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Radioactive oxide removal by XeCl laser

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Abstract

We present the results on cleaning, achieved in nuclear facilities, of metallic surfaces polluted by radioactive oxides. Experimental results of excimer laser decontamination have been obtained for different radionuclides (Cs, Co, Eu, etc.) deposited under various conditions (fixed or unfixed contamination). Our laser decontamination prototype is composed of a XeCl laser, a bundle of fibers for beam transmission, optical systems, collection cell with filter for ablated particle recovery, computer control of cleaning efficiency and beam displacement. We show that the use of the excimer laser ablation technique for decontamination of nuclear facilities has many advantages such as the possibility of remote control, dry process, and, especially, the absence of secondary wastes (clean process). Decontamination factors (DF: initial activity/residual activity) higher than 15 for fixed contamination and up to 100 for unfixed contamination are obtained. © 2002 Published by Elsevier Science B.V.

Keywords: UV laser; Ablation; Cleaning; Decontamination; Nuclear

1. Introduction

Decontamination of solid materials is a critical problem in the nuclear field. The surface characteristics (size, shape, structure, etc.) can be extremely variable. The materials are mainly metals (steel, aluminum, compounds, etc.), but can be also polymers, glass, concrete, etc. Radionuclides are usually part of an oxide layer which is formed under various conditions (temperature, pressure, air, water, etc.). Up to now, the processes used to clean large contaminated surfaces are generally based on dry ice blasting, chemical gels or decontaminating foams. These decontamination techniques are, however, not dry and generate secondary wastes for long-term storage. Therefore, there is a great interest in developing an efficient dry decontamination process which does not produce any secondary wastes.

Both Nd:YAG and excimer lasers were shown to be successful tools for surface treatment and surface cleaning [1,2]. To demonstrate the possibility to use the laser ablation process for radioactive oxide removal, we have developed a laser decontamination prototype [3]. Then, experiments have been done in nuclear facilities, at CEA (Commissariat a l'Energie Atomique) Cadarache.

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2. Laser cleaning prototype

Laser irradiation of a polluted surface induces the absorption of the photon energy by a thin oxide layer, and can lead to the ablation of this oxide layer. Because material absorption coefficient is inversely proportional to the wavelength, the use of short wavelengths is more suitable to obtain an efficient ablation process. In addition, the pulse duration influences the thickness of the thermally affected zone and, therefore, a short duration laser pulse is required to reduce the modification of the surface properties. An excimer laser has been thus chosen to develop the decontamination prototype. To reduce the energy losses that occur during beam transportation through optical fibers as a result of the formation of color centers (E'), it is preferable to select XeCl laser instead of KrF or ArF laser. Two XeCl laser systems have been used to study this cleaning process: (i) Lambda Physics EMG 203 MSC with 25 ns pulse duration and 80 W average power (400 mJ, 200 Hz) and (ii) a CILAS UV 635 [4] with 70 ns pulse duration and 1.2 kW average power (3 J, 400 Hz).

Laser ablation process induces the ejection of material, including radioactive particles, and formation of a plasma which expands perpendicularly to the surface. The particles need to be collected to prevent their redeposition on the treated surface. For this purpose, a collecting cell has been designed. A coaxial nozzle with a 10 mm diameter aperture is set close (several mm) to the surface. The cell is connected to a vacuum pump through a filter to ensure a sufficient depression under the irradiated zone and to trap the particles. A lens is mounted in the cell and focuses the laser beam onto the surface through the nozzle. Gas flows in front of the optic system to protect it against the plasma deposition and to drag the particles towards the filter.

A beam delivery system must be used to guide the beam from the output of the laser to the surface and to move it along the surface by remote control. A 5 m long bundle of 90 fibers allowed to deliver 150 W (at 308 nm) with a 50% overall efficiency [5].

To allow computer control of the full process, real time measurements of the cleaning quality are needed. A spectroscopic method allows to analyze the fluorescence emission intensity of the radionuclides (for example Cs at 852 nm) in the ablation plume via an optical fiber. While this emission is detected, the laser is running at the same surface zone and when the emission disappears the robot moves the laser beam. A second technique based on the detection and analysis of the plasma electric field has also been developed [6].

3. Experimental setup

For the process validation, experiments have been performed with the 80 W Lambda Physics excimer laser (EMG 203 MSC) set close to a glove bag. The laser beam goes through a silica window into the bag, and the samples are small enough to be mounted on XY tables and moved in front of the beam. The optical system allows to get a laser beam with a uniform fluence on the surface. Samples, motorized tables, collection cell and filter are set into the glove bag. The samples are continuously moved and the XY table speed is computer controlled and is adjusted together with the laser repetition rate to define the number of laser shots received by each point of the surface.

Three kinds of samples have been used to test the laser decontamination process. Stainless steel (316L) flat samples and inconel half tubes come from the vapor generator of a nuclear reactor. Oxides have been created and activated under high pressure (170 bar) and temperature (300 °C) of water. The main radionuclide is Co^{60}. The oxide layer thickness is around 10 μm. Aluminum samples come from used fuel retreatment facility and the oxide layer was deposited at the surface at room temperature and atmospheric pressure. The main radionuclides are Cs^{137}, $Eu^{154,\ 155}$ and Sb^{125}. The sample activity measurements have been performed by gamma spectrometry. The detection is made with a germanium detector.

