation (1) must be re-written into the form containing the feed mass F only:

$$\Delta U = F \cdot [\theta \cdot V(X_P) + (1-\theta) \cdot V(X_W) - V(X_F)]. \qquad (2)$$

2. The separation potential functions $V(X_P)$ and $V(X_W)$ must be expanded in the Taylor series in the vicinity of X_F point including only the first three terms of the expansion.

Then, by assuming that the single-stage separative work is independent on the feed concentration X_F, the following second-order differential equation can be obtained for the separation potential function:

$$\frac{d^2V(X)}{dX^2}=\frac{1}{X^2\cdot(1-X)^2};$$

with the solution:

$$V(X)=(2X-1)\ln\frac{X}{1-X}.$$

Derivation of mathematical formula for the separation potential function

The feed mass F comes to the single-stage inlet, and two new materials leave the single-stage outlet, namely the product $P=\theta\cdot F$ and the waste $W=(1-\theta)\cdot F$. As a result, equation (2) was obtained.

If the separation potential functions $V(X_P)$ and $V(X_W)$ are expanded in the Taylor series in the vicinity of X_F point by such a way:

$$V(X_P)\approx V(X_F)+\frac{dV}{dX}\cdot(X_P-X_F)+0,5\cdot\frac{d^2V}{dX^2}\cdot(X_P-X_F)^2;$$

$$V(X_W)\approx V(X_F)+\frac{dV}{dX}\cdot(X_W-X_F)+0,5\cdot\frac{d^2V}{dX^2}\cdot(X_W-X_F)^2;$$

and substituted into equation (2), then the following equation is obtained:

$$\begin{aligned}\Delta U &= V(X_F)\cdot[\theta\cdot F+(1-\theta)\cdot F-F]+\\ &+\frac{dV}{dX}\cdot[\theta\cdot F\cdot(X_P-X_F)+(1-\theta)\cdot F\cdot(X_W-X_F)]+\\ &+0,5\cdot\frac{d^2V}{dX^2}\cdot[\theta\cdot F\cdot(X_P-X_F)^2+(1-\theta)\cdot F\cdot(X_W-X_F)^2].\end{aligned}\qquad(3)$$

By using the mass balance relationships, it is easy to show that the first two terms of equation (3) are equal to zero. Indeed:

$$F \cdot \theta + (1-\theta) \cdot F - F \equiv 0;$$

$$F \cdot \theta \cdot (X_P - X_F) + F \cdot (1-\theta) \cdot (X_W - X_F) = P \cdot (X_P - X_F) + W \cdot (X_W - X_F) \equiv 0.$$

Then

$$\Delta U = 0,5 \cdot \frac{d^2V}{dX^2} \cdot [\theta \cdot F \cdot (X_P - X_F)^2 + (1-\theta) \cdot F \cdot (X_W - X_F)^2]. \quad (4)$$

Let assume that very little enrichment and depletion gains occurred at any one of multiple stages of the isotope separation process, i.e. the single-stage enrichment and depletion gains ε' and ε'' are much lower than unity. Then, by using definitions of these gains, the following approximate expressions can be obtained:

$$\varepsilon' = \frac{X_P / (1-X_P)}{X_F / (1-X_F)} - 1 = \frac{X_P - X_F}{X_F \cdot (1-X_P)};$$

$$X_P - X_F = \varepsilon' \cdot X_F \cdot (1-X_P) \approx \varepsilon' \cdot X_F \cdot (1-X_F);$$

$$\varepsilon'' = \frac{X_F / (1-X_F)}{X_W / (1-X_W)} - 1 = \frac{X_F - X_W}{X_W \cdot (1-X_F)};$$

$$X_F - X_W = \varepsilon'' \cdot X_W \cdot (1-X_F) \approx \varepsilon'' \cdot X_F \cdot (1-X_F).$$

These expressions being substituted into equation (4) for ΔU can transform the equation into the following:

$$\Delta U = 0,5 \cdot \frac{d^2V}{dX^2} \cdot X_F^2 \cdot (1-X_F)^2 \cdot F \cdot [\theta \cdot \varepsilon'^2 + (1-\theta) \cdot \varepsilon''^2].$$

One else assumption must be used, namely the single-stage separative work is independent on the feed concentration X_F and defined only by the stage design and by the applied technology. If so, the following second-order differential equation is obtained:

$$\frac{d^2V}{dX^2}\cdot X^2\cdot(1-X)^2=1,\quad \text{or}\quad \frac{d^2V}{dX^2}=\frac{1}{X^2\cdot(1-X)^2}.$$

General solution of this equation can be written in the following form:

$$V(X)=(2X-1)\cdot\ln\left(\frac{X}{1-X}\right)+A\cdot X+B.$$

It can be easily shown that any values of A and B factors do not change the separative works scope at all because the (A and B)-related terms can produce no effect on ΔU value:

$$\Delta U_{A,B}=A\cdot(P\cdot X_P+W\cdot X_W-F\cdot X_F)+B\cdot(P+W-F)\equiv 0.$$

Therefore, the following last assumption can be accepted: $A=B=0$. Finally, the separative works scope can be calculated by using the formula:

$$\Delta U=P\cdot V(X_P)+W\cdot V(X_W)-F\cdot V(X_F);$$

where

$$V(X)=(2X-1)\ln\left(\frac{X}{1-X}\right).$$

If kilograms are chosen as the feed, product and waste mass units, then the separative works scope can be also measured in the SW-kilograms, and, by definition, 1 SW-kilogram = 1 SWU (separative work unit).

Specific scope of the separative works η_{SWU} can be defined as the works scope needed to produce 1 kg of enriched uranium:

$$\eta_{SWU}=\frac{\Delta U}{P},\text{SWU / kg.}$$

As it was shown above:

$$F = P \cdot \frac{X_P - X_W}{X_F - X_W}; \quad W = P \cdot \frac{X_P - X_F}{X_F - X_W}.$$

So:

$$\Delta U = P \cdot V(X_P) + P \cdot \frac{X_P - X_F}{X_F - X_W} \cdot V(X_W) - P \cdot \frac{X_P - X_W}{X_F - X_W} \cdot V(X_F);$$

and

$$\eta_{SWU} = V(X_P) + V(X_W) \cdot \frac{X_P - X_F}{X_F - X_W} - V(X_F) \cdot \frac{X_P - X_W}{X_F - X_W}.$$

The concepts of the separative works and their units of measure have been developed at Oak Ridge National Laboratory (USA) to provide a scientific foundation for prices and commercial accounts to be paid for the offered enriching services. All the expenses related with uranium enrichment are referred to the really performed separative works. Dependencies of the separative works needed to produce 1 kg of enriched uranium from natural uranium are available now in a tabular form as functions of relative ^{235}U content in the feed and waste uranium.

Some data on the single-stage separation factor and specific energy consumption are presented in Table 1.4 for different uranium enrichment technologies.

Table 1.4

Comparison of uranium enrichment technologies on the separation factor and specific energy consumption

Technology	Separation factor	Energy consumption, kWh/SWU
Electromagnetic	20-40	4000
Gas diffusion	1,0043	2300-2600
Gas centrifuges	1,25	100-300
Separation nozzle	1,025	3000-3500
Laser	3-15	10-50
Chemical	1,0025	400-700
Plasma	3,5-10	200-600

1.2.2. Properties of uranium hexafluoride and technologies for its production

The most of the uranium enrichment technologies apply gaseous uranium hexafluoride UF_6 as an initial (feeding) material. This uranium compound is characterized by a series of very attractive properties, especially important for the uranium enriching process:

1. Natural fluorine is a one-isotope element containing only one stable isotope ^{19}F. If natural fluorine would contain one else stable isotope (^{18}F, for instance), then the isotope separation process would deal with four components ($^{235}U^{18}F_6$, $^{238}U^{18}F_6$, $^{235}U^{19}F_6$ and $^{238}U^{19}F_6$) with molecular masses of 343, 346, 349 and 352 a.m.u., respectively. This means that the lighter fraction (343 and 346 a.m.u.) would contain some amount of ^{238}U while the heavier fraction (349 and 352 a.m.u.) would contain some amount of ^{235}U.

2. Fluorine is a comparatively light chemical element. Relative difference of molecular $^{235}UF_6$ and $^{238}UF_6$ masses is equal to $3/349 \approx 0{,}0086$. This value is lower than relative difference of atomic ^{235}U and ^{238}U masses ($3/235 \approx 0{,}0128$) but not very much.

3. Uranium hexafluoride can exist in the solid, liquid and gaseous states under moderate temperature and pressure conditions (Fig. 5). Triple point at UF_6 state diagram corresponds to the temperature of 64^0C and the pressure of 1138 mmHg (about 1,5 atmosphere).

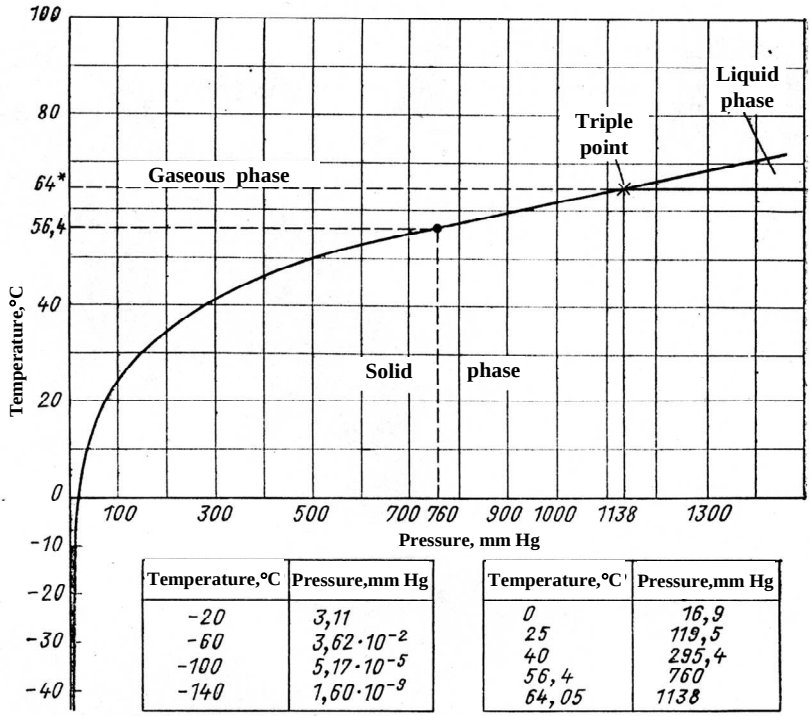
4. Uranium hexafluoride can be sublimated from the solid state into the gaseous state omitting the liquid state by a slight warming-up. And vice versa, gaseous uranium hexafluoride can be condensed into the solid stat by a slight cooling-down.

Thus, physical properties of uranium hexafluoride are very suitable to develop sufficiently simple in design, comfortable and compact facilities for the uranium isotope enrichment.

However, uranium hexafluoride is characterized by the following disadvantages:

1. Strong chemical activity. Uranium hexafluoride can intensely interact with air and water vapor with the formation of uranium tetrafluoride UF_4 as a powder that can deposit on inner surfaces of technological circuitry.

2. As a consequence, a necessity arises to use only tightly hermetical pipes and vessels, maintain their dehydration, degreasing and the surgical-like cleanness. The most stable structural materials for operations with gaseous uranium hexafluoride are nickel, aluminum, magnesium, copper, and their alloys, teflon of organic materials.



Temperature,°C	Pressure,mm Hg
-20	3,11
-60	$3,62 \cdot 10^{-2}$
-100	$5,17 \cdot 10^{-5}$
-140	$1,60 \cdot 10^{-9}$

Temperature,°C	Pressure,mm Hg
0	16,9
25	119,5
40	295,4
56,4	760
64,05	1138

Fig. 1.1. Diagram of uranium hexafluoride states

Conversion of uranium oxides into uranium hexafluoride

In the open NFC the uranium concentrate U_3O_8, product of the extraction affinage, is an initial material for its conversion into uranium hexafluoride.

Uranium concentrate U_3O_8 is usually fluorinated by means of the following two-step process:

1. Reaction of U_3O_8 with gaseous fluorine at 350-370^{0}C that leads to the formation of uranyl-fluoride UO_2F_2:

$$U_3O_8 + 3F_2 \rightarrow 3UO_2F_2 + O_2.$$

2. Reaction of uranyl-fluoride with gaseous fluorine at slightly reduced temperature (~270^{0}C):

$$UO_2F_2 + 2F_2 \rightarrow UF_6 + O_2.$$

Another one-step process is feasible too. The one-step process is based on the direct high-temperature fluorination technology. However, the process is feasible only at excess amounts of gaseous fluorine and at substantially higher temperatures (900-1000^{0}C):

$$U_3O_8 + 9F_2 \rightarrow 3UF_6 + 4O_2.$$

In the closed NFC with recycling of the regenerated uranium, uranium dioxide UO_2 extracted from spent fuel assemblies is an initial material for its conversion into uranium hexafluoride. In this case the following two-step fluorination process is usually applied:

1. Reaction of uranium dioxide with hydrofluoric acid at 500-600^{0}C that leads to the formation of uranium tetra-fluoride UF_4:

$$UO_2 + 4HF \rightarrow UF_4 + 2H_2O.$$

2. Reaction of uranium tetra-fluoride with gaseous fluorine at 400^{0}C:

$$UF_4 + F_2 = UF_6$$

Afterwards, uranium hexafluoride is condensed at -15^{0}C and can be transported in the containers made of nickel-based alloys.

1.2.3. Uranium enrichment by gas diffusion technology

Gas diffusion (GD) is a physical phenomenon of mass transport in a mixture of different gases caused by their thermal movements.

The GD-technology of material separation is based on different thermal velocity of light and heavy molecules, and on different penetrability of light and heavy molecules through porous walls (membranes).

In binary mixture of light and heavy gases both components have the same temperature and, thus, the same kinetic energy:

$$m_{LIGHT} \cdot V_{LIGHT}^2 = m_{HEAVY} \cdot V_{HEAVY}^2.$$

So, the light molecules can move with higher velocity and, as a consequence, can penetrate through a porous wall with larger probability:

$$\frac{V_{LIGHT}}{V_{HEAVY}} = (m_{HEAVY} / m_{LIGHT})^{1/2}.$$

In principle, it can be shown that the maximal, theoretically achievable ideal separation factor α_0 for two gases diffusing through a porous wall is equal to:

$$\alpha_0 = \frac{V_{LIGHT}}{V_{HEAVY}} = \left(m_{HEAVY}/m_{LIGHT}\right)^{1/2} \approx 1 + \frac{\Delta m}{2m_{LIGHT}}.$$

Molecular masses of $^{235}UF_6$ (the light gas) and $^{238}UF_6$ (the heavy gas) are equal to 349 a.m.u. and 352 a.m.u., respectively. Thus:

$$\alpha_0 = 1,0043; \quad \varepsilon_0' = \alpha_0 - 1 = 0,0043.$$

Efficiency of the GD-technology can be upgraded if the mean free path λ of UF_6 molecules is much longer than typical size of pores a ($\lambda >> a$) because main mechanism of thermal movements must be molecule-pore interactions, not inter-molecular collisions. The mean free path of any gaseous molecules is inversely proportional to the pressure. For example, the mean free path of UF_6 molecules is equal to ~1 micron at atmospheric pressure and to ~700 microns at 1 mm Hg.

Manufacturing of the GD-membranes with the micron-level sizes of pores is a very complicated and the most classified problem. The porous walls must be:

1. thin (well below 1 mm);
2. strong (under the pressure drop up to 0,3 atmosphere);
3. corrosion-resistant in the UF_6 environment.

Currently, the porous tubular elements for the GD-technology are being made of the following materials:

1. sintered powders of alumina and nickel oxide;
2. sintered nickel powder;
3. porous aluminum produced by the electrical etching technology.

Typical parameters of the GD-technology: the temperature range – $65 \div 110^0$C, the pressure - ~0,35 atmosphere, the pressure drop - ~0,3 atmosphere.

Cascading of the GD-process. Since the single-stage enrichment gain that can be achieved by the GD-technology is very small (0,0043), many successive GD-stages have to be used to reach necessary values of uranium enrichment (for example, up to 5% ^{235}U for nuclear power reactors or above 90% ^{235}U for nuclear weaponry). System of the successively linked GD-stages constitutes the GD-cascade with two different separation branches (Fig. 1.2): the depleting branch where relative ^{235}U content reduces from 0,71% in natural uranium (the feed material) down to 0,2-0,3% in the depleted uranium (the waste material or tails), and the enriching branch where relative ^{235}U content increases from 0,71% in natural uranium up to the necessary values (5%÷90% ^{235}U).

Experimental studies have shown that the best arrangement for the successive GD-stages is that in which half the gas flow pumped into each stage diffuses through the porous wall to the next higher (enriching) stage, the other half being returned to the feed material of the lower stage.

Evidently, the numbers of the GD-stages and UF_6 flow rates are different in the enriching and the depleting branches. These values depend on ^{235}U content in the product and in the waste. It is clear that relatively small number of the depleting stages is required to reduce ^{235}U content from 0,71% in natural uranium down to 0,2-0,3% in the waste. On the contrary, relatively large number of the enriching stages is required to upgrade ^{235}U content from 0,71% in natural uranium up to 5% ^{235}U (re-

actor-grade uranium) or 90% ^{235}U (weapon-grade uranium) in the product. The flow rates of uranium hexafluoride successively reduce in both branches but the reduction of UF_6 flow rate is sharper in the enriching branch. The sharper reduction can be explained by the following consideration. In the extreme case of producing 100% ^{235}U in the product and 0% ^{235}U in the waste from 1000 kg of natural uranium, final stages of the enriching branch would handle with 8-10 kg of the enriched uranium while final stages of the depleting branch would handle with ~990 kg of the depleted uranium (Fig. 1.3).

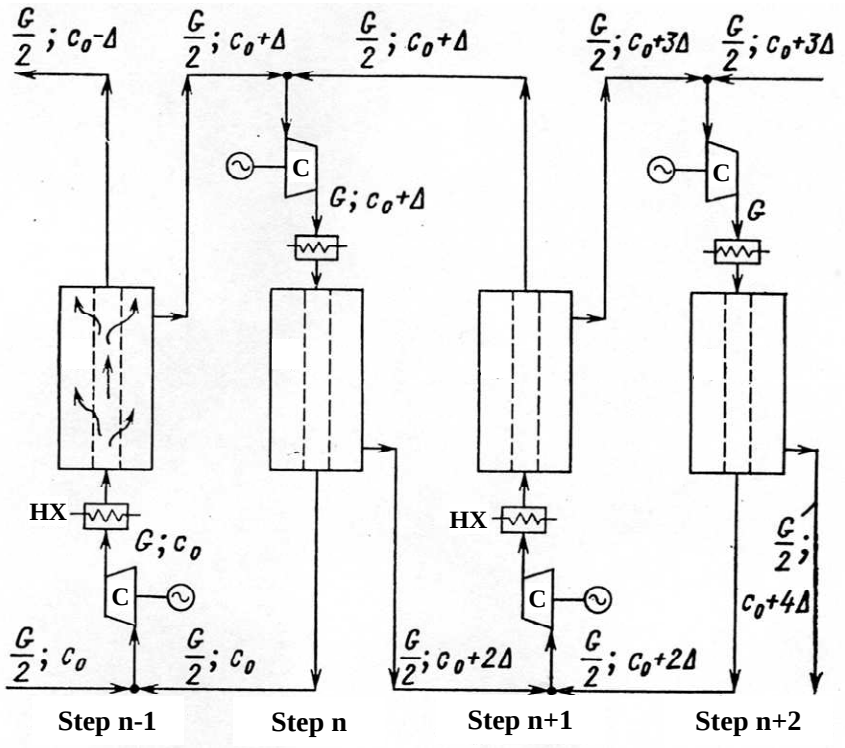


Fig. 1.2. Layout of the GD-cascade

The numbers of the enriching and the depleting stages in the GD-cascade can be evaluated following form the definitions of the single-stage enrichment ε' and depletion ε'' gains.

The following expressions can be written for uranium enrichment after the first enriching stage and, then, after N_P enriching stages:

$$\frac{X_P}{1-X_P}(1) = (1+\varepsilon')\cdot\frac{X_F}{1-X_F};$$

$$\frac{X_P}{1-X_P}(N_P) = (1+\varepsilon')^{N_P}\cdot\frac{X_F}{1-X_F}.$$

So,

$$N_P = \frac{\ln\dfrac{X_P/(1-X_P)}{X_F/(1-X_F)}}{\ln(1+\varepsilon')} \approx \frac{1}{\varepsilon'}\cdot\ln\frac{X_P/(1-X_P)}{X_F/(1-X_F)}.$$

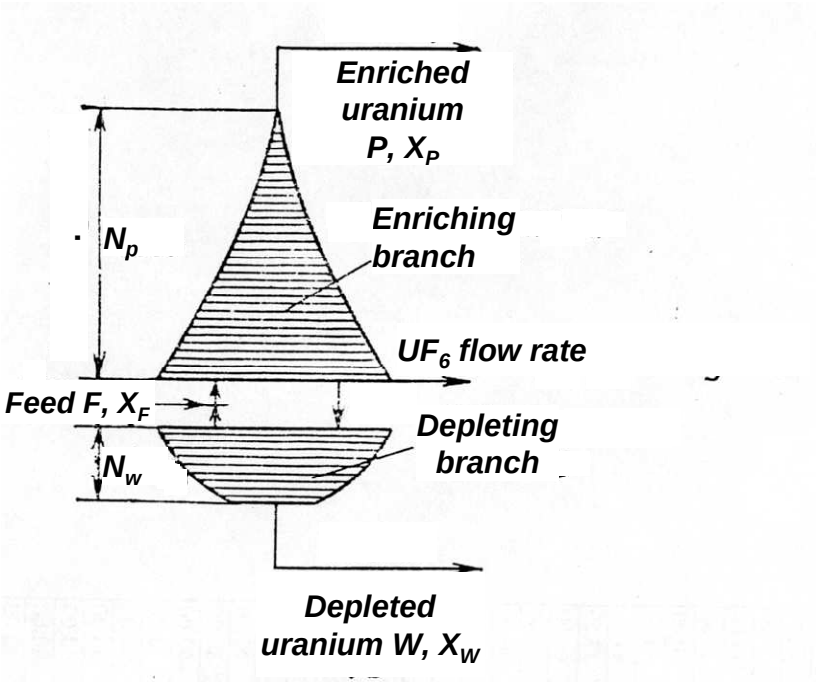


Fig. 1.3. Reduction of the stage quantity and uranium flow rate in the GD-branches

Similar expressions can be obtained for the depleting branch:

$$\frac{X_W}{1-X_W}(1)=\frac{1}{1+\varepsilon''}\cdot\frac{X_F}{1-X_F};$$

$$\frac{X_W}{1-X_W}(N_W)=\frac{1}{(1+\varepsilon'')^{N_W}}\cdot\frac{X_F}{1-X_F}.$$

So,

$$N_W=\frac{\ln\dfrac{X_F/(1-X_F)}{X_W/(1-X_W)}}{\ln(1+\varepsilon'')}\approx\frac{1}{\varepsilon''}\cdot\ln\frac{X_F/(1-X_F)}{X_W/(1-X_W)}.$$

Let assume that it is necessary to produce weapon-grade uranium ($X_P=90\%\ ^{235}U$) from natural uranium ($X_F=0,71\%\ ^{235}U$) by the GD-technology ($\varepsilon'=\varepsilon''=0,0043$) with ^{235}U content in the waste $X_W=0,2\%\ ^{235}U$. Then, the numbers of the enriching stages and the depleting stages are equal $N_P\approx1660$ and $N_W\approx290$, respectively. If reactor-grade uranium ($X_P=4\%\ ^{235}U$) must be produced from natural uranium with the same ^{235}U content in the waste (0,2%), then the number of the enriching stages decreases to $N_P\approx410$ at the same number of the depleting stages ($N_W\approx290$).

1.2.4. Uranium enrichment in gas centrifuges

If a cylindrical vessel (centrifuge) containing a binary mixture of light and heavy gases rotates with angular velocity ω, then the centrifugal force acts on the elementary volume of the gaseous mixture:

$$F_{1,2}(r)=\gamma_{1,2}\cdot\omega^2\cdot r;$$

where $\gamma_{1,2}$ – densities of the gaseous components; r – distance from center of the vessel.

The pressures on the gaseous components can be determined with application of the following differential equation:

$$\frac{dP_{1,2}(r)}{dr} = F_{1,2}(r) = \gamma_{1,2} \cdot \omega^2 \cdot r. \qquad (5)$$

By using the Mendeleev-Clapeyron equation:

$$P_{1,2}(r) \cdot V = \frac{m}{M_{1,2}} \cdot R \cdot T;$$

(M_1, M_2 – molecular masses of the gaseous components), densities of the gaseous components can be determined:

$$\gamma_{1,2}(r) = \frac{m}{V} = \frac{P_{1,2}(r) \cdot M_{1,2}}{R \cdot T}.$$

Then, differential equation (5) can be re-written

$$\frac{dP_{1,2}(r)}{dr} = \frac{P_{1,2}(r) \cdot M_{1,2}}{R \cdot T} \cdot \omega^2 \cdot r;$$

and solved:

$$P_{1,2}(r) = P(0) \cdot \exp\left(\frac{M_{1,2} \cdot \omega^2 \cdot r^2}{2 \cdot R \cdot T}\right) = P(0) \cdot \exp\left(\frac{M_{1,2} \cdot V^2}{2 \cdot R \cdot T}\right);$$

where V – linear velocity.

Evidently, content of the light and heavy components in the gaseous mixture are proportional to the spatial pressure distribution:

$$X_{235}(r) = X_{235}(0) \cdot \exp\left(\frac{M_{LIGHT} \cdot V^2(r)}{2 \cdot R \cdot T} \right);$$

$$X_{238}(r) = X_{238}(0) \cdot \exp\left(\frac{M_{HEAVY} \cdot V^2(r)}{2 \cdot R \cdot T} \right).$$

These formulas demonstrate that content of the heavy component (depleted uranium) is larger in peripheral region of the centrifuge, and, vice versa, content of the light component (enriched uranium) is larger in central region of the centrifuge. In this case, the single-stage enrichment factor can be determined from the following expressions:

$$\alpha(r) = \frac{X_{235}(0) / X_{238}(0)}{X_{235}(r) / X_{238}(r)} = \frac{\exp\left(-M_{LIGHT} \cdot V^2(r) / 2 \cdot R \cdot T\right)}{\exp\left(-M_{HEAVY} \cdot V^2(r) / 2 \cdot R \cdot T\right)} =$$

$$= \exp\left(\Delta M \cdot V^2(r) / 2 \cdot R \cdot T\right);$$

$$\varepsilon'(r) = \alpha(r) - 1 \approx \frac{\Delta M \cdot V^2(r)}{2 \cdot R \cdot T}).$$

As is seen, the single-stage enrichment gain ε' of the GC-technology depends only on absolute, not relative like in the GD-technology, difference of molecular masses of the light and heavy gas components. Also, the single-stage enrichment gain is proportional to the squared linear velocity of the centrifuge rotation. The centrifuges of contemporary designs can rotate with linear velocities up to 500-700 m/s, i.e. near to the velocity of a bullet outgoing from the rifle tube. According to many numerical evaluations, the GC-technology can provide the following velocity-dependent values of the single-stage enrichment gain at the outer centrifuge radius r_0:

$$\varepsilon'(r_0) = 0,068 \text{ at } V = 330 \text{ m/s};$$
$$\varepsilon'(r_0) = 0,098 \text{ at } V = 400 \text{ m/s};$$
$$\varepsilon'(r_0) = 0,152 \text{ at } V = 500 \text{ m/s};$$
$$\varepsilon'(r_0) = 0,300 \text{ at } V = 700 \text{ m/s}.$$

The gas centrifuges are currently being made of the following structural materials:

1. Aluminum-based alloys for linear velocities $V \leq 350$ m/s.
2. Titanium-based alloys for linear velocities $V \leq 450$ m/s.
3. Alloyed steels for linear velocities $V \leq 500$ m/s.
4. Graphite-reinforced glass-fiber plastics for linear velocities $V = 500$-700 m/s.

If vertical gas circulation can be arranged in the centrifuge (for example, by thermal convection caused by temperature gradient between top and bottom parts of the centrifuge), then the centrifuge can act as the enriching cascade. The gaseous mixture goes upwards along central axis and then goes downwards along the centrifuge wall. So, the gaseous mixture of $^{235}UF_6$ and $^{238}UF_6$ is being continuously enriched with the heavy component in the peripheral bottom region while the gaseous mixture is gradually enriched with the light component in the central top region of the centrifuge.

The gas flow going upwards in the center and downwards at the periphery can be formed by an insignificant warming-up of the central region. The warming-up effect can be produced by a small electrically heated rod placed in the centrifuge center.

1.2.5. Separation nozzle technology

The separation-nozzle (SN) technology has been developed at the Karlsruhe Nuclear Research Center (Germany) as an alternative to the GD- and GC-technologies. The gaseous mixture UF_6 and hydrogen (or helium) expands along a bent wall. The centrifugal deflection force can split the flow into the light and heavy fractions by means of a slimmer (Fig. 1.4).

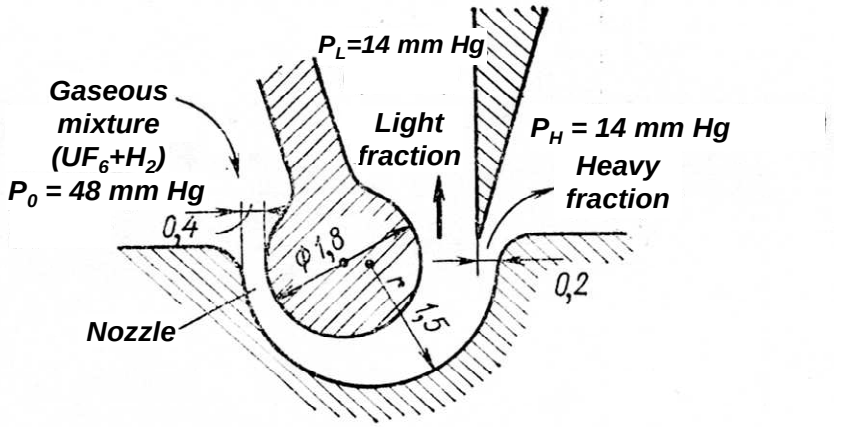


Fig. 1.4. Layout of the separation-nozzle technology

The hydrogen or helium auxiliary gas (gas-carriers) increases the flow velocity and, hence, it increases the centrifugal forces defining efficiency of the SN-process.

The SN-technology is profitably distinguished from the GC-technology by the absence of the rotating details but it requires a very fine mechanical assemblage because of very little sizes of the splitting slits (decimal fractions of one millimeter). The single-stage enrichment gain in the SN-technology can reach $\varepsilon' \approx 0,025$ at the specific energy consumption about 3000 kWh/SWU.

1.2.6. Laser technologies of isotope separation

The laser technologies of uranium enrichment rely on the slightly different excitation energies of electronic shells that surround ^{235}U and ^{238}U nuclei. Three extra neutrons in ^{238}U nucleus caused the slight shift in the electron excitation energy scheme as compared with ^{235}U nucleus. This energy shift can be used to excite selectively uranium atoms or uranium-containing molecules by the monochromatic laser light properly tuned to the required wavelength. The excited state of electronic shell can selectively enhance some physical or chemical processes with uranium-containing materials and, thus, promote isotope separation.

The following conditions should be satisfied for successful implementation of the laser-induced isotope separation:

1. The energy spectrum of the excited electronic levels must contain a line belonging to one isotope only, and this line must be sufficiently far from other spectral lines of the desirable isotope and from all spectral lines of other isotopes.
2. Physical or chemical processes must be found which are able to separate the excited and non-excited uranium-containing components.
3. Laser-induced impact on the isotopic composition to be separated must be a main excitation mechanism, not inter-atomic or inter-molecular collisions.
4. High-efficiency lasers must be developed and finely tuned to the appropriate wavelength.

Presently, the following two laser isotope separation technologies are under intense development and demonstration, namely atomic vapor laser isotope separation (AVLIS) and molecular laser isotope separation (MLIS).

The AVLIS-technology has been developed at the Lawrence Livermore National Laboratory (USA). The AVLIS technology includes the following stages:

1. Vacuum evaporation of uranium atoms at very high temperature (~2300^{0}C). Beam of accelerated electrons knocks uranium atoms out of uranium-rhenium alloy.
2. Irradiation by xenon laser (λ~3780 Å, ultraviolet range). ^{235}U atoms are selectively excited.
3. Irradiation by krypton laser (λ~3500 Å, ultraviolet range). The excited ^{235}U atoms are selectively ionized.
4. Collection of ^{235}U ions on an electrically charged plate.

The MLIS-technology has been developed at the Los Alamos National Laboratory (USA). The MVLIS technology includes the following stages:

1. Expansion of gaseous uranium hexafluoride – hydrogen composition through a hypersonic nozzle. As a result, uranium hexafluoride cools down to about 30 K but it does not condense.
2. Irradiation by infrared laser (λ~1,6·10^5 Å). Molecules of $^{235}UF_6$ are selectively excited.

3. Irradiation by ultraviolet laser ($\lambda \sim 3{,}08 \cdot 10^4$ Å). The excited molecules of $^{235}UF_6$ are selectively dissociated with the formation of uranium pentafluoride $^{235}UF_5$ and free fluorine:

$$2 \cdot {}^{235}UF_6 \rightarrow 2 \cdot {}^{235}UF_5 + F_2.$$

Uranium pentafluoride $^{235}UF_5$ precipitates from the gas flow as a fine powder (so called, "laser snow") that can be easily collected.

The single-stage enrichment factors are very high for both laser technologies of uranium enrichment. They cover the range from 3 to 15, according to different experimental studies. Such high-efficiency technologies make it possible to use even the waste materials from GD- and GC-processes containing about 0,2% ^{235}U for production of reactor-grade uranium (about 3% ^{235}U) by a single enrichment stage.

1.2.7. Chemical methods of isotope separation

The chemical methods of isotope separation are based on the preferential stability of certain isotopes in various immiscible chemical compounds. The isotope exchange reactions can occur, if two different chemical compounds of one multi-isotope chemical element enter into a contact. The isotope exchange reactions lead to the concentration of isotopes in those compounds where they can be more stable.

The following conditions must be satisfied for feasibility of the chemical isotope separation technologies:

1. The contacting compounds must be chemically stable together.
2. The contacting compounds must be separated by a relatively simple means (for example, organic and inorganic substances).
3. It is desirable for the chemical element to be of different valences in two contacting compounds.

The following examples of the chemical isotope separation are presented below:

1. Boron enrichment with isotope ^{10}B:

$$BF_3 + BF_3O(CH_3)_2 \rightarrow {}^{11}BF_3 + {}^{10}BF_3O(CH_3)_2 ,$$

i.e. isotope ^{10}B passes into the organic compound.

2. Production of heavy water:

$$H_2O + HDS \rightarrow HDO + H_2S.$$

Natural hydrogen contains about 0,015% deuterium. In the isotope exchange reaction between light water and hydrogen sulphide, deuterium passes into the aqueous fraction.

The chemical isotope separation technologies for boron enrichment and heavy water production are characterized by the single-stage separation factor about 1,0025 and specific energy consumption within the range of 400-700 kWh/SWU.

Presently, the advanced chemical uranium enrichment technology is under development and testing in the USA and Japan. The technology applies UF_6 and $NOUF_6$ as the contacting compounds. The process is called as "reduction-oxidation (redox) chromatography". The redox chromatography consists in alternating the reduction reaction with hydrogen and the oxidation reaction with oxygen. The process results in separation of the chemical compound containing UO_2^{++} ions (six-valence uranium where ^{235}U is more stable) and U^{4+} ions (four-valence uranium where ^{238}U is more stable). Some experimental studies demonstrated sufficiently good parameters of the redox chromatography: the single-stage separation factors are about 1,08 and specific energy consumption is about 150 kWh/SWU.

1.2.8. Plasma method of isotope separation

The plasma technology of isotope separation is based on the effect of ion cyclotron resonance. The effect is described below.

If any charged particles (ions, for instance) pass through a constant magnetic field B, they begin rotating along spiral orbits around force lines of the magnetic field under action of the centrifugal force $\vec{F}$:

$$\vec{F} = q \cdot [\vec{V} \times \vec{B}];$$

where q –electrical charge of ions; $\vec{V}$ - velocity of ion movement.

Orbital radius R and angular frequency of spiral rotation can be derived from the following relationships:

$$F = q \cdot V \cdot B = \frac{m \cdot V^2}{R};$$

$$R = \frac{m \cdot V}{q \cdot B} = \frac{(2 \cdot m \cdot E)^{1/2}}{q \cdot B};$$

$$\omega = \frac{V}{R} = \frac{q \cdot B}{m}.$$

The angular frequency ω is called as an ion cyclotron frequency (ICF) of isotope with mass m.

Isotope collectors	Isotope separation area	Microwave ion source

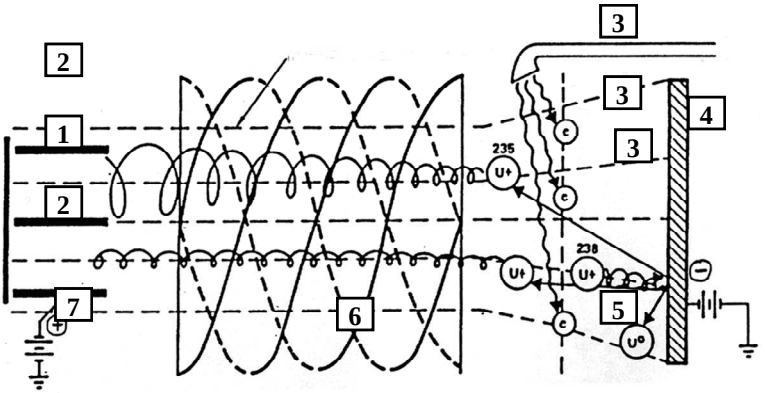


Fig. 1.5. Layout of the plasma isotope separation
1 – collectors of the waste; 2 – collectors of the product; 3 – force lines; 4 – metal plate (source of neutral particles); 5 – electrical heating area; 6 – antenna of the alternating electrical field; 7 – charging of the collectors to enhance isotope separation efficiency.

A principal layout of the plasma isotope separation is shown in Fig. 1.5.

If the alternating electrical field with the frequency equaled to the ICF value of ^{235}U ions, for instance, is applied to the flow of spirally rotating ions, then energy of the alternating electrical field can be absorbed by ^{235}U ions only. Just this is the effect of ion cyclotron resonance. Selective increasing the energy of ^{235}U ions can extend their spiral trajectories and, thus, create the opportunity for spatial separation of ^{235}U and ^{238}U ions. The ICF values of two main uranium isotopes differ from each other on about 1,2%. The difference can allow it to arrange selective acquisition of ^{235}U and ^{238}U ions on the properly placed and charged collectors.

1.3. Technologies for fabrication of fuel rods and fuel assemblies

Presently, uranium dioxide UO_2 is the most widely used type of ceramic nuclear fuel. Uranium dioxide fuel (UOX-fuel) is currently loaded into practically all types of nuclear power reactors including thermal light-water and heavy-water reactors as well as fast breeder reactors).

Uranium dioxide is a dark-brown, highly hard and brittle substance. Uranium dioxide does not interact with alkaline and aqueous solutions up to 300^0C but it can be well dissolved by acidic solutions (nitric acid and mixture of nitric acid with hydrochloric or hydrofluoric acid).

Main advantages of uranium dioxide:

1. High melting temperature (2780^0C).
2. High chemical stability in contacts with main coolants of nuclear power reactors (light water, heavy water, sodium and carbon dioxide).
3. Satisfactory compatibility with main cladding materials of nuclear power reactors (stainless steels, zirconium-based alloys) within the reactor temperature ranges.
4. Acceptable radiation resistance under high neutron fluxes ($\sim 10^{14}$ $n/cm^2 \cdot s$) and fluences (up to $\sim 10^{22}$ n/cm^2, i.e. for about three years).
5. Manufacturing feasibility of high-density UOX-fuel pellets (up to 95% of its theoretical density that equals 10,96 g/cm^3).

6. Isotropy of crystalline lattice that can simplify the process of high-temperature sintering.

Main shortcomings of uranium dioxide:

1. Low hat conductivity and its sharp reduction at the elevated temperatures (8,4 W/m·K at 45^0C and 2,4 W/m·K at 1327^0C). Such low values and temperature dependency of UOX heat conductivity results in very large temperature gradients inside of very thin (R ~ 3 mm) fuel pellets (ΔT ~ 1500^0C at the distance of 3-4 mm).
2. Intense oxidation ability by wet air at ambient temperature (hygroscopicity). This effect requires an inert dry environment 0r vacuum for UOX-fuel pellets manufacturing. Otherwise, superficial layers of UOX-fuel pellets can be saturated with water and oxygen. Later on, during the reactor operation, the moisture released from the pellet surface can cause hydration of the cladding materials and destruction of fuel rods.
3. The presence of oxygen in UOX-fuel composition softens neutron spectrum and, thus, decreases the secondary fuel production rate.

1.3.1. Pelletization of uranium dioxide

The following processes are used now to produce UOX-fuel pellets:

1. *Conversion of uranium hexafluoride into uranium dioxide.* Two conversion technologies have been developed and currently used:

a. "Wet" technology of the AUC-process:

- Barbotage of gaseous uranium hexafluoride through aqueous solution of ammonium carbonate $(NH_4)_2CO_3$ followed by precipitation of solid insoluble deposit of ammonium-uranyl-carbonate (AUC) - $(NH_4)_4UO_2(CO_3)_3$.
- Heat treatment of AUC at 550-650^0C followed by thermal AUC dissociation with the formation of finely dispersed UOX powder.

b. "Dry" technology:

- Hydrolysis of uranium hexafluoride by water vapor at 150-300^0C with the formation of uranyl-fluoride UO_2F_2:

$$UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF.$$

- Pyrohydrolysis of uranyl-fluoride by hydrogen at ~550^0C with the formation of finely dispersed UOX powder and hydrofluoric acid.

$$UO_2F_2 + H_2 \rightarrow UO_2 + 2HF.$$

Thus, the finely dispersed UO_2 powder is produced. Unfortunately, such small-sized powder is unsuitable for manufacturing of UOX-fuel pellets by pressing because of too small dimensions of the powder particles (below 0,5 micron). The following procedures should be performed to enlarge the powder particles:

2. *Mixing of UO_2 powder with an organic plasticizer* (polyvinyl, glycidol and so on).
3. *Hydro-compaction of the powder-plasticizer mixture:* the mixture is placed into a plastic form; the plastic form is placed into a reservoir filled up with water, uniform omni-directional pressing, and production of the powder-plasticizer briquettes.
4. *Granulation of the briquettes by milling.*
5. *Annealing* at 600-800^{0}C for removal of organic plasticizers.
6. *Cold pressing of pellets* (p = 1500-2000 atmospheres).
7. *Sintering of UOX-fuel pellets* at 1600-1700^{0}C.
8. *Quality control of UOX-fuel pellets (*sizes, content of carbon as a residual of organic plasticizers, stoichiometry).

