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Original Article

Studies of laser cleaning and decontamination in liquid and polymerized media

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ABSTRACT

The study highlights the importance of developing new methods for decontaminating surface radioactive contamination caused by the accumulation of substances such as uranium and plutonium on equipment. Pulsed laser cleaning, which is particularly effective in combination with liquid media, is seen as a promising solution. To safely test the technology, preliminary experiments were carried out on model samples of AISI 321 stainless steel contaminated with cerium oxide, and then on real contaminants of uranium and plutonium salts. Different liquids were used to minimize aerosol release and prevent surface overheating: water, acetone, isopropyl alcohol and polyvinyl alcohol (PVA) solution. Laser cleaning in PVA showed high efficiency, removing more than 90 % of the contaminants in a single pass. Optimal laser modes minimized structural changes to the material surface. The PVA reliably fixes contaminants and, after polymerization, is easily removed from the sample as a solid film.

1. Introduction

At present, nuclear power programs are being actively developed and new nuclear power plants are being planned to meet the growing demand for energy. Operation of nuclear power plants and industry facilities inevitably generates radioactive surface contamination. The most important element of the operating technology of such facilities is the large-scale decontamination of the surfaces of structures and equipment that have accumulated uranium and plutonium compounds during their long-term operation. The materials resulting from the decontamination should be technologically used and buried. Due to the high cost of such work, there is a need to develop safe, efficient and low-waste methods for decontaminating surface radioactive contamination. Such methods should focus on maximum compactness of the removed radioactive contamination in a form convenient for further handling.

Studies [1–3] have shown that most of the radioactive contaminations is concentrated in the surface layers of the material and in the oxide film. The use of laser cleaning methods makes it possible to effectively remove such surface layers together with the various contaminants [4–7]. Such methods facilitate the solution of waste compaction tasks, are amenable to automation, provide a high level of safety due to the possibility of removing the operator from the place of work. Pulsed laser cleaning makes it possible to control the volume of material removed and the degree of surface heating. The use of nanosecond pulses reduces the modification of the original surface. The depth of thermal penetration l_t during a pulse τ_p in a material with thermal diffusivity α is determined by Ref. [8]:

$$l_{t=}2\sqrt{\alpha\tau_p}$$

With a pulse duration of 8 ns, the depth of thermal penetration for steel is no more than 1 μm. In this case, evaporation occurs with minimal melting of the substrate material [9–11]. The combination of laser and liquid cleaning methods [12,13] is a particularly promising approach. The use of liquid improves the oxide removal process, reduces thermal damage to the surface [14], and increases the uniformity of cleaning, making the method promising for industrial application. Laser cleaning in liquid media makes it possible to avoid the use of complex, large-scale equipment for filtering aerosols formed during cleaning in gaseous media. Contaminants removed from the surface are captured by the liquid layer and held securely in place. There is also work on the removal of radioactive contamination from surfaces by adsorption of polymerizing compositions for surface decontamination [15–17]. After curing

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and removal from the surface, such compositions are able to retain the removed particles. This facilitates further work with the waste.

This paper presents the results of experimental studies on laser cleaning of AISI 321 stainless steel with simulated non-radioactive contamination [18,19] based on cerium oxide IV (CeO_2). The results of a study on laser decontamination of radioactive contaminants on the surface of stainless-steel containing uranium (U) and plutonium (Pu) salts are also presented. The experiments were carried out under a layer of one of the following liquids: water, acetone, isopropyl alcohol and a decontamination composition based on an aqueous solution of polyvinyl alcohol (PVA). A laboratory YAG:Nd laser with an emission wavelength of 1064 nm, operating in the goodness modulation mode, was used as the laser radiation source. Thus, the key part of the work was to select the optimal laser operating mode for effective cleaning of the contaminated surface by liquid layers of different viscosities. At the same time, the mode should not lead to aerosol escape from the liquid layer, nor to its significant overheating and evaporation. Modification of the surface layer of the cleaned material should be minimal.

2. Experiment, methodology

A laboratory bench (Fig. 1) was constructed to perform laser cleaning experiments. Radiation is output from the YAG:Nd laser (1). This allowed a focusing lens (2) with a focal length of f = 100 mm to be used to inject the radiation into the fiber (3), d = 600 μm. After the fiber, the radiation was collimated using a lens (4) and then then transferred to a galvanometric scanner (5,6). The scanner was then used to direct the radiation onto the surface of the sample (9). The sample was placed in a glass cuvette (7) that was filled with the 8 ml of the liquid to be tested (8). The liquid medium used was one of the following: distilled water, acetone, or isopropyl alcohol. The thickness of the liquid layer in front of the target was minimized to avoid chromatic dispersion and spatial stretching of short intense radiation pulses; its characteristic value was 3 mm.