4. Results and interpretation

A_i and A_r are, respectively, the initial and residual activities. Fig. 1 shows the activity percentage removed ($100(A_i - A_r)/A_i$) as a function of the laser shot number for the stainless steel samples. The initial activity of those samples is around 30,000 Bq/cm^2, and the laser fluence has been varied from 0.5 to 2.5 J/cm^2. At 0.5 J/cm^2, the fluence is too low to get an

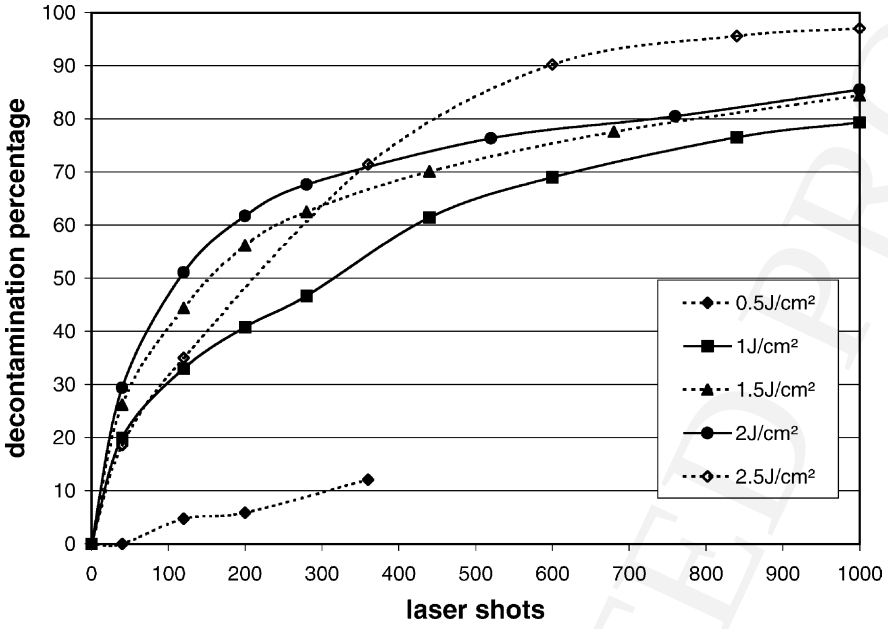

Fig. 1. Percentage of removed activity as a function of number of laser shots for the stainless steel samples.

efficient ablation process. The used fluence is certainly below the oxide ablation threshold, and the weak removed activity is due to unfixed contaminated particles. At a laser fluence higher than 1 J/cm^2, an ablation plasma is clearly visible when the surface is irradiated. All the curves have almost the same evolution when the laser shot number increases. The beginning of the 2.5 J/cm^2 curve is slightly different. Indeed, due to the small amount of contaminated samples we used the sample previously irradiated at 0.5 J/cm^2 and then the unfixed contamination was removed. First, the laser decontamination is very efficient for the first hundred shots (50% at 2 J/cm^2), and then the decontamination rate decreases. After 500 shots, it becomes very difficult to remove the oxide layer. Finally, decontamination percentage between 80 and 97% (2.5 J/cm^2) is obtained after 1000 shots.

Fig. 2 shows the results obtained with two inconel samples irradiated at 1.4 and 3 J/cm^2. Their initial activities were 8000 Bq/cm^2. The curves are similar to the stainless steel ones, but we obtained lower decontamination percentages.

For these two materials, the laser decontamination process can be divided into three steps. During several first shots, the unfixed contamination is easily removed by laser irradiation, even at low fluence. Then, a thin oxide layer is removed by each laser shot. Finally, when the pure oxide layer is carried away, the remaining part of the oxides trapped into the material cracks seems practically impossible to remove. The cracks are created inside the vapor generator because of the temperature and pressure conditions. They are smaller for stainless steel than for inconel, in which they can be deeper than 150 μm. This observation can explain the difference between the laser decontamination efficiency measured for these two materials.

Fig. 3 presents the results obtained for the aluminum cleaning. The initial activities of the three samples vary from 2000 to 12,000 Bq/cm^2. At low fluences (1.4 J/cm^2), no ablation plasma was observed and the measured decontamination percentage can be attributed to the removal of unfixed particles. An intense plasma appears only for fluences higher than the ablation threshold of the oxide layer. In this case, the oxide layer is quickly removed after a few laser shots. The high laser decontamination rate observed for aluminum samples is connected with the nature of the oxides and the way they are formed. In fact, radionuclides are trapped in the natural aluminum oxide layer at room temperature and atmospheric