The manufacturing process of UOX-fuel pellets usually associated with the manufacturing process of mixed oxide (MOX), mainly uranium-plutonium, fuel pellets. In principle, the following three MOX-fuel compositions are feasible:

1. PuO_2 + $^{238}UO_2$, where plutonium is taken from the weapon-grade nuclear materials (weapon-grade plutonium).
2. PuO_2 + $^{238}UO_2$, where plutonium is extracted from spent fuel of nuclear power reactors (reactor-grade plutonium).
3. $^{235}UO_2$ + $^{238}UO_2$, where ^{235}U is taken from the weapon-grade nuclear materials (weapon-grade uranium).

Anyway, there is a distinction of principle in the manufacturing process of UOX-fuel pellets from a single feed flow and the manufacturing process of MOX-fuel pellets from two different feed flows. In the former case, natural uranium is a single feed material which, after a series of technological operations, converts into the enriched uranium dioxide and, then, into UOX-fuel pellets. At all these operations, ^{235}U nuclei were uniformly mixed with ^{238}U nuclei. In the latter case, on the

contrary, the manufacturing process of MOX-fuel pellets is based on technological operations with two different feed flows:
1. The fertile material fraction, i.e. $^{238}UO_2$ powder made of depleted or natural uranium.
2. The fissile material fraction, i.e. PuO_2 powder made of weapon-grade or reactor-grade plutonium, or $^{235}UO_2$ powder made of weapon-grade uranium.

Here, the homogeneity of the fertile-fissile fractional mixture is not guaranteed. So, a high degree of homogeneity must be ensured in the blending process of the fertile and fissile components. The blending process of two different feed flows is the only stage that distinguishes the MOX-fuel manufacturing technology from the UOX-fuel manufacturing technology.

The homogeneous mixture of the fissile and fertile components can ensure the safer operation of nuclear power reactors because fertile isotope ^{238}U and fissile isotopes ^{235}U, ^{239}Pu can cause quite different reactivity effects under accidental conditions. If the reactor power increased, then both fuel components warmed up but fissile isotopes, main contributors into the chain fission reaction, warmed up in the first turn. Fertile isotopes can warm up with some time delay, and the better homogeneity of fuel composition results in the shorter time delay of the fertile component warming up. Temperature increasing of the fissile component causes the Doppler effect that leads to the energy extension of the resonances in neutron capture and fission cross-sections. As a rule, the Doppler effect of fissile isotopes can cause a relatively small but positive reactivity change (increment). As a rule too, the Doppler effect of fertile isotopes can cause a large and negative reactivity change (decrement). If the Doppler effects of fissile and fertile isotopes occur simultaneously (the best case) or with only short time delay, then the reactivity stabilization effect of fertile isotopes can be in a due time for neutralization of the positive reactivity change caused by fissile isotopes warming up. If the time delay between actuation of the reactivity increment caused by fissile isotopes and the reactivity decrement caused by fertile isotopes would be remarkably long (the worst case), then the reactivity increment of fissile isotope can have a sufficiently long time interval to increase the reactor power up to an unacceptably high level

till the stabilizing reactivity decrement of fertile isotopes would begin acting.

At initial stage of nuclear power development in the world some two-purpose thermal reactors were fueled with metal natural uranium (the UK "Magnox" reactors, for instance). These reactors were characterized by relatively low values of fuel burn-up and thermal energy generation rate.

Metal uranium has the following advantages:

1. High density (18,7 g/cm^3 via 10,96 g/cmм^3 of uranium dioxide).
2. The better neutron balance in the reactor core leads to the lower values of uranium enrichment and annual uranium consumption.
3. High heat conductivity (30 W/m·K via 3 W/m·K of uranium dioxide).
4. High heat generation rate and, thus, small sizes of the reactor core.
5. The higher values of breeding ratio.
6. Simplicity and cheapness of metal uranium fuel manufacturing.

However, the developers had to decline the further usage of metal uranium fuel in contemporary designs of nuclear power reactors and decided to use UO_2-based fuel preferentially. This decision was validated by the following shortcomings of metal uranium fuel:

1. Incompatibility with light-water coolant. If some defects in the fuel cladding appeared, then metal uranium is intensely dissolved by hot water, and radioactive fission products can release from fuel meat and spread into NPP circuitry.
2. Instability of fuel sizes under high values of fuel burn-up, neutron flux and fluence. The radiation damages of metal uranium fuel pass the following three consecutive phases as the fuel temperature increases:

a. Irradiation-induced anisotropic growth of the metal grain sizes and irradiation creep at temperatures below 500^0C.

b. Cavitation swelling within the temperature range from 370^0C to 500^0C. The cavitation swelling is caused by the formation of irregular pores within the temperature range where mechanical stresses caused by the irradiation-induced growth of the metal grain sizes still take place also. As a consequence, mechanical strength of metal uranium fuel is substantially weakened, especially along the metal grain boundaries.

c. Gas swelling at temperatures above 500^0C. The gas swelling effect of metal uranium fuel is caused by gaseous fission products which are preferentially accumulated on the grain boundaries.

Specific volumetric swelling $\Delta V/V$ of metal uranium fuel covers the range of 4-6% per one percent of fuel burn-up. Typical values of maximal fuel burn-up in thermal reactors are equal to 4-5% HM. Therefore, initial porosity of metal uranium fuel must be equal to ~25% for neutralization of the swelling effect. In fast reactors maximal fuel burn-up can reach 10% HM. So, initial porosity of metal uranium fuel must be increased up to ~50% for the same purpose. It seems unreasonable to deal with so porous fuel. The high-density advantage of metal uranium fuel practically disappears.

Uranium dioxide is superior to metal uranium in specific swelling values. Specific swelling of uranium dioxide is about 1,4-1,5% $\Delta V/V$ only per one percent of fuel burn-up. At 10% fuel burn-up in fast reactors initial porosity of uranium dioxide fuel can be below 15%. For comparison, specific swellings of uranium nitride and uranium carbide are equal to 1,5-1,6% $\Delta V/V$ and 1,7-1,8% $\Delta V/V$ per one percent of fuel burn-up, respectively, i.e. only slightly larger than that of uranium dioxide.

A lot of experimental studies have been carried out to eliminate this shortcoming of metal uranium by its alloying. Some promising results were obtained with the alloying components such as molybdenum, zirconium, silicon, iron, aluminum and fissium (imitator of FP composition). Metal uranium alloying with molybdenum and zirconium (up to 10%) allowed it to upgrade corrosion resistance in water and stability of the grain sizes for temperatures up to 600^0C.

Metal uranium is produced in reaction of uranium tetra-fluoride UF_4 with high-purity metals (calcium or magnesium):

$$UF_4 + 2\ Mg \rightarrow 2\ MgF_2 + U.$$

Magnesium fluoride as a light slag is easily removed from surface of a metal uranium ingot.

The next step is a vacuum melting of the uranium ingot for removal of volatile impurities and introduction of the alloying components,

which are able to enhance radiation-resistance of metal uranium fuel, and, finally, casting of uranium rods.

Some data on physical properties of uranium-based fuels (density, melting temperature, heat conductivity and volumetric swelling) are presented in Table 1.5.

Table 1.5

Physical properties of uranium-based fuels

Fuel type	Density, g/cm^3	Melting temperature, ^oC	Heat conductivity, W/m·K	Swelling ΔV/V, %
Metal	18,67	1130	28 (27^{o}C) 44 (727^{o}C)	4-6
UO_2	10,96	2780	8,4 (45^{o}C) 2,4 (1327^{o}C)	1,4-1,5
UC	13,63	2350	32,7 (45^{o}C) 7,3 (500^{o}C)	1,7-1,8
UN	14,32	2650	16 (200^{o}C) 21 (800^{o}C)	1,5-1,6

1.3.2. Fabrication of fuel rods and fuel assemblies

The following requirements must be satisfied by the manufacturing technologies of fuel rods and fuel assemblies:

1. Designs of fuel rods and fuel assemblies, physical properties of fuel and structural materials must be able to ensure long-term mechanical strength, stability of forms and sizes during a reactor lifetime.
2. Materials of fuel rods (fuel meat, cladding, fuel-cladding gap) must be chemically compatible and mutually stable, i.e. any fuel-cladding interactions that can cause radiation embrittlement and plasticity loss must be excluded.
3. The cladding materials must be insoluble and corrosion-resistant in cladding-coolant interactions.
4. Structural materials of fuel rods and fuel assemblies must be sufficiently weak neutron absorbers (minimal cross-sections of neutron radiative capture).

5. Designs and the manufacturing technologies of fuel rods and fuel assemblies must exclude any possibilities of local overheating. This exclusion can be achieved by undertaking the following countermeasures:
a. Uniform distribution of fissile isotopes in fuel rods.
b. Availability of the contact interlayer (fuel-cladding gap) filled up with helium or sodium for intensification of heat removal processes and for prevention of fuel-cladding interactions.
c. Strict spatial separation of fuel rods from each other by special spacers with proper accounting for potential shortening of the inter-rod gap in the process of the reactor operation.
6. Designs and the manufacturing technologies of fuel rods and fuel assemblies must be sufficiently simple for their mass production.
7. Designs, the manufacturing technologies and selection of materials for fuel rods and fuel assemblies must take into account a feasibility of sufficiently simple dismantling procedures for spent fuel reprocessing.

The manufacturing process of UO_2-based fuel rods and fuel assemblies includes the following stages:
1. Preparation of nuclear fuel (conversion of uranium hexafluoride into uranium dioxide powder, granulation and sintering of fuel pellets).
2. Preparation of fuel cladding (flaw detection and quality control).
3. Preparation of the completing details for mounting of fuel assemblies (wrappers, end caps, spacers).
4. Manufacturing of fuel rods: insertion of fuel pellets into tubular claddings, installation of end caps, filling up with helium (as a fuel-cladding gap and a flaw detector thanks to high permeability of helium), sealing of fuel rods by welding, quality control.

Recently, the Russian R&D Institute of Atomic Reactors in Dimitrovgrad has developed the fuel rod manufacturing technology that constitutes an alternative to the technology based on fabrication of fuel pellets. UOX- or MOX-fuel granules are used as the feed material. Some amount (up to 5% HM) of metal natural uranium powder (so called getter) is blended with other fuel granules. Main mission of the getter consists in absorption of oxygen atoms released from fuel particles in fission reactions. As a result, oxygen atoms can not move to the fuel cladding, their corrosion activity is neutralized. The fuel-getter blend is introduced into tubular cladding and packed by vibration. Sufficiently

high fuel density (above 90% of its theoretical value) can be achieved. The experiences gained during pilot usage of the vibration-compacted fuel rods in the research fast reactor BOR-60 demonstrated a high efficiency of such technology. The record values of fuel burn-up were reached (about 32%HM) with specific volumetric swelling at the level of 0,6% ΔV/V per one percent of fuel burn-up. For comparison, metal uranium fuel and UOX-fuel can swell up with the rate of 4-6% ΔV/V and 1,4-1,5% ΔV/V, respectively, per one percent of fuel burn-up.

5. Assemblage of fuel rods into a single fuel assembly, quality control and testing.

The following concluding remarks can be made on the manufacturing technologies of fuel rods and fuel assemblies:

a. The manufacturing technology is a mass production and highly automated process.

b. The manufacturing technology is a high-precision process.

The manufacturing process of RBMK-1000 core requires about 200 thousand completing details, 14 million fuel pellets and 240 thousand welded joints.

All the manufacturing procedures must be put under strict quality control with application of the computer-aided NM control and accountability network. The nuclear fuel fabrication plant is a very significant area for functioning of a reliable and highly effective NM physical protection, control and accountability (MPC&A) system.

In the future, when nuclear fuel cycle will be closed, even fresh nuclear fuel is characterized by intense radioactivity and residual heat generation. This will require applying only the newest remote technologies for manufacturing of fuel rods and fuel assemblies. Accordingly, all components of MPC&A system become more complicated and must be more sophisticated.

1.4. Use of nuclear fuel in nuclear power reactors

1.4.1. Purposes of nuclear reactor refueling

The major NFC stage is an energy utilization of nuclear fuel in nuclear power reactors.

In order to provide sufficiently long operation of nuclear power reactors (up to 18 months as in some advanced LWR), appropriate amount of nuclear fuel must be loaded and properly arranged in the reactor core. Neutron-physical properties of the loaded fuel must ensure large enough reactivity margin to compensate negative reactivity effects caused by depletion of fissile isotopes and build-up of fission products, parasitic neutron absorbers. This means that, before the reactor operation starts up, the reactor is essentially supercritical but the reactivity margin (K_{EFF} - 1) must be suppressed by the regulatory mechanisms: control rods made of natural (or enriched with ^{10}B isotope, very strong neutron absorber) boron carbide, boric acid dissolved in light-water coolant, burnable poisons (gadolinium, erbium) introduced into fuel compositions.

As content of fissile isotopes in fuel decreases and content of fission products (parasitic neutron absorbers) in fuel increases, the moment comes when the reactor can not be longer critical. All control rods are already withdrawn from the reactor core, all boric acid is removed from light-water coolant. Nevertheless, the reactor becomes sub-critical ($K_{EFF} < 1$).

For the reactor operation to continue, NPP operator has to undertake some corrective measures to return the reactor to the supercritical state ($K_{EFF} > 1$). The following actions can be performed:

1. Full or partial substitution of fresh fuel assemblies for irradiated ones.
2. Partial transpositions (shuffling) of irradiated fuel assemblies from one region of the reactor core to another.
3. Any combinations of two aforementioned actions.

A set of these actions undertaken to restore the reactor ability for long-term operation is named the refueling strategy.

Thus, the first and major mission of the refueling is to replenish the reactivity margin and enable the reactor to continue a long enough operation at nominal power level during a certain time interval.

The second, also important but supplementary, mission of the refueling is to ensure as flat as possible spatial shape of heat generation rate. If spatial distribution of heat generation rate in the reactor core is uniform enough, then all fuel assemblies are used under maximal acceptable level of energy production. So, maximal value of total energy out-

put can be obtained from the reactor core. In addition, uniform spatial distribution of heat generation rate ensures uniform fuel burn-up, i.e. all fuel assemblies discharged from the reactor core are characterized by the same (or nearly the same) isotope compositions.

However, these benefits (maximal energy output and identical isotope compositions of spent fuel) can not be obtained in a uniformly fueled reactor. In such reactors heat generation rate is highest in the core center and drops down nearly to zero at the core periphery. The average value of heat generation rate is approximately three times lower than its maximal value in the core center. Non-uniform fuel loading is required to flatten spatial shape of heat generation rate and fuel burn-up. For example, uranium fuel of the lower enrichment may be placed in central region of the reactor core while uranium fuel with relatively higher enrichment may be placed at the core periphery. As a rule, main control rods are also placed in central region of the reactor core. That is why neutron flux and heat generation rate in the core center are lower than those at the core periphery.

Thus, formation of such a fuel loading that is characterized by as flat as possible spatial shape of heat generation rate in the reactor core constitutes a supplementary but very important mission of the refueling strategies.

1.4.2. Strategies of nuclear reactor refueling

There are many various refueling strategies in nuclear power reactors.

1. The simplest refueling strategy presumes a uniform fuel loading with its complete removal and replacement with fresh fuel loading when the reactor criticality can not be longer maintained. This refueling scheme is called **the "batch irradiation"** and not used now because of the following serious drawbacks:

a. Spatial shape of heat generation rate in the reactor core is quite non-uniform with the peaking factor (peak-to-average ratio) above three.

b. Content of fissile isotopes decreases more intensely in the central core region than at the core periphery. So, very uneven fuel burn-up takes place in the uniformly fueled reactor core. After each irradiation

cycle, central fuel assemblies have reached their maximal burn-up while peripheral fuel assemblies have no delivered completely their reactivity and energy potential.

c. Large reactivity margin must be formed in the central core region. Hence, control rods with appropriately large total reactivity worth must be placed in the core center. This can worsen neutron economy during the reactor operation cycle.

2. **The "partial batch"** refueling strategy presumes that only those fuel assemblies, which have reached their maximal acceptable fuel burn-up, must be removed after the irradiation cycle and replaced with fresh fuel assemblies. At the next refueling, fuel assemblies with the highest fuel burn-up are replaced again, and so on. If the reactor core is divided into several concentric layers, then each layer, starting from the innermost layer and proceeding outward, is replaced with fresh fuel in several successive refuelings.

Main advantage of the "partial batch" refueling strategy is a fairly uniform fuel burn-up in each concentric layer. However, central layers are refueled more frequently than peripheral layers because maximal fuel burn-up is reached by central fuel assemblies for a relatively shorter time interval. Hence, spatial shape of heat generation rate can be shifted towards the core center, and the peaking factor increases.

3. **The "scatter refueling"** strategy was developed to solve the high peaking factor problem of the "partial batch" refueling scheme. The "scatter refueling" technology presumes that the reactor core is divided into small local groups containing an equal number of fuel assemblies (four-assembly groups, for instance). At the first refueling, all fuel assemblies labeled 1 are removed and replaced with fresh fuel assemblies. At the second refueling, all fuel assemblies labeled 2 are removed and replaced, and so on. Thus, each fuel assembly is completely utilized for four irradiation cycles. Fresh fuel assemblies are not concentrated in the core center but they are scattered throughout the reactor core as a whole. That is why the peaking factor can be substantially reduced.

4. **The "out-in"** refueling strategy presumes that the reactor core is again divided into several concentric layers containing an equal number of fuel assemblies. At the refueling, only central fuel assemblies have reached their maximal fuel burn-up, and they must be removed from the reactor core. Fuel assemblies from the next outer layer are inserted into

the central (inner) layer, and so on, i.e. fuel assemblies from the outer layer are moved to the neighboring inner layer in the common direction from the core periphery to the core center. Fresh fuel assemblies are loaded into the outermost layer. At the next refuelings, the same operations of central layer removal, inward movement of partially spent fuel assemblies and placement of fresh fuel assemblies into the outermost layer are repeated.

According to the “out-in” refueling strategy, fresh fuel assemblies with the highest reactivity potential are loaded into the core periphery. So, spatial shape of heat generation rate is depressed in the core center, and the peaking factor can increase.

5. **The “modified scatter”** refueling strategy is a combination of the “scatter” and “out-in” refueling schemes. The reactor core is divided into an outermost layer containing one-fifth part of total fuel assemblies, and the inner zone containing four-fifths of total fuel assemblies. The inner zone is subdivided into small local groups, like in the “scatter refueling” strategy (four-assembly groups). At the first refueling, fuel assemblies with the highest fuel burn-up are removed from each four-assembly group and replaced with fuel assemblies from the outermost layer. Thus, the outermost layer is emptied and filled up with fresh fuel assemblies. The “modified scatter” scheme is characterized by the flattened spatial shape of heat generation rate in central core region without high peaking factors in the “scatter refueling” strategy and without the central depression of heat generation rate in the “out-in” refueling strategy.

6. **The “uniformly partial”** refueling strategy is based on the following assumptions. Let spatial shape of heat generation rate be flat enough in the reactor core. It means that maximal values of fuel burn-up can be reached by all fuel assemblies simultaneously. The following case can be considered as an example. The time interval needed to reach maximal fuel burn-up equals 3 years, and the refueling is performed once a year. Then, the “uniformly partial” refueling strategy consists of the following steps:

a. At the first refueling, one-third fraction of fuel assemblies is replaced with fresh fuel assemblies, i.e. the discharged fuel has reached 33% of acceptable fuel burn-up.

b. At the second refueling, one-third fraction of fuel assemblies is again replaced with fresh fuel assemblies, i.e. the discharged fuel has reached 67% of acceptable fuel burn-up.

c. At the third refueling, one-third fraction of fuel assemblies is again replaced with fresh fuel assemblies, i.e. the discharged fuel has reached 100% of acceptable fuel burn-up.

Beginning from the fourth refueling, an equilibrium refueling regime has been established. The equilibrium regime is characterized by quite similar compositions of the reactor core at the beginning and at the end of irradiation cycle:

a. At the beginning of each irradiation cycle, the reactor core contains one-third fraction of fresh fuel assemblies, one-third fraction of fuel assemblies with 33% of acceptable fuel burn-up and one-third fraction of fuel assemblies with 67% of acceptable fuel burn-up.

b. At the end of each irradiation cycle, the reactor core contains one-third fraction of fuel assemblies with 33% of acceptable fuel burn-up (the former fresh fuel assemblies), one-third fraction of fuel assemblies with 67% of acceptable fuel burn-up (the former 33%-fuel assemblies) and one-third fraction of fuel assemblies with 100% of acceptable fuel burn-up (the former 67%-fuel assemblies).

Main advantage of the “uniformly partial” refueling strategy is the same number of the discharged fuel assemblies with maximal acceptable fuel burn-up. Main drawback of the “uniformly partial” refueling strategy is a removal of only partially burnt up fuel assemblies at the first and second refuelings (at initial stage of the reactor operation, in general).

Unfortunately, real spatial shape of heat generation rate is not so flat that fuel assemblies in different core regions could reach maximal fuel burn-up for the same time interval. Under these conditions, the reactor core can be subdivided into several concentric layers, within each of them spatial shape of heat generation rate can be regarded as a flat enough. Then, basic ideology of the “uniformly partial” refueling strategy can be applied to each concentric layer separately.

1.4.3. Technologies of nuclear reactor refueling

All the refueling strategies listed above must be supplied with appropriate technological tools for conduction of the refueling operations. In principle, the reactor refueling can be performed:

a. after the reactor shutdown, cooldown, depressurization and removal of the reactor head;

b. after the reactor shutdown but without cooldown and removal of the reactor head;

c. at the reduced or full power level, i.e. without the reactor shutdown, cooldown, depressurization and removal of the reactor head.

In practice, light-water reactors are usually refueled only with application of the first scheme, i.e. after the reactor shutdown, cooldown, depressurization and removal of the reactor head. Once a year (or 18 months) the reactor is shutdown for 4-6 weeks, the reactor head is removed, some spent fuel assemblies are transferred to the fuel storage pool, the remaining fuel assemblies are reshuffled, and fresh fuel assemblies are introduced into the reactor core. All the refueling operations are performed under sufficiently thick water layer.

In contrast to LWR, refueling of liquid-metal fast breeder reactors (LMFBR) is done without removing the head of the reactor vessel. There are three areas involved into the refueling process: the reactor vessel, the fuel transfer chamber (FTC) and the ex-vessel storage tank (EVST). An in-vessel transfer machine (IVTM) can transfer fuel assemblies inside the reactor vessel only. Fuel assemblies are transferred between the reactor vessel and the EVST in a transfer bucket by means of a special hoist. The transfer ports are located between the reactor vessel and the FTC and between the FTC and the EVST. Fuel assemblies remain under sodium throughout the fuel transfer process.

Consider the replacement of spent fuel assembly with fresh fuel assembly starting with fresh fuel assembly. The fresh fuel assembly is lifted out by the EVST handling arm and placed into the transfer bucket. The transfer bucket has a space for one fresh fuel assembly and for one spent fuel assembly. At this step the transfer bucket contains only one fresh fuel assembly, in a vertical position. The transfer bucket is hoisted at an angle through the first fuel transfer port and guided by tracks up into the FTC. The FTC is then guided into the reactor vessel and down through the second fuel transfer port and placed in a vertical position in the region outside the reactor core.

The IVTM is moved to the position directly above the spent fuel assembly to be replaced, and the assembly is grappled by the IVTM manipulator. The spent fuel assembly is raised above the remaining assemblies and transferred through the sodium pool to the open space in the transfer bucket, into which it is then lowered. The fresh fuel assembly is next withdrawn from the transfer bucket by the IVTM manipulator and transferred to the position in the reactor core from which the spent fuel assembly was just removed. The spent fuel assembly is then transferred through the FTC to the EVST, and the refueling process is ready to be repeated for the next fuel assemblies.

Contemporary LMFBR designs use a rotating plug concept, in which several (three, as a rule) rotating plugs are located in the reactor head, and the IVTM is mounted on the smallest plug. The largest plug is concentric with the reactor vessel while the smaller plugs are eccentric ones. Each plug can rotate independently so that the IVTM manipulator can be placed in any position above any fuel assembly inside the reactor vessel.

Heavy-water CANDU-type reactors have a distinct advantage over LWR of the same power due to their on-line, continuous refuelings. Natural uranium is used here as a fuel material thus elimination a need for uranium enriching services but excluding the use of light water as a coolant and neutron moderator material because of high neutron absorption cross-sections. Heavy water is substituted for light water. Annually, about one ton of expensive heavy water is needed per one megawatt of electrical output.

Fuel consists of 0,5-m-long fuel bundles inserted into horizontal pressure tubes that run through a thin-walled tank (calandria) filled up with heavy-water moderator. Each fuel channel contains twelve fuel bundles. The refueling process is done on a daily basis. Two refueling machines are connected to a fuel channel, one on each side of the horizontally placed reactor. Each refueling machine is equipped with a barrel that attaches onto a fuel channel, unlocks the end plug, removes and replaces it by itself. Up to twelve fuel bundles (one fuel channel) can be inserted or removed during one visit of the refueling machines to a fuel channel. One refueling machine inserts fresh fuel bundles while another refueling machine, at the opposite side, receives spent fuel bundles as they are ejected into its barrel. The fuel motion takes pun the direction

of coolant flow which alternates between adjacent fuel channels. So, the refueling process in two adjacent channels is done in two mutually opposite directions. The insertion of fresh fuel bundles into peripheral regions of the reactor core from two opposite sides can upgrade heat generation rate at the reactor periphery and, thus, flatten spatial distribution of heat generation rate.

Russian RBMK-type reactors (light-water cooled, graphite moderated reactors, LWGR) can be also refueled in a continuous on-line manner, like CANDU-type reactors because both reactor types are channel reactors that made it possible to arrange refueling of any fuel channel individually. A dedicated loading-unloading machine (LUM) containing one fresh fuel assembly and space for disposition of one spent fuel assembly can do the following refueling operations:

1. The LUM filled up with the warm condensate (30^0C) attaches onto the fuel channel to be refueled.
2. Pressure in the fuel channel and pressure in the LUM cask used for disposition of spent fuel assembly are equalized (~75 atmospheres).
3. The fuel channel and the LUM form a single circuit. The warm condensate is pumped into the circuit.
4. Spent fuel assembly is grappled by the LUM manipulator and withdrawn from the fuel channel.
5. Passability of the fuel channel is checked up with a fuel assembly imitator.
6. Fresh fuel assembly is inserted into the fuel channel.
7. The fuel channel is locked, pressure in the LUM decreases to the ambient level, the LUM and the fuel channel are disconnected.

It is evident that those nuclear power reactors which can be refueled continuously, without the reactor shutdown for several weeks, i.e. CANDU and RBMK reactors represent a particular threat to nuclear non-proliferation regime because of the following reasons:

1. Operators of CANDU and RBMK reactors are able, in principle, to conduct relatively short-term (two-three months) irradiation of uranium fuel assemblies for unauthorized build-up of weapon-grade plutonium.
2. To prevent the unauthorized short-term irradiations of uranium fuel assemblies, a continuous (not periodical) presence of the IAEA inspectors is required at the operating CANDU and RBMK reactors.

3. All the MPC&A-related measures are more difficult for undertaking at the reactors with continuous refueling operation mode.

Spent fuel assemblies are intense radiation and decay heat sources. That is why all NPP reactors are provided with spent fuel water pools where spent fuel assemblies are stored until their radioactivity and residual heat generation rate drop below the acceptable levels for transportation, reprocessing or ultimate disposal.

The spent fuel storage pools must be equipped with the following auxiliary systems:

1. Residual heat removal system.
2. Ion-exchange installation for water purification and removal of solid radioactive particles.
3. Ventilation installation for air purification and retention of gaseous radioactive materials by special super-filters.

Interim storage of LWR spent fuel assemblies in NPP water pools can last up to 10 years. The same storage time is chosen for LMFBR spent fuel assemblies. Previously, it was thought that the backend part of the closed NFC including LMFBR spent fuel reprocessing and plutonium recycle must be as short as possible (6-12 months as a target value) to supply the developing nuclear power industry with plutonium-based fuel. Nowadays, there are no imperative reasons for the world-wide deployment of LMFBR-based NPP. So, the same storage time was adopted for spent fuel assemblies discharged from LWR- and LMFBR-type reactors.

Upon the expiry of the interim storage time, spent fuel assemblies can be transported to the deep underground repositories for ultimate disposal (the open NFC option) or to the spent fuel reprocessing plants for plutonium recovery and energy utilization (the closed NFC option). Relatively long interim storage time and weak development of nuclear technologies intended for ultimate disposal or radiochemical reprocessing of spent fuel assemblies resulted in gradual exhaustion of the water pools capacity. To neutralize these negative effects, the following countermeasures are undertaken:

1. The tighter positioning of spent fuel assemblies inside the water pools under the stricter nuclear safety control.
2. Partitioning of the water pools by metal structures containing strong neutron absorbers (boron, for instance).

3. Build-up of the centralized large spent fuel storages.

1.4.4. Transportation of spent nuclear fuel

Transportation is a necessary link between all NFC stages, and is especially significant for transportation of spent fuel assemblies. Spent fuel assemblies may be shipped by all transport means (trucks, railroad, river boats or sea-going ships) except of aircrafts. According to the RF regulations, all shipments of nuclear materials with specific radioactivity above 2 μCi/kg are regarded as radiation shipments. Specific radioactivity of spent fuel equals about 1 MCi/kg.

The spent fuel transport casks can weigh about 100 tons. Total weight of spent fuel assemblies in the transport casks is about 2-5% from total weight of the transport cask. The remaining 95-98% of total weight belongs to the transportation safety systems.

A typical spent fuel transport cask looks as follows:

1. Large hollow thick-walled cylinder in a vertical or horizontal (preferentially) position (1,5-2 m in diameter, 4-6 m in length, wall is about 40 cm thick) made of steel, cast iron or concrete.
2. Outer surface of the transport cask is covered by special fins for extension of the heat removal area (~30 m^2). The finned outer surface extends the heat removal area approximately twice.
3. Inner surface of the transport cask is lined with stainless steel to enhance corrosion-resistance. The inner liners can include some layers of neutron moderators and neutron absorbers (borated polyethylene, for instance).
4. Metal shelves for disposition of spent fuel assemblies are placed in the inner cavity of the transport cask. During shipment, the inner cavity is filled up with coolant. Decay heat is removed from spent fuel assemblies either by natural convection or forced circulation depending on the value of heat generation rate.
5. The transport casks are hermetized with application of the reinforced densifiers.
6. The transport casks are equipped with control systems for permanent monitoring of the inner cavity parameters (radioactivity, decay heat generation rate, temperature and pressure of coolant) and with accidental decontamination system.

The following requirements must be satisfied by designers of the spent fuel transport casks:

1. Reliable radiation protection of the staff involved, population and the environment against neutron and gamma-emissions (metal vessel containing high-efficiency neutron moderators and neutron absorbers).
2. Reliable nuclear safety ensuring (metal shelves containing strong neutron absorbers, limitations on the number of spent fuel assemblies to be loaded into the transport casks).
3. Reliable removal of decay heat (the finned outer surface, forced circulation of coolant in the inner cavity of the transport cask).
4. Reliable hermetization of the transport casks even under severe accidental conditions.

To check up hermeticity of the transport casks, they must undergo the following severe tests:

a. Drop test from 9-m height onto a steel plate.
b. Puncture test from 1-m height onto a vertical metal rod (15 cm in diameter).
c. Immersion test in light water (depth - 15 m, duration – 8 hours).
d. Fire test – staying in flame at 800^0C for 30 minutes plus 2-hour staying without a forced cooldown.

The IAEA has elaborated the following requirements to thermal parameters of the transport casks in operation:

a. Temperature of the cask surface must be below 82^0C at the ambient air temperature of 38^0C.
b. Internal coolant pressure in the cask cavity must be below 7 atmospheres.
c. The outer surface of the cask must be extended by fins. Additional 30 m^2 of the heat removal area would be large enough for safe shipment of spent fuel assemblies with total power from 25 to 30 kW (~30 spent fuel assemblies from VVER-440 after 3-year cooling period) without a forced cooldown.

1.5. Radiochemical reprocessing of spent nuclear fuel

The following aims are pursued by technologies intended for spent nuclear fuel (SNF) reprocessing:

1. Recovery of accumulated plutonium and residual uranium for the repeated use (recycle) as fissile and fertile materials.
2. Separation of fission products and transuranium elements for further treatment as radioactive wastes.

The IAEA has worked out the following recommendations on the reprocessing quality of spent fuel assemblies discharged from power LWR after the standard operation cycle (fuel burn-up - 33 GWd/t, the cooling time – 10 years):

1. Extent of plutonium and uranium recovery – above 99,9%.
2. Decontamination factors, i.e. ratios of an impurity content before treatment to an impurity content after treatment:
   a. Uranium from plutonium – at the level of 10^7.
   b. Uranium from fission products – at the level of 10^7.
   c. Plutonium from uranium – at the level of 10^6.
   d. Plutonium from fission products – at the kevel of 10^8.

Such a fine purification gave a foundation to call this approach as the “clean fuel – dirty waste” concept. The concept is very attractive for NFC closure but it can produce some difficulties for nuclear non-proliferation regime. Therefore, when some advanced SNF reprocessing technologies have been developed, which deliberately left some remarkable quantity of radioactive fission products in the recycled fuel to enhance nuclear non-proliferation regime, a new approach arose, namely the “dirty fuel – clean waste” concept.

Nearly 7000 t SNF are discharged annually from the world NPP. Capabilities of the existing and under construction facilities for SNF reprocessing are shown in Table 1.6. As is seen, the existing SNF reprocessing plants are not able to deal with full annual SNF amount discharged from all NPP in operation throughout the world.

SNF reprocessing plant is a rather expensive enterprise. Approximate specific cost of SNF reprocessing is evaluated as ~500 US dollars/kg SNF.

Table 1.6

Capabilities of SNF reprocessing plants

Country	Annual throughput, t SNF
Great Britain	1500 1200
France	900 800
Russia	400
India	2 × 100
Japan	90
China	50
Under construction	
Russia	1500
Japan	800

Total: ~ 5100 t/year in operation, 2300 t/year under construction.

1.5.1. Classification of SNF reprocessing technologies

1. Aqueous (wet) reprocessing technologies:

a. Solvent-extraction processes which are based on selective recovery of uranium and plutonium compounds from fuel-containing solutions by organic extractants.

b. Precipitation processes which are based on the formation of insoluble uranium and plutonium compounds with their further deposition on a vessel bottom by introducing appropriate precipitants.

2. Non-aqueous (dry) technologies:

a. Pyrochemical processes: for example, the fluoride volatility process that is based on different volatility and sorption of uranium, plutonium and FP fluorides.

b. Pyrometallurgical processes: for example, the electrochemical refinement process that is based on different transport properties of uranium, plutonium and fission products in molten salts.

The most widely used and industrially matured technologies are the aqueous solvent-extraction processes. The PUREX (Plutonium and Uranium Recovery by Extraction) technology is the most representative example of these processes.

1.5.2. Main stages of aqueous solvent-extraction technology PUREX

Main stages of the PUREX-process

1. Dismantling of spent fuel assemblies and chopping of spent fuel rods.
2. Preliminary oxidation (voloxidation) of spent fuel.
3. Dissolution of spent fuel and preparation of the fuel solution for uranium and plutonium extraction.
4. The extraction – re-extraction cycles.

These stages of the PUREX-process are described in more details below.

Dismantling of fuel assemblies and chopping of fuel rods. Thus, spent UOX-fuel assemblies, after lengthy cooldown period, were shipped to radiochemical plant for reprocessing. At first, the following initial procedures must be carried out with spent fuel assemblies:

1. Removal of end caps, wrappers and spacers, dismantling of fuel lattices.
2. Shearing operation that chops long fuel rods into short (2,5-5 cm) pieces.

Dismantling of fuel assemblies and chopping of fuel rods must be done in the closed hot cells with inert atmosphere (nitrogen or argon).

Preliminary SNF oxidation (voloxidation). The next stage of the PUREX-process consists in preliminary oxidation of spent fuel pieces by oxygen as elevated temperatures (~600^0C). Uranium dioxide UO_2 converts into uranium octa-oxide U_3O_8:

$$3\,UO_2 + O_2 \rightarrow U_3O_8.$$

The UO_2–to-U_3O_8 conversion leads to the following positive effects:

a. Fuel density decreases on ~30% due to so different densities of initial and final reagents: $\gamma(UO_2) \approx 11$ g/cm^3, $\gamma(U_3O_8) \approx 8{,}3$ g/cm^3. As consequences, fuel volume increases according to this difference, fuel meat becomes more porous and friable than can facilitate the further dissolution of fuel pieces.

b. Fuel crystalline lattice undergoes substantial changes.

c. Both these effects create the favorite conditions for intense release of tritium, gaseous and volatile fission products. If the voloxidation tem-

perature is kept at 650^0C for 12 hours, then up to 99,96% tritium, 70% ^{85}Kr, 40% ^{129}I and 90% ^{106}Ru can escape the fuel pieces.

Dissolution of fuel pieces. Spent UOX-fuel pieces are dissolved by boiling (t ~ 100^0C) nitric acid HNO_3 concentrated up to 6-12 M for 4-6 hours:

$$UO_2 + HNO_3 \rightarrow UO_2 (NO_3)_2 + NO_X + H_2O.$$

Simultaneously, nitric acid can be recombined in the dissolver where air or oxygen flow is pumped to intensify the regeneration process:

$$NO_X + H_2O + O_2 \rightarrow HNO_3.$$

The use of nitrogen oxides for nitric acid regeneration promotes forming a smoke-free process, i.e. without any release of gaseous nitrogen oxides into the environment.

In the process of SNF dissolving, metal claddings (zirconium-based alloys or stainless steels) of fuel rods remain non-dissolved and can be easily removed for further treatment as solid radioactive wastes.

Preparation of SNF solution to the extraction – re-extraction process consists of the following steps:

1. Clarification of SNF solution:

a. Filtration for removal of small (~3 microns) solid particles through metal-ceramic filters or porous polypropylene with application of some coagulants for enlarging the particles.

b. Centrifugation with application of some coagulants for removal of the smaller (~1 micron) solid particles.

2. Removal of some gaseous and volatile fission products from SNF solution:

a. Barbotage of SNF solution by air flow for removal of radioactive iodine ions I^- and IO_3^-. Then, the Barbotage off-gases are pumped through the filters impregnated with silver nitrate $AgNO_3$:

$$3 I_2 + 6 AgNO_3 + 3 H_2O \rightarrow 5 AgI + AgIO_3 + 6 HNO_3.$$

b. Barbotage of SNF solution by ozone flow for removal of radioactive ruthenium ions Ru^{4+}:

$$Ru^{4+} + 2\ O_3 + 2\ H_2O \rightarrow RuO_4 + 2\ O_2 + 2\ H_2.$$

Ruthenium oxide RuO_4 is then removed from the barbotage off-gases by reaction with sodium hydroxide NaOH.

c. Removal of inert gases (Kr, Xe) by the barbotage process with further their sorption in zeolite or activated charcoal at reduced temperatures. Then, the barbotage off-gases are diluted with air flow down to the acceptable concentrations and released into the environment.

3. Correction of SNF solution acidity by water dilution or evaporation to reach the level of 2-4 M HNO_3.

Extraction. The solvent-extraction technology of SNF reprocessing is very similar to the extraction affinage technology which was applied at the NFC front-end for removal of neutron-absorbing impurities from natural uranium concentrate. The extraction affinage process presumes dissolving uranium concentrate U_3O_8 by nitric acid, recovery of uranyl-nitrate $UO_2(NO_3)_2$ by organic extractant tri-butyl-phosphate (TBP), re-extraction of uranyl-nitrate from organic fraction by hydrogen peroxide H_2O_2 or ammonium bicarbonate NH_4HCO_3 and precipitation of insoluble uranium compounds.

The following significant distinctions can be seen between the extraction affinage of natural uranium concentrate and the solvent-extraction reprocessing of spent UOX-fuel:

1. Acidic SNF solution contains residual amount of enriched uranium, reactor-grade plutonium, fission products and minor actinides where FP and MA play a very similar role that was played by neutron-absorbing impurities in the extraction affinage process, i.e. they must be removed.
2. The extraction affinage process dealt with weakly radioactive materials (natural uranium and impurities) while the solvent-extraction SNF reprocessing technology deals with FP and MA, intense radiation and heat sources.

The solvent extraction process, like the extraction affinage, applies the same organic extractant TBP in mixture with an inert organic dilutant for reduction of TBP viscosity. Density of the “TBP-dilutant” mixture covers the range of 0,8-0,9 g/cm^3 while density of acidic SNF solution covers the slightly heavier range of 1,1-1,2 g/cm^3.

Thus, the solvent-extraction process is a separation of uranyl-nitrate between two immiscible fractions, namely the light organic fraction

(TBP plus dilutant) and the heavy aqueous fraction (acidic SNF solution).

Layout of the extraction – re-extraction process. The extraction process takes place in two connected vessels, mixer and settler. Acidic SNF solution and extractant TBP are pumped into the mixer. Their intense mixing leads to tight SNF-TBP contacts, uranyl and plutonyl nitrates receive an opportunity to form stable solvates with TBP molecules. Then, plutonium and uranium accumulate in the light organic fraction while fission products and minor actinides accumulate in the heavy aqueous fraction. Afterwards, the mixture is pumped into the settler where the light organic fraction (extract) and the heavy aqueous fraction (raffinate) can be easily separated. The extraction process is over.

The-re-extraction process takes place also in two connected vessels, mixer and settler. The extract and the aqueous washing solution are pumped into the mixer. Their intense mixing leads to the uranium and plutonium compounds are washed off the extract. Then, the mixture is pumped into the settler where the light organic fraction (extractant) and the heavy aqueous fraction (re-extract) can be easily separated. The extractant can be used again in the extraction process, after purification and regeneration procedures. The re-extraction process is over.