The conditions for cleaning rough surfaces have been greatly improved by the use of a fiber. A large number of modes propagating at different angles to the fiber axis are created when laser radiation is propagated through a multimode fiber. This trend continues after focusing. Such a fan-shaped beam is able to penetrate well into narrow slots and cavities in rough surfaces [20].

Three scanning modes have been employed throughout the study, depending on the type of the overlap between the adjacent irradiated regions that correspond to the laser spot on the surface: S = d, S = d/2 and S = d/4, where S is the step between the centers of the adjacent spots, d is the beam diameter determined by the standard technique at the level of $1/e^2$ of the peak intensity (Fig. 2). The laser beam parameters were measured at the output of the Galvano scanner (Table 1).

A polymerization gel was also used in the laser cleaning experiments. This gel is a 10 % aqueous solution of polyvinyl alcohol (PVA). In the case of the PVA, the cleaning procedure was as follows: first, the sample was placed vertically in the polytetrafluoroethylene holder instead of in the cuvette. PVA was applied to its surface with a spatula. Then cleaning with a laser beam was performed. Next, the sample was removed from the holder and sent for drying in a horizontal position (Fig. 3, a). At the same time, most of the PVA remained on the surface of the sample and polymerized completely within 10–12 h at room temperature. The film was uniform in thickness and easily removed from the sample surface without damage after drying and polymerization (Fig. 3, b).

3. Results and discussion

3.1. Model contaminants

To thoroughly and safely investigate the processes involved in laser decontamination, it was decided to conduct experiments using non-radioactive coatings simulating uranium-plutonium radioactive contamination. To achieve the most realistic simulation of hard-to-remove contamination, the modelling element should be in a non-water-soluble oxide form and carefully incorporated into the oxide film on the metal surface [21].

The selection of the material to simulate the fouling was based on the proximity of melting and boiling temperatures and heat capacity to the real fouling. Cerium dioxide (CeO_2) of natural isotopic composition was chosen to simulate uranium (UO_2) and plutonium (PuO_2) dioxide contamination, since its thermophysical properties are like those of these radioactive materials [22–25].

The model specimens were prepared in the form of AISI 321 chromium-nickel steel plates with dimensions of 30 × 30 mm and thickness of 2 mm. The preparation is carried out in 4 stages according to the developed methodology [26]: 1) surface preparation by anodic electrochemical etching of each sample in a 5 % HCl solution (current density of 20–25 A/dm2); 2) maintenance with an aqueous solution of cerium (III) nitrate at a concentration of 0.5 mol/l; 3) annealing in an air atmosphere at a temperature of 750 C; 4) ultrasonic cleaning, natural drying. The character of the distribution of the initial model impurities on the surface of the samples was investigated by the EDX method on the SEM using a Tescan MAIA-3 SBH microscope (Fig. 4). Cerium was found in the form of individual deposits of 0.5–1 μm in size or in the form of

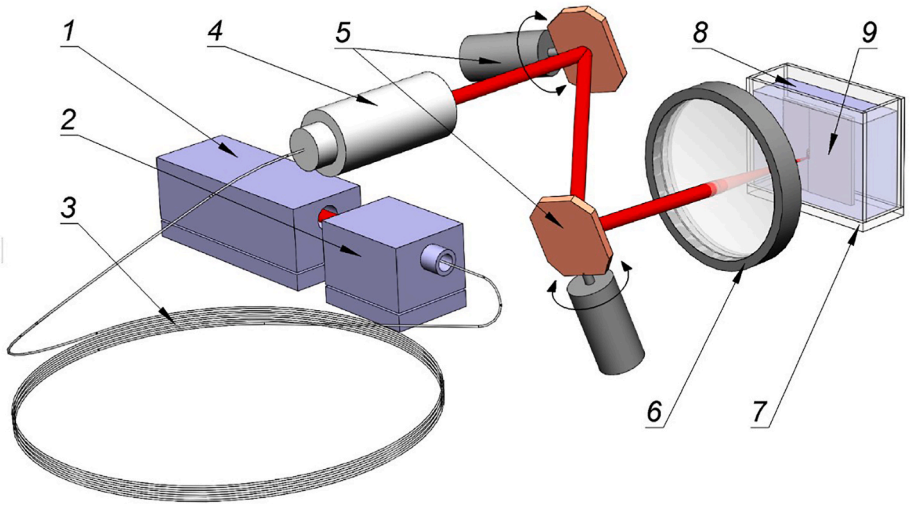

Fig. 1. Optical scheme of the lab bench (1 - YAG:Nd laser, 2 - focusing system, 3 - optical fibre, 4 - collimator, 5 - galvanic mirror, 6 - f-theta lens, 7 - cuvette, 8 - liquid, 9 - sample).