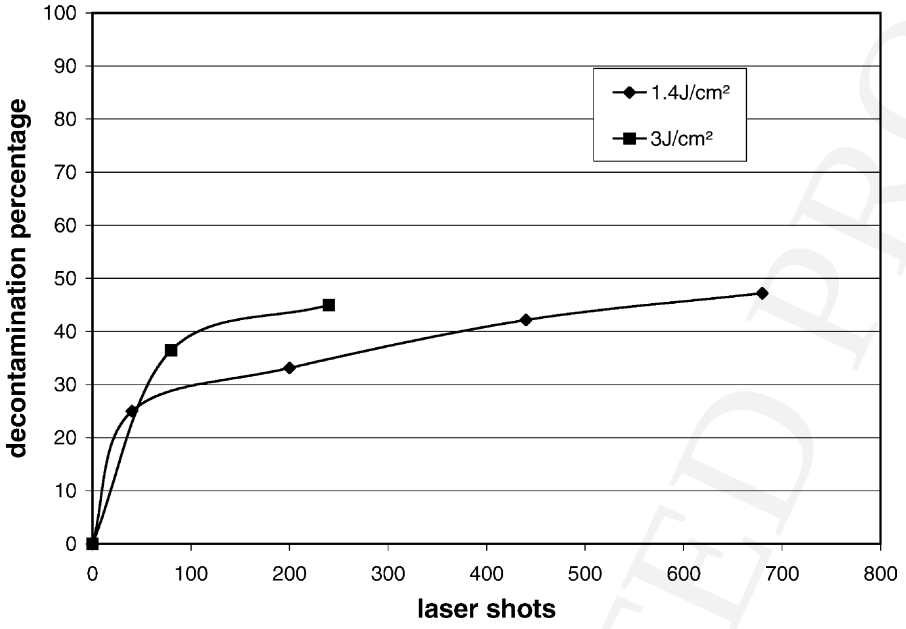

Fig. 2. Percentage of removed activity as a function of number of laser shots for the inconel samples.

pressure. So, the oxide layer is very thin and there is almost no penetration of the oxides inside the material. The experimental setup used to hold aluminum samples did not allow the irradiation of the full surface and, therefore, a complete decontamination is not obtained in this case.

For natural oxide removal, the laser process allows to get the best cleaning efficiency of the dry processes

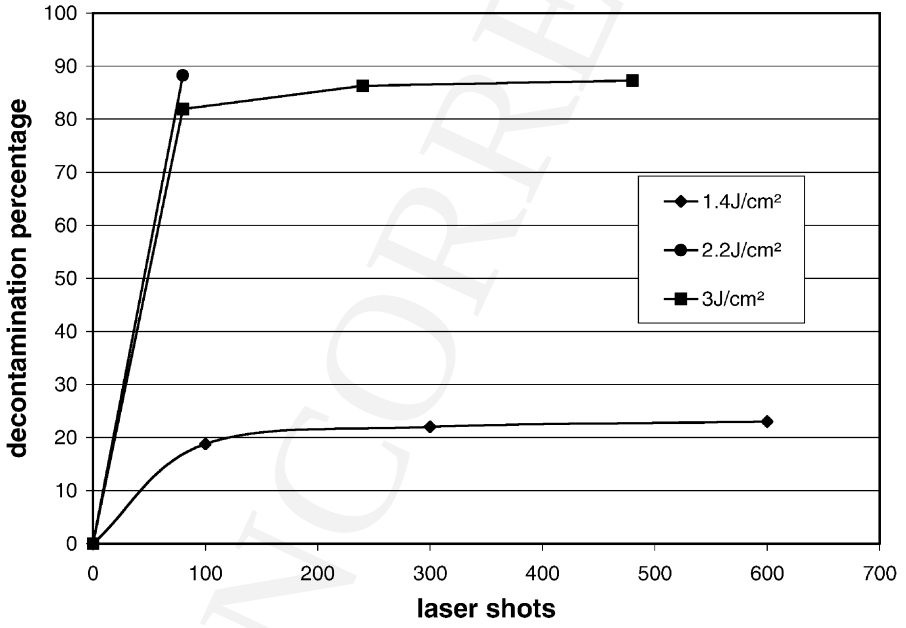

Fig. 3. Percentage of removed activity as a function of number of laser shots for the aluminum samples.

(electro-decontamination allow to reach DF $>$ 10, but it is not adapted for large surface cleaning). In case of deeply fixed contamination like vapor generator, the laser process is still one of the best dry technique, but chemical gels or decontaminating foams allow to reach higher cleaning efficiency (>95%).

5. Conclusion

Excimer laser decontamination has been demonstrated to be efficient in nuclear facilities. For unfixed contamination or oxide layers deposited under normal conditions of pressure and temperature, a few laser shots are sufficient to get an efficient decontamination. For surface activation and oxides created under severe conditions, a few hundreds of shots allow the removal of the oxide layer. Radionuclides trapped inside cracks of the materials are, however, very difficult and sometimes impossible to remove. The used laser decontamination process is dry, clean, well adapted for large area surfaces (rate of 1 m^2/h for aluminum), and the decontamination factors obtained are high enough to use this process (can be used) for the removal of most nuclear contaminations.

Acknowledgements

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