Thus, uranium and plutonium compounds are partially separated from fission products and minor actinides. Multiple application of the extraction – re-extraction process can separate them completely. General layout of the extraction – re-extraction process is shown in Fig. 1.6.

The coefficient D (Distribution Ratio) characterizes distribution of elemental concentrations C between the light organic fraction and the heavy aqueous fraction:

$$D = \frac{C_{element}(\text{organic fraction})}{C_{element}(\text{aqueous fraction})}.$$

Evidently, if the distribution ratio of a certain element $D > 1$, then this element concentrates in the light organic fraction, and, vice versa, if $D < 1$, then the element concentrates in the heavy aqueous fraction.

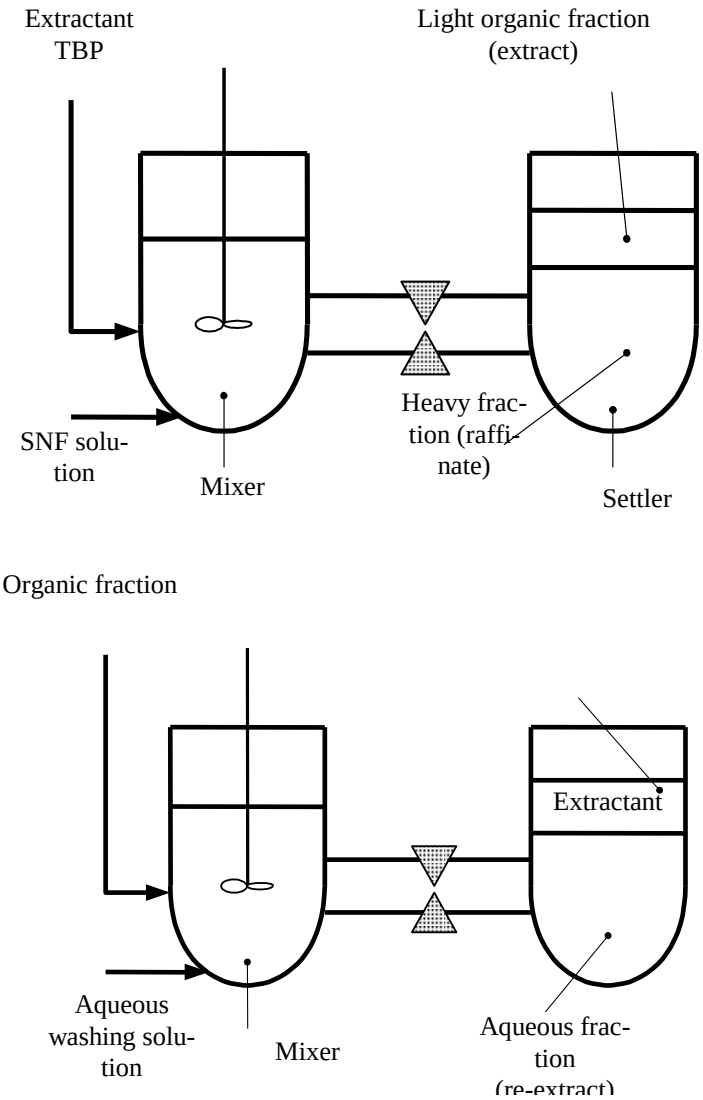


Fig. 1.6. The extraction – re-extraction process

The well-known quadratic dependencies of the distribution ratios on the SNF solution acidity are shown in Fig. 1.7 for uranium (a typical

representative of fuel materials) and for zirconium (a typical representative of fission products).

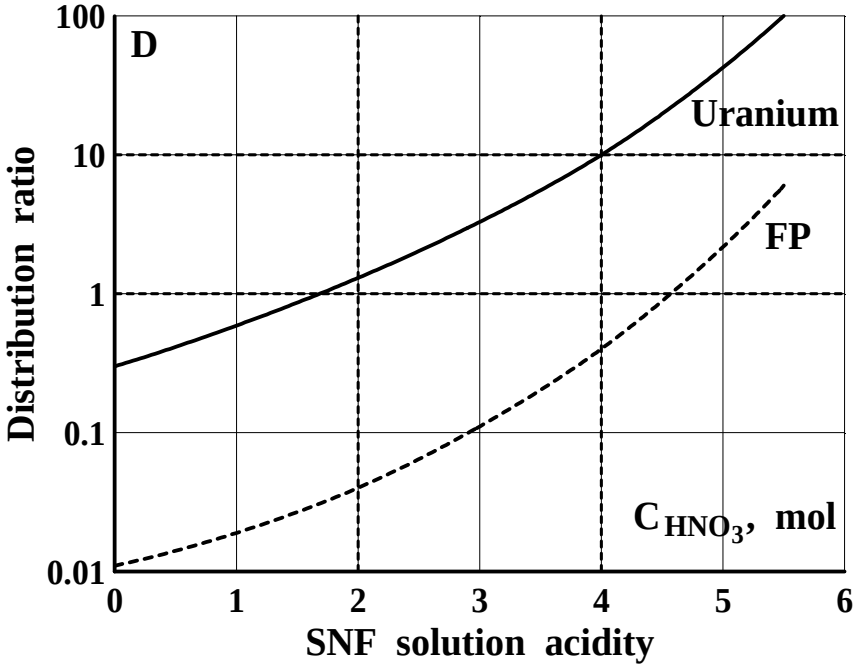


Fig. 1.7. Dependencies of the distribution ratios on the SNF solution acidity

As it is seen, if nitric acid concentration C(HNO_3) belongs to the acidity range from 2 M to 4 M, then the uranium distribution ratio is larger than unity, and fuel materials seek to concentrate in the light organic fraction. Within this acidity range the zirconium distribution ratio is lower than unity, and fission products seek to concentrate in the heavy aqueous fraction. This consideration explains why the SNF solution acidity was corrected to be within the 2-4 M range at the preparatory stage, before the extraction – re-extraction process started. In addition, these dependencies of the distribution ratios on the SNF solution acidity can explain the preferential accumulation of fission products in the aqueous washing solution during the re-extraction process. In the

latter case, the uranium distribution ratio D(U) ≈0,1 at C(HNO_3) near to zero, and uranium seeks to concentrate in the aqueous washing solution.

Let assume that D(U) = 9 and 100 g U in the acidic SNF solution come to the extraction – re-extraction process. Then, after the first cycle, 90 g U were recovered while 10 g U remained in the aqueous fraction. After the second cycle, additional 9 g U were recovered while 1 g U remained in the aqueous fraction. So, two-three cycles of the extraction – re-extraction process are able to recover up to 99-99,9% U from the initial SNF solution.

TBP radiolysis. The most substantial disadvantage of the solvent-extraction technology consists in intense radiolysis of organic extractants under ionizing irradiation. The higher radioactivity of the acidic SNF solution, the more intense chemical dissociation of organic extractants occurs. Specific radioactivity of the SNF solutions can reach the level of 500 Ci/l for spent fuel discharged from thermal reactors and the level of 1000 Ci/l for spent fuel discharged from fast reactors. That is why reprocessing of spent fuel with high values of fuel burn-up is a very complicated technology.

Radiolysis of tri-butyl-phosphate can cause the following negative effects:

1. Chemical dissociation of TBP molecules occurs according to the scheme with breaking C_4H_9-O links:

$$\begin{matrix} C_4H_9-O \\ C_4H_9-O-P=O \\ C_4H_9-O \end{matrix} \rightarrow \begin{matrix} C_4H_9-O \\ C_4H_9-O-P=O \\ H-O \end{matrix} \rightarrow \begin{matrix} C_4H_9-O \\ H-O-P=O \\ H-O \end{matrix} \rightarrow H_3PO_4$$

i.e. TBP as an organic salt of phosphoric acid transforms, at first, into di-butyl-phosphoric (DBP) acid, then – into mono-butyl-phosphoric (MBP) acid, and, finally, into phosphoric acid H_3PO_4: TBP → H(DBP) → H_2(MBP) → H_3PO_4.

2. Appearance of chemically active acids results in forming salts of DBP-, MBP and phosphoric acids containing fission products as metal components. These FP-containing organic salts can concentrate in the light organic fraction and, thus, worsen the SNF reprocessing quality.

3. TBP radiolysis creates the conditions needed to form the third fraction on the interface between the light organic and heavy aqueous frac-

tions. The third fraction includes some jelly-like insoluble (or ill-soluble) materials which can block technological pipelines at SNF re-processing plant. The following materials can be components of the third fraction:

a. Compounds of fissile isotopes with TBP radiolysis products.

b. Compounds of fission products with TBP radiolysis products.

c. Products of radiation-induced polymerization of TBP dilutant in the form of stable jelly-like emulsions.

TBP regeneration. When TBP contacts with the SNF solution, salts of DBP-, MBP- and phosphoric acids can appear and contain metal fissile materials and fission products. The contaminated TBP can be cleaned with application of the carbonate-alkaline washing-out process. Usually, mixture of soda Na_2CO_3 with caustic soda NaOH is applied as a washing-out solution. Sodium, as the more chemically active element, substitutes itself for all other metal components in salts of DBP-, MBP- and phosphoric acids. The sodium-based salts are well-soluble by water, and they can be easily removed by aqueous washing-out solutions.

The carbonate-alkaline washing-out regeneration process has the following drawbacks:

1. Large volume of middle-level radioactive wastes.
2. Residual plutonium in TBP can concentrate in the washing-out solution, undergo radiation-induced polymerization and fall out as sediment.
3. The process is not able to reach complete TBP purification.

Separation of plutonium from uranium. The uranium-plutonium mixture produced by the solvent-extraction technology contains the following uranium-TBP and plutonium-TBP solvates:

one six-valent uranium-TBP solvate $UO_2(NO_3)_2 \cdot 2TBP$;

three plutonium-TBP solvates with different plutonium valencies:

trivalent $Pu(NO_3)_3 \cdot 3$ TBP, four-valent $Pu(NO_3)_4 \cdot 2$ TBP and six-valent $PuO_2(NO_3)_2$ 2 TBP.

Separation of uranium-plutonium mixture is based on experimental fact that trivalent plutonium-TBP solvate $Pu(NO_3)_3$ 3 TBP is characterized by its minimal solubility in the light organic fraction as compared with solubilities of other uranium and plutonium solvates. Therefore, if all plutonium-TBP solvates are converted into trivalent state by the aqueous reducing solution, then trivalent plutonium-TBP solvate can

concentrate in this solution while uranium-TBP solvate remains in the organic fraction.

When separating plutonium from uranium, six-valent plutonium-TBP solvate is reduced, at first, up to four-valent state, then – up to trivalent state and washed out of the organic fraction.

Six-valent plutonium solvate can be reduced up to four-valent state by reaction with potassium nitrite KNO_2:

$$PuO_2(NO_3)_2 + KNO_2 \rightarrow Pu(NO_3)_4 + KNO_3.$$

Afterwards, four-valent plutonium solvate is reduced up to trivalent state by means of the following methods:

1. Reactions with bivalent iron compounds:

$$Pu^{4+} + Fe^{2+} \rightarrow Pu^{3+} + Fe^{3+}.$$

Iron gives one valent electron to plutonium.

2. Reactions with four-valent uranium compounds:

$$Pu^{4+} + U^{4+} + 2\ H_2O \rightarrow Pu^{3+} + UO_2^{2+} + 2\ H_2.$$

3. Electrochemical plutonium reduction.

If the light organic fraction is washed out by the aqueous reducing solution, then trivalent plutonium concentrates in the aqueous fraction while uranium remains in the organic fraction. Afterwards, the re-extraction process is used to recover uranium from the organic fraction by low-concentrated nitric acid HNO_3. Uranium transfers into the aqueous fraction (re-extract). This is a final stage of the extraction – re-extraction process. Thus, one cycle of this process consists of the following stages:

1. SNF dissolution by nitric acid.
2. Extraction of uranium and plutonium from the acidic SNF solution by organic extractant TBP. Uranium and plutonium are jointly separated from fission products.
3. Re-extraction of plutonium from the organic fraction by the aqueous reducing solution. Six- and four-valent plutonium solvates are reduced

up to trivalent state and transferred into the aqueous fraction. Plutonium is separated from uranium.

4. Re-extraction of uranium from the organic fraction by diluted nitric acid. Uranium transfers into the aqueous fraction.

No more than three cycles of the extraction – re-extraction process are traditionally used. The number of the cycles can substantially change the content of radioactive fission products in the reprocessed fuel materials, i.e. proliferation-resistance of the extracted plutonium can be changed. If the extracted plutonium is remarkably contaminated with radioactive and heat-generating fission products, then plutonium becomes unsuitable material for manufacturing of a weapon-grade nuclear explosive device. The low number of the extraction – re-extraction cycles well corresponds with the "dirty fuel – clean waste" concept.

Specific features in reprocessing of spent fuel assemblies discharged from fast reactors. Spent nuclear fuel discharged from fast reactors (SNF-FR) is characterized by the higher values of fuel burn-up in comparison with spent nuclear fuel discharged from thermal reactors (SNF-TR). In fast reactors fuel burn-up can reach 100 GWd/t HM via 40-50 GWd/t HM in thermal reactors. As a consequence, SNF-FR contains the larger quantities of plutonium (up to 20% via ~0,7% in SNF-TR) and fission products (up to 10% via 4-5% in SNF-TR). That is why SNF-FR reprocessing encounters the following technological challenges:

1. The more intense radiolysis of the SNF solutions and organic extractants.
2. The larger content of volatile fission products (I, Kr, Xe, T) requires applying the advanced gas-absorption systems at SNF chopping and dissolving.
3. The larger plutonium content can degrade TBP efficiency due to the lower solubility of plutonium dioxide.
4. The more intense radioactivity requires applying the advanced systems for remote control of the extraction – re-extraction process.
5. Volumes of liquid high-level wastes (HLW) are about five times larger than those released in the SNF-TR reprocessing. Nearly 10 m^3 HLW per one SNF ton are produced in the SNF-FR reprocessing via 1-3 m^3 HLW per one SNF ton in the SNF-TR reprocessing.

1.6. Treatment and ultimate disposal of radioactive wastes

All nuclear technologies are related with use or generation of radioactive substances. For example, fresh fuel assemblies of nuclear reactors contain radioactive isotopes of uranium; spent fuel assemblies contain radioactive isotopes of uranium, plutonium, transuranium elements and fission products. Some radioactive isotopes can be recovered from spent fuel and profitably used. Fissile isotopes can be repeatedly used (recycled) in fresh fuel compositions. Some fission products and transuranium elements are widely applied as heat sources, sources of ionizing radiation in medicine and various industrial branches. The remaining radioactive substances, whose profitable applications are unfeasible yet, are usually regarded as radioactive wastes (RAW). Thus, RAW are those radioactive substances whose profitable applications are unfeasible now.

Therefore, the following materials and products can be included into RAW composition:

1. Those products of nuclear technologies which are unsuitable now for any profitable applications.

2. All the materials and products which are contaminated with radioactive substances before their decontamination.

Specific peculiarity of RAW is a principal impossibility of their extermination by means of any traditional technology (incineration, conversion into any other chemical form). RAW remain to be radioactive in any chemical forms. Traditional technologies can only transform RAW into the forms suitable for ultimate disposal in deep underground geological repositories. Non-traditional methods of RAW extermination presume construction of the dedicated nuclear facilities where RAW are bombarded by ionizing radiation (neutrons or gamma-rays) with the only aim to transmute (convert) long-lived radioisotopes into short-lived or stable isotopes.

The most dangerous RAW are by-products of spent fuel reprocessing. These RAW are dangerous materials both in respect of their quantity and intensity of radiation emitted mainly by fission products. FP quantity in SNF discharged from thermal and fast reactors are equal to about 40-50 kg/t and up to 100 kg/t, respectively. Appropriate specific

radioactivities of SNF discharged from thermal and fast reactors are equal to ~6 MCi/t and 20 MCi/t, respectively.

For comparison:

1. Total release of radioactive materials after Chernobyl accident is evaluated as 90 MCi.

2. Total release of radioactive materials after Kyshtym accident (explosion of liquid RAW storage) is evaluated as 20 MCi.

1.6.1. Classification of RAW

Classification of radioactive wastes

RAW are classified depending on their state of aggregation (liquid, gaseous and solid RAW) and on their specific radioactivity (low-level, middle-level and high-level RAW). The norms used by Russian regulatory bodies for classification of RAW are presented in Tables 1.7, 1.8.

Table 1.7

Classification of liquid and gaseous RAW

Category	Specific activity, Ci/l	
	Liquid	Gaseous
Low-level	$\leq 10^{-5}$	$\leq 10^{-13}$
Middle-level	$10^{-5} – 1$	10^{-13} - 10^{-9}
High-level	> 1	$> 10^{-9}$

Table 1.8

Classification of solid RAW

Category	Dose rate, R/h	Type of radiation		
		α, Ci/kg	β, Ci/kg	γ, Gr/h
Low-level	< 0,2	$2 \cdot 10^{-7}$-10^{-5}	$2 \cdot 10^{-6}$-10^{-4}	$3 \cdot 10^{-7}$-$3 \cdot 10^{-4}$
Middle-level	0,2-2	10^{-5}-10^{-2}	10^{-4}-10^{-1}	$3 \cdot 10^{-4}$-10^{-2}
High-level	> 2	$> 10^{-2}$	$> 10^{-1}$	$> 10^{-2}$

Main mission of RAW treatment is to protect humans and the environment against negative effects of radioactive materials. The most significant negative effects include ionizing radiation, heat generation and chemical toxicity.

1.6.2. Treatment of high-level RAW

There are the following two main forms of HLW:

1. HLW from radiochemical SNF reprocessing facilities.

These wastes are mainly liquid RAW because the industrial-scale SNF reprocessing is primarily based on the aqueous solvent-extraction PUREX-like technologies. As is known, the solvent-extraction reprocessing of SNF discharged from nuclear power reactors can produce about 45 m^3 of liquid HLW, 150 m^3 of liquid middle-level wastes (MLW) and up to 2000 m^3 of liquid low-level wastes (LLW) per one ton of spent fuel.

2. Spent fuel assemblies discharged from nuclear power reactors.

In the USA, where the moratorium has been decreed on radiochemical reprocessing of spent fuel from commercial NPP, these assemblies are considered as a form of the transport RAW containers completely ready for interim storage and, further, for ultimate disposal in deep underground geological repositories.

Main stages of the HLW treatment

1. Interim storage:

a. Spent fuel assemblies are placed into the water storage pools at NPP or at SNF reprocessing plants.

b. Liquid HLW are poured into the steel storage tanks. The storage tanks are put under strict control of heat generation rate (if necessary, forced heat removal must be provided) and elemental composition of the gas cushion over the HLW level (if necessary, air blowing-through is carried out to remove explosive hydrogen produced by water radiolysis).

2. Evaporation of liquid HLW.

The HLW evaporation process provides ~200-fold reduction of the HLW volume. However, the following negative effects arise:

a. Specific radioactivity of the evaporated HLW increases.

b. Specific heat generation rate of the evaporated HLW increases too. The larger heat generation rate warms up the evaporated HLW.
c. Corrosion activity of the evaporated HLW intensifies due to the higher corrodent concentrations and to the elevated temperature.
d. Gas release intensifies too due to the radiolysis of water and some liquid HLW components.

The following countermeasures are usually undertaken:
a. Control of explosive hydrogen content in the gas cushion above the HLW level in the storage tanks.
b. Periodical air blowing-through for dilution and removal of explosive hydrogen.
c. Control of the gas cushion temperature ($< 60^0$C).
d. Forced heat removal.
e. Application of corrosion-resistant alloys and stainless steels as structural materials of the HLW evaporation facilities and the HLW storage tanks.
f. Introduction of the corrosion inhibitors into the evaporated HLW.
g. Disposition of the HLW storage tanks below the earth level on the concrete saucers.

3. Solidification of the evaporated HLW.

Main mission of the HLW solidification is to implant the HLW into a stable inert material (matrix) that can reliably prevent the HLW release into the environment and, finally, into the food chains. Migration ability of the HLW must be substantially weakened, or a reliable HLW immobilization must be guaranteed.

At present, the HLW implantation into some glass compositions, or the HLW vitrification, is considered as the most suitable form for the HLW immobilization. The following two technologies of the HLW vitrification are the most well-known:

1. One-step technology.

The liquid concentrated HLW are poured into a refractory crucible together with the glass-producing additives. Under gradual warming up, the mixture undergoes the following changes:
a. Ultimate HLW evaporation.
b. Calcination of dried HLW at 300-400^{0}C.
c. Glass-mass melting at 1100-1150^{0}C.

After relatively short cooldown, the crucible with all its content is transported to the ultimate disposal site.

2.Two-step technology.

The French AVM-process can be considered as a typical example of the two-step HLW vitrification technologies.

Main stages of the AVM-process:

a. Calcination of the evaporated HLW at 300-400^{0}C.

b. Mixing the calcination product with the glass-producing additives.

c. The mixture is poured into a melting furnace.

d. Gradual warming up and formation of the glass-mass at 1100-1150^{0}C.

e. Periodical drainage of the glass-mass into steel containers.

f. Interim storage and ultimate disposal of the HLW containers.

Some alternative versions of the HLW vitrification technologies have been developed till now. The alternative technologies presume the HLW implantation into other stable materials, such as ceramics, glass-ceramics or mineral-like SYNROC materials. The term SYNROC is an abbreviated form from the words “Synthetic Rocks”, i.e. artificial but natural rock-like materials. Development of the SYNROC materials and the technology for the HLW implantation into them (the SYNROC technology) is based on the hope that the SYNROC materials could be characterized by the same physical and chemical properties (primarily, high long-term stability) as their natural analogues.

The SYNROC technology includes the following main stages:

1. Mixing the evaporated HLW with predecessors of the SYNROC materials. These predecessors are, as a rule, various refractory oxides. One typical example of the SYNROC predecessor composition is as follows: TiO_2(71%), CaO(11%), ZrO_2(7%), BaO(6%), Al_2O_3(5%).

2. Calcination of the mixture at 650-750^{0}C.

3. Hot pressing of the mixed powder into the SYNROC pellets (temperature - 1100-1200^{0}C, pressure - 150-200 atmospheres).

4. Filling up the steel containers with the SYNROC pellets, interim storage and ultimate disposal of the HLW containers.

Multiple tests were carried out with the HLW-containing SYNROC materials, and the following main results were obtained:

1. Physical, chemical and corrosion-resistance properties of the SYNROC materials appeared to be very similar with those of natural

rock minerals, i.e. the SYNROC materials are able to maintain their stability under any environmental impacts for sufficiently long time periods.

2. The SYNROC materials can retain up to 20% HLW.

3. The water-leaching rate of the SYNROC materials covered the range of $10^{-6} \div 10^{-5}$ gram from 1 cm^2 of the sample surface a day ($g/cm^2 \cdot day$).

The achievable HLW contents and the HLW leaching rates from the SYNROC materials are inferior to analogous properties of the borosilicate glass. The borosilicate glasses can retain up to 30% HLW. In general, the glasses are characterized by intrinsically disordered molecular lattice and, therefore, the glasses are able to keep wide spectrum of various radioisotopes. The SYNROC materials with their finely ordered crystalline lattice are able to keep only the radioisotope compounds with certain atomic dimensions and with certain valencies. The water-leaching rate of the vitrified HLW is evaluated as $10^{-8} \div 10^{-7}$ $g/cm^2 \cdot day$.

So, the SYNROC materials are inferior only to the glasses in respect to the achievable HLW content and the water-leaching rate but, nevertheless, they remain to be the second candidate for the HLW immobilization.

After the HLW are immobilized in the glass-mass or in the SYNROC pellets, these solidified HLW forms are placed into the steel containers. The further HLW management foresees sufficiently long (up to 50 years) interim storage in the near-to-surface storage points with air or water cooling. The containers can be periodically retrieved to investigate the current state of the solidified HLW and, if necessary, to perform their additional treatment.

1.6.3. Ultimate disposal of high-level RAW in geological formations

The next stage is an ultimate disposal of the HLW containers in deep underground geological repositories. Geological formation can be regarded as a suitable place for ultimate disposal of the HLW containers only if the formation satisfies the following requirements:

1. *Geographical properties of the formation:*

a. Far distance from the densely populated areas.

b. Low seismicity and low probability of earthquakes.

c. Far distance from the level of ground waters.

d. The geological stratum must not enter the earth surface.

2. *Physical properties of the formation:*

a. Good heat conductivity and heat capacity.
b. Good mechanical strength and plasticity.
c. Good chemical stability and retentivity of radioisotopes.

The following three geological formations are estimated now as the most promising candidates for ultimate disposal of the HLW containers in deep underground repositories:

1. Salt mines.
2. Sedimentary clayey formations.
3. Rocky formations.

Unfortunately, it appeared impossible to distinguish one the most suitable geological formation from these candidatures even basing only on their physical properties. All the candidates are characterized by their own advantages and drawbacks.

Salt mines

Advantages:

1. Far distance from ground waters, i.e. hydrological conditions of the salt mines were so stable that the salts remained in their initial state for a geological-scale time period (some millions or even milliards of years) despite of their good solubility by light water.
2. Good plasticity.
3. High heat conductivity.

Drawbacks:

1. Good solubility by light water.
2. Potential usefulness for many industrial branches.
3. Radiolysis by ionizing radiations with intense release of harmful gaseous substance (chlorine, for instance).

Sedimentary clayey formations

Advantages:

1. Full water impermeability.
2. High retentivity of radioactive fission products (with the exception of ^{129}I and ^{99}Tc).
3. Good plasticity.

Drawbacks:

1.Low retentivity of ^{129}I and ^{99}Tc, radioisotopes with high migration ability.

2. Low heat conductivity.
3. Proximity to the earth surface.

Rocky formations

Advantages:

1. High water impermeability.
2. Good mechanical strength and chemical stability.

Drawbacks:

1. Low plasticity, i.e. high probability for the cracks to appear as potential pathways for the HLW migration into the biosphere.
2. Low heat conductivity.

The most advanced all over the world project of the deep underground HLW repository is the Yucca Mountain project (Nevada, USA). Schematic layout of the Yucca Mountain repository is shown in Fig. 1.8.

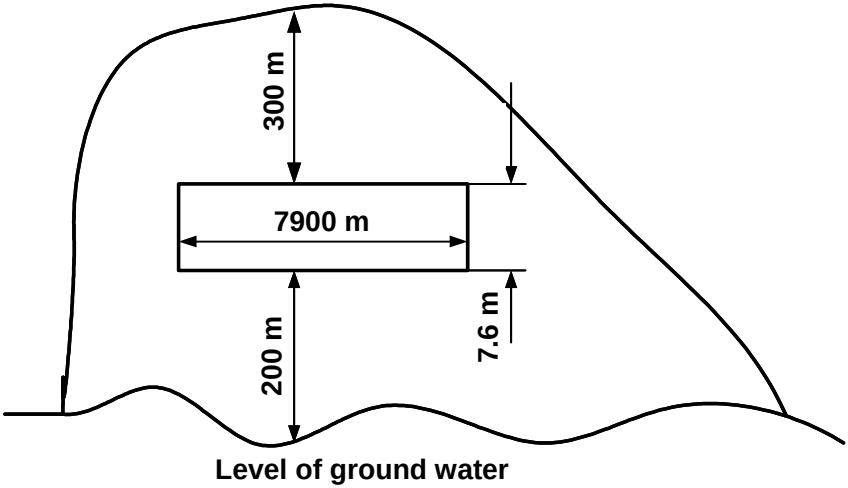


Fig. 1.8. Layout of the Yucca Mountain repository

Construction of the Yucca Mountain repository was begun in 1994. By April 1997 the main drifting works were finished with the following dimensions of the major tunnel: length – 7900 m; height – 7,6 m; distance from the level of ground water – 200 m downwards; distance from the mountain top – 300 m upwards. About $20 milliards were already

spent for these drifting works. In 2002 all the studies on geological, hydrological, geochemical and geothermal properties of the repository site were completed, and the US Nuclear Regulatory Commission received the application on start-up of the repository operation. As was previously planned, the loading process of the repository with the HLW containers must be begun in 2010 (total capacity of the Yucca Mountain repository was evaluated as 77,000 t HLW). However, the license on the repository operation was not issued by the NRC till now. Moreover, federal funding of the Yucca Mountain project was ended just in 2010.

The geological formation of the Yucca Mountain repository is a rocky tuff with large quantity of cracks. Vertical infiltration of light water from upwards into the major tunnel was measured and appeared equal to approximately one liter per one square meter of the tunnel bottom annually, i.e. 1 mm-thick water layer a year.

Some numerical evaluations demonstrated that major effect on potential contacts of ground water with radioisotopes and probability of their release into the biosphere from the fully loaded HLW repository Yucca Mountain is mainly defined by residual heat generation. The mountain part adjacent to the major tunnel can be warmed up to ~130^0C, i.e. above boiling temperature of ground water. So intense warming up can create the closed circuit of natural water convection from hot HLW repository to relatively cold rocks. There, water vapor condenses and flows down. That is why hydrological conditions of fully loaded repository cardinally differ from those in empty repository.

The following changes can occur in hydrological conditions of the fully loaded HLW repository:

1. Formation of the condensed water layer above the HLW repository by natural convection of hot vapor and cold water.
2. The rocky area adjacent to the HLW repository is impregnated with water.
3. Intense cracking of the tuff layers adjacent to the HLW repository by hot vapor and temperature gradient.
4. Chemical activity of hot water enhances. Consequently, corrosion rate of the HLW containers and solubility of radioisotopes can increase.

Central zone of the HLW repository can remain relatively dry because of maximal heat generation rate and rapid evaporation of the flowing down water. Atmosphere in peripheral zone of the HLW re-

pository can be more humid and, thus, it can intensify corrosion of the HLW containers. So, internal heat generation can be a serous capacity-limiting factor for the very expensive HLW repositories. The heat generation is mainly caused by radioactive decays of some long-lived fission products (^{137}Cs, ^{90}Sr) and minor actinides (radioisotopes of neptunium, americium and curium). In a relatively short-term perspective (100-200 years), fission products are main contributors into the internal heat generation. In a longer perspective (t > 1000 years) the dominant role in the decay heat generation passes from fission products to minor actinides.

Therefore, some advanced alternative approaches to the HLW management are under thorough studies now throughout the world. These approaches presume various options for extraction and partitioning of long-lived fission products (LLFP) and minor actinides. The separated radioisotopes can be further used as heat sources and sources of ionizing radiation in many industrial branches.

Minor actinides are able to enhance nuclear non-proliferation regime because neutron irradiation of ^{237}Np and ^{241}Am in nuclear reactors can transform them into plutonium isotope ^{238}Pu, intense source of decay heat and spontaneous fission neutrons. Plutonium with high enough content of ^{238}Pu becomes completely unsuitable for manufacturing of any nuclear explosive devices.

Some other approaches presume neutron transmutation of LLFP and minor actinides in the dedicated irradiation facilities (nuclear reactors, accelerator-driven systems, thermonuclear installations) where the most harmful radioisotopes can be converted into short-lived or stable nuclides.

1.6.4. Treatment of liquid middle- and low-level RAW

The following procedures are used to treat liquid MLW and LLW:

1. Precipitation and removal of solid particles from the MLW and LLW solutions.
2. Ion-exchange purification of the clarified solutions.
3. Evaporation up to the dry sediment.
4. Immobilization by bituminization or cementation.
5. Placement of the solidified radiowastes into the steel containers.

6. Interim storage and ultimate disposal of the steel containers.

Bitumen as a material for RAW immobilization can offer the following advantages:

1. Low leaching rate by light water.
2. Suitability for immobilization of any chemical RAW forms (salts, hydroxides, organics).
3. Good radiation resistance.

However, bitumen is an inflammable material as a by-product of natural oil reprocessing, and bitumen softens under warming up.

The alternative option to the RAW bituminization is a cementation process, i.e. RAW implantation into the concrete blocks.

Concrete a material for RAW immobilization can offer the following advantages:

1. Low cost and simplicity of the cementation process.
2. Good radiation resistance.
3. High heat conductivity.
4. Concrete is not an inflammable material and does not soften when warmed up.

Unfortunately, concrete is very sensitive to the water leaching. Comparative data on the water leaching rates of the most widely known materials for RAW immobilization are presented below:

1. Glass: $10^{-8} \div 10^{-7}$ g/cm^2·day;
2. SYNROC: $10^{-6} \div 10^{-5}$ g/cm^2·day;
3. Bitumen: $10^{-6} \div 10^{-4}$ g/cm^2·day;
4. Concrete: $10^{-3} \div 10^{-2}$ g/cm^2·day.

That is why glasses and the SYNROC materials are preferentially used to immobilize HLW while bitumen and concrete – for immobilization of MLW and LLW.

Chemical stability of the concrete blocks can be enhanced by impregnating the cement mixture with some organic monomers. At solidification, the monomers are polymerized, and chemical stability of the concrete blocks improves substantially.

1.6.5. Treatment of gaseous RAW

Gaseous RAW can produce the following negative effects on human organism:

1. Direct external irradiation and irradiation by the fallen out radioactive particles.
2. Internal irradiation at inhalation of air contaminated with gaseous RAW.
3. Chemical toxicity of gaseous RAW at inhalation.

The following radioisotopes are main components of gaseous RAW:
1. Radioactive noble gases (radioisotopes of krypton and xenon).
2. Iodine radioisotopes.
3. Carbon radioisotope ^{14}C.
4. Tritium.

After lengthy (5-10 years) staying in the SNF storage pool at NPP only the following relatively long-lived gaseous radioisotopes remained in RAW composition:
1. Of noble gases – only ^{85}Kr (half-live $T_{1/2}$ = 10,7 years).
2. Of iodine radioisotopes – only ^{129}I ($T_{1/2} = 1{,}6 \cdot 10^7$ years).
3. Radiocarbon ^{14}C ($T_{1/2}$ = 5730 years).
4. Tritium ^{3}H ($T_{1/2}$ = 12,3 years).

Removal of ^{85}Kr. The following methods are used to remove ^{85}Kr from gaseous RAW composition:
1. Cryogenic adsorption by activated charcoal or molecular sieves as ultra-filters.
2. Cryogenic adsorption by liquid carbon dioxide or liquid fluorocarbons.

Removal of ^{129}I. In gaseous RAW composition the radioiodine can be in the forms of molecular iodine I_2, iodides (I^-) and iodates (IO_3^-). The following methods are used to remove ^{129}I from gaseous RAW composition:
1. Absorption of the radioiodine by alkaline or acidic solutions in scrubbers where the radioiodine is oxidized up to solid insoluble compound HI_2O_8.
2. Chemisorption of the radioiodine onto the zeolite impregnated with silver nitrate $AgNO_3$. Molecular radioiodine is bound into insoluble silver iodide and silver iodate through the following chemical reaction:

$$2\ AgNO_3 + I_2 + H_2O + O_2 \rightarrow AgI + AgIO_3 + 2\ HNO_3\ .$$

Removal of ^{14}C. Gaseous RAW contains radiocarbon ^{14}C in its oxide forms ^{14}CO and $^{14}CO_2$. Radiocarbon is a product of neutron $^{14}N(n,p)^{14}C$ reaction with nitrogen that is contained in air, in coolant and structural materials as an impurity.

Unfortunately, till now no any industrial-scale technologies have been developed to catch ^{14}CO or $^{14}CO_2$ efficiently. In laboratorial studies some fluorocarbons demonstrated highly efficient absorption of ^{14}C (up to 99,9% ^{14}C) within low temperature range from -40^0C to $+4^0C$.

Removal of tritium. In nuclear reactors tritium can be produced by neutron reactions with coolant and some impurities (hydrogen, lithium) in structural materials, In addition, tritium is a product of very rare ternary fission reactions with emission of three (not usual two) fission products. Since the ternary fissions can occur with very low probability, spent fuel contains only about $2 \cdot 10^{-5}$ % ^3H.

The following properties of tritium should be taken into account:

1. Tritium is a source of low-energy β-radiation.
2. Tritium can actively enter into the isotope exchange reaction with light water thus forming tritium water HTO or T_2O at the SNF reprocessing. Therefore, tritium is present in all liquid RAW produced by aqueous technologies of the SNF reprocessing.

The following methods are used to remove tritium from gaseous RAW composition:

1. Voloxidation of spent fuel before its dissolution: oxidation by air at the elevated temperatures, $450\text{-}650^0C$. The air humidity can bind tritium into tritium water for further treatment as liquid RAW.
2. Chemisorption of tritium water by zeolite.
3. Light-water washing-out of organic TBP-containing fraction after the solvent-extraction process.

Treatment of volatile aerosols and dust. Volatile aerosols and dust are gas-like materials with specific content of solid and liquid particles within the range from 10^{-2} g/m^3 to 10 g/m^3. The following methods are used to treat volatile aerosols and dust:

1. Gravitational deposition in the dust-collecting chambers.
2. Centrifugal removal of solid and liquid particles in cyclones. The gas flow enters into a cylindrical vessel at an angle to its vertical axis. Solid and liquid particles can strike against the wall and drop out of the gas flow.

3. Electrostatic deposition (imparting an electrical charge to solid and liquid particles and their deposition by electrical field).
4. Gas washing-out in scrubbers.
5. Ultra-filtration with application of the dedicated filters made of fiber-glasses, polymers, metal-tissue and metal-ceramic materials.

1.6.6. Treatment of solid RAW

The following materials and products are the main components of solid RAW:
1. Details of nuclear equipment, construction materials, rubbish and working clothes before their decontamination.
2. Ion-exchange resins and filters.
3. Metal claddings of fuel rods.
4. Deposits on internal wall surfaces of technological equipment.

The following methods are used to treat solid RAW:
1. Reduction of RAW volume:
   a. Incineration with up to 100-fold reduction of RAW volume.
   b. Pressing with up to 10-fold reduction of RAW volume.
   In total, RAW volume can be decreased by a factor of about 1000.
2. Placement of the remaining RAW into the steel containers, interim storage and ultimate disposal.

A special technology is used to treat radioactive claddings of fuel rods. The following processes caused their radioactivity:
1. Neutron irradiation in nuclear power reactors can transform some stable isotopes of iron, chromium, nickel, molybdenum and other constituents of stainless steels into appropriate radioisotopes.
2. Fission products and minor actinides can migrate from fuel meat to fuel cladding. That is why fuel claddings can contain α-active radioisotopes.
3. Residuals of undissolved spent fuel.

Treatment of fuel claddings includes the following main stages:
1. Interim storage in concrete shelters under water layer (zirconium particles are pyrophoric in air).
2. Chemical treatment by hydrofluoric acid HF at the elevated temperatures (550-600^{0}C). This treatment can form superficial friable films on

the cladding surface. Then, these MA-containing films can be dissolved and removed by alkaline or acidic solutions.
3. Re-melting of fuel claddings into metal ingots in electrical furnaces.
4. Placement of metal ingots into the steel containers, interim storage and ultimate disposal.

One else form of solid RAW is constituted by radioactive deposits on internal walls of technological equipment units (mixers, settlers, connecting pipelines, etc.) at the SNF reprocessing plants. The radioactive deposits can be formed by the following processes:
1. Sorption of radioisotopes from the SNF solutions. Radioactivity of the walls gradually increases and can reach the values comparable with radioactivity of the SNF solutions.
2. The walls are gradually saturated with radioisotopes. After each decontamination the process of radioisotope sorption and gradual saturation reiterates.
3. Time-dependent evolution of basic thermal, physical and chemical conditions causes hardening the radioactive deposits. Only RAW components with the highest mechanical strength, chemical, radiation and thermodynamic resistance can remain on internal walls of technological equipment units. The most significant radioactive deposits contain zirconium- and silicon-based compounds (zirconates and silicates of fission products and fuel components) which are able to polymerize with formation of jelly-like materials (salts of silicic acid H_2SiO_3, oxides of silicon, magnesium, calcium and some alkaline metals).

Technological equipment of the SNF reprocessing plants is mainly decontaminated by the chemical desorption of solid radioactive deposits with liquid desorbing reagents. Radioactivity of the walls must be reduced to the levels acceptable for the repair or dismantling works. The desorbing process can transform solid deposits into liquid solutions for their further treatment as liquid RAW. The desorbing process is usually performed by the multiple washing-outs of technological units. At first, low concentrated solution of nitric acid HNO_3 is used to dissolve and remove spent fuel residuals. Afterwards, the multiple alternation of the walls treatment by liquid desorbing solutions is undertaken to weaken the deposits and, then, to dissolve and remove them.

The most widely applied decontamination technology is based on the multiple alternation of the washing-out processes by alkaline solutions

(chemical dissociation of ill-soluble deposits, hydration of high-density oxides and salts) and by acidic solutions (dissolution of the most friable deposits and removal them from technological units for further treatment as liquid RAW).

Control questions to Chapter 1

1. Call main categories of natural uranium resources.
2. Call main stages of hydro-metallurgical treatment of uranium ore.
3. What is the sorption process of uranium recovery based on?
4. What is the extraction process of uranium recovery based on?
5. What is the chemical precipitation process of uranium recovery based on?
6. What is the in-situ leaching process of uranium ore based on? Call main conditions for the ISL applicability.
7. What is the aqueous extraction affinage process? Call its main stages.
8. Write the material balance relationships for isotope uranium enrichment.
9. What are the basic ideological propositions for determination of the separative work scope and for determination of the separation potential function?
10. Call properties of uranium hexafluoride which are the most important for isotope uranium enrichment.
11. What technologies are used to convert uranium oxides into uranium hexafluoride?
12. What is the gas diffusion technology of isotope uranium enrichment based on?
13. What is the gas centrifuge technology of isotope uranium enrichment based on?
14. What is the separation-nozzle technology of isotope uranium enrichment based on?
15. Call main stages of the atomic vapor laser isotope separation process.
16. Call main stages of the molecular laser isotope separation process.
17. How can the chemical methods of isotope separation be used?
18. What is the plasma technology of isotope separation based on?
19. Call main advantages and shortcomings of uranium dioxide fuel.