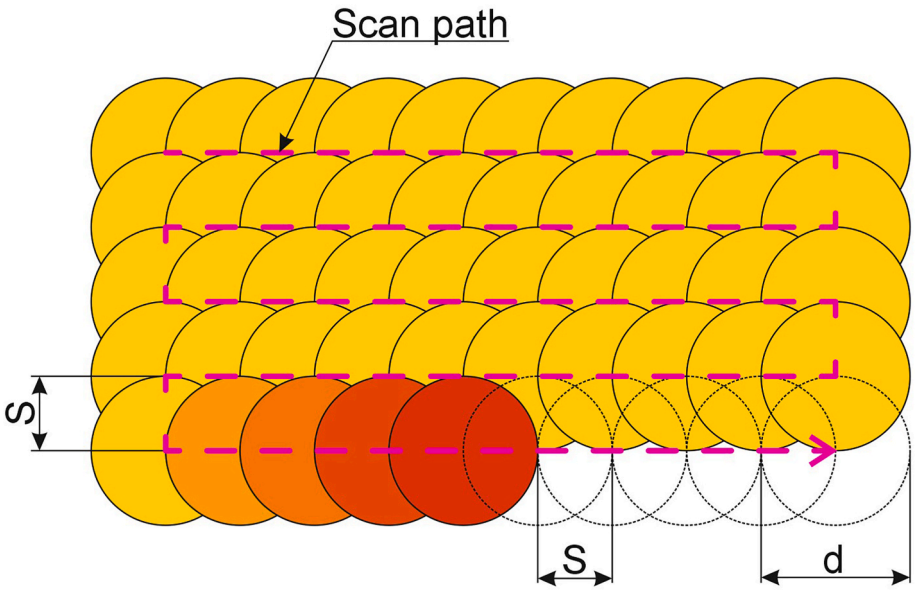

Fig. 2. Scanning trajectory.

Table 1
Parameters of laser radiation.

Parameter	Value
Spot diameter, mm	0,8
Pulse duration, ns	8
Pulse repetition frequency, Hz	100
Pulse energy, mJ	11,9

agglomerates of up to 10 μm in size.

X-ray fluorescence analysis (XRF) using a Shimadzu Lab Centre XRF-1800 wave dispersion spectrometer was used for relative quantification of the content of model impurities on the surface of the samples. The cleaning coefficient (CC) counted the ratio of outgoing secondary radiation atomic cerium.

Analysis of the data obtained (Fig. 5) shows that a significant proportion of the contamination is removed after the first pass. This indicates the high efficiency of the process in the initial stage. An additional mechanism that contributes to the removal of contaminants is the process of cavitation bubble collapse. The efficiency of this mechanism is largely determined by the viscosity of the fluid used in the cleaning process. Thus, when considering fluids such as acetone (viscosity at 25 °C - 0.295 mPa s) and water (viscosity at 25 °C - 0.89 mPa s), it can be observed that isopropyl alcohol (viscosity at 25 °C - 2.43 mPa s) showed significantly lower residual contaminant levels in all overlap modes compared to them.

When cleaning in liquid media (acetone, water, isopropyl alcohol), the ablated particles settle to the bottom of the cuvette, which determines their influence on further processing cycles. As a result, in liquids, a significant increase in the cleaning coefficient is observed as the number of passes increases. After only two cycles of treatment in isopropyl alcohol, 89.2 % of the contaminants were removed, and when the number of cycles was increased to five, this indicator rose to 94.3 %. It can be concluded that the number of cycles and the properties of the liquids play a key role in achieving a high level of cleaning.

And in the case of using PVA (viscosity at 25 °C - 21 mPa s), the values of the cleaning coefficient are achieved in the first pass - C_c = 90.7 % (at S = d), C_c = 91.2 % (at S = d/2), C_c = 90.1 % (at S = d/4). The removed contaminants were well fixed by the film. The boundary between the treated and untreated areas is clearly visible. If the surface is treated several times (2 and 5 passes), a slight improvement in cleanliness may be observed. However, the effect is usually not as significant as it may appear, especially if a certain level of cleaning has already been achieved on the first pass. This is due to the overlap of the beam with the ablated particles in the highly viscous PVA volume. This may also explain the lack of increase in the cleaning factor with increasing overlap factor.

