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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 à l'Energie Atomique) Cadarache.

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## 52 2. Laser cleaning prototype

53 Laser irradiation of a polluted surface induces the  
54 absorption of the photon energy by a thin oxide layer,  
55 and can lead to the ablation of this oxide layer.  
56 Because material absorption coefficient is inversely  
57 proportional to the wavelength, the use of short wave-  
58 lengths is more suitable to obtain an efficient ablation  
59 process. In addition, the pulse duration influences the  
60 thickness of the thermally affected zone and, there-  
61 fore, a short duration laser pulse is required to reduce  
62 the modification of the surface properties. An excimer  
63 laser has been thus chosen to develop the decontami-  
64 nation prototype. To reduce the energy losses that  
65 occur during beam transportation through optical  
66 fibers as a result of the formation of color centers  
67 ( $E'$ ), it is preferable to select XeCl laser instead of KrF  
68 or ArF laser. Two XeCl laser systems have been used  
69 to study this cleaning process: (i) Lambda Physics  
70 EMG 203 MSC with 25 ns pulse duration and 80 W  
71 average power (400 mJ, 200 Hz) and (ii) a CILAS UV  
72 635 [4] with 70 ns pulse duration and 1.2 kW average  
73 power (3 J, 400 Hz).

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

89 A beam delivery system must be used to guide the  
90 beam from the output of the laser to the surface and to  
91 move it along the surface by remote control. A 5 m  
92 long bundle of 90 fibers allowed to deliver 150 W (at  
93 308 nm) with a 50% overall efficiency [5].

94 To allow computer control of the full process, real  
95 time measurements of the cleaning quality are needed.  
96 A spectroscopic method allows to analyze the fluor-  
97 escence emission intensity of the radionuclides (for  
98 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

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

119 Three kinds of samples have been used to test the  
120 laser decontamination process. Stainless steel (316L)  
121 flat samples and inconel half tubes come from the  
122 vapor generator of a nuclear reactor. Oxides have been  
123 created and activated under high pressure (170 bar)  
124 and temperature (300 °C) of water. The main radio-  
125 nuclide is  $\text{Co}^{60}$ . The oxide layer thickness is around  
126 10  $\mu\text{m}$ . Aluminum samples come from used fuel  
127 retreatment facility and the oxide layer was deposited  
128 at the surface at room temperature and atmospheric  
129 pressure. The main radionuclides are  $\text{Cs}^{137}$ ,  $\text{Eu}^{154, 155}$   
130 and  $\text{Sb}^{125}$ . The sample activity measurements have  
131 been performed by gamma spectrometry. The detec-  
132 tion is made with a germanium detector.