20. Call main stages of the process applied for manufacturing of UOX-fuel pellets.
21. What specific features can you call in the manufacturing process of MOX-fuel pellets?
22. Call main advantages and shortcomings of metal uranium fuel.
23. Call main missions of refueling in nuclear power reactors.
24. Call main stages of the batch irradiation, partial batch and uniformly partial refueling strategies.
25. Call main stages of the scatter and modified scatter refueling strategies.
26. Call main stages of the out-in refueling strategy.
27. How are the refueling works performed in thermal LWR-type reactors?
28. How are the refueling works performed in fast LMFBR-type reactors?
29. How are the refueling works performed in thermal CANDU-type reactors?
30. How are the refueling works performed in thermal RBMK-type reactors?
31. Describe briefly a typical transport cask for spent fuel assemblies.
32. What requirements must the transport cask design satisfy?
33. What tests must a typical transport cask undergo?
34. Call main missions of spent fuel reprocessing.
35. Call main categories of the spent fuel reprocessing technologies.
36. Call main stages of the aqueous solvent-extraction technology.
37. How does TBP radiolysis occur? What negative consequences does TBP radiolysis lead to?
38. What technology is applied to separate plutonium from uranium?
39. Call main RAW components.
40. Call main RAW categories.
41. Call main stages of high-level RAW treatment.
42. What requirements must geological formations satisfy to be suitable for ultimate disposal of RAW?
43. What geological formations are under estimation now as potential candidates for ultimate disposal of RAW? Call their main advantages and drawbacks.

44. What are the main difficulties for ultimate disposal of RAW in the Yucca Mountain geological repository?
45. Call main stages of middle-level and low-level RAW treatment.
46. What materials are currently used for RAW immobilization? Range them according to their relative immobilization quality and explain it.
47. Call main stages of solid RAW treatment.
48. What technology is used to decontaminate technological equipment of the SNF reprocessing plants?

CHAPTER 2. NEGATIVE ECOLOGICAL EFFECTS AT VARIOUS STAGES OF THE CLOSED NUCLEAR FUEL CYCLE

All enterprises of nuclear fuel cycle deal with nuclear materials which are potentially able to impact negatively on the environment and human organisms. Even the least harmful nuclear material – uranium ore – is a radioactive substance, weak emitter of α-particles and spontaneous fission neutrons from decays of uranium isotopes ^{235}U and ^{238}U.

Acceptable specific activity of α-emitters equals 0.2 μCi/kg. As it was noted above, the mined uranium ore contains, in average, about 0.1% U, i.e. 1 kg of uranium ore contains 1 g U which, in its turn, contains 993 mg ^{238}U, 7 mg ^{235}U and 0.05 mg ^{234}U. Radioactivity A of radionuclide with mass m (grams), atomic weight M (a.m.u.) and half-life $T_{1/2}$ (s) can be calculated by using the formula:

$$A(\mu Ci) = \frac{m}{M} \cdot N_a \cdot \frac{\ln 2}{T_{1/2}} \cdot \frac{10^{-4}}{3.7};$$

where N_a – the Avogadro number, $6.023 \cdot 10^{23}$ nuclei per a mol.

It is easy to determine the following contributions of uranium isotopes into total specific radioactivity of uranium ore:

$$A(^{238}U) \approx 0.332 \text{ μCi/kg};$$
$$A(^{235}U) \approx 0.015 \text{ μCi/kg};$$
$$A(^{234}U) \approx 0.332 \text{ μCi/kg}.$$

As is seen, contribution of only uranium isotopes (~0.7 μCi/kg) into specific radioactivity of uranium ore exceeds the acceptable level by a factor of 3.5. However, uranium ore contains radioactive daughter products of ^{235}U and ^{238}U decay families. The chains of radioactive decays start from ^{235}U and ^{238}U, many new radionuclides are produced which, in their turn, undergo decays via their own channels (α and β-decays, isomeric transitions) and so on. Final products of ^{235}U and ^{238}U decay chains are stable lead isotopes ^{207}Pb and ^{206}Pb, respectively. The decay rate (radioactivity) of each intermediate radionuclide equals the decay rate (radioactivity) of its predecessor since for the elapsed many billion years the so-called eternal equilibrium established between all members of decay families. So, radioactivity of any intermediate radionuclide equals radioactivity of start radionuclide. Thus, total uranium radioactivity can be calculated as radioactivity of ^{238}U multiplied by the number of its daughter

products plus radioactivity of ^{235}U also multiplied by the number of its daughter products. ^{234}U can be omitted because ^{234}U is a member of ^{238}U decay family. In reality, different migration ability of decay products (especially, gaseous radionuclides) in geological environment can tear the decay chains and decrease significantly specific radioactivity of uranium ore. With proper accounting for migration ability of some radionuclides, specific radioactivity of uranium ore containing 0.1% U is equal to about 4 μCi/kg, i.e. twenty times higher than acceptable level. Natural uranium, recovered from uranium ore and chemically separated from its decay products, contains three uranium isotopes - ^{235}U, ^{238}U and ^{234}U. Specific radioactivity of natural uranium is evaluated to be 700 μCi/kg, i.e. 3500 times higher than acceptable level. This means that already at initial stages of nuclear fuel cycle its enterprises have to deal with materials whose specific radioactivity exceeds significantly the sanitary norms.

Nuclear enterprises are able to produce the following negative effects on the environment and human organisms:

1. Radiation injuries since nuclear technologies deal with materials – emitters of ionizing radiation.
2. Thermal impact caused by hot-water effluents from NPP steam turbines into the air atmosphere and other objects of hydrosphere.
3. Chemical impact of toxic materials contained in tails from hydrometallurgical processing of uranium ore at initial NFC stage, in wastes from radiochemical SNF reprocessing at final NFC stage and in common domestic wastes from routine activity of nuclear enterprises.
4. Alienation of large territories that takes place mainly at initial NFC stage related with mining and primary processing of uranium ores.

Although each of these negative effects deserves an individual analysis, radiation impact calls the most public concerns. That is why main attention will be given below to radiation effects produced by routine operation of nuclear enterprises on the environment and human organisms.

2.1. Natural radiation background

In order to evaluate the radiation threats from nuclear enterprises, the following question should be answered: in what radiation field created by natural sources of ionizing emissions do the environment and

the Earth population reside in the present time? The Earth's biosphere had existed for many billions of years. During such a lengthy time period all representatives of the Earth's biosphere had cultivated some adaptive mechanisms which allowed them to accommodate and survive under all natural negative conditions including natural radiation background. The negative effects produced by natural radiation background can be used to estimate ecological threats from various nuclear technologies. The problem of the humankind is to form the situation when nuclear technologies do not produce so severe radiation impact on the environment that exceeds substantially the radiation impact produced by natural background.

The following main contributors into natural radiation background can be noted:

- cosmic rays;
- radioactivity of rocky minerals (uranium, thorium, their decay products, isotopes from middle part of Mendeleev's table – ^{40}K, ^{48}Ca, ^{87}Rb);
- radioactivity of soil, atmosphere, hydrosphere and biosphere.

Statistical information about natural radiation background in various world regions, including Russia, can be presented as average annual individual effective doses from natural sources of ionizing radiation.

1. Cosmic rays.
   Range over the world regions – from 300 to 1000 μSv/year.
   Average over the world – 390 μSv/year.
   Average over Russia – 460 μSv/year.
2. External irradiation by terrestrial radionuclides.
   Range over the world regions – from 300 to 600 μSv/year.
   Average over the world – 480 μSv/year.
   Average over Russia – 440 μSv/year.
3. Internal irradiation by terrestrial radionuclides.
   Range over the world regions – from 400 to 10000 μSv/year.
   Average over the world – 1570 μSv/year.
   Average over Russia – 1470 μSv/year.

Total radiation impact from natural background:
Average over the world – 2440 μSv/year.
Average over Russia – 2370 μSv/year.

Thus, it may be concluded that radiation impact from natural sources of ionizing radiation on human organism can be evaluated at an approximate level of 2,4 mSv/year. This value corresponds to the collective effective dose received by full the Earth's population at the level of $1{,}5 \cdot 10^7$ person·Sv/year.

Theses two values, namely 2,4 mSv/year per a person and $1{,}5 \cdot 10^7$ person·Sv/year, will be used further .to compare radiation threats at various NFC stages with natural background of ionizing radiation.

According to the RF regulations, additional radiation burden from nuclear enterprises must be well below 1 mSv/year for population and 20 mSv/year for staff.

2.2. Ecological effects of various NFC stages

2.2.1. Mining and hydrometallurgical treatment of uranium ore

At this NFC stages the staff members of uranium mines, HM-plants and population of adjacent regions can suffer from both external and internal irradiation since large amounts of radioactive materials are brought onto the Earth surface and can migrate into the environment. The main striking effects are caused by radioactive dust, aerosols and some gaseous radionuclides from U and Th decay families, namely ^{226}Rn (radon) from ^{238}U decay family and ^{220}Rn (thoron) from ^{232}Th decay family.

The following values can be taken from statistical information about radiation doses at the stages of uranium ore mining and its HM-treatment:

1. Average annual effective dose from external irradiation is evaluated as 10 mSv/year for the staff members involved into uranium ore mining.
2. Roughly the same radiation dose (about 10 mSv/year) is received in the form of internal irradiation. Thus, collective effective dose received by the staff members involved into uranium ore mining is evaluated as 1,7 person·Sv/GWe·year.
3. Average-world individual effective dose received by the staff members involved into operation of HM-plants is evaluated as 12 mSv/year for external irradiation and 5 mSv/year for internal irradiation. Thus,

collective effective dose received by the staff members of HM-plants is evaluated as 0,4 person·Sv/GWe·year.
4. Collective effective dose received by population of adjacent regions is evaluated as 0,2 person·Sv/GWe·year, i.e. by one order of magnitude lower that that for the staff members involved into uranium ore mining.

It is noteworthy that these NFC stages are maximally related with alienation of large territories for disposition of HM-plants and their wastes (tails). According to the RF regulations, two alienation forms may be applied< namely temporary alienation (with possibility for future re-cultivation) and permanent alienation (for disposition of radioactive tails): 60 ha/GWe·year for temporary alienation and 2 ha/GWe·year for permanent alienation.

2.2.2. Affinage of uranium concentrate, isotopic uranium enrichment and fabrication of fuel rods

At these NFC stages collective effective doses received by the staff members are evaluated as 0,1 person·Sv/GWe·year at the stages of uranium concentrate affinage and uranium isotope enrichment, and as 0,6 person·Sv/GWe·year at the fabrication stage of fresh fuel assemblies.

Collective effective doses received by population of adjacent regions are evaluated as $2 \cdot 10^{-4}$ person·Sv/GWe·year from liquid effluents, and as $19 \cdot 10^{-4}$ person·Sv/GWe·year from gaseous effluents.

Thus, initial (front-end) part of nuclear fuel cycle (mining and HM-treatment of uranium ores, affinage of uranium concentrate, uranium isotope enrichment and fabrication of fresh fuel assemblies) can be characterized by the following radiation impacts:
1. Collective effective dose received by the staff members is evaluated as 4 person·Sv/GWe·year.
2. Collective effective dose received by population of adjacent regions is evaluated as 0,25 person·Sv/GWe·year.

Total capacity of the world nuclear power system is about 375 GWe in 2014. So, collective effective dose can be calculated as $1{,}6 \cdot 10^{3}$ person·Sv/year, i.e. about 0.01% from radiation impact of natural background ($1.5 \cdot 10^{7}$ person·Sv/year).

2.2.3. Use of nuclear fuel at NPP

Even normal NPP operation mode is accompanied by accumulation of radiowastes in nuclear fuel and in some other NPP components. Main radiation sources in the wastes are fission products and minor actinides in nuclear fuel, corrosion and activation products in structural materials of fuel and in other metal in-pile structures of nuclear reactors. Penetration of radiowastes into the environment and biosphere encounters a series of protective barriers including fuel pellets and their metal claddings, the reactor vessel, circuits for coolant circulation and NPP containment. Nevertheless, some small fraction of radiowastes is able to come into the environment with coolant leakages, at the repair and maintenance works, at replacement of defected equipment units. Besides, some radionuclides can escape the spent fuel storage pool as liquid effluents from the washing-out process of contaminated filters and ion-exchange purification systems.

NPP with RBMK-type reactors is usually characterized by relatively higher radiation impact than NPP with VVER-type reactors. The closed primary circuit of VVER-type reactors promotes the longer retention of radionuclides in comparison with the open primary circuit of RBMK-type reactors where boiling light-water coolant can circulate. Gaseous and volatile radiowastes can rapidly escape the boiling coolant with vapor and release into the environment through the condenser's ejector.

Collective effective doses received by NPP staff are evaluated as 3,9 person·Sv/GWe·year for VVER-type reactors and 20,3 person·Sv/GWe·year for RBMK-type reactors.

Under nominal operation conditions of Russian NPP, collective effective doses received by population of adjacent regions are evaluated as 0,13 person·Sv/GWe·year for VVER-type reactors and 0,6 person·Sv/GWe·year for RBMK-type reactors. As total capacity of Russian nuclear power system is equal to about 23 GWe in 2014 with approximately the same contributions from VVER and RBMK-type reactors, collective effective doses 8 person·Sv/year for population of adjacent regions and 450 person·Sv/year for NPP staff members. These values are negligibly small as compared with natural radiation background ($1.5 \cdot 10^7$ person·Sv/year).

Thermal pollution of the environment

As is known, cooling of the condensers at NPP with electrical output of 1 GWe requires water flow rate at the level of 50 m^3/s and warms up water on 10^0C. Following from these NPP operation data, minimal area of water-table in NPP heat-sink pool is evaluated as 6 km^2/GWe. If the heat-sink pool is also used for some other purposes (fish breeding, tourism, rest of people), then the minimal area must be roughly doubled, up to 13 km^2/GWe.

The water warming-up effect can change water density, viscosity and gas solubility. The warmed water evaporates quicker and, thus, promotes forming the damper climate.

Thermal pollution of hydrosphere caused by hot-water effluents into NPP heat-sink pool can lead to appearance of vertical water stratification with relatively hotter superficial layers. Consequently, oxygen concentrations in cold, near to the bottom, layers can remarkably decrease. General water warming-up effect intensifies decomposition of organic residuals and water-plants, upgrades demands of fishes for oxygen. All these effects can provoke creating the areas of mass fish starvation.

Mechanical traumatism of fishes by the water-intaking tools in combination with toxicity of chlorine that is applied to prevent overgrowing of these tools with water-plants can result in even more negative effect as compared with that of hot-water effluents only.

2.2.4. Radiochemical SNF reprocessing

Radiochemical reprocessing of spent fuel assemblies differs two possible organizational options of nuclear fuel cycle, namely the closed NFC and open (once-through) NFC. SNF reprocessing extracts valuable and relatively low-active nuclear materials (uranium and plutonium) and returns (recycles) them to the fabrication stage of fresh fuel rods and fuel assemblies for their further usage in nuclear power reactors. Unfortunately, radiochemical SNF reprocessing is inevitably accompanied by accumulation of liquid, solid and gaseous radiowastes in very large quantities. These radiowastes have to be treated and ultimately disposed of in stable geological formations.

According to the IAEA information, collective effective doses received by the staff members of the SNF reprocessing plants differ each

other depending on the country where these plants are put into operation:

France – 0,6 person·Sv/GWe·year;

Great Britain – 11,0 person·Sv/GWe·year;

Russia – 1,8 person·Sv/GWe·year.

Analogous values for population of adjacent regions (within radius of 50 km) cover the range from 20 to 30 person·Sv/GWe·year. If a mean value of collective effective dose received by the staff members, 10 person·Sv/GWe·year , for instance, as in Great Britain, is adopted as a reference point, then total collective effective dose (global nuclear power capacity is ~375 GWe in 2014) can be determined as $3.75 \cdot 10^3$ person·Sv/year, i.e. two orders of magnitude higher than that for population of adjacent regions. However, even this high value equals about 0.025% from collective effective dose produced by natural sources of ionizing radiation ($1.5 \cdot 10^7$ person·Sv/year).

2.2.5. Treatment and disposal of radiowastes

The currently world-wide adopted strategy of radiowastes management includes the following main stages:

1. Long-term controlled storage of radiowastes at nuclear enterprises.
2. Conditioning of radiowastes, i.e. the radiowastes treatment in order to reduce their volume and transform them into any forms suitable for containerization, transportation and ultimate disposal in stable geological formations which are able to decrease drastically any probability for radionuclides to penetrate into the environment.
3. Ultimate disposal of steel containers filled up with solidified radiowastes in stable geological formations (salt mines, sedimentary clayey and rocky formations).

Full absence of real geological repositories and, as a consequence, full absence of experiences on their long-term operation and maintenance leads to a necessity to use only theoretical evaluations of the radiation effects produced by radiowastes on the environment and human organism. These evaluations presume that steel containers filled up with radiowastes could fail after 1000 years of their disposal, and the solidified radiowastes could escape their inert matrices and penetrate into the environment after 10000 years of their disposal.

The following conclusions can be derived from these theoretical evaluations:

1. Ultimate disposal of spent fuel assemblies discharged from power light-water reactors (no reprocessing option) leads to collective effective dose at the level of 270 person·Sv/GWe for 10000 years.
2. Ultimate disposal of radiowastes produced by radiochemical reprocessing of spent fuel assemblies from power LWR (reprocessing option) can reduce collective effective dose down to the level of 50 person·Sv/GWe for 10000 years.

It seems very improbable that direct disposal of spent fuel assemblies without their reprocessing will be practiced for a sufficiently long time period. General trend in nuclear power development consists in implementing the closed NFC option, i.e. radiochemical reprocessing of spent fuel assemblies, recycle of fertile and fissile materials, ultimate disposal of radiowastes from SNF reprocessing. Consequently, average annual collective effective dose from geological repositories of radiowastes can be evaluated as ~0,2 person·Sv/year.

2.2.6. Total ecological effects of NFC

By summing up the results obtained in analysis of various NFC stages from standpoint of their radiation impact on the environment and human organism, the following collective effective doses can be presented:

1. At initial (front-end) part of NFC (mining and primary treatment of uranium ore, affinage of uranium concentrate, uranium isotope enrichment, fabrication of fresh fuel assemblies) collective effective dose is evaluated as $1{,}6 \cdot 10^3$ person·Sv/year, i.e. ~0,01% from radiation impact of natural background ($1{,}5 \cdot 10^7$ person·Sv/year).
2. At main NFC stage (use of nuclear fuel at Russian NPP) collective effective dose received by NPP staff is evaluated as ~450 person·Sv/year while collective effective dose received by population of adjacent regions is evaluated as ~8 person·Sv/year. Both values are negligibly small as compared with natural radiation background.
3. Radiochemical SNF reprocessing (back-end part of NFC) can produce collective effective dose received by the staff members at the level

of $3{,}7 \cdot 10^3$ person·Sv/year, i.e. ~0,025% from radiation impact of natural background.

4. Ultimate disposal of radiowastes (back-end part of NFC) in stable geological formations can produce, according to some theoretical evaluations, collective effective dose at the level of ~0,2 person·Sv/year.

So, by summing up the collective effective doses received by staff members and population of adjacent regions, it can be concluded that total radiation impact of nuclear enterprises on the environment and human organism does not exceed 0,04% from radiation impact of natural radiation background. Of course, all these evaluations are correct only under normal operation conditions of nuclear enterprises. Under some accidental conditions, radiation impact of nuclear enterprises can be substantially higher as we have seen on the examples of well-known catastrophic events at Chernobyl and Fukushima NPP, at Kyshtym repository of radiowastes.

2.3. Some ecological problems in the USA

According to publications of American environmentalists, natural radiation background on the USA territory slightly differs from that on the RF territory and from the world-averaged values. The following statistical data can characterize contributions from main components into average natural radiation background in the USA:

1. Radon in structural materials – 2 mSv/year.
2. Radioactivity of the Earth's crust (soils and rocks) – 0,28 mSv/year.
3. Cosmic rays – 0,27 mSv/year.
4. Radioactivity of human body (human blood contains radionuclide ^{40}K with half-life of $1{,}25 \cdot 10^9$ years) – 0,40 mSv/year.

Subtotal – 2,95 mSv/year.

As for artificial (anthropogenic, or man-made) irradiation, American environmentalists presented the following statistical information:

Artificial irradiation

1. Medical procedures – 0,53 mSv/year.
2. Consumer products – 0,10 mSv/year.
3. Weapon-test fallout – 0,01 mSv/year.
4. Nuclear power industry – 0,01 mSv/year.

Subtotal – 0,65 mSv/year.

Elective irradiation

1. Use of natural gas – 0,06 mSv/year.
2. Airline flights – 0,02 mSv/year.
3. Watching color TV – 0,01 mSv/year.
4. Sleeping with another person – 0,01 mSv/year.

Subtotal – 0,10 mSv/year.

By summing up all the subtotals, total average annual radiation effect produced by natural, artificial and elective exposures of American citizens can be evaluated as:

2,95 mSv/year + 0,65 mSv/year + 0,1 mSv/year = 3.7 mSv/year.

This dose is remarkably higher than the world-averaged value (2,4 mSv/year).

One else very interesting approach has been developed by the US scientists to evaluation of the average time period by which a human life could be shortened through encountering a particular risk. This average time period is defined as the "Loss-of-Life-Expectancy" and obtained from analysis of broad statistical information. Under the approach, many harmful factors which are able to shorten a human life have been investigated, including radiation impact produced by natural background and routine operations of nuclear enterprises. The American specialists wished to answer the following question: how long could the Loss-of-Life-Expectancy be depending on personal life choices, occupations, type of diseases, circumstances and, at last, on electricity generation technologies?

The following unexpected results have been obtained.

Loss-of-Life-Expectancy for life choices

1. Unmarried males – 3000 days.
2. Smoking males – 2590 days.
3. Unmarried females – 1600 days.
4. Smoking females – 1530 days.
5. Alcohol abuse – 365 days.
6. Vehicular accidents – 207 days.

Loss-of-Life-Expectancy for various occupations

1. Living in poverty – 3500 days.
2. Low social-economic status – 1670 days.

3. Coal miners – 1100 days.
4. Grade school drop-out – 800 days.
5. Unemployment – 500 days per one year of unemployment.

Loss-of-Life-Expectancy for health ailments

1. Heart diseases – 1607 days.
2. Cancer – 1247 days.
3. Overweight – 1020 days.
4. Stroke – 510 days.
5. Pneumonia, influenza – 105 days.

Loss-of-Life-Expectancy for circumstantial situations

1. Being male versus female – 2800 days.
2. Being black versus white – 2000 days.
3. All accidents – 360 days.
4. Married to smoker – 50 days.
5. Ionizing irradiation:
   a. Radon in house - 29 days.
   b. Living near NPP – 10 hours.
6. Nuclear power industry:
   a. Evaluated by the Union of Concerned Scientists – 2 days.
   b. Evaluated by the Nuclear Regulatory Commission – 1 hour.

Loss-of-Life-Expectancy for electricity generation technologies

1. Coal-fired plants – 23 days.
2. Oil-fired plants – 4 days.
3. Natural gas-fired plants – 2,5 days.
4. Nuclear power plants:
   a. Evaluated by the Union of Concerned Scientists – 2 days.
   b. Evaluated by the Nuclear Regulatory Commission – 1 hour.

So, it becomes evident that natural radiation background and nuclear power plants are very far from being the main factors for a human life shortening of American people.

2.4. Principle of radiation equivalency

Russian nuclear specialists put forward the principle of radiation equivalency between natural uranium, on the one hand, consumed by nuclear power plants for energy generation and the radiowastes, on the another hand, as harmful by-products of this process.

The essence of the radiation equivalency principle can be defined by the following considerations. Natural uranium is used to fabricate nuclear fuel. Nuclear fuel burning-up at NPP is able to yield the wholesome products (electrical and thermal energy, secondary fuel) as well as the undesirable by-products (radiowastes with fission products and minor actinides as main components). So, initial material is a natural uranium which can be converted into nuclear fuel and burnt-up in nuclear power reactors. The burning-up process is accompanied by generation of the wholesome and harmful products. The problem is to organize the nuclear fuel burn-up and the radiowastes management by such a way that ecological danger from the radiowastes became approximately the same as ecological danger from natural uranium for a relatively short time period. In other words, if M_U mass of natural uranium is burnt-up at NPP with undesirable accumulation of the radiowastes (M_{RAW} – mass of the radiowastes), then potential biological danger of both masses must be made equivalent to each other by using a certain scheme of NM flows within a nuclear energy system. Since radioactivity is the most dangerous property of the radiowastes, the radiation equivalency principle is proposed, i.e. radioactivities (or radiotoxicities in a more general sense) of natural uranium and the radiowastes accumulated in the uranium energy utilization should be balanced. This principle can keep total radioactivity of the Earth at a constant, unchangeable level.

Large-scale system of lead-cooled fast reactors fueled with mixed uranium-plutonium nitride (BREST-type reactors) is under intense development now in Russia as one of the most promising options for real implementation of the radiation equivalency principle. Numerical and theoretical studies have demonstrated that fast BREST-type reactors possess the property of inherent safety, i.e. any reactivity-induced accidents (like Chernobyl catastrophe) can be excluded deterministically. No physical phenomena and erroneous actions of NPP operators are able to initiate these severe accidents.

Unique neutron abundance and high neutron flux in fast BREST-type reactors make it possible to organize full recycle of main fissile materials (uranium and plutonium isotopes), effective transmutation of minor actinides (neptunium, americium and curium isotopes) and some long-lived fission products (99Tc and 129I). Optimal configuration of material flows in the emerging nuclear energy system of fast BREST-type reactors and the SNF reprocessing facilities can reach the balanced

situation between potential biological danger of the accumulated radio-wastes and potential biological danger of the consumed natural uranium after 200-300 years of the system operation.

2.5. Two approaches to evaluating the radiation effects on human organism

There are two contrary approaches to determining the dependency of harmful irradiation consequences for a human health on the absorbed radiation dose.

The first approach presumes that any level of the radiation dose, no matter how small, absorbed by a human body can contribute to the risk of health ailments, and the amount of radiation damages is directly proportional to the radiation dose. Presently, the overwhelming majority of environmentalists has adopted the linear hypothesis for dependency of radiation damage on the radiation dose. This approach is named as the "non-threshold" theory. Though the scope of experimental data on radiation damages from small radiation doses is evidently insufficient for unconditional corroboration of the non-threshold theory, nevertheless, the non-threshold approach is regarded now as a basic one and widely used in the regulatory radiation safety guidelines for all operations with radioactive materials. Foundation for such a conservative (i.e. with a certain reserve) approach is quite clear and still acceptable now: even if we are mistaken, we are mistaken in the better direction, namely in the direction towards the stronger (maybe, excessively stronger) protection of the humankind against any negative consequences of any ionizing irradiation.

The weakest point of the non-threshold theory is a shortage or even full absence of reliable experimental data about radiation effects from small radiation doses. Available now experimental information allows us to say with a great confidence about linear dependency of radiation damages on the radiation doses only for the doses above 1 Sv. The dose range from zero to one sievert remains an unexplored area, field for a lot of various guess-works and speculations.

Opponents of the non-threshold theory put forward the following counter-arguments:

1. Some experimental studies with animals have revealed that complete isolation from natural radiation background increased their mortality.

Moreover, the low-dose irradiation can be actually beneficial for some animals and plants.

2. Many low-level impacts can produce a favorable effect on a human health while the same but high-level impacts can be very harmful. For example, one pellet of a soporific drug saves from insomnia while taking of many the same pellets is the most widely used form of suicide. The morning cold-water douche can trigger the restorative mechanisms in a human body, and they overcome the initial negative cold effect with a great reserve. At the same time, even relatively short staying in cold winter water can lead to the lethal outcome.

The following conclusions can be derived from these counter-arguments.

1. Full exclusion of natural radiation background can produce a negative biological effect on a human organism. Probably, low-dose irradiation is able to suppress some pathological phenomena and intensify some restorative forces.

2. If the radiation dose increases from zero to the level of natural radiation background, then a certain strengthening effect (like cold-water douche) arises, the harm caused by complete exclusion of natural background drops down and, maybe, changes its sign. In other words, low-level radiation doses can produce a positive healthy effect. It is interesting to note here that mountain-dwellers who live under substantially higher natural radiation background (radioactive rocky formations plus cosmic rays) distinguish, as a rule, with their robust health and enviable longevity.

3. If the radiation dose continues increasing above the level of natural background, then its sanitary effect is accompanied by the gradually enhancing negative consequences. At a certain dose, the positive impact reaches its maximal value, the negative factors begin prevailing. The "dose-damage" curve (Fig. 2.1) crosses the zero value, and the corresponding dose is a true radiation threshold beyond which ionizing radiation can produce negative effects only.

4. When the radiation dose increases up to one sievert, the "dose-damage" curve will coincide with linear dependency of the non-threshold theory.

So, the "dose-damage" curve can be presented in the form shown in Fig. 2.1.

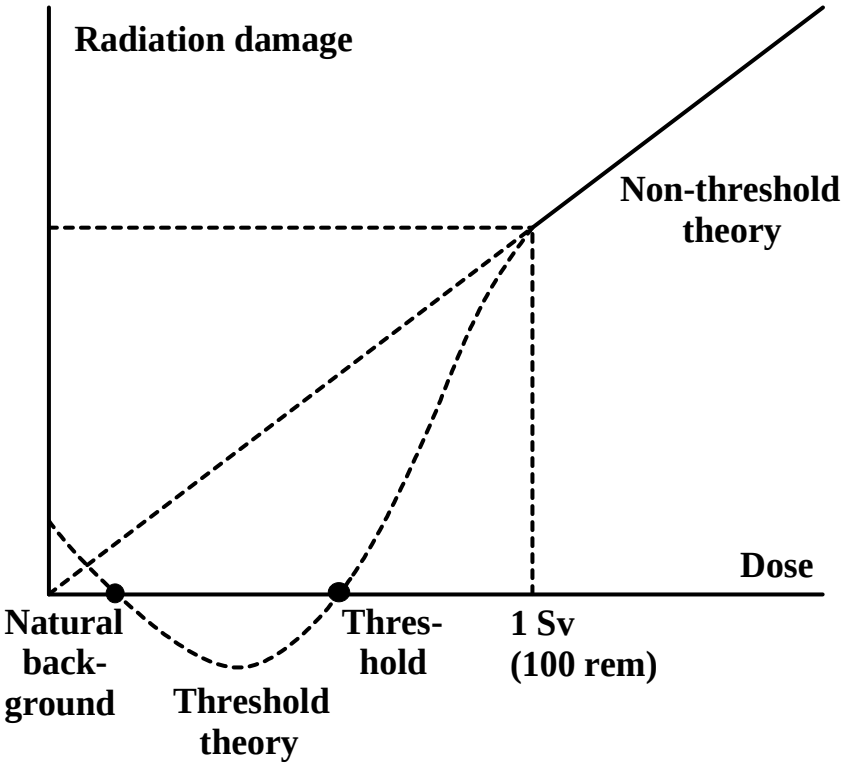


Fig. 2.1. Two approaches to evaluating the radiation damage

Control questions to Chapter 2

1. What ecological effects can nuclear enterprises produce?
2. Call main components of natural radiation background.
3. Call main ecological effects produced by mining and HM-treatment of uranium ore.
4. Call main ecological effects produced by nuclear power plants during their normal operation.
5. What is the main ides of the radiation equivalency principle?
6. What are the differences between the threshold and non-threshold approaches to evaluating the radiation damages for a human organism?

CHAPTER 3. PROBLEMS OF FISSILE MATERIALS NON-PROLIFERATION AT VARIOUS STAGES OF THE CLOSED NFC

Nuclear technologies deal with fissile nuclear materials which could be used as a charge of a nuclear explosive device. Therefore, one of the main tasks in development and routine application of nuclear technologies is a strict NM control at all NFC stages in order to prevent NM usage in any illegal actions.

The following three ways towards NM diversion from a peaceful civilian (mainly, energy) use to any illegal (mainly, military of terrorist) applications are estimated as possible ones:

1. Forcible theft of nuclear materials resulted from a terrorist attack on a nuclear object or a transportation tool.

Prevention of the forcible NM theft is a major mission of NM physical protection system (PPS).

Main PPS components:

a. System of physical barriers (fences) to prevent any intrusion of potential proliferators (or terrorists) onto the territory of a nuclear object or into the premises where NM can be placed.

b. Systems of exterior and interior sensors to detect any intrusion of potential proliferators through the outer perimeter of a nuclear object and through the inner barriers towards NM storage and utilization points.

c. System of TV-surveillance for the area adjacent to the outer perimeter and for the inner premises where NM can be stored or utilized.

d. System of special means to delay movement of potential proliferators through the territory of a nuclear object.

e. System of the armed guard forces for detection, interception and detention of potential proliferators.

2. Covert theft of nuclear materials by staff members (internal adversaries) of a nuclear object (maybe, by gradual theft of very small, undetectable NM quantities for a long time).

Prevention of the covert NM theft is a major mission of NM control and accountability (MC&A) system.

Main components of MC&A system:

a. Video-surveillance of NM stored in a form of accountable items (containers).
b. Administrative measures on access control to NM storage and utilization points.
c. Division of a nuclear object into a series of NM balance areas (MBA) equipped with sensors capable to detect NM movements between different MBA.
d. Computerized NM accounting system equipped with remote terminals in key MBA points which can transmit NM-related information to a central computer with application of the information security software tools.
e. Periodical NM physical inventory taking for NM in a form of accountable items and for NM in a bulk-form with application of temper-indicating devices (TID).
f. Selective examination of NM containers and NM in a bulk-form with application of destructive and non-destructive experimental methodologies.

3. Covert NM switching over to an illegal military purpose sanctioned by national government.

Prevention of the covert NM switching over to any military aims sanctioned by national government is a major mission of the international treaties intended to control peaceful use of nuclear energy.

The following main international treaties on peaceful use of nuclear energy were concluded and signed:
a. The Non-Proliferation Treaty (NPT), or the Treaty on the Non-Proliferation of Nuclear Weapons, was signed on July 1, 1968, and entered into force on March 5, 1970. On May 11, 1995, the Treaty was extended indefinitely.
b. The EURATOM Treaty, i.e. the Treaty establishing the European Atomic Energy Community, was signed in Rome, on March 25, 1957.
c. Nuclear Suppliers Group (NSG) was founded in November 1975 as a response to the Indian nuclear test. The NSG is a multi-national body concerned with reducing nuclear proliferation by controlling the export and re-transfer of materials that may be applicable to nuclear weapons development.
d. The Treaty of Tlatelolco, i.e. the Treaty for the prohibition of nuclear weapons in Latin America and Caribbean, was signed on February 14,

1967, in Mexico City. Under the Treaty, the states-parties agreed to prohibit and prevent the "testing, use, manufacture, production or acquisition by any means whatsoever of any nuclear weapons" and the "receipt, storage, installation, deployment and any form of possession of any nuclear weapons".

e. The Treaty of Rarotonga, i.e. the South Pacific Nuclear-Free Zone Treaty, was signed on August 6, 1985, on the Rarotonga Island (Cook Islands). The Treaty bans the use, testing and possession of nuclear weapons within the borders of the zone.

f. The Comprehensive Nuclear Test Ban Treaty (CTBT) is a multilateral treaty by which the states-parties agreed to ban all nuclear explosions in all environments for military or civilian purposes. The Treaty was adopted by the UN General Assembly on September 10, 1996.

The main State-level mechanism of nuclear non-proliferation control is based on regular inspections performed by the IAEA experts who examine independently the really available NM quantities at nuclear objects under the IAEA jurisdiction.

3.1. Factors of NM attractiveness at various NFC stages

Applicability of nuclear materials to their use in nuclear explosive devices can be characterized by the following factors of NM attractiveness:

1. Quantity and quality of nuclear materials needed to produce a nuclear explosive device.

Minimal mass of fissile NM in which the chain fission reaction can take place is a critical mass. For example, critical masses of ^{235}U, ^{239}Pu and ^{233}U are equal approximately to 50 kg, 15 kg and 17 kg, respectively. These values were obtained for metallic spheres without neutron reflector. Effective neutron reflector can reduce the critical mass nearly twice.

The IAEA introduced a special unit for measuring NM mass, namely "Significant Quantity" (SQ). Detection of NM disbalance exceeding 1 SQ is a reason for the IAEA inspectors to initiate a special investigation and, if necessary, appeal to the UN Security Council for application of appropriate sanctions. The Significant Quantity is nearly half the critical mass of fissile materials in the form of non-reflected metallic sphere.

$$1\ SQ\ (^{239}Pu,\ ^{233}U) = 8\ kg;\ 1\ SQ\ (^{235}U) = 25\ kg.$$

The critical masses of uranium and plutonium dioxides are larger than those for metal uranium and plutonium by a factor about 1,5.

2. NM accessibility, simplicity of NM theft, detectability of NM theft.

3. Simplicity of NM conversion into charge of a nuclear explosive device. Is it sufficient to use mechanical or chemical treatment only? Is it necessary to apply some sophisticated technologies of isotope separation?

These factors define the following values:

1. Duration of the time interval needed to manufacture a nuclear explosive device by the adversaries or to undertake proper countermeasures by the security forces.

2. Scale of material, industrial and financial resources needed to manufacture a nuclear explosive device.

All the factors should be taken into account to estimate various NFC stages from the standpoint of nuclear non-proliferation, i.e. NM attractiveness for theft and further manufacturing of nuclear explosive devices. It is a difficult task to give an exact quantitative estimation of the NFC stages on their attractiveness for potential nuclear proliferators but some qualitative estimates were made by the US experts (Fig. 3.1).

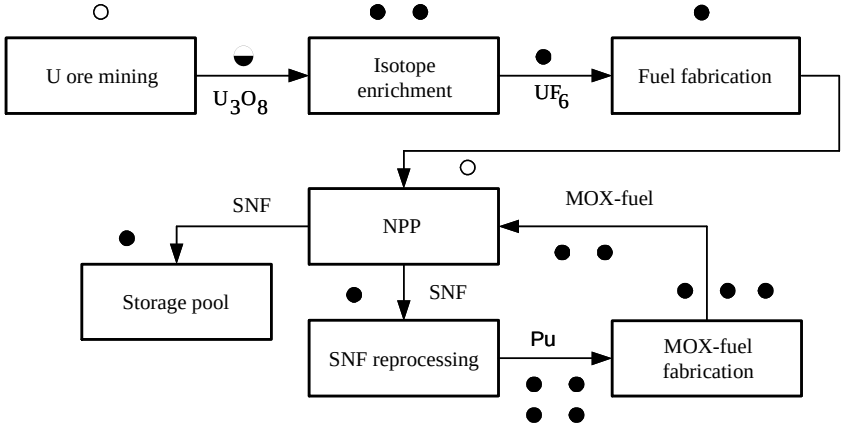


Fig. 3.1. Attractiveness of the NFC stages on the reasonability of NM theft

The number of black points at each NFC stage characterizes its attractiveness for nuclear proliferators. Really, the NFC stages are estimated in a four-point scale. As is seen, the most attractive NFC stages are related with isotope uranium enrichment, SNF reprocessing, plutonium extraction and fabrication of mixed uranium-plutonium oxide fuel.

The following features of all NFC stages must be considered from nuclear non-proliferation point of view:

1. Mining and primary treatment of uranium ore.

NM vulnerability to theft (VT).

In order to produce 25 kg of weapon-grade uranium (> 90% ^{235}U), it is necessary to use nearly 5000 kg of natural uranium, or about 5000 t, in average, of uranium ore. Imperceptible theft of so large amount of uranium ore is a quite impossible event. Thus, the VT value is low.

NM vulnerability to diversion (VD)

Uranium mines and plants for primary treatment of uranium ore are outside of the IAEA safeguards. Thus, the VD value is high.

Risk of nuclear weapon proliferation (RP)

The RP value is low because natural uranium can not be used as a charge of a nuclear explosive device.

2. Production of uranium hexafluoride for isotope enrichment

NM vulnerability to theft (VT)

The VT value is low like at the mining of uranium ore.

NM vulnerability to diversion (VD)

The VD value can cover the range from low to high depending on applications of the IAEA safeguards, i.e. L/H(IAEA).

Risk of nuclear weapon proliferation (RP)

The RP value is low because natural uranium can not be used as a charge of a nuclear explosive device.

3. Uranium enrichment with isotope ^{235}U

NM vulnerability to theft (VT)

The VT value is high. Relatively small amount (~25 kg) of weapon-grade uranium is required to manufacture a nuclear explosive device. Even one man is able to handle with so small mass.

NM vulnerability to diversion (VD)

The VD value can cover the range from low to high depending on applications of the IAEA safeguards, i.e. L/H(IAEA).

Risk of nuclear weapon proliferation (RP)

The RP value is high. The Nuclear Suppliers Group put an informal embargo on export of the isotope separation technologies.

4. Fabrication of nuclear fuel (fuel rods and fuel assemblies)

NM vulnerability to theft (VT)

The VT value is low. One fuel assembly weighs 300-500 kg depending on the reactor type. So, a special transportation tool has to be used for theft of even one fuel assembly.

NM vulnerability to diversion (VD)

The VD value can cover the range from low to high depending on applications of the IAEA safeguards, i.e. L/H(IAEA).

Risk of nuclear weapon proliferation (RP)

The RP value can cover the range from low to high depending on the value of uranium enrichment, i.e. L/H(Enrichment).

5. Use of nuclear fuel at NPP

NM vulnerability to theft (VT)

The VT value is low because of large weight, radioactivity and disposition of fuel assemblies in a nuclear reactor core.

NM vulnerability to diversion (VD)

The VD value can cover the range from low to high depending on applications of the IAEA safeguards, i.e. L/H(IAEA).

Risk of nuclear weapon proliferation (RP)

The RP value can cover the range from low to high depending on the value of uranium enrichment, i.e. L/H(Enrichment).

6. Interim storage of SNF

NM vulnerability to theft (VT)

The VT value is low because of large weight, radioactivity and residual heat generation of spent fuel assemblies.

NM vulnerability to diversion (VD)

The VD value can cover the range from low to high depending on applications of the IAEA safeguards, i.e. L/H(IAEA).

Risk of nuclear weapon proliferation (RP)

The RP value can cover the range from low to high depending on the value of uranium enrichment, i.e. L/H(Enrichment).

7. SNF reprocessing

NM vulnerability to theft (VT)

The VT value is high. SNF reprocessing technologies deal with highly radioactive and heat-generating materials. That is why only remote equipment is used to separate spatially staff members and dangerous nuclear materials. However, at some steps of the SNF reprocessing, plutonium-containing materials may be more accessible for theft.

NM vulnerability to diversion (VD)

The VD value can cover the range from low to high depending on applications of the IAEA safeguards, i.e. L/H(IAEA).

Risk of nuclear weapon proliferation (RP)

The RP value is high. SNF reprocessing plants can produce either weapon-grade plutonium or, at least, reactor-grade plutonium with relatively worse isotope composition but also suitable for manufacturing of a nuclear explosive device with significantly lower energy yield. The Nuclear Suppliers Group put an informal embargo on export of the SNF reprocessing technologies.