During the experiments, it was found that PVA forms bubbles under laser irradiation, which leads to foaming of the polymer. The result was an inhomogeneous structure of the final film after polymer solidification. The gassing was caused by the overheating of the polymer, both due to its own absorption of radiation and due to heat from the surface of the sample. It was decided to modify the beam trajectory to avoid foaming, the formation of structural defects and the likely violation of

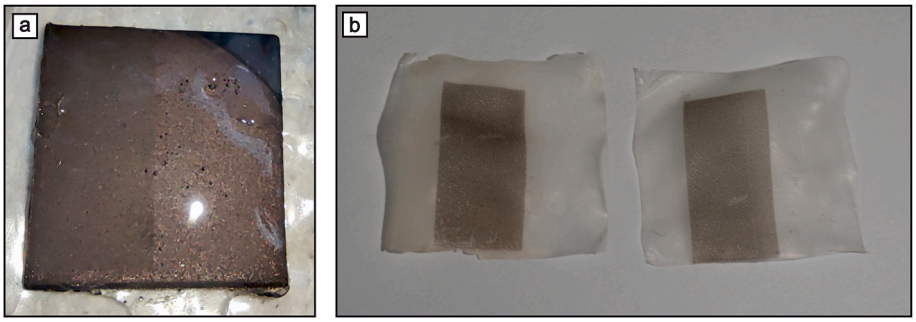

Fig. 3. PVA: (a) on the sample surface after laser cleaning, (b) dried film removed from the sample surface.

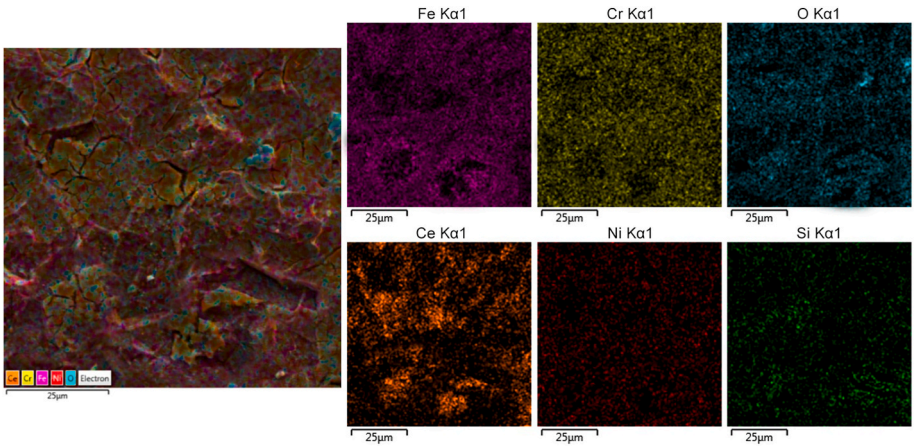

Fig. 4. SEM of the model sample surface and mapping obtained by the EDX method.

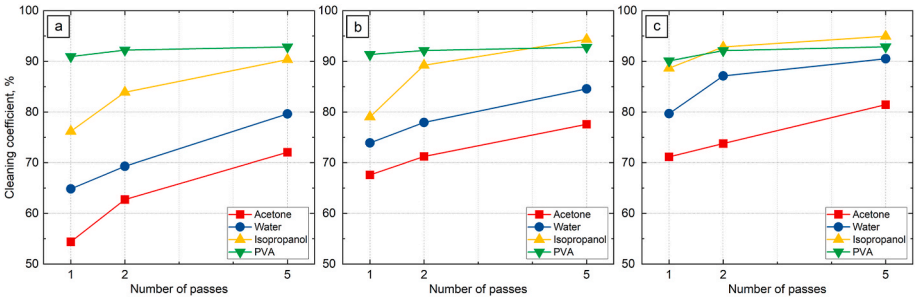

Fig. 5. Dependence of cleaning coefficient on fluid viscosity, number of passes and overlapping mode: (a) S = d, (b) S = d/2, (c) S = d/4.

continuity. The overlapping of the spots was maintained, but the distance between adjacent rows was first increased, and then the gap between the rows was filled during the next beam passes. This technique reduced the local temperature of the PVA and minimized bubble formation.