## 4. Results and interpretation

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

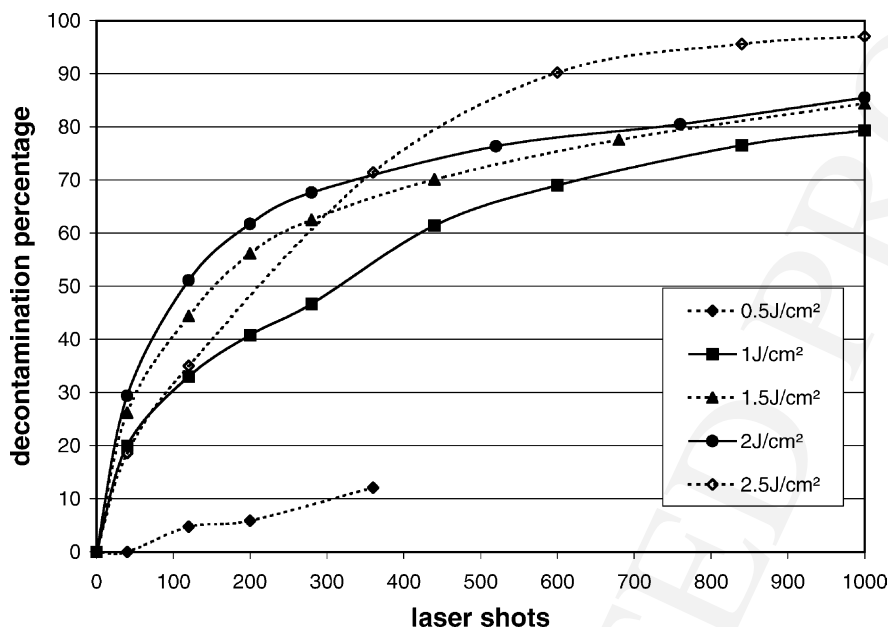


Fig. 1. Percentage of removed activity as a function of number of laser shots for the stainless steel samples.

141 efficient ablation process. The used fluence is certainly below the oxide ablation threshold, and the  
 142 weak removed activity is due to unfixed contaminated particles. At a laser fluence higher than  $1 \text{ J/cm}^2$ , an  
 143 ablation plasma is clearly visible when the surface is irradiated. All the curves have almost the same evolu-  
 144 tion when the laser shot number increases. The beginning of the  $2.5 \text{ J/cm}^2$  curve is slightly different.  
 145 Indeed, due to the small amount of contaminated samples we used the sample previously irradiated at  
 146  $0.5 \text{ J/cm}^2$  and then the unfixed contamination was removed. First, the laser decontamination is very  
 147 efficient for the first hundred shots (50% at  $2 \text{ J/cm}^2$ ), and then the decontamination rate decreases.  
 148 After 500 shots, it becomes very difficult to remove the oxide layer. Finally, decontamination percentage  
 149 between 80 and 97% ( $2.5 \text{ J/cm}^2$ ) is obtained after 1000 shots.

159 Fig. 2 shows the results obtained with two inconel samples irradiated at 1.4 and  $3 \text{ J/cm}^2$ . Their initial  
 160 activities were  $8000 \text{ Bq/cm}^2$ . The curves are similar to the stainless steel ones, but we obtained lower decon-  
 161 tamination percentages.

164 For these two materials, the laser decontamination process can be divided into three steps. During several  
 165 first shots, the unfixed contamination is easily  
 166

167 removed by laser irradiation, even at low fluence. Then, a thin oxide layer is removed by each laser  
 168 shot. Finally, when the pure oxide layer is carried away, the remaining part of the oxides trapped into the  
 169 material cracks seems practically impossible to remove. The cracks are created inside the vapor gener-  
 170 ator because of the temperature and pressure conditions. They are smaller for stainless steel than for  
 171 inconel, in which they can be deeper than  $150 \mu\text{m}$ . This observation can explain the difference between  
 172 the laser decontamination efficiency measured for these two materials.

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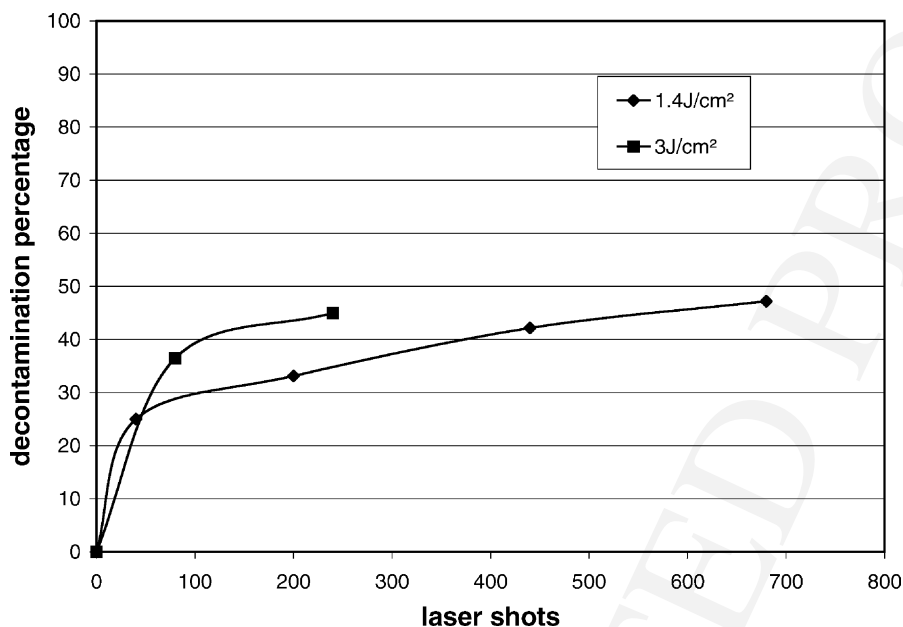