8. Ultimate disposal of radioactive wastes

NM vulnerability to theft (VT)

The VT value is low because of intense radioactivity, residual heat generation and small content of fissionable nuclides.

NM vulnerability to diversion (VD)

The VD value is low because of small content of fissionable nuclides.

Risk of nuclear weapon proliferation (RP)

The RP value is low because of intense radioactivity, residual heat generation and small content of fissionable nuclides.

The factors defining threats from all the NFC stages to nuclear nonproliferation regime are gathered in Table 3.1.

3.2. Comparison of reactor types from viewpoint of nuclear nonproliferation

In addition to the NFC stages, different types of nuclear reactors can be characterized by different values of NM attractiveness from nonproliferation point of view. The following fuel parameters can be helpful for estimating attractiveness of nuclear reactors from this viewpoint:

1. Quantity and quality of fresh fuel loaded into the reactor cores.
2. Quantity and quality of spent fuel unloaded from the reactor cores.

For the beginning, the reactors fueled with highly-enriched, weapon-grade uranium (above 90% ^{235}U) are considered below.

Table 3.1

Danger from the NFC stages

NFC stage	Vulnerability to theft	Vulnerability to diversion	Proliferation risk
Mining of uranium ore	Low	High	Low
UF_6 production	Low	L/H(IAEA)	Low
Isotope enrichment	High	L/H(IAEA)	High
Fabrication of nuclear fuel	Low	L/H(IAEA)	L/H(Enrichment)
NPP	Low	L/H(IAEA)	L/H(Enrichment)
Interim SNF storage	Low	L/H(IAEA)	L/H(Enrichment)
SNF reprocessing	High	L/H(IAEA)	High
Ultimate disposal of RAW	Low	Low	Low

1. Research reactors

Some research reactors are still using highly-enriched, weapon-grade uranium fuel in a very attractive form of pure metals or metal alloys. However, thermal power of the research reactors in operation now is relatively low (at the level of several megawatts) and, therefore, total mass of ^{235}U in their cores is well below 10 kg.

According to the IAEA recommendations, the national programs on conversion of the research reactors from highly enriched to middle-enriched (below 20% ^{235}U) uranium fuel are currently underway in some countries. By the way, critical mass of 20%-uranium is evaluated as large as 830 kg. The reduced uranium enrichment can lead to larger sizes of the reactor core, larger amounts of loaded fresh fuel, but total mass of ^{235}U can remain at the same or even lower level thanks to the better neutron economy in the larger reactor cores (lower neutron leakage).

Secondary nuclear fuel is not produced practically by the research reactors in operation now because of low neutron flux and small amount of fertile nuclides.

2. High-Temperature Gas-Cooled Reactors (HTGR)

These reactors are fueled with highly enriched uranium (93% ^{235}U) as a fissile material and natural thorium as a fertile material. HTGR-

type reactors use the dispersed fuel in form of spherical micro-particles (500-800 microns in diameter) inside of multi-layer cladding made of pyrolytic carbon and silicon carbide. The fuel micro-particles are uniformly dispersed in graphite matrix that is used further to fabricate spherical (~6 cm in diameter) or prismatic fuel elements.

TRISO-type micro-particles consist of fuel kernel coated with three-layer cladding (low-density pyrolytic carbon, silicon carbon and high-density pyrolytic carbon).

BISO-type micro-particles consist of fuel kernel coated with two-layer cladding (low-density pyrolytic carbon and high-density pyrolytic carbon).

The HTGR-770 project presumes that initial fuel loading consists of 8100 kg ^{232}Th as thorium dioxide in BISO-type micro-particles and 700 kg ^{235}U as uranium carbide in TRISO-type micro-particles. By the end of irradiation cycle the reactor core contains about 7500 kg ^{232}Th, 40 kg ^{235}U and 180 kg ^{233}U (secondary fuel), or ~230 kg ^{233}U/GWe·year.

3. Light-water reactors (LWR)

3a. VVER-type reactors

Power VVER-type reactors are fueled with low-enriched (4-5% ^{235}U) uranium dioxide. As a rule, initial fuel loading of VVER-1000 is equal to about 100 t UO_2. Secondary fuel is produced with a specific rate ~200 kg Pu/GWe·year.

However, isotope composition of the produced plutonium extracted from spent fuel is far from optimal suitability for manufacturing of a nuclear explosive device. Typical weapon-grade plutonium contains mainly ^{239}Pu and below 7% ^{240}Pu. Typical plutonium extracted from spent fuel of VVER-type reactors (reactor-grade plutonium) contains about 2% ^{238}Pu, 58% ^{239}Pu, 25% ^{240}Pu, 11% ^{241}Pu and 4% ^{242}Pu, i.e. ~71% of fissile plutonium isotopes. Critical mass of metal reactor-grade plutonium is larger on 50% than critical mass of metal weapon-grade plutonium (23 kg via 15 kg). But this is not the most major aspect. Reactor-grade plutonium contains larger ^{240}Pu quantity (by a factor of 4) than weapon-grade plutonium. The larger quantity of ^{240}Pu can sharply reduce (roughly by a factor of 30) energy yield of nuclear explosive devices because ^{240}Pu is an intense emitter of spontaneous fission neutrons. These neutrons can cause untimely premature initiation of the

chain fission reaction in a nuclear charge (the pre-detonation effect) and, thus, energy yield of nuclear explosion will not exceed 3% of nominal energy yield. According to some numerical evaluations, if Hiroshima-type atomic bomb (nominal energy yield - 20 kt TNT) would be made of the reactor-grade plutonium, then the most probable energy yield would be about 600 t TNT. Nevertheless, this value is a high enough energy equivalent. As is known, masses of usual explosives exploded in Moscow and caused many human victims were well below 100 kg TNT.

3b. RBMK-type reactors

In some publications the RBMK reactors are named as Chernobyl-type reactors because the world-wide known Chernobyl accident (1986) occurred in the RBMK reactor. The RBMK reactors use reactor-grade graphite as a neutron moderator, and boiling light water as a coolant. The light-water coolant circulates in vertical technological channels that transpierce through the graphite stack of the reactor core (diameter of the graphite stack - ~12 m, height - ~8 m). The heat-generating cassettes consisting of two consecutively coupled fuel assemblies (length – 3,5 m each) are inserted into the technological channels.

RBMK-type reactors are fueled with low-enriched (1,8-2% ^{235}U) uranium dioxide. As a rule, initial fuel loading of RBMK-1000 is equal to about 150-180 t UO_2. Secondary fuel is produced with a specific rate ~250 kg Pu/GWe·year. Isotope composition of reactor-grade plutonium extracted from SNF of the RBMK-type reactors is inferior to reactor-grade plutonium extracted from SNF of the VVER-type reactors in respect of fissile isotopes content and in respect of ^{240}Pu content. Typical plutonium extracted from spent fuel of RBMK-type reactors contains about 45% ^{239}Pu, 36% ^{240}Pu, 11% ^{241}Pu and 8% ^{242}Pu, i.e. ~56% of fissile plutonium isotopes.

A particular threat of the RBMK-type reactors to nuclear non-proliferation is caused by their principal capability to work in the continuous refueling operation mode without reactor outages for refueling. Under this operation mode, fuel exposure time may be chosen short enough to produce plutonium with isotope composition very suitable for manufacturing of a nuclear explosive device.

Heavy-water CANDU-type reactors

The CANDU-type reactors are able to use even natural uranium containing only 0,72% ^{235}U as fuel material. Initial fuel loading of CANDU-600 is equal to about 100 t UO_2. Secondary fuel is produced with a specific rate ~350 kg Pu/GWe·year. Isotope composition of reactor-grade plutonium extracted from SNF of the CANDU-type reactors is very close to reactor-grade plutonium extracted from SNF of the VVER-type reactors in respect of fissile isotopes content and in respect of ^{240}Pu content. Typical plutonium extracted from spent fuel of CANDU-type reactors contains about 66% ^{239}Pu, 27% ^{240}Pu, 5% ^{241}Pu and 2% ^{242}Pu, i.e. the same 71% of fissile plutonium isotopes and almost the same content of ^{240}Pu (25% in VVER via 27% in CANDU).

The CANDU-type reactors, quite like the RBMK-type reactors, can represent a potential threat to nuclear non-proliferation regime because their operation modes with continuous refuelings can be easily re-tuned (by proper selection of fuel irradiation time, for instance) to form the best conditions for wide-scale production of weapon-grade plutonium. Besides, the operation mode with continuous refueling can require a permanent presence of the IAEA inspectors to control proper utilization of primary fuel and accumulation of secondary fuel, potentially dangerous material for non-proliferation of nuclear weapons.

By the way, plutonium for the first atomic bombs exploded in July 1945 in the USA and in August 1945 over Japan was produced by heavy-water reactors for about half a year.

5. Liquid-metal fast breeder reactors (LMFBR)

Currently, the LMFBR-type reactors are still loaded with uranium oxide (UOX) fuel, not mixed uranium-plutonium oxide (MOX) fuel as it was anticipated earlier. The UOX fuel is based on middle-enriched uranium (15-25% ^{235}U). Initial fuel loading of LMFBR-1000 is equal to about 10-15 t UO_2. Secondary fuel is produced with a specific rate ~1500 kg Pu/GWe·year in the once-through NFC option. If the NFC becomes closed, then large fraction (up to 80%) of the produced plutonium is recycled to provide fuel self-sustainability of the LMFBR-producer, and net rate of plutonium production for other purposes is equal to ~250 kg Pu/GWe·year.

There are no intense neutron absorbers among fission products within high-energy range of the LMFBR-type reactors. That is why

typical values of fuel burn-up in the LMFBR-type reactors can reach ~100 GWd/t, or 10% HM, i.e. roughly twice higher than acceptable values of fuel burn-up in LWR. Thanks to the higher values of fuel burn-up and, as a consequence, longer fuel lifetimes, plutonium produced by the LMFBR-type reactors is characterized by such isotopic composition which is low suitable for manufacturing of nuclear explosive devices.

Some data on consumption of fresh primary fuel and production of secondary fuel are gathered in Table 3.2 for various reactor types.

Table 3.2

Loaded and unloaded fuel of nuclear reactors

Reactor type	Primary fuel	Secondary fuel, kg/GWe·year	Comments
Research reactors	5-10 kg (90% ^{235}U)	—	Small power
HTGR-770	8,1 t ThO_2 0,7 t UC(93%^{235}U)	230	—
VVER-1000	100 t UO_2 (3-5% ^{235}U)	200 (25% ^{240}Pu)	—
RBMK-1000	150-180 t UO_2 (1,8-2% ^{235}U)	250 (36% ^{240}Pu)	Continuous refuelings
CANDU-600	100 t UO_2 (0,7% ^{235}U)	350 (27% ^{240}Pu)	Continuous refuelings
LMFBR-1000	10-15 t UO_2 (15-25% ^{235}U)	1500-Open NFC 250-Closed NFC	—

3.3. Advanced proliferation-resistant SNF reprocessing technologies

3.3.1. Aqueous SAFAR reprocessing technology

Main idea of nuclear non-proliferation ensuring within the frames of the SAFAR (Safeguarded Fabrication and Reprocessing) technology consists in incomplete separation of uranium, plutonium and fission products. Consequently, at any stage of the SAFAR-technology, pluto-

nium can not be extracted in the form suitable for its diversion and manufacturing of nuclear explosive devices.

The following specific features can distinguish the SAFAR-technology from traditional PUREX-technology:

1. Incomplete separation of plutonium from uranium and fission products. Plutonium and uranium are recovered jointly (co-extraction) by using only two cycles of the extraction - re-extraction process. As a result, plutonium is deliberately contaminated with uranium and radioactive fission products (~1% of their initial content). The decontamination factors are about 100 instead of 10^6-10^7 in traditional PUREX-technology.
2. Pure uranium and plutonium dioxides are not produced. Final products of the SAFAR-technology are spherical micro-granules of mixed oxide uranium-plutonium (MOX) fuel. These micro-granules are formed by using the sol-gel process that is described below.
3. The re-fabricated MOX-fuel is characterized by the elevated radioactivity due to the relatively large content of fission products. The elevated radioactivity of the MOX-fuel can be estimated as a certain positive factor from nuclear non-proliferation point of view: the radiation barrier against the MOX-fuel diversion for manufacturing of nuclear explosive devices; unattractiveness for thefts; easy control (high detectability) of any MOX-fuel movements. However, some additional countermeasures must be undertaken to enhance radiation protection of the staff involved.

The sol-gel process, as a key stage of the SAFAR-technology, should be described in more details. Sol is a suspension-like substance, gel is a colloid, or a jelly-like substance. So, the “sol-gel” term means a gradual densification of the SNF solution through consecutive transformations from the liquid SNF solution into the SNF suspension, then into the SNF colloid and, ultimately, into the solid MOX-fuel granules followed by the fuel pelletization. Main mission of the sol-gel process is to avoid technological operations with finely dispersed powders of uranium and plutonium dioxides and to work with sufficiently large MOX-fuel granules.

The sol-gel process uses the acidic SNF solution after two cycles of the extraction – re-extraction process for partial removal of fission products as an initial feed material. So, the SAFAR-technology can be

regarded as an advanced version of the solvent-extraction PUREX-technology. Then, the following operations are performed:

1. Addition of the chemical reagents which are able to upgrade alkaline properties of the SNF solution (urea $(NH_2)_2CO$, for example).
2. Infusion of the SNF solution into a water-absorbing organic material (ethyl-benzoate, for example). This infusion converts the SNF solution into the colloid-like substance $(U,Pu)O_2(OH)_{0,4}(NO_3)_{1,6}$.
3. Injection of the colloid-like substance into an ammonia-based organic material for further gradual dehydration. This injection converts the colloid-like substance into the jelly-like spherical granules $(U,Pu)O_2(OH)_2$ 0,5 NH_3 0,5 H_2O with typical sizes within the range of 40-100 microns, i.e. they are large enough for further pelletization (cold pressing and sintering).
4. Thermal treatment of the jelly-like granules with gradual elevation of temperature. Residual ammonia-based organics is removed at 95^0C. Ultimate dehydration occurs at 125-200^0C with the formation of $(U,Pu)O_2(OH)_4$. All residual organic substances are completely evaporated at 300-400^0C. Ultimately, solid MOX-fuel granules are calcined at 400-500^0C.
5. Fabrication of fresh MOX-fuel rods and fuel assemblies.

The SAFAR-technology can be estimated as a well proliferation-protected spent fuel reprocessing technology because of the following main reasons:

1. Uranium and plutonium dioxides are extracted from the acidic SNF solution jointly. Plutonium dioxide is never separated from uranium dioxide.
2. The MOX-fuel granules are characterized by the enhanced radioactivity due to residual content of radioactive fission products after only two cycles of the solvent-extraction process.

Evolutionary progress of aqueous solvent-extraction technologies is being achieved now in following main directions:

1. Application of new, more effective and radiation-resistant organic extractants for selective removal of minor actinides from the raffinate produced at the extraction stage.
2. Implementation of some new stages intended to extract the most harmful fission products (extraction of radioiodine by volatilization, and technetium by the raffinate radiolysis).

3. Co-extraction of uranium, plutonium, neptunium and some fission products in order to enhance the barriers against unauthorized NM diversion to any undeclared applications.

According to these directions, some new advanced processes have been developed in France (COEX), Japan (NEXT), USA (family of UREX+ technologies) and so on. These technologies can apply new, more effective extractants, use new organizational schemes for material flows and the reprocessing operations. For example, all SNF reprocessing technologies from UREX+ family apply the following sequence of actions: at first, uranium is extracted from acidic SNF solution and, then, the residuals are treated to co-extract plutonium and minor actinides for further recycling in nuclear reactors.

For the last time the US specialists have developed a whole series of SNF reprocessing technologies under the common name UREX+ (Uranium Recovery by Extraction) family. This name underlines uranium extraction at the very initial stage of all UREX+ technologies. One else common property of the UREX+ technologies is the absence of pure plutonium among all the SNF reprocessing products.

Таблица 3.3

Products at various stages of the UREX+ technologies

Technology	Stages						
	1	2	3	4	5	6	7
UREX+1	U	Tc	Cs, Sr	MA, RE	FP		
UREX+1a	U	Tc	Cs, Sr	MA	FP		
UREX+2	U	Tc	Cs, Sr	Pu, Np	Am,Cm, RE	FP	
UREX+3	U	Tc	Cs, Sr	Pu, Np	Am, Cm	FP	
UREX+4	U	Tc	Cs, Sr	Pu, Np	Am	Cm	FP

Non-aqueous pyrochemical and pyrometallurgical technologies are currently considered as advanced and very promising options of SNF reprocessing but they are not ready yet for wide industrial usage.

3.3.2. Non-aqueous (dry) technologies for SNF reprocessing

The pyrochemical gas-fluoride technology

Main mission of the gas-fluoride technology is to provide SNF reprocessing without application of any liquid reagents (dissolvents, extractants and so on) and, as a consequence, without large volumes of liquid HLW. The gas-fluoride technology is based on different boiling temperatures, different volatilities and different abilities to be adsorbed by some adsorbents of uranium, plutonium and FP fluorides. At normal atmospheric pressure, uranium hexafluoride begins boiling at 56^0C, plutonium hexafluoride – at 62^0C, i.e. the boiling temperatures differ insignificantly. At these temperatures, main mass of fission products can form only non-volatile or weak-volatile fluorides.

The gas-fluoride technology includes the following main stages:

1. Thermal melting of fuel claddings at 1600^0C.
2. The SNF fluorination by gaseous fluorine-nitrogen mixture (20% F_2 and 80% N_2 for corrosion protection of technological vessels and pipelines) at 400^0C:

$$(U,Pu)O_2 + 4\ F_2 + 3\ H_2 \rightarrow (U,Pu)F_6 + 2\ HF + 2\ H_2O.$$

Main mass of FP fluorides (up to 85%) remains in the non-volatile sediment while well-volatile fluorides of uranium, plutonium and some fission products together with gaseous fission products (Xe, Kr, I) go out from spent fuel.

3. Freezing of FP fluorides in the fore-condensers at 27^0C. The fore-condenser is a cylindrical vessel into which the gas flow is introduced at an angle to vertical axis of the cylinder. Solid particles can strike against the cylinder wall and drop out of the gas flow. Weak-volatile fluorides of some fission products (Cs, Ru, Zr, and Nb) can be removed.
4. The gas flow passes through the column filled up with solid granules of sodium fluoride NaF at elevated temperature. At this stage, different sorption ability of NaF granules in respect of uranium, plutonium and FP fluorides is used to separate them. Uranium, neptunium and technetium fluorides are preferentially sorbed by NaF granules at 100^0C. Plutonium, ruthenium, zirconium and niobium fluorides are preferentially sorbed by NaF granules at 400^0C.

5. Desorption of uranium and plutonium hexafluorides from the surface of NaF granules by gaseous mixture (10% F_2 and 90% N_2) at 400^0C.

The following main drawbacks of the gas-fluoride technology can be mentioned:

1. Incomplete purification of uranium hexafluoride UF_6 from some FP fluorides. About 99,5% of uranium is extracted from spent fuel but uranium content in the recovered uranium hexafluoride flow equals 96% only. Thus, technological vessels and pipelines are contaminated with the remaining 3,5% of uranium.
2. Plutonium volatilization takes place with the lower efficiency than uranium volatilization. So, plutonium can contaminate technological equipment units too.
3. The gas-fluoride technology is not able to reprocess spent MOX-fuel because of large plutonium content.

FLUOREX technology

Japanese specialists have developed a new original FLUOREX technology that combined some stages of aqueous solvent-extraction PUREX technology with some stages of non-aqueous gas-fluoride technology.

The FLUOREX technology includes the following main stages:

1. Dismantling of fuel assemblies and chopping of fuel rods.
2. Decladding of fuel rods by alternating the oxidation reaction with oxygen and the reduction reaction with hydrogen. Uranium dioxide transforms into uranium octa-oxide via the oxidation reaction and returns into its initial state via the reduction reaction. The alternation continues until the extended fuel meat throws the weakened cladding off and becomes powder-like material,
3. Fluorination of spent fuel powder. Main uranium mass converts into gaseous uranium hexafluoride and escapes spent fuel together with some volatile and gaseous FP fluorides. Uranium (partially), practically full plutonium amount, main mass of fission products and minor actinides remain in the non-volatile sediment.
4. Uranium hexafluoride is separated from the accompanying substances by adsorption on NaF granules with high values of the decontamination factor ($10^6 \div 10^7$). Final product of this stage is a pure uranium hexafluoride separated from any impurities. Such uranium

hexafluoride can be then used to produce low enriched uranium fuel for light-water and heavy-water power reactors. Also, uranium hexafluoride can be re-enriched up to any desirable level.

5. Residuals of the fluorination process (small uranium quantity, almost full plutonium amount, fission products and minor actinides) are dissolved in nitric acid followed by the extraction - re-extraction treatment, quite like in traditional PUREX technology. Final product of this stage is the co-extracted uranium and plutonium which can be used to produce MOX-fuel pellets.

The pyrometallurgical technology for SNF reprocessing

The pyrometallurgical technology was initially intended for reprocessing of spent mixed metal uranium-plutonium fuel discharged from advanced fast breeder reactors with high breeding gain.

Till recently, the research fast reactor EBR-II was operated in Argonne National Laboratory (USA). The reactor was loaded with metal U-Zr fuel enriched up to 50% ^{235}U. The pyrometallurgical electrochemical refining technology was worked out just to reprocess SNF discharged from the EBR-II reactor. General scheme of the electrochemical refining facility is presented in Fig. 3.2.

The electrochemical refining facility represents a cylindrical vessel filled up with liquid cadmium in the bottom part and molten salts (mixture of potassium, sodium, calcium and barium chlorides) above the liquid cadmium layer (anode). From the top part, iron rod (cathode) is introduced into the molten chloride layer.

The electrochemical refining process includes the following main steps:

1. Spent fuel rods are chopped into short pieces and loaded into a perforated graphite basket.

2. The graphite basket with spent fuel pieces is loaded into the liquid cadmium layer.

3. Spent fuel is dissolved by liquid cadmium. Fuel claddings and some insoluble fission products can be removed for further treatment as solid radioactive wastes.

4. The dissolved SNF and fission products are distributed in layers of liquid cadmium and molten salts by such a way:

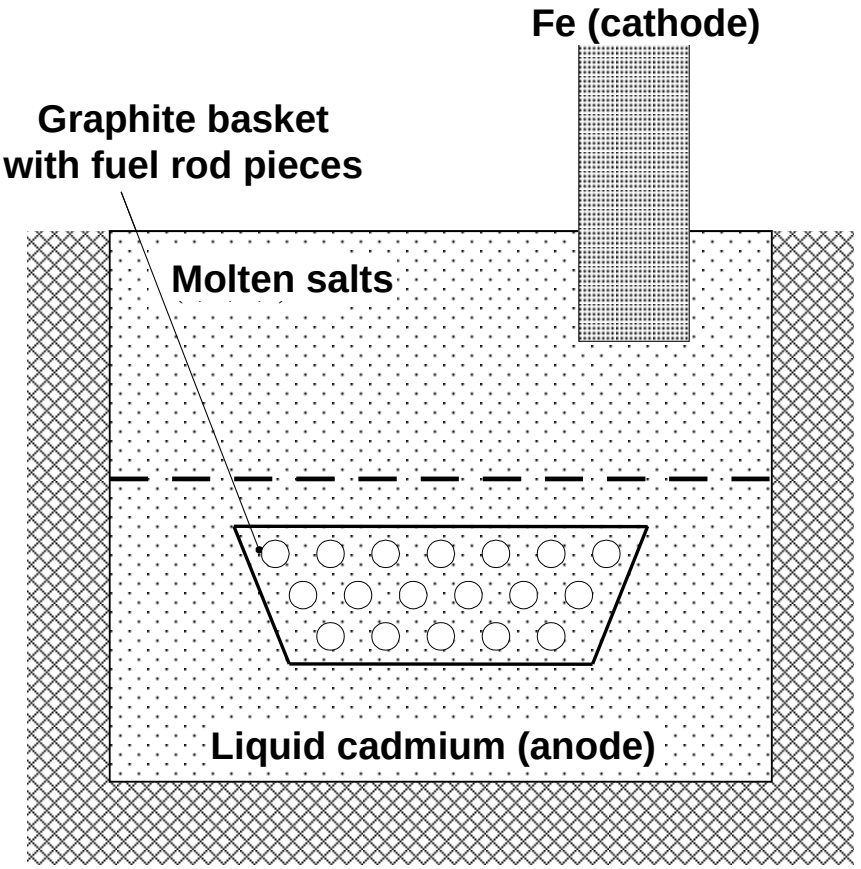


Fig. 3.2. General scheme of the electrochemical refining facility

a. Gaseous and volatile fission products escape the molten materials and enter into a gas cushion above the molten salt layer.
b. Alkaline-earth, rare-earth and alkaline fission products escape the liquid cadmium layer and enter into the molten salt layer.
c. Uranium and plutonium are contained in both layers.
5. When electrical current is switched on between the liquid cadmium anode and the iron cathode, some fission products, uranium, and plutonium escape the molten layers and precipitate on the iron cathode.

The cathode deposition is periodically taken off and melted down into a fresh nuclear fuel. The vacuum melting and casting of fuel rods are used at this step. The molten U-Pu-Zr alloy is poured into a cylindrical central hole of a quartz glass block. Upon completion of the cool-down phase, the quartz glass and the metal rod can be easily separated, and the metal rod is ready for manufacturing of a fuel element.

The finer purification of mixed uranium-plutonium fuel can be achieved by using the halide-slagging process. The following chemical reaction of the cathode deposition with magnesium chloride

$$2(U,Pu) + 3MgCl_2 \rightarrow 3Mg + 2(U,Pu)Cl_3;$$

can transform metal uranium and plutonium into their chlorides. Then, the uranium and plutonium chlorides can be returned into the molten salt layer, and the electrochemical refining is repeated. Even if the halide-slagging process is applied, the decontamination factors in respect of some undesirable FP can be increased up to 10^2-10^3 only via 10^6-10^8 in the solvent-extraction PUREX-technology.

The Integrated Fast Reactor concept

Nuclear specialists from Argonne National Laboratory (USA) have developed the project of a modular fast reactor with the integrated nuclear fuel cycle, where the pyrometallurgical electrochemical refining technology could be used for SNF reprocessing. The project was named Integrated Fast Reactor (IFR).

The IFR project can be characterized by the following specific features:

1. Modular small-power (170 MWe) fast reactor with metal U-Pu-Zr fuel and liquid-metal (sodium) coolant.
2. Small sizes of the reactor core and small fuel volume with increased uranium enrichment.
3. Co-allocation of NPP and pyrometallurgical facility for SNF reprocessing in a single site.

Advantages of the IFR project:

1. Passive nuclear safety thanks to excellent thermophysical properties of sodium coolant and large neutron leakage from the small-sized reactor core.
2. Factory-based manufacturing of the small-sized reactor with the subsequently high fabrication quality, reliability of the reactor operation, the subsequently low financial expenses, possibility of standardization.
3. Relatively short construction time with subsequently low risk of financial investments.
4. Possibility for a step-wise upgrading the NPP power on 170 MWe at each next step.
5. Enhanced proliferation resistance because of the following reasons:

a. Uranium and plutonium co-extraction without any their separation.

b. Weak purification of uranium and plutonium from fission products and minor actinides. Subsequently, the re-fabricated fuel is characterized by intense radioactivity, residual heat generation and intense emission of spontaneous fission neutrons.

c. No long-distant SNF transportation from the reactor to the SNF reprocessing facility is required here since they are allocated in a single site.

The only drawback of the IFR concept is related with its small power. Evidently, one large-scale power reactor is more economical than a system of small-scale reactors with the same total power.

DUPIC-technology

Name of the DUPIC-technology is an abbreviation from the “Direct Use of spent PWR fuel in CANDU reactors”. The DUPIC-technology is a product of the collaborative efforts undertaken by nuclear specialists from the USA, Canada and South Korea. The DUPIC-technology is a non-aqueous process with enhanced proliferation resistance.

As it follows from its name, main mission of the DUPIC-technology consists in the repeated use of spent fuel discharged from light-water power reactors of PWR-type in heavy-water power reactors of CANDU-type. Reasonability of this approach is based on the fact that spent PWR fuel can contain the amount of fissile isotopes large enough for further use as a fresh fuel composition of CANDU-type reactors. As

is known, the standard spent PWR fuel contains partially burnt-up uranium with residual enrichment at the level of ~0,9% ^{235}U and ~0,6% of reactor-grade plutonium with about 70%-content of fissile isotopes ^{239}Pu and ^{241}Pu. Thus, in total, the standard spent PWR fuel contains ~1,3% of fissile isotopes ^{235}U, ^{239}Pu and ^{241}Pu. Fortunately, heavy-water CANDU-type power reactors are able of functioning even if they are fueled with natural uranium (0,7% ^{235}U). So, spent PWR fuel can provide the twofold amount of fissile isotopes to make CANDU-type reactor operation feasible.

The DUPIC-technology provides spent PWR fuel reprocessing with application of thermal and mechanical procedures only. No components of aqueous, solvent-extraction, Pyrochemical and pyrometallurgical technologies are applied here.

Main stages of the DUPIC-technology:

1. Dismantling of spent fuel assemblies, withdrawal of spent fuel rods.
2. Transversal chopping of fuel rods into small pieces (~20 cm).
3. Longitudinal slitting of fuel claddings to weaken them.
4. Voloxidation, i.e. thermal treatment of fuel pieces in oxygen at 400^0C. Uranium dioxide UO_2 converts into uranium octa-oxide U_3O_8. This conversion causes increasing volume of fuel meat on ~30%, and fuel pieces throw their previously weakened cladding. In addition, fuel meat becomes more porous, partially transforms into a powder-like substance, some gaseous and volatile fission products (nearly all tritium, up to 40% ^{129}I, 70% ^{85}Kr and 90% ^{106}Ru) escape the porous fuel meat.
5. Treatment by the OREOX-process (Oxidation-Reduction of Oxide fuel). The OREOX is an oxidizing-reducing process with multiple interchange of the following reactions:

a. Oxidation by air at 450^0C. Uranium dioxide UO_2 converts into uranium octa-oxide U_3O_8, like the voloxidation reaction.

b. Reduction by (Ar - 4% H_2) gaseous mixture at 700^0C. Uranium octa-oxide U_3O_8 returns into uranium dioxide UO_2.

The multiple alternations of the oxidizing and reducing reactions can produce the dispersed UO_2 powder, and result in complete release of all gaseous and volatile fission products. Only solid fission products remain in the powder particles.

6. Manufacturing of UO_2 pellets from the dispersed UO_2 powder with sintering up to the pellet density about 96% of its theoretical value.

7. Manufacturing of fresh fuel rods and fuel bundles for CANDU-type reactors by using the traditional technology but all the manufacturing operations must be performed in hot cells, behind thick enough radiation shielding.

Specific features of the DUPIC-technology:

1. Full absence of any liquid solvents and extractants. Consequently:

a. Small volumes of radioactive wastes (gaseous and volatile FP, metal claddings of spent fuel rods).

b. Compact reprocessing facility and, therefore, a real possibility for co-allocation of NPP and the reprocessing facility in a single site.

2. No uranium – plutonium separation. No complete separation of uranium and plutonium from radioactive fission products. Only gaseous and volatile fission products can be released. Solid fission products remain in the reprocessed fuel.

3. Enhanced proliferation resistance of the DUPIC-technology because of the following reasons:

a. Intense radioactivity of fuel materials containing solid fission products.

b. No technological operations with separation of plutonium from uranium.

c. No long-distant transportations of fissile materials as NPP and the reprocessing facility can be co-allocated in a single site.

3.4. Control of NM non-proliferation at SNF reprocessing plants

The spent fuel reprocessing plant (SFRP) is one of the most sensitive NFC part from the viewpoint of nuclear non-proliferation ensuring. Main difficulty here is a plutonium non-proliferation control. In general, plutonium control and accountability at the SFRP encounters the following main challenges:

1. Large plutonium amounts. Throughputs of French SFRP are at the level of 800-900 SNF tons a year, at English SFRP - 1200-1500 SNF tons a year. In average, one SNF-TR ton contains 6-7 kg of plutonium, i.e. 5-10 tons of plutonium can go through the SFRP annually.

2. High required accuracy of the plutonium control. The significant plutonium quantity SQ(Pu) was adopted by the IAEA as 8 kg. The US Nuclear Regulatory Commission (NRC) has adopted even the stricter

constraint for the plutonium non-proliferation control, SQ(Pu) = 2 kg. Consequently, the available plutonium quantity must be controlled by the SFRP staff with accuracy about 1 kg of plutonium or below. At average annual SFRP throughputs of 10 plutonium tons, the accuracy of the control plutonium measurements must be at the level of ~0,01%. Really achievable accuracies of plutonium measurements are at the level of ~0,1%.

According to the requirements developed by Russian and American nuclear regulatory bodies, the maximal allowable plutonium disbalance at the SFRP must be equal to 0,1%, i.e. at the utmost achievable level of the measuring capabilities. The problem of so high required accuracy can be solved by using the following two ways:

1. The plutonium balance can be summed up at several time points a year, not once a year, when 5-10 plutonium tons have to be measured. The reasonable chosen number of physical inventory takings can decrease appropriately quantity of the Pu-bearing materials to be assayed. This way can be named as a time sharing.
2. The plutonium balance can be summed up at several material balance areas (MBA) of the SFRP. The reasonable chosen number of MBA can decrease appropriately quantity of the Pu-bearing materials available at each MBA to be measured. This way can be named as a space sharing.

Application of both ways (several physical inventories a year at several MBA separately) opens an opportunity to sum up the plutonium balance at the SFRP as a whole with the accuracy required by the nuclear regulatory guidelines.

3. Different states of the Pu-bearing materials. At the SFRP plutonium can be in various aggregate states (solid fuel and liquid SNF solution), in the aqueous and organic fractions, in solvates with different plutonium valencies. Different Pu-bearing materials can be characterized by different attractiveness for potential nuclear proliferators.

The following factors can be used to evaluate relative attractiveness of the Pu-bearing materials:

1. The density factor f_1 depends on the specific volume V of the Pu-bearing material per one gram of contained plutonium. Metal plutonium is chosen as a reference material with the highest relative attractiveness for potential proliferators of nuclear weapons, i.e. $f_1(V_{Pu\text{-}metal}) = 1$. As

density of metal plutonium equals 19,8 g/cm^3, the specific volume $V_{Pu\text{-}metal} \approx 5 \cdot 10^{-5}$ l/g. Thus, initial point of f_1 dependency on V equals unity at $V = 5 \cdot 10^{-5}$ l/g. The specific volumes of all other Pu-bearing materials are substantially larger, and their relative attractiveness appropriately diminishes (Fig. 3.3).

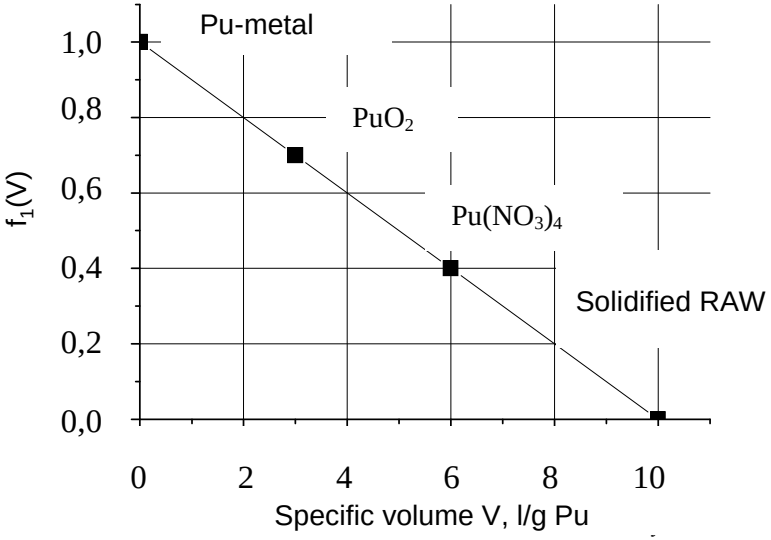


Fig. 3.3. Dependency of the density factor
on the specific volume of the Pu-bearing materials

2. The time factor f_2 depends on duration of the time interval needed to convert the Pu-bearing material into a charge of a nuclear explosive device by well-skilled specialists equipped with the most updated technical tools. Metal plutonium is chosen again as a reference material with the top relative attractiveness. It is assumed that one-week time interval would be required by the specialists to make a nuclear explosive device with metal plutonium as a charge material, i.e. f_2 (7 days) = 1. The time intervals needed for the specialists to convert all other Pu-bearing materials into a charge of a nuclear explosive device are substantially longer, and their relative attractiveness appropriately diminishes (Fig. 3.4).

3. The radiation factor f_3 (A) depends on radioactivity of the Pu-bearing materials per one gram of contained plutonium. The radiation factor of metal plutonium is assumed as unity.

The generalized attractiveness factor of various Pu-bearing materials is defined as a product of three aforementioned factors (the density, time and radiation factors). The generalized attractiveness factors of various Pu-bearing materials are presented in Table 3.4.

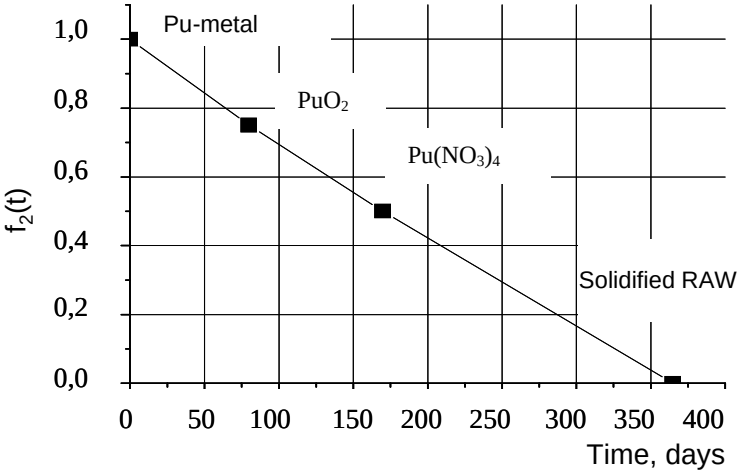


Fig. 3.4. Dependency of the time factor on duration of the time interval

Table 3.4

Attractiveness factors of the Pu-bearing materials

Material	$f_1(V)$	$f_2(t)$	$f_3(A)$	$f_1 \cdot f_2 \cdot f_3$
Pu-metal	1	1	1	1
PuO_2	0,70	0,90	1	0,63
$(U,Pu)O_2$	0,40	0,65	1	0,26
$Pu(NO_3)_4$	0,25	0,80	1	0,20
SNF solution	0,06	0,35	0,004	$8 \cdot 10^{-5}$
Spent fuel assembly	0,08	0,10	0,004	$3 \cdot 10^{-5}$
Solidified HLW	0,05	0,02	0,001	$1 \cdot 10^{-6}$

It is noteworthy that the density and radiation factors characterize the difficulty of obtaining the Pu-bearing materials while the time factor characterizes the difficulty of converting the Pu-bearing materials into a charge of a nuclear explosive device.

Control questions to Chapter 3

1. What stages of nuclear fuel cycles are the most dangerous for non-proliferation of nuclear weapons?
2. What types of nuclear reactors are the most dangerous for non-proliferation of nuclear weapons?
3. Call main stages of the SAFAR reprocessing technology.
4. Call main stages of the gas-fluoride reprocessing technology.
5. Call main stages of the electrochemical refining technology.
6. Call main stages of the DUPIC-technology.
7. Call and describe the attractiveness factors of plutonium-bearing materials.

CHAPTER 4. EXPLOSIVE PROPERTIES OF REACTOR-GRADE PLUTONIUM

Plutonium is an artificial chemical element that has no one stable isotopes. Fortunately, some radioactive plutonium isotopes can decay with half-lives of several thousand and tens of thousand years, i.e. they are sufficiently long-lived radionuclides for their civilian and military applications.

Plutonium was first synthesized in 1940 by the research team from the California University (USA) headed by G.T. Seaborg in experimental studies on bombarding of natural uranium by accelerated deuterons. It is interesting to tell about history of plutonium name. The next, after uranium, chemical element - neptunium – was named as the planet Neptune, the next, after the planet Uranus, planet in the solar system. Similarly, plutonium, the next chemical element after neptunium, was named as the planet Pluto, the next, after the planet Neptune, planet in the solar system.

There are six metal plutonium allotropes within six temperature ranges where plutonium density varies from 15,9 g/cm^3 to 19,6 g/cm^3. The most known δ-phase of metal plutonium (γ = 15,9 g/cm^3) exists within the temperature range from 310^{0}C to 450^{0}C but this phase may be stabilized at the room temperature by plutonium alloying with Ga, Al or Ce.

Table 4.1

Some physical properties of plutonium isotopes

Isotope	$T_{1/2}$, years	Decay heat, W/kg	Spontaneous fission neutrons, n/(s·kg)	Critical mass, kg
^{238}Pu	87.7	560	$2.6\cdot10^6$	10
^{239}Pu	24100	1.9	22	10
^{240}Pu	6560	6.8	$9.1\cdot10^5$	40
^{241}Pu	14.4	4.2	49	10
^{242}Pu	376000	0.1	$1.7\cdot10^6$	100

Main channels of plutonium radioactivity are alpha-decays and spontaneous fission reactions. Retardation of heavy α-particles by plutonium

mass defines its decay heat generation while spontaneous fission reactions define neutron activity of plutonium isotopes. Some physical properties of main plutonium isotopes are presented in Table 4.1.

Plutonium is already used for a sufficiently long time period as a basic component of mixed uranium-plutonium oxide (MOX) fuel loaded into nuclear power reactors in France and Belgium.

As is known, consecutive neutron captures by main uranium isotope ^{238}U can build-up, at first, well-fissile plutonium isotope ^{239}Pu and, then the heavier plutonium isotopes. If neutron irradiation lasts a relatively short time, then fraction of the heavier plutonium isotopes (next after ^{239}Pu) can be very small (several percents of ^{240}Pu and much lower contents of ^{241}Pu and ^{242}Pu). Plutonium with such isotope composition is the most suitable fissile material for manufacturing of nuclear explosive devices (NED) and, therefore, this plutonium was named as weapon-grade plutonium (WG-Pu). Under long irradiation time and high fuel burn-up, plutonium isotope composition shifts towards the larger fraction of the heavier plutonium isotopes. Such plutonium was named as reactor-grade plutonium (RG-Pu). Typical isotope compositions of RG-Pu are presented in Table 4.2 for various values of fuel burn-up in light-water PWR-type reactors.