The thickness of the polymerized film was approximately 60 μm. Inspection of the inner surface of the cured PVA film by SEM (Fig. 6) revealed ablated fragments. The image shows a clear boundary between the treated and untreated surfaces. In the laser-affected area, the particles are uniformly distributed over the film surface. Individual particles are also present in the untreated area, but in smaller numbers. These particles were probably detached from the surface after PVA polymerization.

Optical microscopy (Fig. 7) further showed that the removed particles were not only distributed on the inner surface of the film, but also penetrated into its volume in the laser interaction region. This indicates that not only surface but also volume redistribution of the material occurred under the influence of laser radiation, which may be related to local thermomechanical effects in the ablation process.

The number of bubbles and defects inside the sample and in the polymer volume near the surface of the sample was higher with increasing laser spot overlap. The outside of the film was smooth and free of defects. Even when the target was significantly overheated and the PVA began to boil, the continuity of the film was not affected. The film reliably trapped all laser ablation products without allowing them to pass through itself into the surrounding space.

3.2. *Real contaminants*

Decontamination experiments were carried out on the laboratory bench. Decontamination studies were carried out on stainless steel samples of radioactively contaminated AISI 321 stainless steel tray. Such equipment was used for 12 years for work with aqueous solutions of uranium and plutonium salts. The exact isotopic composition of the contamination was not determined. The distribution of the contamination over the surface was uneven. The level of initial and residual

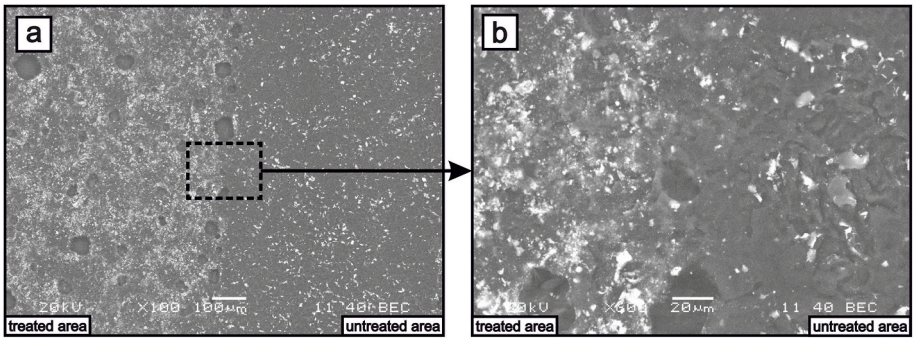

Fig. 6. SEM images of the inner surface of the PVA film.

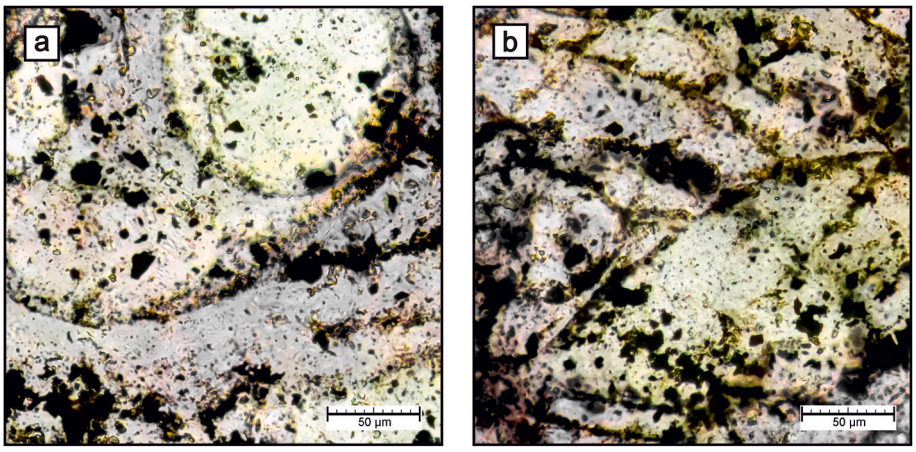

Fig. 7. Optical microscopy images of the PVA film.

contamination of the surface was determined by radiometric control using a dosimeter-radiometer MKS-AT1117 with a detection unit BDPA-01 to measure the flux density of alpha particles. A DKS 96 dosimeter-radiometer with a BDZB-96 detection unit was used to measure the flux density of beta particles. The initial contamination level of the laboratory tray was 600 count/(min·cm^2) for alpha radiation and 2300 count/(min·cm^2) for beta radiation. Six consecutive measurements of background beta radiation were taken. The average background beta level at the measurement site was 17 count/(min·cm^2). Before the samples were prepared, the pallet was mechanically cleaned of weakly fixed contaminants and then cut into separate samples (Fig. 8, a). Since the maximum values of cleaning of model contaminants were obtained exactly when using PVA, it was decided to conduct experiments in it when cleaning samples with radioactive contaminants.