Fig. 2. Percentage of removed activity as a function of number of laser shots for the inconel samples.

193 pressure. So, the oxide layer is very thin and there is  
 194 almost no penetration of the oxides inside the material.  
 195 The experimental setup used to hold aluminum samples  
 196 did not allow the irradiation of the full surface

and, therefore, a complete decontamination is not  
 197 obtained in this case.

For natural oxide removal, the laser process allows  
 199 to get the best cleaning efficiency of the dry processes  
 200

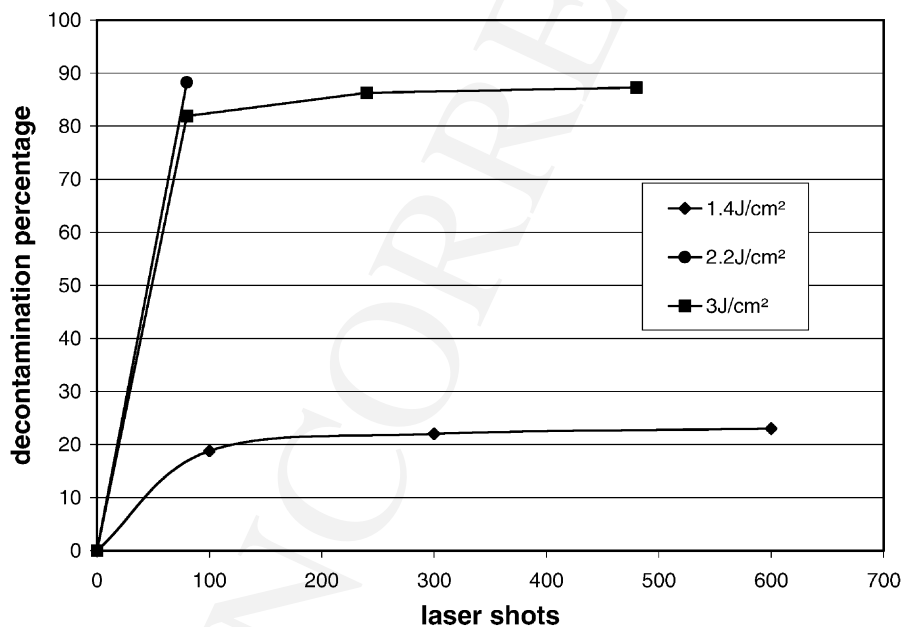


Fig. 3. Percentage of removed activity as a function of number of laser shots for the aluminum samples.

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

## 207 5. Conclusion

208 Excimer laser decontamination has been demon-  
209 strated to be efficient in nuclear facilities. For unfixed  
210 contamination or oxide layers deposited under normal  
211 conditions of pressure and temperature, a few laser  
212 shots are sufficient to get an efficient decontamination.  
213 For surface activation and oxides created under severe  
214 conditions, a few hundreds of shots allow the removal  
215 of the oxide layer. Radionuclides trapped inside cracks  
216 of the materials are, however, very difficult and some-  
217 times impossible to remove. The used laser deconta-  
218 mination process is dry, clean, well adapted for large  
219 area surfaces (rate of  $1 \text{ m}^2/\text{h}$  for aluminum), and the  
220 decontamination factors obtained are high enough to  
240

use this process (can be used) for the removal of most  
nuclear contaminations.

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