Table 4.2

Isotope compositions of RG-Pu and WG-Pu (for comparison)

Parameter	WG-Pu	RG-Pu at fuel burn-up, GWd/t			
		30	50	60	72
^{238}Pu, %	0.01	1.6	2.9	3.8	5.0
^{239}Pu, %	93.8	56.5	53.3	51.7	50.1
^{240}Pu, %	5.8	23.8	23.3	23.1	22.6
^{241}Pu, %	0.35	12.8	13.9	14.2	14.5
^{242}Pu, %	0.02	5.3	6.6	7.2	7.8
Critical mass, kg	7.35	9.24	9.84	9.85	9.88
Decay heat, W	16.6	112	187	243	311
Spontaneous fission neutrons, n/s	$3.9\cdot10^5$	$2.9\cdot10^6$	$4.1\cdot10^6$	$4.2\cdot10^6$	$4.6\cdot10^6$

As is seen, RG-Pu is inferior to WG-Pu in neutron-multiplying properties (the lower content of main fissile isotope ^{239}Pu and the larger con-

tents of weakly fissionable isotopes). In addition, the increased contents of ^{238}Pu and ^{240}Pu, intense heat sources and emitters of spontaneous fission neutrons, can upgrade these RG-Pu properties in comparison with those of WG-Pu. Advanced LWR projects presume further increasing the fuel burn-up. If so, then RG-Pu from the advanced LWR will be more inferior to WG-Pu in neutron-multiplying properties and in additional difficulties to handle with it because of the upgraded decay heat rate and generation rate of spontaneous fission neutrons. The higher fuel burn-up, RG-Pu is the lower suitable material both for civilian nuclear power industry and for illegal military applications.

4.1. Applicability of reactor-grade plutonium for NED manufacturing

Despite RG-Pu is evidently less suitable material for military applications than WG-Pu, nevertheless, the following three questions about RG-Pu applicability for NED manufacturing can be asked and should be answered:

1. What RG-Pu mass must be obtained to manufacture one NED?
2. Can a terrorist group manufacture NED charged with RG-Pu that was extracted from SNF with high fuel burn-up?
3. What energy yield can be expected from NED charged with RG-Pu?

Critical mass of RG-Pu is remarkably larger than that of WG-Pu because of the aforementioned differences between plutonium isotope compositions. Experimental studies and numerical evaluations have revealed that critical mass of WG-Pu was practically the same with that of ^{239}Pu (about 10 kg) while critical mass of RG-Pu tool an intermediate position between critical masses of ^{239}Pu and ^{240}Pu, (about 16 kg, in average). All these critical masses have been determined for bare (unreflected) metal spheres. If an effective neutron reflector surrounds plutonium charge, then critical mass roughly halves. Consequently, 9-10 kg of RG-Pu is a mass large enough for manufacturing of one NED. Approximately such plutonium amount is contained in one ton of spent fuel discharged from power LWR. Solvent-extraction SNF reprocessing for the design-basis value of fuel burn-up costs about $500/kg SNF. Thus, production of RG-Pu mass that is large enough to manufacture one NED will cost about $500,000.

4.2. Energy yield of NED with reactor-grade plutonium

There are two different, in principle, types of NED.

One of them is based on the implosion effect and can be named as the implosive NED. In the implosive NED a spherical charge (metal δ-plutonium, for instance) is surrounded by annular layer of high chemical explosive acting, in addition to its main mission, as a neutron reflector and reducing the critical mass by a factor of two, or so, below the bare critical mass. The ingoing shock wave from the high explosives compresses the central plutonium charge, the charge-reflector system becomes supercritical, and nuclear explosion can occur. So, the inward-directed shock wave from the high explosives creates the conditions for initiation and fast propagation of the chain fission reaction (CFR) accompanied by release of huge energy amount (energy yield).

Another type of NED, so-called the gun-type NED, is based on fast joining of two sub-critical masses into one supercritical system where the CFR could be initiated and propagated.

These two NED types, in addition to their evident constructive peculiarities, are characterized by substantially different times needed to transform the NED into the state with maximal super-criticality. In this respect the implosive NED demonstrates an obvious superiority over the gun-type NED. The implosive NED can transform itself into the maximal super-criticality state for a substantially shorter time interval, by one order of magnitude, as compared with the gun-type NED. In the implosive NED the inward-directed shock wave from the high explosives moves with mean velocity of 5 km/s and compresses the plutonium charge (~5 cm in diameter) for about 10^{-5} s. In the gun-type NED two sub-critical masses can move to meet with mean velocity of 300 m/s, and maximal super-criticality, after completion of the assemblage process, can be achieved for about 10^{-4} s, i.e. ten times slower. The slower actuation of the gun-type NED can produce a negative effect on the CFR initiation and propagation.

Mathematical model for the CFR initiation and propagation

Let assume that time dependency of the NED reactivity can be described by a simple linear function. Initially, the NED is a sub-critical

system with effective neutron multiplication factor K_{EFF} is near but less than unity ($K_{EFF} \approx 0.99$). When the NED is activated (high chemical explosive begins compressing the central plutonium charge or two sub-critical masses begins moving to join), the system gradually becomes super-critical. Let $t = t_1$ be the time moment when the system becomes critical ($K_{EFF} = 1$), and $t = t_0$ be the time moment when the system reaches maximal super-criticality ($K_{EFF} = 2$): the shock wave came to the center, or two sub-critical masses formed a single mass. Since initial state is very close to criticality, i.e. $t_1 \approx 0$ and

$$K_{EFF}(t) = 1 + \frac{t}{t_0}.$$

At $t \geq 0$ the CFR can be initiated by the appearance of some neutrons. In principle, the CFR can not be initiated at all, if no neutrons appeared in the system during the time interval $[0, t_0]$. At $t \geq t_0$ the system would be destructed and flown away.

The flying-away stage can begin when the fissile material overheats, vaporizes and creates so high internal pressure that can stop the further compression or mutual rapprochement. Numerical evaluations have demonstrated that e^{42} fission reactions could produce дthermal energy that is large enough to overheat and vaporize fissile materials. Unfortunately, these evaluations could give only qualitative results. That is why the further analysis was oriented on somewhat larger value of e^{45} fission reactions as a threshold that defines transition of the system from the compression stage to the flying-away stage, i.e.

$$e^{45} = \int_{t_i}^{t_F} N_f(t)dt;$$

where t_i – time moment of the CFR initiation; t_f – time moment when the CFR finishes because e^{45} fission reactions already occurred; $N_f(t)$ – fission rate.

Time dependency of the fission rate in a nuclear charge can be written as follows:

$$N_f(t) = N_f(0) \cdot exp(\int_0^t \frac{K_{\Im\Phi}(t') - 1}{l_{prompt}} dt') = N_f(0) \cdot exp(\frac{t^2}{2l_{prompt} \cdot t_2});$$

where $N_f(0) = \Sigma_f \cdot \Phi(0)$; $\Phi(0)$ – integral neutron flux in volume of a nuclear charge; l_{prompt} – prompt neutron lifetime (~10^{-8} s).

Then:

$$e^{45} = \Sigma_f \cdot \Phi(0) \cdot \int_{t_i}^{t_F} exp(\frac{t^2}{2l_{prompt} \cdot t_2}) dt.$$

If the integral in the right part is replaced by its approximate value, then:

$$e^{45} = (t_f - t_i) \cdot \Sigma_f \cdot \Phi(0) \cdot exp\left[\frac{t_f^2 - t_i^2}{2l_{prompt} \cdot t_2}\right].$$

By using the following identity:

$$(t_f - t_i) \cdot \Sigma_f \cdot \Phi(0) \equiv \exp\{\ln[(t_f - t_i) \cdot \Sigma_f \cdot \Phi(0)]\},$$

the balance equation for the required quantity of fission reactions can be re-written:

$$e^{45} = exp\left\{ln\left[(t_f - t_i) \cdot \Sigma_f \cdot \Phi(0)\right] + \frac{t_f^2 - t_i^2}{2l_{prompt} \cdot t_2}\right\}.$$

The summand $\ln[(t_f - t_i) \cdot \Sigma_f \cdot \Phi(0)]$ in the exponential function defines the contribution given by neutrons-initiators of the CFR into

total quantity of fission reactions. It seems evident that this contribution is negligibly small in comparison with total contribution from all fission reactions taking place during the CFR propagation. If the summand is removed, then we can derive the equation that links the time moments related with the CFR initiation and completion:

$$\frac{t_f^2 - t_i^2}{2l_{prompt} \cdot t_2} = 90. \qquad (4.1)$$

Maximal and minimal energy yields

Energy yield of a nuclear explosion could achieve its maximal value only if the initiated CFR would not be able to prevent the nuclear charge from reaching maximal super-criticality. In other words, if e^{45} fission reactions can occur only by the time moment $t_f \geq t_0$, then the CFR can not stop the compression stage. As it follows from equation (4.1), the CFR must not be initiated too early, i.e.

$$t_i \geq t_0 \cdot (1 - \frac{90 \cdot l_{prompt}}{t_0})^{1/2}.$$

In the implosive NED $t_0 = 10^{-5}\ s$. It is easy to calculate that, in order to produce maximal energy yield, the CFR must be initiated at $0.954 \cdot 10^{-5}$ s. In this case, e^{45} fission reactions can occur only for the remaining $0.046 \cdot 10^{-5}$ s, and effective neutron multiplication factor K_{EFF} can reach its maximal value.

Minimal energy yield ("fizzle") could be produced if the CFR is initiated just at the time moment when the system became critical, i.e. at $t_i = 0$. This is the worst moment for neutrons-initiators to appear in the system. As it follows from equation (4.1), in this case the CFR lasts about $3 \cdot 10^{-6}$ s. The necessary quantity of fission reactions (e^{45}) occurs in the system, and effective neutron multiplication factor increases up to $K_{EFF} = 1,3$.

Similar evaluations can be carried out for the gun-type NED where $t_0 = 10^{-4}\ s$. In order to produce maximal energy yield, the CFR must be initiated at $0.995 \cdot 10^{-4}$ s. Then, e^{45} fission reactions can occur for the remaining $0.005 \cdot 10^{-4}$ s, and effective neutron multiplication factor K_{EFF} can reach its maximal value. Minimal energy yield could be produced if the CFR is initiated at $t_i = 0$. Then, the CFR lasts $9{,}5 \cdot 10^{-6}$ s, and effective neutron multiplication factor increases up to $K_{EFF} = 1{,}095$ only.

Premature initiation of the CFR by neutrons that appeared in the system before an optimal time moment is called as the pre-detonation regime. It is evident that energy yield from the pre-detonation regime is lower than maximal (nominal) value of energy yield, and minimal energy yield ("fizzle yield") is produced if the CFR is initiated at the very inappropriate time moment (at $t_i = 0$).

As is known, main fraction of energy yield Y releases at the flying-away stage, and Y is defined by maximal value of effective neutron multiplication factor achieved at the time moment when the CFR propagation finishes, i.e. energy yield is directly proportional to α^3 where $\alpha = \dfrac{K_{EFF}(t_f) - 1}{l_{prompt}}$. Consequently, minimal energy yield in the pre-detonation regime Y_{FIZZLE} and nominal energy yield Y_{NOM} are linked by the following relationship:

$$\frac{Y_{FIZZLE}}{Y_{NOM}} = \left(\frac{K_{EFF}(t_f)_{FIZZLE} - 1}{K_{EFF}(t_f)_{NOM} - 1}\right)^3;$$

where $K_{EFF}(t_f)_{NOM} = K_{EFF}(t_2) = 2$, $K_{EFF}(t_f)_{FIZZLE} = 1.3$ for the implosive NED and $K_{EFF}(t_f)_{FIZZLE} = 1.095$ for the gun-type NED. It

is easy to calculate that $\frac{Y_{FIZZLE}}{Y_{NOM}} = 0{,}027$ for the implosive NED and $\frac{Y_{FIZZLE}}{Y_{NOM}} = 8{,}57 \cdot 10^{-4}$ for the gun-type NED.

Let assume that nominal energy yield corresponds to the energy yield released by plutonium bomb exploded over Nagasaki (Japan) on August 8, 1945, i.e. about 20 kt TNT. Then,

Y_{FIZZLE} = 0.027·20 kt = 540 t TNT for the implosive NED;
Y_{FIZZLE} = 8.57·10^{-4}·20 kt = 17 t TNT for the gun-type NED.

Despite energy yields in the pre-detonation regime are substantially lower than nominal energy yield, nevertheless, the fizzle yields are sufficiently large and dangerous. Energy yields of chemical high explosives in many terrorist attacks were well below 100 kg TNT.

Probability of maximal energy yield

As is known, probability for the CFR to be initiated prematurely, at the time moment $t < T$, in the system containing internal neutron sources (for example, radionuclides-emitters of spontaneous fission neutrons) can be determined by using the formula:

$$P(t < T) = 1 - \exp\left[- N_{SFN} \cdot T \cdot (\overline{K}_{EFF} - 1)\right];$$

where N_{SFN} – generation rate of spontaneous fission neutrons; $\overline{K}_{EFF}$ – mean value of effective neutron multiplication factor K_{EFF} within the time interval $[0, T]$. Under assumption on linear time dependency $K_{EFF}(t)$ mean value of K_{EFF} can be determined as $\overline{K}_{\Im\Phi} = 1 + 0.5T / t_0$. Then

$$P(t<T)=1-\exp\left[-\frac{N_{SFN}\cdot T}{2}\cdot(\frac{T}{t_0})\right].$$

Nominal energy yield can be obtained if the CFR is not initiated before a certain minimal time moment, i.e.

$$t_i \geq t_{i,\min}=t_0\cdot(1-\frac{90\cdot l_{\text{prompt}}}{\text{t}_0})^{1/2}.$$

Consequently, $P(t<t_{i,\min})$ is a probability for the CFR to be initiated before the time moment that can guarantee nominal energy yield, and $1-P(t<t_{i,\min})$ is a probability for the NED to produce nominal energy yield:

$$P(Y_{NOMi})=1-P(t<t_{i,\min})=\exp\left[-\frac{N_{SFN}\cdot t_{i,\min}}{2}\cdot\frac{t_{i,\min}}{t_0}\right]=$$

$$=\exp\left[-\frac{N_{SFN}\cdot t_0}{2}\cdot(1-90\cdot\frac{l_{prompt}}{t_0})\right]. \qquad (4.2)$$

Experimental information about generation rate of spontaneous fission neutrons by plutonium isotopes (Table 4.1), about isotope compositions of WG-Pu and RG-Pu U (Table 4.2) can be used to determine total generation rate of spontaneous fission neutrons in critical masses of WG-Pu (~5 kg) and RG-Pu (~8 kg):

$$N_{SFN}(WG-Pu)=3.9\cdot10^5\ n/s.$$
$$N_{SFN}(RG-Pu,PWR)=2.9\cdot10^6\ n/s.$$

By substituting these values into equation (4.2), the following probabilities can be calculated. Probability of nominal energy yield produced by the implosive NED ($t_0=10^{-5}\ s$) charged with WG-Pu is

equal to 23,5%. Similar probability for the implosive NED charged with RG-Pu from heavy-water CANDU reactor (minimal value of N_{SFN}) is considerably lower, about $1.2 \cdot 10^{-3}$ %.

Probability of nominal energy yield produced by the gun-type NED ($t_0 = 10^{-4}\ s$) even charged with WG-Pu is evaluated as a very small value, about $1.3 \cdot 10^{-5}$ %.

Consequently, RG-Pu of any isotope composition is able to produce nuclear explosion only with minimal energy yield ("fizzle yield") even in the implosive NED.

So, the following conclusions can be made:

1. Reactor-grade plutonium extracted from SNF of nuclear power reactors is a potentially dangerous material from the standpoint of its military applications.
2. Critical mass of reactor-grade plutonium is insignificantly larger than critical mass of weapon-grade plutonium (9-10 kg via 5-6 kg).
3. Main factor that can complicate any military application of reactor-grade plutonium is an intense generation of spontaneous neutrons by plutonium isotope ^{240}Pu. These neutrons are able to initiate the CFR prematurely and, thus, substantially reduce the expected energy yield.
4. Nevertheless, even minimal energy yield from any NED charged with reactor-grade plutonium can reach tens or hundreds of TNT tons. Therefore, reactor-grade plutonium must be put under strict control to prevent its diversion from civilian to military purposes.

Control questions to Chapter 4

1. Characterize main physical properties of plutonium isotopes.
2. By what does reactor-grade plutonium differ from weapon-grade plutonium?
3. What are main peculiarities of the implosive and gun-type nuclear explosive devices?
4. What conditions must be satisfied for a nuclear explosion to produce minimal and maximal energy yields?
5. Why is reactor-grade plutonium regarded as a dangerous material for nuclear non-proliferation regime?

CHAPTER 5. UTILIZATION OF REACTOR-GRADE PLUTONIUM AND WEAPON-GRADE PLUTONIUM IN NUCLEAR POWER REACTORS

As it was already mentioned above, theoretical evaluations have demonstrated that reactor-grade plutonium (RG-Pu) can be used as a charge only in primitive and low-efficient nuclear explosive devices (NED) with relatively small energy yield (at the level of several hundreds of TNT tons). Nevertheless, even so small energy yield represents a serious military threat. Therefore, these theoretical conclusions about RG-Pu applicability for military and terrorist purposes have been supported by the IAEA. In 1972 the IAEA issued the document that confirmed that plutonium of any isotope composition, including RG-Pu extracted from spent fuel of nuclear power reactors, must be regarded as a weapon-usable material, quite like weapon-grade plutonium (WG-Pu). This statement means that strict national requirements to physical protection, control and accountability of WG-Pu must be spread on RG-Pu. Only plutonium containing above 80% ^{238}Pu can be excluded from the IAEA safeguards because ^{238}Pu is an intense heat source (570 W/kg) and intense emitter of spontaneous fission neutrons ($2.6 \cdot 10^6$ n/(s·kg), i.e. ^{238}Pu is more intense neutron source than ^{240}Pu by a factor of 2,5) Therefore, radiochemical plants for SNF reprocessing have been built and put in operation only in those countries which already possessed nuclear weaponry. Also, the IAEA has worked out very strict limitations on accuracy of physical inventory taking for plutonium-bearing materials at the SNF reprocessing plants.

5.1. Utilization of weapon-grade plutonium in nuclear power reactors

In 2007 global stockpiles of WG-Pu were evaluated as ~230 tons (Russia – 120 t, USA – 90 t, Great Britain – 8 t, France – 5 t and China – 4 t). In 1993-2010 the USA and Russia have undertaken some steps towards nuclear disarmament.

In 1993 Russia has decided to withdraw 500 tons of weapon-grade uranium (WG-U) from its nuclear arsenals, dilute WG-U with low-enriched uranium and sell this reactor-grade uranium to the USA for

energy utilization at commercial NPP. In 1994 the USA administration declared that 174 tons of WG-U are no longer required for national defense purposes. So, this amount of WG-U was diluted with low-enriched uranium and used at commercial NPP. In 2005 the USA decided to withdraw additional 200 tons of WG-U from nuclear arsenals and use them at NPP.

In 1995 the USA and Russia declared that 50 tons (in each country) of WG-Pu are no longer required for national defense purposes. The USA and Russia agreed to use some excessive amounts (34 t in each country) of their WG-Pu in nuclear power industry.

By 2010 about 2300 RG-Pu tons were accumulated in SNF discharged from commercial NPP all over the world. Only ~30% of these RG-Pu FK 2010 годуamounts were extracted from SNF and used in European LWR. So, addition of 34 WG-Pu tons converted into spent RG-Pu after energy utilization in commercial LWR can not significantly increase the scope of works on plutonium utilization.

The US National Academy of Sciences has analyzed some potential ways for safe management and energy utilization of those WG-Pu amounts which have been declared as excessive for national defense purposes. The following four strategies of WG-Pu management were regarded as the most promising options:

1. Incorporation of WG-Pu into SNF compositions.

The SNF-incorporation option presumes introducing WG-Pu into fresh fuel compositions of power LWR and energy utilization of such fuels in the once-through fuel cycle. In this case spent WG-Pu containing fuel is characterized by practically insurmountable physical, chemical and radiation barriers radiochemical extraction and military application of “dirty” plutonium.

2. Short-term irradiation of WG-Pu.

This strategy presumes incorporation of WG-Pu into MOX-fuel composition and short-term irradiation of so modified MOX-fuel in power LWR. The strategy can form the remarkably weaker physical, chemical and radiation barriers against military applications of irradiated WG-Pu than the SNF-incorporation option but time expenses and the number of nuclear power reactors involved into the WG-Pu utilization program could be substantially shortened.

3. Full extermination of WG-Pu.

This strategy presumes practically complete incineration of WG-Pu in nuclear power reactors with application of uranium-free but plutonium-bearing fuel compositions. This strategy is able to burn-up about 80% WG-Pu under conditions of the once-through fuel cycle and practically full WG-Pu amounts under conditions of the closed fuel cycle. However, very large financial and time expenses would be required to develop advanced or upgrade the existing designs of nuclear power reactors.

4. Mixing of WG-Pu with radiowastes.

This strategy presumes co-immobilization of WG-Pu and radiowastes from radiochemical SNF reprocessing. WG-Pu and radiowastes must be jointly incorporated into stable inert matrices for long-term interim storage and ultimate disposal in geological repositories. This strategy and the SNF-incorporation option pursue just the same aim, namely formation of inherent physical, chemical and radiation barriers against potential use of WG-U as a charge of NED.

Thus, the USA and Russia have declared that 50 WG-Pu tons (in each country) were no longer required for national defense needs. Till now, the main strategy for excess WG-Pu management presumes its energy utilization in the existing, evolutional and innovative LWR.

Belgium and France have gained the largest experience in LWR-SNF reprocessing, extraction of RG-Pu, fabrication of fresh MOX-fuel assemblies and multiple recycle of MOX-fuel in power LWR. It was found that, under stationary conditions of MOX-fuel recycling, plutonium can be loaded only into one-third part of the reactor core. The remaining part of the reactor core has to be occupied by the standard UOX-fuel assemblies with low-enriched uranium. One-third part of the reactor core occupied by MOX-fuel assemblies is a practically achievable limit of feasibility for transition of the LWR designed on utilization of UOX-fuel only to the LWR with mixed fuel loading pattern with MOX-fuel assemblies (one-third) and UOX-fuel assemblies (two-thirds) because of the following reasons:

1. The lower fraction of delayed neutrons emitted by WG-Pu.
2. The higher-energy neutron spectrum in the part of the reactor core occupied by MOX-fuel assemblies.
3. The larger effect of resonance neutron absorption by ^{239}Pu within the energy range around 0,3 eV.

These three circumstances already require introducing substantial changes into design of the LWR control systems.

4. More intense gamma-radiation emitted by MOX-fuel assemblies can enhance radiation damages and degradation of strength properties of structural materials in MOX-fuel assemblies and in=pile metal components.

5. More intense radioactivity of spent MOX-fuel assemblies requires the longer cooling time after their withdrawal from the reactor core.

As it follows from some American publications, many (although far from all) power light-water reactors in the USA can be easily transferred on 100% loading with MOX-fuel assemblies (System 80 and System 80+ Projects of the Combustion Engineering Company).

Energy utilization rate of WG-Pu in the once-through fuel cycle is defined by the following LWR operation parameters:

- thermal power of the reactor;
- installed capacity utilization factor;
- fraction of MOX-fuel assemblies in the reactor core;
- WG-Pu fraction in MOX-fuel;
- average value of spent fuel burn-up.

Capabilities of American LWR on energy utilization of WG-Pu wee evaluated by many numerical studies, and the following results were obtained:

1. The existing power reactors of PWR-1000 type with the following operation parameters:

- thermal efficiency – 34,2%;
- installed capacity utilization factor – 0,7;
- fraction of MOX-fuel assemblies in the reactor core – 33%;
- WG-Pu fraction in MOX-fuel – 2,5%;
- average value of spent fuel burn-up – 30,4 GWd/t;

are able to exterminate 50 tons of excessive WG-Pu .for about 250 reactor·years.

2. The evolutional power reactors of PWR-1200 type (System 80 Project) with the following operation parameters:

- thermal efficiency – 34,2%;
- installed capacity utilization factor – 0,75;
- fraction of MOX-fuel assemblies in the reactor core – 100%;
- WG-Pu fraction in MOX-fuel – 4,0%;

- average value of spent fuel burn-up – 42,0 GWd/t;
are able to exterminate 50 tons of excessive WG-Pu .for about 50 reactor·years.
3. The advanced power reactors of PWR-1200 type (System 80+ Project) with the following operation parameters:
- thermal efficiency – 34,2%;
- installed capacity utilization factor – 0,75;
- fraction of MOX-fuel assemblies in the reactor core – 100%;
- WG-Pu fraction in MOX-fuel – 6,8%;
- average value of spent fuel burn-up – 42,0 GWd/t;
are able to exterminate 50 tons of excessive WG-Pu .for about 30 reactor·years.

So, energy utilization of excessive WG-Pu in the evolutional and advanced LWR designs requires relatively small number of the reactors involved and relatively short operation time with WG-Pu-based MOX-fuel assemblies.

5.2. Recycle of reactor-grade plutonium in nuclear power reactors

If nuclear fuel cycle becomes closed, then spent fuel assemblies, after withdrawal from the reactor core ands long enough (up to 10 years) cooling in SNF storage pools at NPP, are transported to the SNF reprocessing plants in order to extract the accumulated plutonium and the remained uranium, fabricate fresh MOX-fuel assemblies and recycle them in power LWR or LMFBR. Multiple recycling of fissile and fertile isotopes can produce some additional energy and, thus, reduce industrial demands for natural uranium.

By blending the recycled plutonium from MOX-fuel assemblies with “fresh” plutonium from spent UOX-fuel assemblies, we can find so optimal contents of fissile plutonium isotopes ^{239}Pu and ^{241}Pu in MOX-fuel that three-fold plutonium recycling becomes feasible in LWR with 100% MOX-fuel loading. Main safety parameters (reactivity coefficient on coolant temperature, for instance) can be kept within the acceptable constraints with application of so optimal MOX-fuel composition.

MOX-fuel assemblies have the same design as UOX-fuel assemblies, the only difference consists in composition of fuel pellets. Aver-

age plutonium content in MOX-fuel is determined so that MOX-fuel burn-up at the moment of its withdrawal from the reactor core would be the same as that for UOX-fuel. If content of fissile plutonium isotopes must not exceed a certain value (6%, for instance) because of some safety considerations, then small quantity of ^{235}U is added to MOX-fuel to maintain the reactor criticality. Peaks of heat generation rate in fuel rods placed on periphery of MOX-fuel assemblies are suppressed by decreasing plutonium content in peripheral MOX-fuel rods.

The process of gradual conversion of PWR-type reactors from UOX-fuel to MOX-fuel was numerically analyzed for a hypothetical nuclear energy system. These numerical studies were carried out under the following assumptions:

1. Total power of nuclear energy system – 10 GWe.
2. PWR-type reactors are initially loaded with low enriched (4,5% ^{235}U) UOX-fuel assemblies.
3. Then, RG-Pu extracted from spent UOX-fuel is introduced into fresh fuel composition, and plutonium gradually supplants ^{235}U from new fuel composition.
4. Time between two consecutive refuelings – 20 months.
5. Full fuel lifetime – 10 years, i.e. six irradiation cycles.
6. Maximal fuel burn-up – 50 GWd/t.
7. Maximal content of fissile plutonium isotopes in MOX-fuel composition must be below 6% (if necessary for criticality, ^{235}U can be added to MOX-fuel composition).
8. The out-of-pile fuel cycle takes 10years including 7-year cooling time in SNF storage pools at NPP plus 3 years for SNF reprocessing and fabrication of fresh MOX-fuel assemblies.

As far as RG-Pu was accumulated, extracted and recycled, some fraction of UOX-fueled PWR became MOX-fueled PWR (up to 25%). Isotope composition of the recycled plutonium was gradually shifted towards the decreased share of fissile plutonium isotopes ^{239}Pu, ^{241}Pu and the increased share of even plutonium isotopes ^{238}Pu, ^{240}Pu, ^{242}Pu. The system appeared able to incinerate about 420 kg RG-Pu/GWe·year. If fast reactor are introduced instead of MOX-PWR, then RG-Pu incineration rate increased up to 570 kg RG-Pu/GWe·year. The highest rate

of RG-Pu incineration could be reached if accelerator-driven facilities were introduced into the nuclear energy system instead of MOX-PWR: the incineration rate could be upgraded to 700 kg RG-Pu/GWe·year.

Besides, the results demonstrated that, plutonium, after three RG-Pu recycles in MOX-PWR, became quite unsuitable for further usage in PWR-type reactors. It concerns, in the first turn, reactivity coefficient on coolant temperature that becomes unacceptably large. Some data for comparison of neutron-multiplying properties, heat generation and emission of spontaneous fission neutrons by RG-Pu after three recycles in MOX-PWR with analogous properties of RG-Pu extracted from spent UOX-PWR fuel after reaching various values of fuel burn-up are presented in Table 5.1.

Table 5.1

Isotope compositions of RG-Pu and WG-Pu (only for comparison)

Parameter	WG-Pu	RG-Pu, 30 GWd/t	RG-Pu, 60 GWd/t	RG-Pu after 3 recycles
^{238}Pu, %	0.01	1.6	3.8	5.5
^{239}Pu, %	93.8	56.5	51.7	34.1
^{240}Pu, %	5.8	23.8	23.1	31.1
^{241}Pu, %	0.35	12.8	14.2	10.6
^{242}Pu, %	0.02	5.3	7.2	18.7
Critical mass, kg	7.35	9.24	9.85	12.9
Heat generation, W	16.6	112	243	445
Spontaneous fission neutrons, n/s	$3.9\cdot10^5$	$2.9\cdot10^6$	$4.2\cdot10^6$	$9.6\cdot10^6$

Control questions to Chapter 5

1. What amounts of weapon-grade plutonium and reactor-grade plutonium are currently stockpiled in the world?
2. Call main strategies for energy utilization of weapon-grade plutonium.

3. What peculiarities of MOX-fuel should be taken into account for its loading into power light-water reactors?
4. What rates can be reached by power light-water reactors in energy utilization of reactor-grade plutonium?
5. What reasons can limit the number of MOX-fuel recycles in power light-water reactors?

CHAPTER 6. URANIUM AND PLUTONIUM DENATURING AS A WAY TOWARDS NUCLEAR WEAPONS NON-PROLIFERATION

This chapter is devoted to the problems related with protection of the existing and advanced nuclear fuels against uncontrolled proliferation by introducing some additives into fuel compositions. These additives must create certain almost insuperable internal barriers any possible use of nuclear fuel for NED manufacturing, and, at the same time, these additives must preserve (or even upgrade) energy potential of nuclear fuels for their peaceful utilization at civilian NPP. Such an approach to the proliferation protection of nuclear materials is often called as an isotope denaturing. The term "isotope denaturing" means any changes in natural isotope compositions of chemical elements with ultimate goal to give them some new and desirable properties.

6.1. NM proliferation protection in NFC

As is known, natural uranium and thorium are basic materials of contemporary nuclear energy systems. Natural uranium contains only 0.71% ^{235}U, the sole natural fissile isotope, and the self-sustaining chain fission reaction (CFR) on thermal neutrons can be initiated in large volumes of natural uranium, graphite or heavy water. As for natural thorium, this chemical element does not contain at all any fissile isotope that could be regarded as an analogue of ^{235}U. Therefore, manufacturing of small-sized, uranium-based NED requires applying some difficult operations on uranium isotope enrichment in order to produce significant quantity of highly enriched (or weapon-grade) uranium (WG-U) containing up to 95% ^{235}U. The other way towards NED manufacturing is the use of artificial fissile materials (plutonium and ^{233}U) but their wide-scale production could be organized in the dedicated nuclear reactors, by neutron irradiation of natural uranium and thorium.

Nuclear reactors of contemporary civilian NPP are mainly loaded with low-enriched uranium fuel. So, from the standpoint of nuclear non-proliferation, fresh nuclear fuel takes up an intermediate position between natural uranium and WG-U. Very sophisticated and very expensive technologies of isotope separation should be applied to increase

uranium enrichment (relative content of ^{235}U) up to the weapon-usable level and, thereby, convert reactor-grade uranium into WG-U.

Another weapon-usable material – plutonium – is accumulated in nuclear fuel in the process of nuclear reactor operation. As is known, weapon-grade plutonium (WG-Pu) is composed of the following components: about 93% ^{239}Pu, up to 7% ^{240}Pu and very small fraction of the heavier plutonium isotopes. WG-Pu can be built-up in the dedicated (plutonium-producing) nuclear reactors with rather short irradiation cycle. At the same time, nuclear power reactors with substantially longer irradiation cycle and, as a consequence, substantially higher values of fuel burn-up can build-up plutonium with significantly larger contents of heavy plutonium isotopes. This plutonium is usually called as reactor-grade plutonium (RG-Pu). Lengthy neutron irradiation of uranium-based fuel can change considerably uranium isotope composition by appearance of isotopes ^{236}U and ^{232}U. For example, if uranium fuel with initial enrichment of 4.4% ^{235}U was irradiated in LWR to fuel burn-up of 4% HM, then its isotope composition would be substantially changed (see Table 6.1).

Table 6.1

Uranium and plutonium isotope compositions
in fresh and spent LWR fuel

Fresh fuel	Spent fuel composition, %					
4.4% ^{235}U, 95.6% ^{238}U	Uranium	^{232}U	^{235}U	^{236}U	^{238}U	
		$1.4 \cdot 10^{-9}$	1.26	0.59	98.15	
	RG-Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
		1.7	58.0	22.3	12.3	5.7

So, uranium fuel of nuclear power reactors can be used, in principle, to produce weapon-usable materials but their extraction requires applying some additional and very complicated operations.

Neutron irradiation of natural thorium in nuclear power reactors can build-up well-fissile and weapon-usable isotope ^{233}U. However, the process of ^{233}U build-up is accompanied by accumulation of other uranium isotopes ^{234}U, ^{235}U, ^{236}U and the lightest uranium isotope ^{232}U

which is able to complicate handling with uranium extracted from spent thorium-based fuel.

As it was already mentioned above, SNF reprocessing with removal of radioactive fission products, whose ionizing radiation created a protective barrier against uncontrolled NM proliferation, can be regarded as the operation which can completely remove or substantially weaken the radiation protective barrier. SNF partitioning into separated chemical elements or groups of elements is able to facilitate NM diversion from peaceful to military applications. Appropriate countermeasures on NM physical protection, control and accountability must be undertaken to eliminate or, at least, mitigate this threat. In addition to some organizational actions related with the security forces and strict control of nuclear technological processes, the following barriers can be established to prevent diversion of fissile weapon-usable materials:

1. Radiation barrier can be formed by radioactive fission products. This barrier can be naturally set up in the process of long-term fuel irradiation in nuclear power reactors or by a special short-term irradiation of fresh fuel. Additional inherent component of the radiation barrier is a natural radiation background produced by fissile isotopes.
2. Incomplete removal of radioactive fission products from spent fuel that keeps high-intensity radiation fields at further operations with regenerated fuel.
3. Isotope dilution of ^{235}U, i.e. the use of low-enriched uranium.
4. Incomplete separation of uranium-plutonium mixture, or even full exclusion of any technological operations able to separate plutonium from uranium. Co-extraction of uranium and plutonium makes it impossible to use such nuclear materials as a charge of NED.
5. The use of automatic distant technologies for manufacturing of nuclear fuel, fuel rods and fuel assemblies that can complicate access to fissile nuclear materials.
6. Dilution (denaturing) of fissile isotopes by other isotopes of the same chemical element. It is preferable to apply those isotopes whose properties can complicate using the denatured materials in NED. The well-known dilution of fissile isotope ^{235}U by fertile isotope ^{238}U in. low-enriched uranium is a particular case of isotope denaturing. One else example of isotope denaturing is the spent mixed thorium-uranium fuel

where fissile isotope ^{233}U can be also diluted (denatured) by fertile isotope ^{238}U.

As for ^{239}Pu, the heavier plutonium isotopes (^{240}Pu, in the first turn) can perform the denaturing mission. A particular place is taken up by the lightest plutonium isotope ^{238}Pu, intense source of decay heat ($T_{1/2}$ = 87.7 years) and spontaneous fission neutrons. These properties of ^{238}Pu can substantially complicate (or even make it unfeasible) using RG-Pu with large ^{238}Pu content as a charge of NED.

Similar role can be played by the lightest uranium isotope ^{232}U ($T_{1/2}$ = 68.9 years) in thorium-uranium fuel cycle.

In practice, the countermeasures listed above are already used in some combinations. For example, the once-through NFC of power LWR can perform the following proliferation-proof functions:

- Isotope uranium dilution (low-enriched uranium as a fuel).
- High radiation barrier created mainly by radioactive fission products.
- SNF contains uranium-plutonium mixture that is very difficult to separate.

Just the combined proliferation-protective barriers constituted a basis for the Spent Fuel Standard adopted in the USA.

Thus, uranium-based fuel can be proliferation protected by the lightest uranium isotope ^{232}U. So, it seems reasonable here to consider main nuclear properties of ^{232}U and its nearest neutron predecessor ^{231}Pa in more details.

6.2. Nuclear properties of ^{232}U and ^{231}Pa

Some relevant nuclear properties of main uranium isotopes are presented in Table 6.2. As is seen, unique properties of ^{232}U make its a valuable material for protection of uranium-based fuel against uncontrolled proliferation.

^{232}U is a starting isotope for rather long chain of radioactive decays. Some daughter products of ^{232}U can emit high-energy γ-rays (mainly, ^{208}Tl and ^{212}Bi with E_γ = 2,6 MeV and 1,8 MeV, respectively). These γ-rays can improve detectability of uranium-bearing materials and complicate any unauthorized actions. Main parameters of ^{232}U decay products are gathered in Table 6.3.

Similarly to ^{238}U, isotope ^{231}Pa is a fertile isotope that can not be fissioned by thermal neutrons but it can breed fissile materials. Energy dependencies of neutron radiative capture cross-sections are shown in Fig. 6.1 for ^{238}U and ^{231}Pa.

Table 6.2

Nuclear properties of uranium isotopes

Properties	^{232}U	^{234}U	^{235}U	^{238}U
Half-life, years	68.9	$2.45 \cdot 10^{5}$	$7.04 \cdot 10^{8}$	$4.47 \cdot 10^{9}$
Yield of α-particles, 1/(g·s)	$8.0 \cdot 10^{11}$	$2.3 \cdot 10^{8}$	$7.9 \cdot 10^{4}$	$1.2 \cdot 10^{4}$
Mean energy of α-particles, MeV	5.3	4.76	4.4	4.19
Yield of spontaneous fission neutrons, 1/(g·s)	1.3	$5.02 \cdot 10^{-3}$	$2.99 \cdot 10^{-4}$	$1.36 \cdot 10^{-2}$
Fission cross-section (E_n = 0.0253 eV)	77.15	0.465	583.2	$1.2 \cdot 10^{-5}$

Table 6.3

^{232}U decay products (α-emitters)

Properties	^{228}Th	^{224}Ra	^{220}Rn	^{216}Po	^{212}Bi	^{212}Po
Half-life	1.91 years	3.62 d	55.6 s	0.145 s	1.01 h	$3 \cdot 10^{-7}$ s
E_α, MeV (relative intensity)	5.42 (71.7%) 5.34 (27.6%)	5.69 (94.9%) 5.45 (5.1%)	6.29 (100%)	6.78 (100%)	6.09 (9.7%) 6.05 (25.2%)	8.78 (100%)

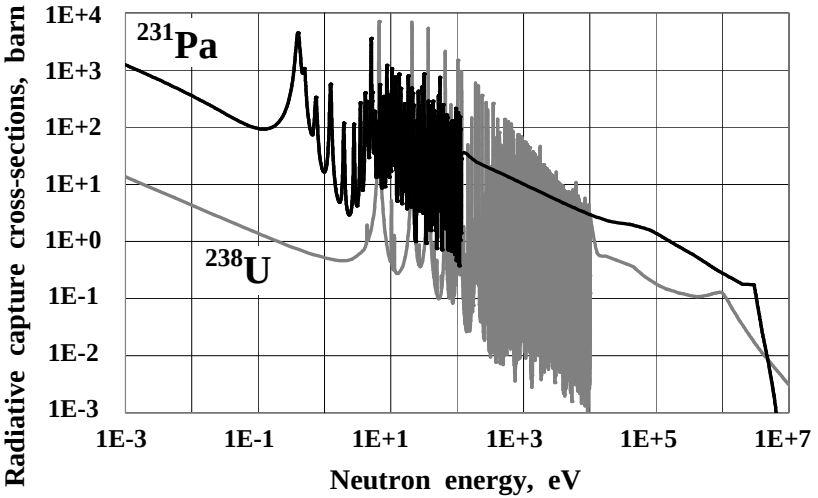


Fig. 6.1. Energy dependencies of neutron radiative capture cross-sections for ^{238}U and ^{231}Pa.

The following two important aspects should be noted here. Firstly, within thermal energy range, ^{231}Pa is a superior neutron absorber as compared with ^{238}U. For example, radiative capture cross sections of thermal neutrons (E_n = 0.0253 eV) for these two isotopes are equal to: $\sigma_c(^{231}\text{Pa})$ = 227 barns, $\sigma_c(^{238}\text{U})$ = 3 barns. So, the presence of ^{231}Pa in fuel composition can promote effective generation of fissile isotopes ^{232}U and ^{233}U. Secondly, there is a rather large energy distance between capture resonances of ^{238}U and ^{231}Pa. Capture resonances of ^{231}Pa belong to relatively low energies, below 100 eV (Fig. 6.1). This means the presence of ^{231}Pa in fuel composition can depress thermal region in energy spectrum of neutrons (Fig. 6.2).

It can be seen that, although neutron energy spectrum in VVER-1000 contains a certain fraction of thermal neutrons, introduction of ^{231}Pa into fuel composition can remove the thermal fraction completely. That is why stainless steel may be used here as a structural material. Indeed, the absence of thermal fraction in neutron spectrum does not result in additional neutron loss but fuel rods can keep their ability for working up to the higher values of fuel burn-up than those with zirconium-based alloys.