The results of the experiments on laser cleaning of radioactive contaminants in aqueous solution of PVA are given in Table 2. In the overlapping modes, where the distance between adjacent spots was half and quarter of the diameter, the alpha and beta particle flux densities were recorded at background levels. These results show that the laser modes used allow the removal of most of the radioactive contamination from the surface of stainless steel in aqueous PVA solution.

Table 2
Results of laser cleaning of radioactive contamination.

Overlap Mode	Number of passes	α, count/(min·cm^2)		β, count/(min·cm^2)	
		Before	After	Before	After
S = d	1	25	2	65	background level
	3	17	2	60	background level
	5	19	<1	66	background level
S = d/2	1	18	<1	65	background level
	3	20	<1	45	background level
	5	24	<1	70	background level
S = d/4	1	9	<1	70	background level
	3	11	<1	28	background level
	5	11	<1	32	background level

4. Conclusion

The results of laser cleaning of model and real contaminants were comparable, confirming the effectiveness of the method. In both cases, laser exposure resulted in a significant reduction in the number of contaminants and a reduction in radiation levels to acceptable levels. The highest CC values were obtained with PVA. In all overlap modes with model samples, the CC levels above 90 % were achieved in one pass. With real contaminants, the best and sufficient values of alpha and beta particle flux reduction were obtained in overlapping modes - S = d/2 and S = d/4 in one pass. This was confirmed by a decrease in alpha and beta radiation levels to room background levels in the experiments. However, the same experiments showed that increasing the number of

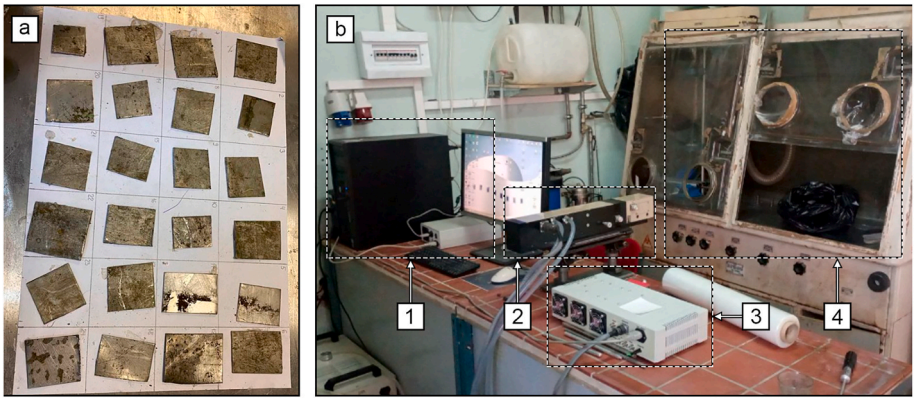

Fig. 8. a – Contaminated samples before cleaning, b – Laser decontamination bench (1 - scanner control station, 2 - Nd:YAG laser, 3 - laser power supply, 4 - fume hood).

runs did not lead to a significant increase in cleaning quality.

The use of a polymerizable film in laser cleaning is a very promising method of capturing ablation products, especially when these products are hazardous to the environment and personnel. The PVA film reliably captured the ablated contaminants and could be removed from the surface of the sample after it had dried. In none of the experiments was there any evidence of the film penetrating or leaking contaminants to the external surface. It is necessary to prevent overheating of the PVA to avoid bubble formation by optimizing the beam trajectory, spot overlap and pulse repetition rate.

CRediT authorship contribution statement

Maxim Cheban: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Konstantin Scherbakov:** Writing – review & editing, Visualization, Supervision, Resources, Project administration, Investigation, Formal analysis, Data curation, Conceptualization. **Dmitry Mamonov:** Writing – review & editing, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Pavel Parabin:** Software, Resources, Formal analysis, Data curation, Conceptualization. **Serafima Filatova:** Writing – review & editing, Supervision, Resources. **Yaroslav Kravchenko:** Writing – review & editing, Resources, Methodology, Data curation, Conceptualization. **Sergey Klimentov:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Conceptualization. **Maxim Chichkov:** Resources, Methodology, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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