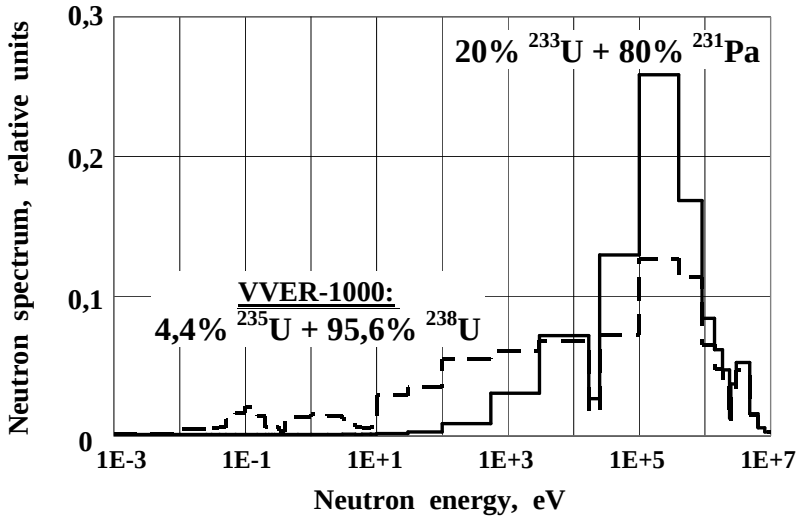


Fig. 6.2. Effect of ^{231}Pa on neutron spectrum

Now let us consider nuclear properties of ^{232}U, product of neutron capture by ^{231}Pa and rapid β-decay of ^{232}Pa ($T_{1/2}$ (^{232}Pa) = 1,3 days). Like ^{235}U, isotope ^{232}U is a fissile nuclide. Dependencies of fission cross-sections on neutron energy are presented in Fig. 6.3 for ^{235}U and ^{232}U.

It can be seen that, within thermal energy range, fission cross-sections of ^{232}U are substantially lower than those for ^{235}U while radiative capture cross-sections of these isotopes are comparable each other. For example, radiative capture cross sections of thermal neutrons (E_n = 0.0253 eV) for these two isotopes are equal to: $\sigma_c(^{232}U)$ = 73 barns, $\sigma_c(^{235}U)$ = 99 barns. So, neutron-multiplying properties of ^{232}U are inferior to those of ^{235}U within thermal energy range.

This conclusion can be confirmed by Fig. 6.4 which demonstrates energy dependency of (ν_{ef} - 1), i.e. the number of excess fission neutrons per one absorbed neutron. ^{235}U looks superior to ^{232}U within thermal energy range but quite another situation takes place in resonance range. So, it may be expected that introduction of ^{231}Pa into uranium-based fuel composition with aim to increase fuel burn-up will be more efficient just in resonance neutron spectrum.

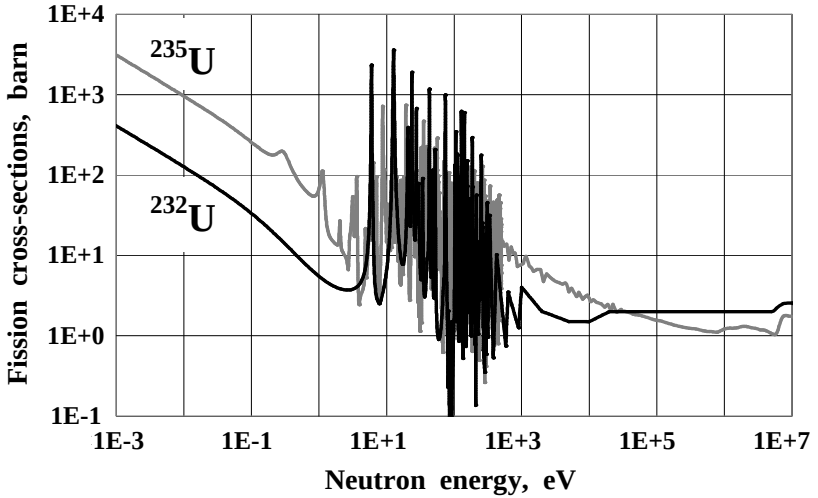


Fig. 6.3. Dependency of fission cross-sections on neutron energy for isotopes ^{235}U and ^{232}U

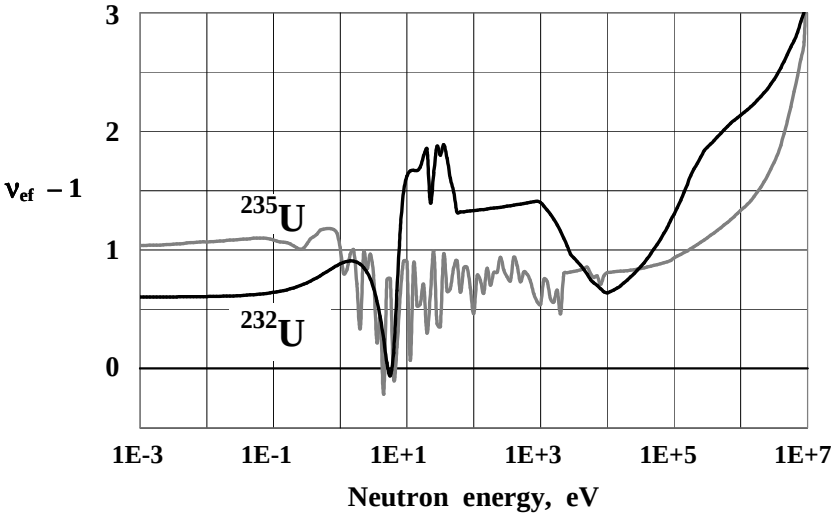


Fig. 6.4. Energy dependency of the number of excess fission neutrons per one absorbed neutron for isotopes ^{235}U and ^{232}U

6.3. NM proliferation protection in open NFC

Presently, there are different points of view on future development of nuclear fuel cycles. Some countries (USA, Canada, Germany and Sweden) are implementing in practice an open (or once-through) nuclear fuel cycle that does not foresee a radiochemical SNF reprocessing in the visible future. One of the reasons for this choice is a wish of decreasing a risk of nuclear weapon proliferation. SNF may be only converted into the forms suitable for long-term controlled storage. However, such a strategy of nuclear power development has already resulted in large SNF stockpiles, potentially dangerous nuclear materials. So, the preferable option for future development of nuclear power consists in transition to the closed fuel cycles with SNF reprocessing, separation of radioactive fission products and recycling of residual fuel.

Low-enriched uranium (LEU) is a fresh fuel for open nuclear fuel cycle. Plutonium in spent fuel assemblies is protected by intense gamma-radiation of fission products. That is why unirradiated nuclear materials are more vulnerable for unauthorized proliferation.

Isotope uranium denaturing may be regarded as an effective method for upgrading self-protection of unirradiated uranium-bearing materials. In principle, uranium may be denatured by the following two ways: direct introduction of intense radioactive isotope ^{232}U into uranium fuel composition or direct introduction of relatively weaker radioactive isotope ^{231}Pa into uranium fuel composition. ^{231}Pa is a neutron predecessor of ^{232}U, main isotope of uranium denaturing. So, only short-term pre-irradiation of fresh fuel assemblies in the research reactors may be sufficient to produce proliferation resistant fuel assemblies, suitable even for export deliveries.

6.3.1. Uranium denaturing as a way for formation of internal α-radiation source

Along with progress in development of high-efficiency enriching technologies, potential threat of LEU diversion and re-enrichment up to the weapon-grade level excites more and more apprehensions. These reasons indicate that, besides reduction of uranium enrichment below

20% ^{235}U, according to the IAEA recommendations, other measures may be also required to upgrade LEU self-protection against its unauthorized re-enrichment. Taking into consideration the growing worldwide scope of LEU utilization, including LEU with enrichment in the vicinity of the upper boundary (~20% ^{235}U), high LEU vulnerability to unauthorized re-enrichment must be recognized. Particular apprehensions are related with 20%-uranium. So, some additional actions should be undertaken to protect LEU against its unauthorized re-enrichment. The effects of ^{232}U introduction into LEU are caused by the following specific properties of this uranium isotope:

1. Good neutron-multiplying properties of ^{232}U and its neutron predecessor ^{231}Pa make it possible to extend time period of continuous reactor operation without refueling up to the values comparable with the reactor lifetime. As a result, unauthorized extraction of plutonium from spent fuel assemblies becomes unfeasible.
2. It is impossible to remove ^{232}U from denatured uranium without application of the very sophisticated and expensive isotope separation technologies.
3. ^{232}U is a neutron source from spontaneous fission reactions and a source of high-energy α-particles. Alpha-particles emitted by ^{232}U are able to dissociate molecules of uranium hexafluoride and, thus, could make it practically unfeasible to re-enrich denatured uranium up to the weapon-grade level. Besides, α-particles are able to initiate (α,n)-reactions with impurities of light elements and, thus, intensify internal neutron generation. Thus, export deliveries of LEU-based fuel assemblies to foreign NPP receive an additional proliferation barrier.

Even if only 10 ppm ^{232}U is introduced into 20%-uranium, then ^{232}U becomes a main contributor to total intensity of α-radiation. This effect plays a definitive role in forming the inherent radiation barrier against unauthorized re-enrichment of 20%-uranium up to weapon-grade level. Since ^{232}U nuclei are lighter than ^{235}U nuclei, ^{232}U content, as far as the re-enrichment proceeds, increases with subsequent intensification of α-radiation. If 0.001÷0.1% ^{232}U is introduced into 20%-uranium, then α-activity of the re-enriched product can increase by two orders of magnitude. Besides, “light” ^{232}U supplants “heavy” ^{235}U and, thereby, limits final ^{235}U content in the r-enriched product. ^{232}U decay products can contribute additionally to total α-activity of the r-enriched uranium.

6.3.2. Evolution of neutron background in re-enrichment of denatured uranium, evaluation of energy yield

Time-dependent evolution of neutron emission rate in the process of 20%-uranium re-enrichment was numerically evaluated, and the following results were obtained.

If ^{232}U is absent at all in 20%-uranium, then ^{234}U is a main neutron emitter through (α,n)-reactions with light impurities (Fig. 6.5). However, in this case, neutron emission rate is lower than neutron emission rate from spontaneous fission reactions of ^{238}U (13,6 n/kg·s) by one order of magnitude. Such low value of neutron yield is quite insufficient for uranium self-protection.

If 20%-uranium was produced from the regenerated LWR uranium, which already contained trace amounts of ^{232}U, then neutron emission rate from (α,n)-reactions with light impurities can increase up to 3,5 n/kg·s in the process of 20%-uranium re-enrichment up to weapon-grade level. ^{234}U remains a main neutron source at negligibly small contributions of ^{232}U and ^{236}U.

If relatively small amount of ^{232}U is introduced into 20%-uranium, regardless of its origin either from natural uranium or from the regenerated LWR uranium, then the situation drastically changes. Amongst all uranium and plutonium isotopes, ^{232}U is the most intense α-emitter. Therefore, (α,n)-reactions with light impurities begin playing a dominant role in formation of neutron background. If only 0.001% ^{232}U is admixed to 20%-uranium, then neutron background upgrades by two orders of magnitude. If 0.1% ^{232}U is admixed to 20%-uranium, then neutron background from the re-enriched product exceeds emission rate of spontaneous fission neutrons of ^{238}U by a factor of 590. This neutron background can continue upgrading thanks to ^{232}U decay products. In a year the neutron background can increase additionally by a factor of 3.

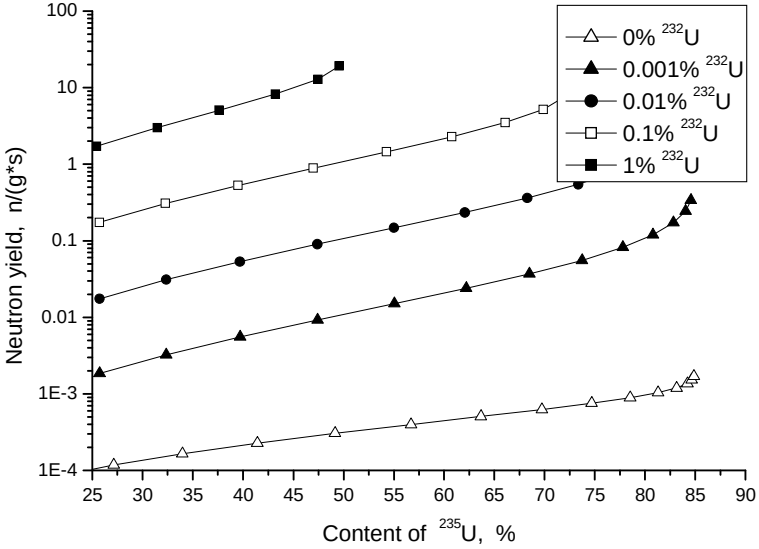


Fig. 6.5. Neutron emission rate by (α,n)-reactions in the re-enrichment process

Threat of any unauthorized applications of uranium-bearing NM is defined by the possibility for the CFR to be initiated and followed by huge energy yield. The effects produced by inherent neutron background, which significantly increased in the re-enrichment process of denatured 20%-uranium up to weapon-grade level, on the CFR energy yield of gun-type NED were numerically evaluated, and some interesting results were obtained under the following assumptions:

- the CFR energy yields were determined for critical mass of weapon-grade uranium;

- central spherical charge was surrounded with neutron reflector made of tungsten carbide, 10-cm thick.

If 20%-uranium contains above 50 ppm ^{232}U, then, even if the assemblage velocity V = 1000 m/s, premature initiation of th CFR (predetonation) occurs with probability above 99%. Main parameters of the CFR initiated in the denatured and re-enriched 20%-uranium are pre-

sented in Table 6.4 for various ^{232}U contents (> 50 ppm) and for various assemblage velocities.

Table 6.4

CFR parameters in the denatured and re-enriched uranium

^{232}U content in 20%-uranium, % HM		0.1%	0.5%	1%
^{235}U content in re-enriched uranium, % HM		72.3	57.4	49.6
Neutron background, 10^6 n/s		0.892	1.85	3.01
K_{max}		1.462	1.424	1.406
V = 300 m/s	t_0 , 10^{-3} s	1.01	1.1	1.17
	t_i / t_0	0.105	0.073	0.057
	t_f / t_0	0.122	0.1	0.091
	Y/Y_{NOM} *), 10^{-3}	1.82	1.0	0.75
V = 600 m/s	t_0 , $\times 10^{-4}$ s	5.07	5.5	5.87
	t_i / t_0	0.149	0.103	0.08
	t_f / t_0	0.173	0.141	0.128
	Y/Y_{NOM} , 10^{-3}	5.18	2.8	2.1
V = 1000 m/s	t_0 , $\times 10^{-4}$ s	3.05	3.3	3.52
	t_i / t_0	0.192	0.133	0.104
	t_f / t_0	0.223	0.182	0.166
	Y/Y_{NOM} , $\times 10^{-3}$	11.1	6.03	4.57

*Relative energy yields Y/Y_{NOM} were evaluated for the confidence probability of 0.9.

As is seen, relative energy yields Y/Y_{NOM} covered the range from 0.001 to 0.01. At moderate values of assemblage velocity (300 m/s) the CFR energy yields were equal tro several thousandth parts of nominal energy yield. If the denatured and re-enriched uranium was stored for one year, then the CFR energy yield dropped down even to the larger degree. In this case, the CFR energy yields decreased to the levels comparable with “fizzle” yields, when the CFR was prematurely initiated just at the moment of the charge criticality.

Thus, growth of neutron background in the re-enrichment process of 20%-uranium denatured by admixing 0.1÷0.5% ^{232}U up to weapon-grade level can decrease the CFR energy yield by three orders of magnitude. In essence, the gun-type NED charged with such denatured and re-enriched uranium is a “dirty” bomb only.

6.3.3. Higher fuel burn-up of proliferation-proof LWR fuel at introducing ^{231}Pa

One of specific features in operation of nuclear power reactors consists in a necessity to perform regular refuelings. This necessity is caused by the following effects: depletion of fissile materials, FP accumulation and possible rupture of fuel cladding with intense release of radioactive materials. LWR, the mostly spread type of power reactors, requires refueling every 1-2 years, when fuel burn-up reaches 4-6% HM.

Extension of fuel life-time up to relatively long time periods (several decades, for instance) can reduce drastically the number of refuelings or exclude them at all. Reduction or full exclusion of refueling procedures decreases the demands for fresh fuel and decreases quantity of SNF discharged per unit of produced energy. Those reactors, which are capable to operate for a sufficiently long time without any refueling, may be used as the only energy source in remote regions, at the floating NPP, as energy source for space investigations (research bases on the Moon or Mars, cosmic flights into the outer space). Our studies demonstrated that introduction of ^{231}Pa into LWR fuel composition could extend significantly the fuel life-time and reach ultra-high fuel burn-up.

It should be noted that achievability of ultra-high fuel burn-up was studied here only from the standpoint of neutron-multiplying properties of advanced fuel compositions. The problems of suitable structural materials, evolution of their strength properties and durability for a long fuel life-time are not analyzed here. At present, maximal fuel burn-up (about 30% HM) was achieved in the research fast reactor BOR-60. It may be expected that the higher values of fuel burn-up could be achieved if the following operations would be multiply carried out: partial fuel burn-up (near to the practically achievable value of 30% HM),

application of DUPIC-technology for removal of gaseous and volatile FP, re-fabrication of fresh fuel pellets.

6.3.4. Neutron-multiplying properties in nuclide transformation chains

In this section we compared time evolutions of neutron-multiplying properties in two isotopic chains: traditional chain that starts from ^{232}Th (^{232}Th → ^{233}U → ^{234}U → …) and nontraditional chain that starts from ^{231}Pa (^{231}Pa → ^{232}U → ^{233}U → …) (see Fig. 6.6). Radiative capture cross-sections σ_c and fission cross-sections σ_f were calculated for a typical neutron spectrum of VVER-1000.

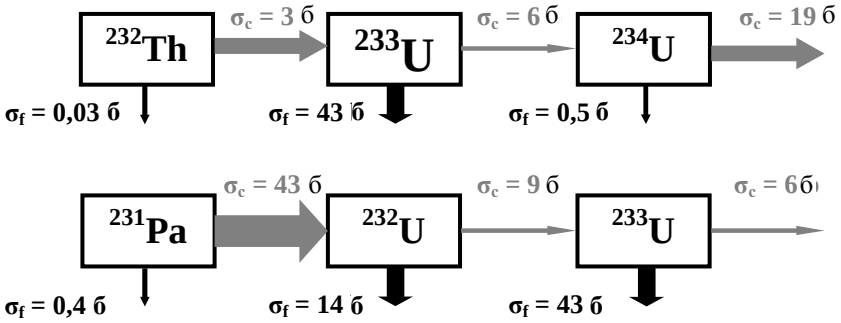


Fig. 6.6. Chains of nuclide transformations

It can be seen that neutron-multiplying properties in non-traditional chain are gradually improved: the starting isotope ^{231}Pa is a neutron absorber, fission cross-section of the second isotope ^{232}U prevails over its capture cross-section, and the third isotope ^{233}U is a well-fissile material. So, non-traditional chain represents the combination of two consecutive fissionable isotopes (^{232}U and ^{233}U) while, in traditional chain, the third isotope ^{234}U is a neutron absorber only.

Thus, in non-traditional chain, parasitic neutron absorption by FP and depletion of fissile materials during the reactor operation can be partially compensated by ^{231}Pa feeding. This makes it possible to talk about a possibility for substantial extension of fuel life-time and achievability of ultra-high fuel burn-up. By the way, in traditional LWR

fuel, the negative effects caused by FP accumulation and depletion of fissile materials are compensated by $^{238}U(n,\gamma)^{239}Pu$ chain significantly weaker than by $^{231}Pa(n,\gamma)^{232}U(n,\gamma)^{233}U$ chain in non-traditional fuel because of lower capture cross-sections: $\sigma_c(^{238}U) = 0.9$ barns, $\sigma_c(^{231}Pa) = 43$ barns.

So, it can be concluded that non-traditional chain ($^{231}Pa \rightarrow {}^{232}U \rightarrow {}^{233}U \rightarrow \ldots$) appears to be more attractive from the standpoint of neutron-multiplying properties (as a consequence, from the standpoint of extended fuel life-time or achievability of ultra-high fuel burn-up) in comparison with traditional chain ($^{232}Th \rightarrow {}^{233}U \rightarrow {}^{234}U \rightarrow \ldots$) due to the following two main reasons:

1. Combination of two consecutive well-fissionable isotopes (^{232}U and ^{233}U).
2. High rate of their generation from the starting isotope ^{231}Pa, whose neutron capture cross-section is larger substantially than that for the starting nuclide ^{232}Th in traditional chain of isotopic transformations.

It is noteworthy that ^{231}Pa may be regarded, to a certain extent, as a burnable neutron poison: for fuel life-time ^{231}Pa is burnt up to 80% and converted into well-fissionable isotopes, neutron capture cross-section of ^{231}Pa is substantially larger than that of fertile isotope ^{232}Th.

As is known, the existing light-water reactors are characterized by thermal neutron spectrum. In advanced LWR designs, for example, in LWR with supercritical coolant parameters (SCLWR), different regions of the reactor core are characterized by different neutron spectra depending on coolant density. Thermal spectrum prevails within the core region containing dense coolant ($\gamma \approx 0.72$ g/cm^3) while resonance neutron spectrum dominates within the core region containing coolant of the lower density ($\gamma \approx 0.1$ g/cm^3).

Reasonability of ^{231}Pa introduction into fuel composition for the cases of thermal and resonance neutron spectra is analyzed in the next section.

6.3.5. Reasonability of ^{231}Pa introduction in a thermal neutron spectrum

Numerical analyses of fuel depletion process were carried out with application of the computer code SCALE-4.3 and evaluated nuclear

data file ENDF/B-V for elementary cells of VVER-1000. The only exception consisted in the use of martensite steel MA956 (elemental composition: 74,5% Fe, 20% Cr, 4,5% Al, 0,5% Ti and 0,5% Y_2O_3) instead of zircalloy as a fuel cladding material. Substitution of martensite steel for zirconium-based cladding is caused by the higher values of fuel burn-up.

Traditional (^{232}Th-^{233}U) and non-traditional (^{231}Pa-^{232}Th-^{233}U) fuel compositions were compared for the case of thermal neutron spectrum (coolant density – 0.72 g/cm^3). Infinite neutron multiplication factor K_∞ is shown in Fig. 6.7 as a function of fuel burn-up. It can be seen that substitution of ^{231}Pa for ^{232}Th decreases K_∞ at the beginning of cycle, i.e. decreases an initial reactivity margin to be compensated. This effect is caused by different capture cross-sections of these isotopes - ^{231}Pa is a significantly stronger neutron absorber than ^{232}Th. In parallel, thanks to the larger capture cross-section of ^{231}Pa, intense breeding of two consecutive well-fissionable isotopes (^{232}U and ^{233}U) takes place. So, gradual introduction of ^{231}Pa into fuel composition results in the smoother relaxation of neutron multiplication factor in the process of fuel burn-up.

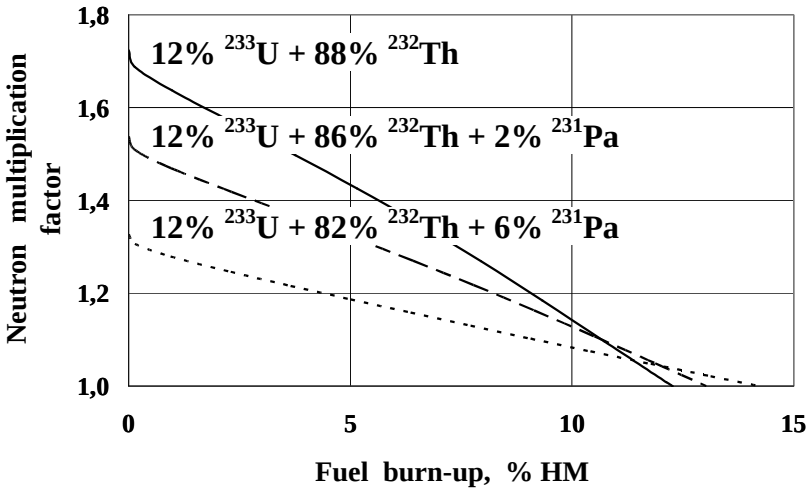


Fig. 6.7. ^{231}Pa effects on fuel burn-up in thermal neutron spectrum

Acceptable fraction of ^{231}Pa in non-traditional fuel composition is limited by the value of neutron multiplication factor (above unity) at the beginning of cycle. So, the effects caused by introduction of ^{231}Pa may take place only in those fuel compositions where fraction of main fissile isotope is sufficiently large. For example, fraction of main fissile isotope ^{233}U may be increased up to the level corresponding to the situation when neutron multiplication factor at the beginning of cycle is equal to about 1.10 at full replacement of ^{232}Th by ^{231}Pa.

The calculations showed that this condition may be satisfied at maximal ^{233}U fraction about 30%. Evolution of neutron multiplication factor in the process of fuel burn-up is presented in Fig. 6.8 for traditional and non-traditional fuel compositions.

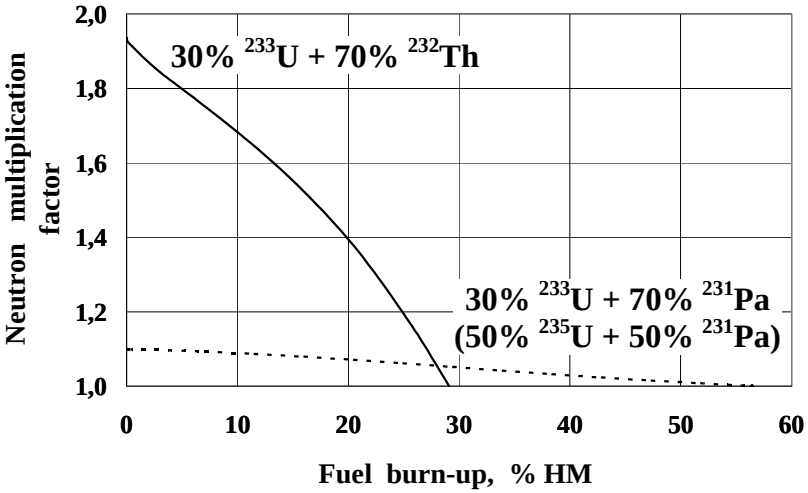


Fig. 6.8. Achievability of ultra-high fuel burn-up by introducing ^{231}Pa (thermal neutron spectrum)

As is seen from Fig. 6.8, traditional thorium-based fuel (30% ^{233}U + 70% ^{232}Th) provides rather high reactivity margin (K_∞(BOC) ≈ 1,9) with achievable value of fuel burn-up about 29% HM. Introduction of

^{231}Pa into fuel composition decreases initial reactivity margin but, at the same time, increases fuel burn-up. If ^{232}Th is completely replaced by ^{231}Pa, i.e. (30% ^{233}U + 70% ^{231}Pa) fuel composition is analyzed, then neutron multiplication factor remains practically unchanged in the vicinity of unity for a full duration of fuel life-time. This means that the negative effects from neutron absorption by FP and depletion of fissile isotope are almost completely compensated by breeding of secondary fissile isotopes from ^{231}Pa. In this case, about 80%-part of ^{231}Pa is converted into secondary fissile isotopes which can provide ultra-high fuel burn-up (near to 57% HM).

If fuel loading in such a reactor is similar to the fuel loading of VVER-1000 (about 66 tons), then achievable value of fuel life-time is near to 40 years for the reactor power of 3000 MWt. It is interesting to note that ^{235}U as well as ^{233}U may be used to achieve ultra-high fuel burn-up. Moreover, ^{235}U option looks very attractive because of two reasons: firstly, ^{235}U resources are more available than resources of ^{233}U, and, secondly, achievement of the same fuel burn-up will require lower quantity of ^{231}Pa, artificial isotope to be produced in the dedicated nuclear power facilities.

6.3.6. Reasonability of ^{231}Pa introduction in a resonance neutron spectrum

Traditional (^{232}Th-^{233}U) and non-traditional (^{231}Pa-^{232}Th-^{233}U) fuel compositions were compared for the case of resonance neutron spectrum (coolant density – 0.1 g/cm^3). Infinite neutron multiplication factor K_∞ is shown in Fig. 6.9 as a function of fuel burn-up.

Comparison of the curves presented in Figs. 6.7 and 6.9 allows us to conclude that introduction of ^{231}Pa into fuel composition is more preferable from the standpoint of higher fuel burn-up in the case of resonance neutron spectrum. This conclusion can be explained by better neutron-multiplying properties of ^{232}U just in resonance neutron spectrum as compared with thermal neutron spectrum (see Fig. 6.4).

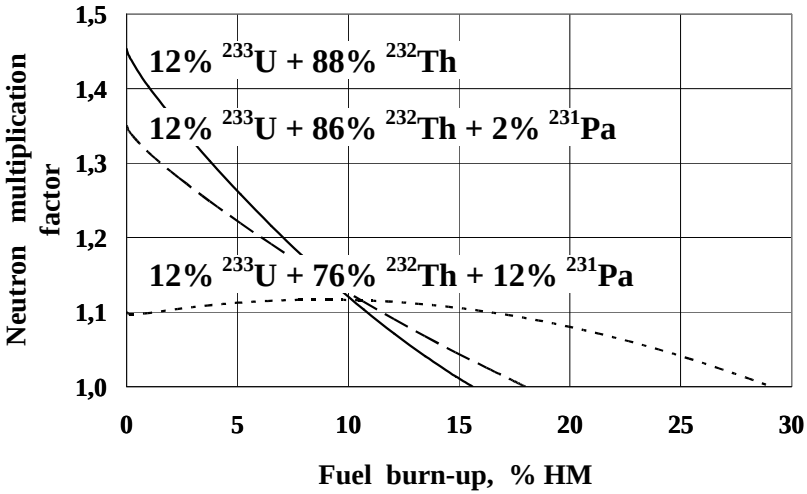


Fig. 6.9. ^{231}Pa effects on fuel burn-up in resonance neutron spectrum

As it follows from Fig. 6.9, introduction of only 12% ^{231}Pa increased fuel burn-up twice. Neutron multiplication factor at the beginning of cycle increased too, i.e. neutron-multiplying properties of fuel composition became better.

Like previous analysis, fraction of main fissile isotope ^{233}U may be increased up to the level corresponding to the situation when neutron multiplication factor at the beginning of cycle is equal to about 1.10 at full replacement of ^{232}Th by ^{231}Pa. In addition, potential use of ^{235}U instead of ^{233}U was analyzed to evaluate a possibility for achieving ultra-high fuel burn-up. So, numerical studies confirmed reasonability for introduction of ^{231}Pa into fuel composition because this introduction results in reduction of initial reactivity margin and in substantial growth of fuel burn-up. Maximal positive effect from introduction of ^{231}Pa may be observed in resonance neutron spectrum. Besides, introduction of ^{231}Pa makes it possible to reach ultra-high fuel burn-up regardless of what main fissile isotope is used, ^{233}U or ^{235}U. In particular, (20% ^{233}U + 80% ^{231}Pa) fuel composition can reach fuel burn-up of 76% HM in resonance neutron spectrum (see Fig. 6.10).

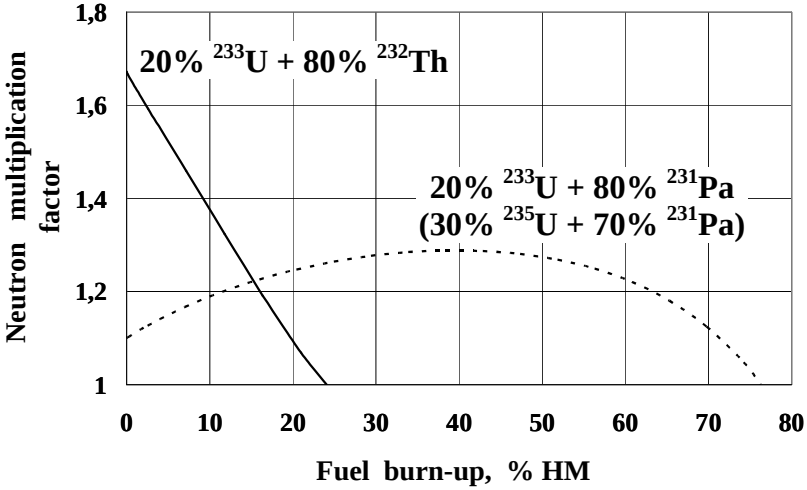


Fig. 6.10. Achievability of ultra-high fuel burn-up by introducing ^{231}Pa (resonance neutron spectrum)

6.3.7. Effects of ^{231}Pa introduction on reactor safety

On the one hand, introduction of ^{231}Pa into fuel composition can provide small value of initial reactivity margin and high value of fuel burn-up. On the other hand, if relatively large ^{231}Pa fraction is introduced into fuel composition, reactivity feedback on coolant temperature becomes positive, and safety of the reactor operation worsens.

Numerical studies demonstrated that, if maintenance of favorable reactivity feedback on coolant temperature during fuel life-time is a mandatory requirement, then, in thermal neutron spectrum, ^{231}Pa fraction in fuel composition is limited by a quite certain value while, in resonance neutron spectrum, introduction of ^{231}Pa is impossible at all. However, this conclusion is correct only for large-sized reactors, where neutron leakage is negligibly small.

So, only thermal neutron spectra should be considered to provide favorable reactivity feedback on coolant temperature. The results presented in Fig. 6.11 demonstrate a possibility for increasing fuel burn-up in thermal neutron spectrum by introducing ^{231}Pa into fuel composition.

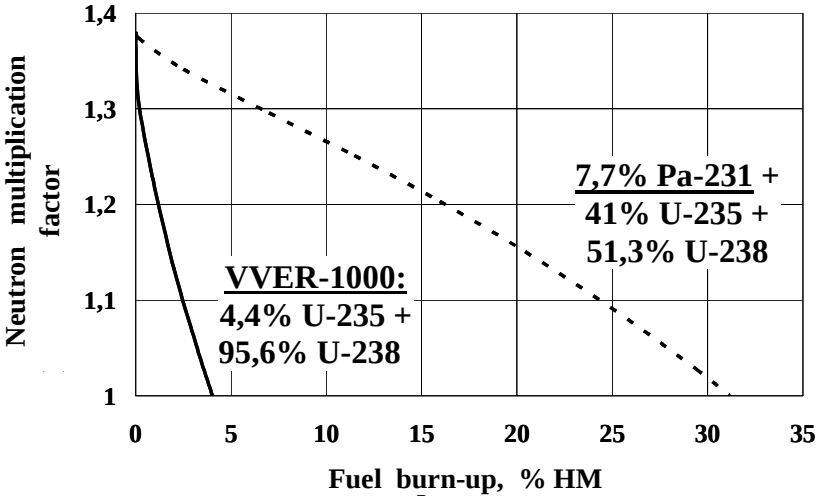


Fig. 6.11. Achievability of ultra-high fuel burn-up by introducing ^{231}Pa with conservation of favorable feedback on coolant temperature (thermal neutron spectrum)

As is known, fuel burn-up in VVER-1000 can reach a value about 4% HM. Introduction of ^{231}Pa and higher contents of ^{235}U can increase fuel burn-up by a factor of 8 with the same initial reactivity margin, i.e. more powerful system of reactivity compensation is not required.

Requirement of favorable reactivity feedback on coolant temperature completely excludes any introduction of ^{231}Pa into fuel composition in the case of large-sized reactors with resonance neutron spectra. However, introduction of ^{231}Pa into fuel composition of small-sized reactors does not worsen safety of the reactor operation because of relatively large neutron leakage. This indicates that the mostly attractive area for ^{231}Pa applications is a small nuclear power including small-sized NPP for remote regions, for the floating NPP, for space stations on the Moon or Mars and for cosmic flights into the outer space.

The following conclusions can be made in respect of potential ^{231}Pa applications:

- Application of ^{231}Pa as a burnable neutron poison can reduce initial reactivity margin and increase fuel burn-up.

- Introduction of ^{231}Pa into fuel composition makes it possible to reach ultra-high fuel burn-up (above 30% HM) both in thermal and resonance neutron spectra.
- The actual problem of ^{231}Pa production in significant amounts should be resolved.

6.4. NM proliferation protection in the closed NFC

NPP operation in open fuel cycle results in accumulation of huge SNF stockpiles that represents a long-term hazard to the humankind. Ultimate SNF disposal is a difficult technical problem requiring large number of practically "eternal" deep underground repositories. That is why many various options for closure of nuclear fuel cycle are currently under research and development including extraction of residual uranium, plutonium and minor actinides from SNF.

As known, the closed uranium-plutonium NFC includes reprocessing and recycling of nuclear fuel and evokes a lot of contradictory opinions with respect to potential risk of plutonium proliferation. This is connected with the following two points:

- Although plutonium extracted from SNF of power reactors (for example, LWR of PWR, BWR or VVER type) is not the best material for nuclear weapons, nevertheless it can be used in NED of moderate energy yield.
- Recycled plutonium will be disposed at the facilities of the closed NFC, and this will increase the probability of it using for illegal aims (diversion, theft).

Under these conditions, the absence of any internationally coordinated plan concerning the utilization or ultimate SNF disposal enforced the leading nuclear countries to undertake the steps directed to strengthening the nonproliferation regime (the IAEA safeguard system, the EURATOM embargo on the export of SNF reprocessing technologies). However, several countries, the USA, in the first turn, refused from deployment of breeder reactors which are intended for operation in the closed NFC, and deliberately focused at once-through NFC. On the other hand, the social demands for solving excess fissile materials (plutonium, the first of all) problem which have both civil and military origins, stimulated carrying out the research on plutonium utilization in

MOX-fuel compositions. At the same time, the studies of advanced NFC protected against uncontrolled proliferation of fissile materials have been initiated.

6.4.1. Radiation protection of MOX-fuel, initiative GNEP

Specialists from Oak Ridge National Laboratory (USA) have investigated the ways for introduction of γ-radiation sources into fresh fuel compositions. Sixty-four γ-active radionuclides were selected and studied as candidates for admixing into fresh fuel compositions. Radionuclides ^{137}Cs ($T_{1/2}$ = 30 years) and ^{60}Co ($T_{1/2}$ = 5.27 years) appeared the most preferable candidates. But cesium is a volatile element, and it can be easily removed from fuel by heating up. Intensity of γ-radiation emitted by ^{60}Co rapidly relaxes.

Specialists from Los Alamos National Laboratory (USA) have proposed the advanced version of the international NFC that enhances proliferation resistance of plutonium. This proposal constituted a basis for the US President's initiative on the Global Nuclear Energy Partnership (GNEP) that was supported by many countries (including Russia) with well-developed nuclear infrastructure. According to this proposal, spent fuel assemblies discharged from power reactors of a country-user must be transported to the Nuclear Club countries for full-scale reprocessing. The extracted plutonium and minor actinides must be incinerated in the reactors placed on the territory of the International nuclear technology centers. Plutonium is not recycled in power reactors of a country-user. The Nuclear Club countries provide fresh LEU fuel deliveries into a country-user.

Upon exhaustion of rich and cheap uranium resources, nuclear power has to use artificial kinds of fresh fuel (plutonium, ^{233}U or their mixtures). The GNEP initiative does not consider this opportunity. It is proposed to use such power reactors which are able to work without refueling for 15-20 years. After this time interval they must be returned to the Nuclear Club countries for SNF discharging, reprocessing and recharging with fresh fuel.

The concentrated incineration of plutonium and minor actinides in the International nuclear technology centers can lead to unacceptably large local release of thermal energy followed by the unpredictable

negative environmental and climatic effects. As to the reactors with long-life cores, these are small and medium-sized power reactors. Besides, during transportation and mounting, they can be very attractive sources of plutonium in amounts large enough for manufacturing of several dozens of nuclear bombs.

6.4.2. Plutonium denaturing as a way for proliferation protection of the closed NFC

Some nuclear properties of ^{238}Pu make this isotope a valuable material for proliferation protection of uranium-plutonium fuel. Firstly, ^{238}Pu is an intense source of thermal energy ($T_{1/2}$ = 87 years, specific heat generation - 570 W/kg). So, introduction of ^{238}Pu into plutonium creates almost insuperable barrier to manufacturing of even primitive implosion-type NED. Plutonium heating up by isotope ^{238}Pu can provoke undesirable phase transitions and thermal pyrolysis of conventional explosives applied for compression of central plutonium charge. Secondly, ^{238}Pu is an intense source of spontaneous fission neutrons, even more intense than ^{240}Pu. As a consequence, probability of premature CFR initiation in NED sharply increases while energy yield of nuclear explosion drastically drops down to the levels comparable with energy yield of conventional explosives. Thus, LWR MOX-fuel cycle with ternary fuel compositions (Np-U-Pu) is characterized by enhanced proliferation resistance.

Like uranium, plutonium can be denatured by the following two ways: either direct introduction of intensely radioactive isotope ^{238}Pu into MOX-fuel composition or introduction of relatively low intense radioactive isotope ^{237}Np into MOX-fuel composition. ^{237}Np is the nearest neutron predecessor of main denaturing isotope ^{238}Pu. So, only short-term pre-irradiation of fresh MOX-fuel assemblies would be sufficient to produce proliferation resistant fuel assemblies, suitable even for export deliveries to any countries.

6.4.3. Effects of ^{237}Np and ^{238}Pu introduction into MOX-fuel composition on proliferation resistance of plutonium

The equilibrium isotope vectors were determined for MOX-fuel circulating between LWR, spent fuel reprocessing plants and fuel manufacturing facilities. It was presumed the fuel feed consisting of ^{237}Np, ^{238}Pu and ^{239}Pu was produced in blanket regions of Hybrid Thermonuclear Installation (HTI). By using the computer code GETERA for cell calculations of fuel burn-up, Pu isotopic compositions of MOX-fueled PWR were determined for moments of the beginning and end of cycle. ^{238}Pu fraction in plutonium was adopted to be an index of Pu protection against uncontrolled proliferation. It means that the impact of the heavier plutonium isotopes on the CFR neutronics in the imploded plutonium charge of NED was not taken into account.

The fuel loaded into PWR core may be considered as material consisting of two parts: the first part includes equilibrium composition of ^{238}U and plutonium isotopes produced by ^{238}U while the second part ("feed part of fuel) includes equilibrium composition of ^{237}Np, ^{238}Pu and other plutonium isotopes produced entirely by the feed. Equilibrium contents of ^{238}Pu in plutonium of PWR fuel depending on ^{238}Pu contents in plutonium of feed (with different ^{237}Np fractions in "feed part of fuel") for equilibrium multi-cycle operation regime are presented in Fig. 6.12.

The plot region situated under the bisectrix B is a region where plutonium protection in feed is higher than plutonium protection in fuel. Respectively, the plot region situated above the bisectrix B is a region where plutonium protection in fuel is higher than that in feed. The curves of this figure characterize the correlation between plutonium protection levels in feed and fuel when the "feed part of fuel" contains ^{237}Np in addition to plutonium. Basing on these data, it is possible to select the appropriate equilibrium regime of NFC. Proper selection of the feed compositions, i.e. fractions of ^{238}Pu and ^{237}Np, makes it possible to attain the same level of fuel plutonium protection for various combinations of ^{238}Pu and ^{237}Np content in feed.

For example, 32%-level of fuel plutonium protection can be attained in case of feed containing (0% ^{237}Np, 52% ^{238}Pu) or (20% ^{237}Np, 43% ^{238}Pu) or (40% ^{237}Np, 32% ^{238}Pu). The latter option corresponds to equal

level of plutonium protection both in fuel and in feed. The line "S" that connects the right ends of the curves shown in Fig. 6.12 may be regarded as an "ultimate option" of the (Np-U-Pu) NFC considered here. The points of this line correspond to particular option of the (Np-U-Pu) NFC where ^{238}U is absent in fuel composition, and its fertile functions passed to ^{238}Pu and ^{237}Np.

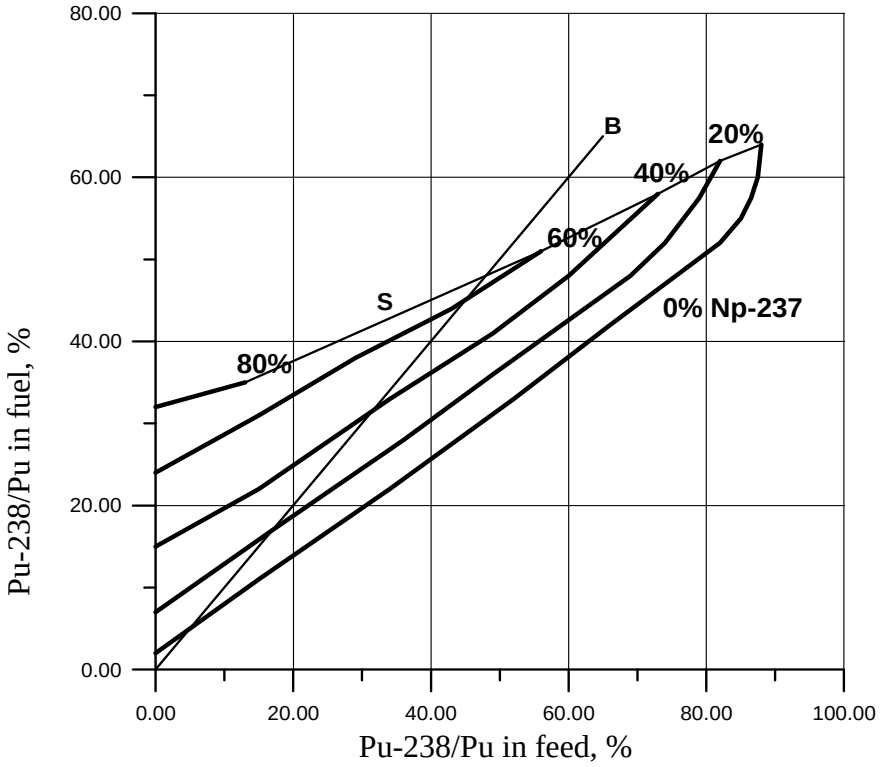


Fig. 6.12. Proliferation protection of plutonium in fuel as function of proliferation protection of plutonium in feed and ^{237}Np content in "feed" part of fuel.

So, this NFC may be called as a (Np-Pu) NFC. In this NFC the highest fuel Pu protection level (65% ^{238}Pu) can be reached with feed Pu

protection of 90% ^{238}Pu. As known, the IAEA safeguards are not applied to plutonium containing 80% ^{238}Pu or more.

Inherent heat generation of plutonium is considered as a significant factor of its protection. The rates of inherent heat generation for various feed compositions are presented in Table 6.5. Here, the rates of specific heat generation for weapons-grade plutonium (WG-Pu) and reactor-grade plutonium (RG-Pu) are presented as well.

Table 6.5

Decay heat generation q(Pu)
and generation of spontaneous fission neutrons n_{sf}(Pu)
in LWR fuel with equal plutonium protection both in fuel and in feed

Plutonium parameters			^{238}Pu/Pu in fuel and in feed		
			Np/(Np + Pu) in feed		
	WG-Pu	RG-Pu	17% (7%)	33% (15%)	44% (19%)
q(Pu), W/kg Pu	2.3	13.	97	186	248
n_{sf},(Pu), 10^6 n/s·kg Pu	0.06	0.38	0.71	1.06	1.30
q (fuel), W/kg fuel	---	---	14.9	41.2	99.5
n_{sf}(fuel), 10^6 n/s·kg fuel	---	---	0.11	0.24	0.53
Feed composition ^{237}Np/^{238}Pu/^{239}Pu, kg/GWe·year	---	---	38/ 82/ 402	103/ 194/ 377	176/ 318/ 421

Basing on the results shown above, it can be concluded that denatured fuel plutonium containing more than 25% ^{238}Pu is characterized by the internal heat generation which exceeds that of RG-Pu by more than order of magnitude and, by the larger extent, that of WG-Pu. In addition, denatured fuel plutonium is characterized by the higher neutron background caused by spontaneous fissions. The factors mentioned above enhance plutonium protection against its utilization in NED. The same factors complicate, to certain degree, the handling procedures with such a fuel in nuclear technologies.

Values of specific heat generation and neutron emission due to spontaneous fission of the loaded MOX-fuel for the equilibrium cycle options analyzed are shown in Table 6.5 too. For comparison, "dry" technology for handling with spent fuel assemblies may be applied if specific heat generation does not exceed 20-35 W/kg fuel. It may be also concluded that plutonium denaturing with ^{238}Pu is restricted by thermal constraints imposed on permissible specific heat generation of fuel.

The same tendency exists in connection with emission of spontaneous neutrons. These constraints need to be taken into account in fuel fabrication, fuel rods and fuel assemblies manufacturing and transport operations. These complications of fuel management may be considered as certain "payment" for proliferation resistance of MOX-fuel cycle.

Actually speaking, the protection of plutonium in (Np-U-Pu)-fuel cycle is supposed to be enhanced due to addition ^{237}Np and ^{238}Pu into fuel. The degree of fissile nuclides protection depends mainly on magnitude of ^{238}Pu fraction in plutonium.

Meanwhile, ^{237}Np itself can be also considered as a potential material for NED. For example, critical mass of ^{237}Np (metal sphere, steel reflector) is about 55 kg. It's ten times more than that of ^{239}Pu. The magnitude of critical mass of ^{237}Np is sensitive with respect of its dilution. For example, minimum critical mass of NpO_2 is as much as 315 kg. Besides, in fuel composition ^{237}Np is present together with plutonium which is characterized by essential neutron source strength due to spontaneous fissions. Therefore, in order to apply extracted ^{237}Np in NED it is needed to perform effective ^{237}Np purification from plutonium (plutonium fraction is restricted by value of 10^{-4} - 10^{-3}.

6.4.4. Proliferation resistance of denatured plutonium

Plutonium can be regarded as a proliferation-proof fissile material only if plutonium is quite unsuitable for manufacturing and military application of even crude and low efficient NED.

As it was already mentioned above, introduction of ^{238}Pu into plutonium isotope composition can provide proliferation protection of the denatured plutonium. Ability of ^{238}Pu to provide a reliable proliferation protection is mainly caused by intense heat generation rate that can warm the plutonium charge in the implosive NED up to unacceptably

high temperatures. The warming-up effect complicates substantially the procedures related with the NED assemblage, storage and transportation. Moreover, the implosive NED can fail as chemical high explosives (HE) can be melted down or destroyed by thermal pyrolysis. So, quantitative evaluations must be carried out to determine ^{238}Pu content in plutonium isotope composition that makes plutonium completely unsuitable for any military applications.

The share of ^{238}Pu required for such a thermal plutonium protection has been evaluated in a series of publications where the implosive NED design consisted of central metal plutonium sphere (charge) surrounded by temper, chemical HE and outer casing. Heat from the outer casing was removed by natural air convection. If HE melting-down is chosen as a criterion of plutonium proliferation protection, then 5% ^{238}Pu is a high enough share. If the HE melting-down criterion is supplemented with the HE self-ignition criterion, then minimal ^{238}Pu content must be increased up to 6%.

One common feature of these publications consisted in assumption that plutonium proliferation protection could be evaluated from equilibrium radial temperature field in the NED components. However, the equilibrium temperature profile establishes after the lapse of a sufficiently long time. Indeed, when the implosive NED is assembled and prepared for application, the NED components begin warming, and the warming-up process can be quick or slow depending on internal heat generation rate. The NED gradually reaches the equilibrium temperature distribution, when internal heat generation rate is completely compensated by external heat removal rate. If the equilibrium state can be reached only after a long enough time interval, then a terrorist could explode the NED before it failed. So, it is evident that the NED failure only after a rather long time interval can not be used as a criterion of plutonium proliferation protection. Only the NED failure for relatively short time interval after its assemblage can confirm sufficiently high level of plutonium proliferation resistance.

Consequently, it is necessary to study non-stationary, time-dependent process of the NED warming-up and evaluate the time intervals till the NED fails for various plutonium isotope compositions and for various mechanisms which can be used to intensify external heat removal. In addition, there are some possibilities to slow down the NED

warming-up process by means, for example, of the following counter-measures:

- ✓ Preliminary cool-down of the NED components.
- ✓ Application of additional heat-conducting layers.
- ✓ Introduction of heat-isolating layers for purposeful re-distribution of radial temperature profile under which the NED could keep its efficiency as long as possible.

These measures can remarkably increase the minimal ^{238}Pu contents for sufficient proliferation protection of the denatured plutonium.

Model of a hypothetical implosive NED

Geometrical model of a hypothetical implosive NED (HNED) is shown in Fig. 6.13.

Central plutonium charge is surrounded by spherical layers of natural uranium, aluminum, chemical HE and outer steel casing. The figure also demonstrates some technology levels (low, medium and high technology) with different thicknesses of spherical layers and with different types of chemical HE. Main relevant properties of effective chemical HE are presented in Table 6.6.

Table 6.6

Thermophysical properties of chemical HE

High explosive	Density, g/cm^3	Heat conductivity, W/m·K	Melting point, ^0C	Self-ignition point, ^0C
Composition B	1,74	0,219	79	214
CYCLOTOL	1,77	0,226	79	208
TNT	1,45	0,259	81	288
HMX	1,84	0,406	256 ÷ 286	259
TATB	1,89	0,544	448	347

As is seen, TATB is characterized by the best heat conductivity and the highest acceptable temperatures. So, it seems reasonable to use TATB as a chemical HE in the HNED. This means that the HNED with TATB as a chemical HE can keep its efficiency in the warming-up process up to the longer time intervals than other chemical HE can pro-

vide. Therefore, the use of TATB in the HNED represents the largest threat from the standpoint of the HNED long-term efficiency.

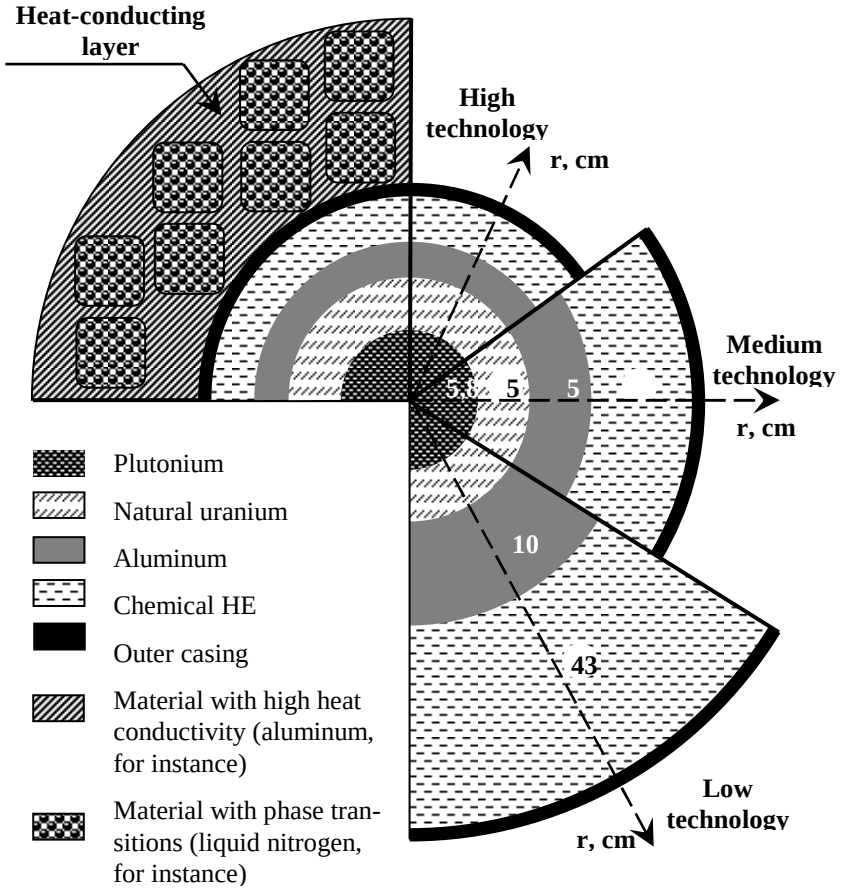


Fig. 6.13. Geometrical model of a hypothetical implosive NED

Thus, to provoke the HNED failure, it is necessary to introduce sufficiently intense internal heat source into its charge, i.e. introduce ^{238}Pu into plutonium isotope composition.

Numerical studies were carried out to assess effectiveness of the countermeasures on prolongation of the time interval till the HNED

failed. This time interval may be called as the HNED lifetime. Content of ^{238}Pu in plutonium charge that provides only very short lifetime can be adopted as a sufficient value for plutonium proliferation protection.

Radial temperature distribution in the geometrical model of the implosive HNED (Fig. 6.13) can be determined through iterative solution of the following differential equation:

$$\text{div}\left[\lambda(r,T)\cdot \text{grad } T(r,\tau)\right] + q_V(r) = c_V(r,T)\cdot\frac{\partial T(r,\tau)}{\partial \tau},$$

where $\lambda(r,T)$, $c_V(r,T)$ – heat conductivity and heat capacity, respectively, which depend on temperature $T(r,\tau)$; $q_V(r)$ – intensity of internal heat source.

Three options for external boundary conditions were used in accordance with different heat removal mechanisms:

1. Ideal heat removal: $T(R_S, \tau) = T_S$, i.e. temperature at outer surface ($r=R_S$) is a time-independent value.

2. Ideal heat isolation: $\left.\frac{\partial T(r,\tau)}{\partial r}\right|_{r=R_S} = 0.$

3. Heat is removed by natural air convection and thermal radiation only.

Criteria for the implosive HNED failure

The most temperature-sensitive component of the implosive HNED is a chemical HE that self-ignites at 347^{0}C. In addition to the melting and self-ignition problems, one else temperature-dependent process can lead to the HNED failure, namely the thermal pyrolysis process. At elevated temperatures the thermal pyrolysis results in intense emission of gaseous products capable to destroy HE. According to some experimental data, chemical HE failed when rather small fraction (0.02%) of the pyrolysis products was accumulated in HE. However, this value can vary within a wide range (from 0.02% to 2%), depending on the HNED design and conditions for HE applications. So, pyrolytic dissociation of 2% HE molecules can be adopted as an upper border of the HE temperature resistance.

Rate of the HE pyrolytic dissociation can be evaluated with application of Arrhenius equation:

$$W(T) = B \cdot \exp\left[-E_{ACT}/\left(R_0 \cdot T\right)\right];$$

where W(T) – dissociation rate at temperature T; B – pre-exponential factor; E_{ACT} – energy of activation; R_0 – universal gas constant (8.31 J/mol·K).

Experimental studies on TATB parameters gave the following results: $\log_{10}B = 11.6\ s^{-1}$, $E_{ACT} = 172.6$ kJ/mol. Fraction of the HE molecules destroyed up to the time moment τ can be calculated by integrating the dissociation rate:

$$\varepsilon(\tau) = \int_0^{\tau} W\left[T(\tau')\right] \cdot d\tau'.$$

At self-ignition point of TATB, i.e. at 347^0C, the upper border of TATB stability (2%) can be reached in eighteen seconds. However, even at the lower temperatures, the upper border can be reached in a rather short time interval (for instance, in five minutes at 300^0C).

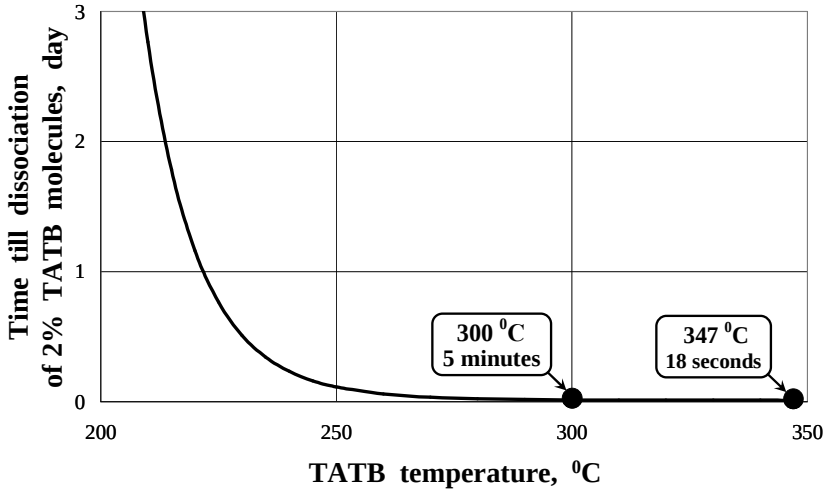


Fig. 6.14. Temperature dependency of TATB dissociation

Thus, the strictest criterion of the implosive HNED efficiency is the thermal dissociation of 2% HE molecules. It is obvious also that the implosive HNED becomes unsuitable for any practical applications if the HNED fails within the time interval needed for its assemblage and transportation. As the HNED assemblage and transportation times can not be evaluated exactly, it seems reasonable to determine the range of the HNED lifetime.

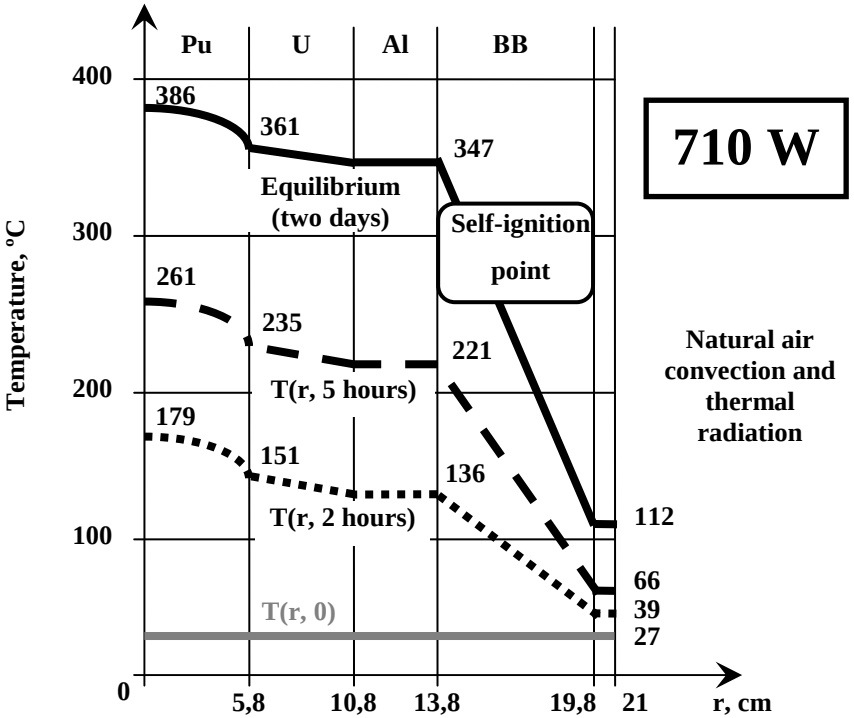


Fig. 6.15. Time evolution of radial temperature profile in the implosive HNED

The necessity to trace the non-stationary warming-up process is illustrated by the curves presented in Fig. 6.15. These curves, time-dependent radial temperature profiles in the high-technology implosive HNED, were calculated under the following assumptions:

- Initial temperature of all the HNED components - 27^0C.
- Plutonium isotope composition corresponds to 710 W of the thermal power generated by plutonium charge.
- External heat removal is provided by natural air convection and thermal radiation only.
- Criterion of the HNED failure is an initiation of the TATB self-ignition process at 347^0C.

As is seen, when radial temperature profiles have reached their equilibrium states (in about two days), maximal TATB temperature has increased up to 347^0C, i.e. the TATB self-ignition could be initiated. However, after the shorter time intervals (two and five hours) the TATB temperatures were well below the self-ignition point. As a consequence, the HNED could keep its efficiency during a relatively long time (almost two days).

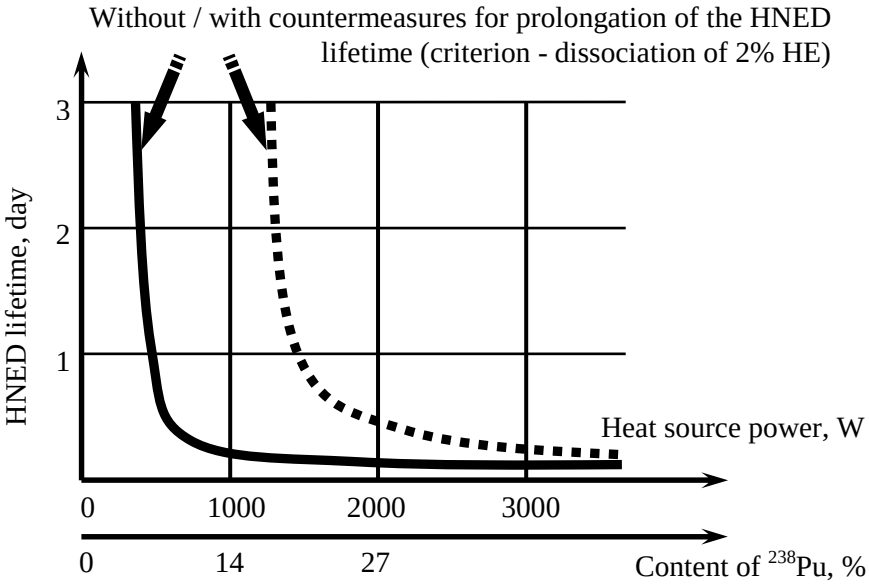


Fig. 6.16. HNED lifetime as a function of heat source power

So, it can not be stated that the denatured plutonium with total thermal power of 710 W is the proliferation-proof fissile material because the HNED failure occurs only at its thermal equilibrium state.

The asymptotic model, which analyzes equilibrium temperature profiles only, can underestimate the thermal power of internal heat source needed to provide proliferation protection of the denatured plutonium.

How short time must elapse prior to the HNED failure for the denatured plutonium to be regarded as a material completely unsuitable for military applications? In other words, how short the HNED lifetime is acceptable from nuclear non-proliferation point of view? Evidently, the countermeasures capable to prolong the HNED lifetime can increase substantially the required intensity of internal heat source, i.e. ^{238}Pu content in the denatured plutonium.

Efficiency of measures for prolongation of the HNED lifetime

Preliminary cool-down of the HNED components

The preliminarily cooled HNED can keep its efficiency for the longer time interval as compared with the HNED without preliminary cool-down. However, heat capacities become very small at cryogenic temperatures. So, the utmost possible cool-down of the HNED components makes no sense for prolongation of the HNED lifetime. For example, if the HNED was preliminarily cooled down to liquid nitrogen point (77 K) instead of liquid helium point (4 K), then the power of internal heat source required for plutonium proliferation protection increased very insignificantly, on 3% only.

The following scenario of preliminary cool-down was studied numerically. All the HNED layers were cooled down to liquid nitrogen point (77 K) with exception of central plutonium charge which was cooled down to 198 K only. At temperatures below 198 K, plutonium in δ-phase (stabilized with molybdenum, for instance) transforms into α-phase, and this irreversible transition is accompanied by remarkable enlargement of plutonium volume. So, such a phase transformation can lead to a partial or full HNED failure. The scenario presumes that, after preliminary cool-down, the HNED is completely isolated from the environment for the utmost long maintenance of the cooled state.

Such a preliminary cool-down resulted in a considerable increase (on 50%) of the heat source power needed for plutonium proliferation protection as compared with the HNED without any cool-down.

Application of additional heat-conducting layer

The option of ideal heat removal can be practically provided by encircling the HNED with a heat-conducting layer that contains a material able to undergo phase transformations. Indeed, such a layer can absorb all the ingoing heat without remarkable warming-up.

The heat-conducting layer can consist of a material with high heat conductivity (aluminum, for instance) and a material with phase transformation at temperatures near to the initial HNED temperature (77 K, liquid nitrogen point).

Numerical evaluations revealed that the heat-conducting layer (22 cm thick) consisting of 25% Al and 75% N_2 can absorb all the ingoing heat without remarkable warming-up. The ideal heat-removal option can increase on 15% the heat source power required for plutonium proliferation protection.

Introduction of heat-isolating layers for re-distribution of temperature profile

As is seen from Fig. 6.15, when the HNED fails due to the unacceptable warming-up of chemical HE, other HNED components (plutonium, uranium, aluminum) are far from their acceptable temperatures. So, it seems reasonable to undertake some measures that could put obstacles in the way of heat transport from inner layers to chemical HE. For example, thin layer of a material with low heat conductivity could be placed between aluminum and chemical HE.

The heat-isolating materials must be characterized by low heat conductivities and high acceptable temperatures. Probably, quartz aerogel is the most attractive material for the heat-isolating layers. Quartz aerogel is characterized by very low heat conductivity (0,004 W/m·K) and sufficiently high acceptable temperature (up to 1200 ^{0}C).

Numerical evaluations revealed that introduction of the heat-isolating layers into the HNED structure for the desirable re-distribution

of radial temperature profiles could lead to the higher (up to 50%) heat source power required for plutonium proliferation protection.

Thus, it may be expected that all three countermeasures on prolongation of the HNED lifetime are able to toughen substantially the requirements to the heat source power (see Fig. 6.16).

Recommendations on proliferation protection of the denatured plutonium

If all the aforementioned countermeasures (preliminary cool-down to cryogenic temperatures, encircling the HNED by the heat-conducting layer, introduction of the heat-isolating layers into the HNED structure) on prolongation of the HNED lifetime are undertaken simultaneously, then some recommendations can be worked out on ^{238}Pu content in the denatured plutonium. ^{238}Pu must provide so intense internal heat source that the HNED lifetime becomes unacceptably short for potential proliferators. For example, 5-hour lifetime may be adopted as a target value because it quite improbable that the HNED assemblage and transportation could be performed for so short time interval.

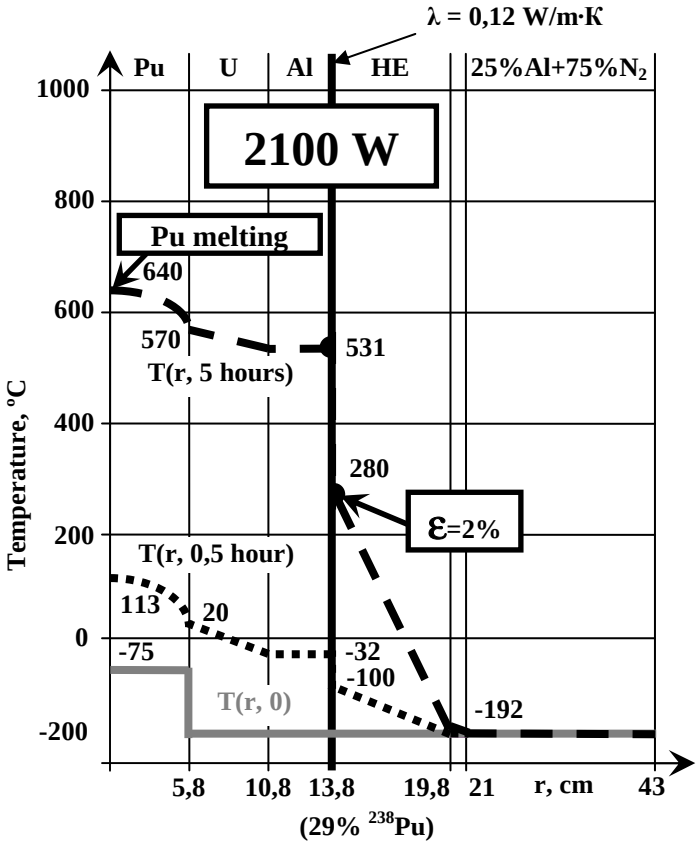
Radial temperature profiles in the high-technology implosive HNED are presented in Fig. 6.17 for the following two cases:

1. Plutonium melting leads to the HNED failure. One heat-isolating layer is introduced into the HNED structure by such a way that plutonium melting and thermal dissociation of 2% HE molecules would occur simultaneously.
2. Plutonium melting does not lead to the HNED failure. Three heat-isolating layers are introduced into the HNED structure by such a way that maximal acceptable temperature of the inmost heat- isolating layer, uranium melting, aluminum melting and thermal dissociation of 2% HE molecules would occur simultaneously.

If plutonium melting is not a reason for the HNED failure, then the required heat source power must be equal to 3100 W (maximal evaluation, see Fig. 6.17, b). This means that only plutonium containing above 42% ^{238}Pu can be regarded as a proliferation-proof material.

If the target value of the HNED lifetime prolonged up to five days, then minimal content of ^{238}Pu for plutonium proliferation protection dropped down to 18%.

Evidently, the numerical evaluations obtained for the high-technology implosive HNED are quite correct for the low- and medium-technology HNED with relatively thicker layers of aluminum and chemical HE. The weaker capabilities of heat removal from these HNED allowed us to conclude that the lower contents of ^{238}Pu (in comparison with the high-technology HNED) could be sufficient to provoke the HNED failure.



a)

Fig. 6.17. Radial temperature profiles in the HNED, when all the measures on prolongation of the HNED lifetime were undertaken
a) Plutonium melting leads to the HNED failure

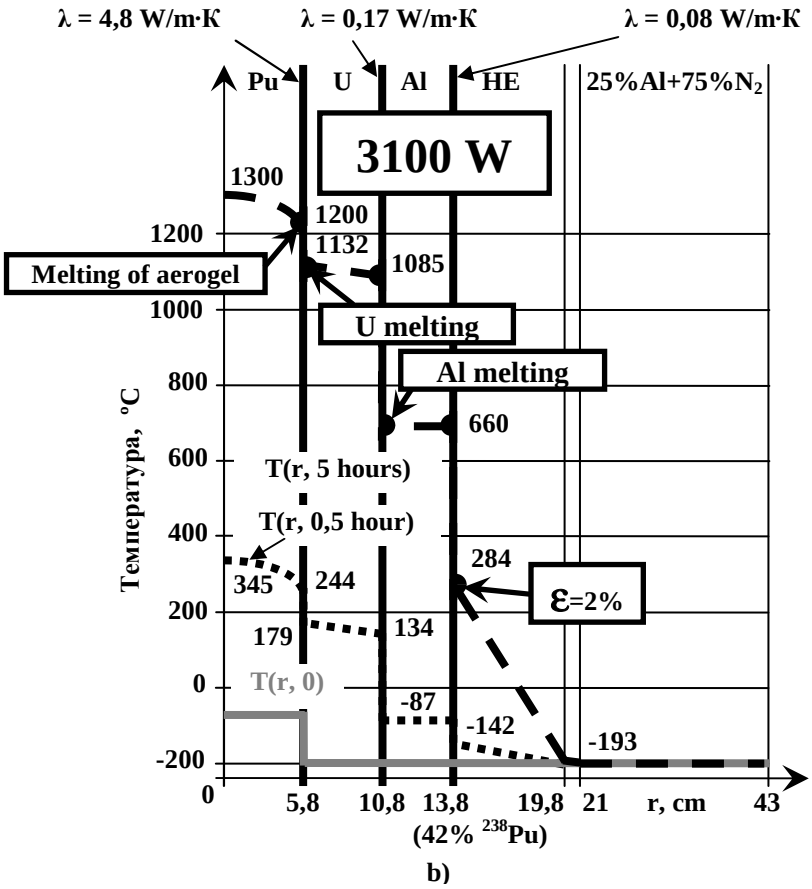


Fig. 6.17. Radial temperature profiles in the HNED, when all the measures on prolongation of the HNED lifetime were undertaken
b) Plutonium melting does not lead to the HNED failure

According to the IAEA documents, plutonium containing above 80% ^{238}Pu.is regarded as a proliferation-protected material. Discrepancy between the IAEA-declared value (80% ^{238}Pu) and the results of previous numerical evaluations (18-42% ^{238}Pu) can be explained by the following considerations:

1. The adopted criterion for thermal stability of chemical HE, i.e. dissociation of 2% HE molecules.

2. The adopted criterion for the HNED failure, i.e. the HNED lifetime must be no longer than five hours. By the way, if plutonium contains 80% ^{238}Pu, then the HNED fails in two hours.
3. The chosen geometrical model of the implosive HNED.
4. The chosen chemical HE with the highest heat conductivity and maximal acceptable temperature.

6.4.5. Higher fuel burn-up in denatured (Np-U-Pu) NFC

Good neutron-multiplying properties of ^{238}Pu and its neutron predecessor ^{237}Np make it possible to extend substantially time period for continuous reactor operation without refuelings. As a consequence, unauthorized extraction of plutonium from SNF becomes practically unfeasible. Indeed, neutron irradiation of (Np-U-Pu) fuel initiates the following “non-traditional” transition chain:

$$^{237}\text{Np}(n,\gamma)^{238}\text{Pu}(n,\gamma)^{239}\text{Pu}(n,\gamma)...$$

A successive transition of these nuclides leads to enhancement of neutron-multiplying properties.

Actually, as it can be seen in Fig. 6.18, excess neutron generation per one absorption (ν_{eff}-1) in ^{237}Np is negative for neutrons of all energy range (excepting fast neutrons), positive for neutrons with $E_n > 1$ keV for ^{238}Pu and, as is known, essential positive one for ^{239}Pu. So, for (Np-U-Pu)-fuel the nuclides we are dealing with can be characterized as follows: ^{237}Np plays a role of “burnable poison", ^{238}Pu is a moderately fissile nuclide by fast neutrons ($E_n > 1$ keV), and ^{239}Pu is a well-known fissile nuclide with excellent neutron-multiplying properties.

At the same time, during irradiation in the reactor core, FP accumulation results in growth of neutron absorption. So, these tendencies can be counterbalanced, and such fuel will be characterized by stabilized neutron-multiplying properties over long fuel lifetime and, as a consequence, high fuel burn-up.

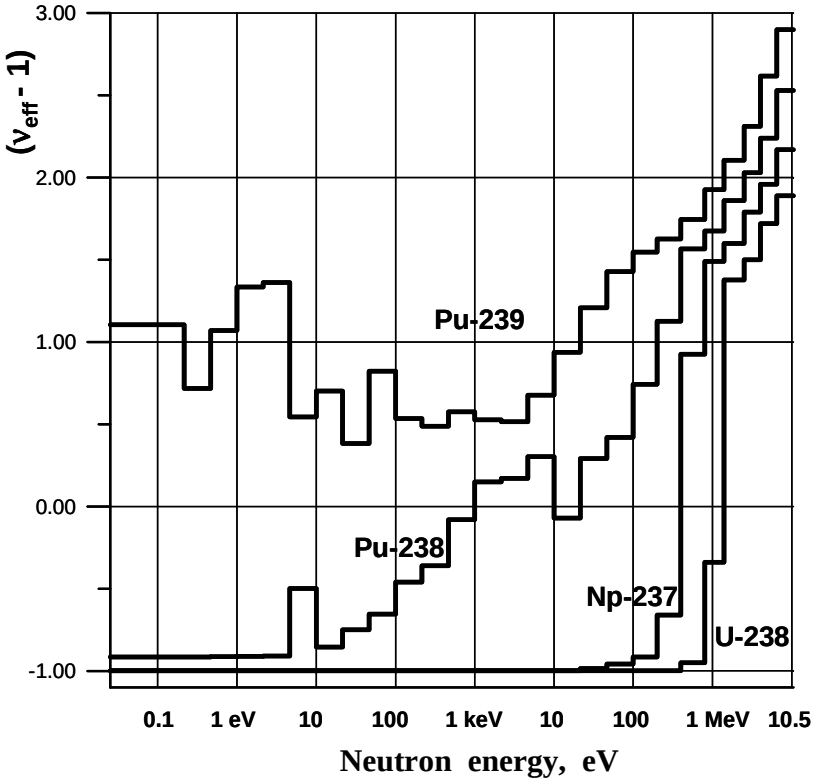


Fig. 6.18. Dependencies of excessive neutron number per one absorption (ν_{eff}-1) on neutron energy for nuclides of (Np-U-Pu) fuel.

Burn-up calculations for mono-nitride (Np-U-Pu) fuel in elementary cell of PWR-type reactor with heavy water as a coolant were performed by using code GETERA. The cell parameters are presented in Table 6.7 (they were similar to those of VVER-1000 cell).

Table 6.7

Cell parameters of PWR-type reactor

Diameter of fuel rod, mm	9.1
Thickness of steel cladding, mm	0.4
Coolant	D_2O
Volume of D_2O / Volume of fuel	1.6
Fuel	(Np-U-Pu)N (porosity - 30%)
Specific heat generation, kW/l	110

The dependencies of K_∞ on fuel burn-up are shown in Fig. 6.19 for various fuel compositions. For comparison it is demonstrated also a similar curve of K_∞ for a standard LWR-UOX reactor. It can be seen that, actually, there is a possibility to attain fuel burn-up of 25-30%HM (corresponding fuel lifetime is about 20-25 years). It is worth-while mentioning that vibration-packed MOX fuel in stainless steel cladding was irradiated in fast reactor BOR-60 (Russia), and it was obtained burn-up of 26% HM on standard fuel assemblies and burn-up of 32% HM in experimental fuel rods. No thermal-mechanical and physical-chemical fuel-cladding interactions were observed in any of the analyzed spent fuel rods.

The results mentioned above referred to so-called "ultimate" fuel compositions which didn't contain ^{238}U at all. Actually speaking, these results can be considered as preliminary ones to demonstrate scale of benefit. Undoubtedly, it is needed to analyze impact of wide fuel compositions (including ^{238}U) on stabilized multiplication properties of ultra long-life cores taking into consideration reactor safety in both critical and sub-critical operation modes. Anyway, application of ultra long-life core concepts based on the denatured (Np-U-Pu) fuel can lead to essential decrease of SNF flow rate, reduction of reprocessing, remanufacturing and shipping operations.

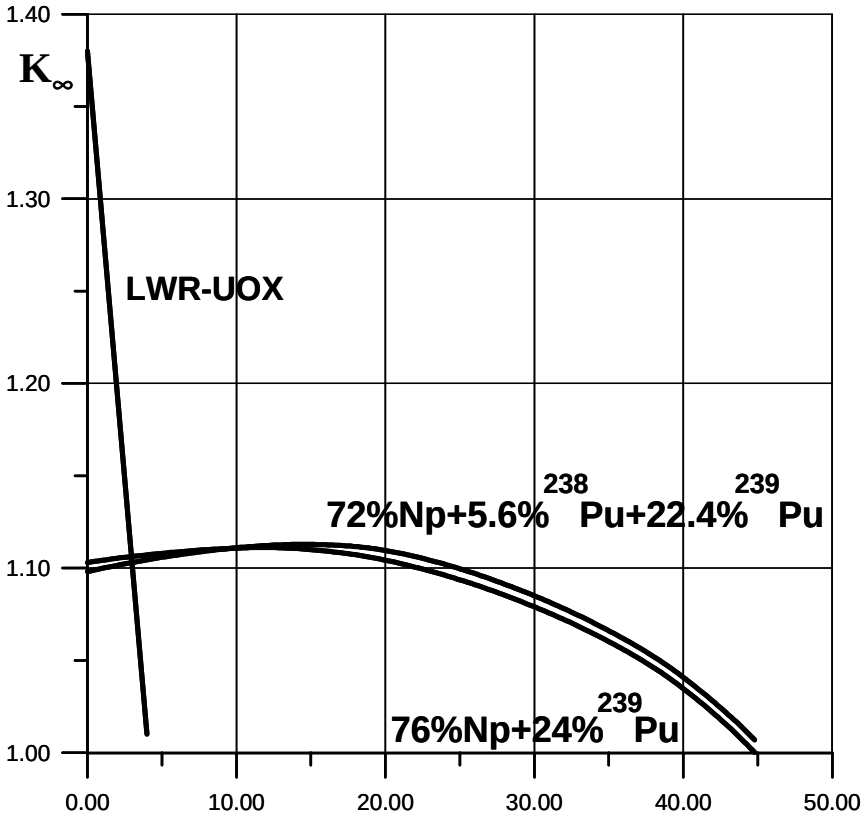


Fig. 6.19. Dependencies of K_∞ on fuel burn-up (% HM) for various fuel compositions

It's a factor for internationalization of Nuclear Energy System fuel cycle. Since fuel cycles under consideration here are "rich" with respect to excess neutron generation in CFR, there is no necessity to perform fine purification of spent fuel by the SNF reprocessing technologies. It's a factor of enhancement of NFC proliferation protection.

Application of NPP with ultra long-life core concepts is expected to be profitable for electricity generation in the developing countries which have not improved nuclear infrastructure.

Control questions to Chapter 6

1. What methods can be used to prevent diversion of fissile NM from peaceful to military purposes?
2. What properties of ^{231}Pa and ^{232}U make it possible to use these isotopes for uranium proliferation protection?
3. What properties of ^{237}Np and ^{238}Pu make it possible to use these isotopes for plutonium proliferation protection?
4. What advantages can be offered by power reactors with ultra-high fuel burn-up for nuclear non-proliferation regime?

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Textbook

Editor I.N. Markina
The original layout is made by V.A. Apse and G.G. Kulikov

Decision on publication 30.06.2014. Format 60x84 1/16
Quires 14. Educational quires 14. Circulation 100 copies
Request №008-3, Order №

National Research Nuclear University MEPhI (Moscow Engineering Physics Institute)
NRNU MEPhI printing house
115409, Moscow, Kashirskoe shosse, 31