

An attempt to use a pulsed CO₂ laser for decontamination of radioactive metal surfaces*

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There is a growing interest in laser radioactive decontamination of metal surfaces. It offers advantages over conventional methods: improved safety, reduction of secondary waste, reduced waste volume, acceptable cost. The main mechanism of cleaning by lasers is ablation. A pulsed TEA CO₂ laser was used in this work for surface cleaning in order to show that ablation of metal surfaces is possible even at relatively low pulse energies, and to suggest that it could be competitive with other lasers because of much higher energy efficiencies. A brief theoretical analysis was made before the experiments. The laser beam was focused using a KBr-lens onto a surface contaminated with ¹³⁷Cs (β^- , $t_{1/2} = 30.17$ y). Three different metals were used: stainless steel, copper and aluminium. The ablated material was pumped out in an air atmosphere and transferred to a filter. The presence of activity on the filter was shown by a germanium detector-multichannel analyzer. The activity levels were measured by a GM counter. The calculated decontamination factors and collection factors showed that ablation occurs with a relatively high efficiency of decontamination. This investigation suggests that decontamination using a CO₂ laser should be seriously considered.

Keywords: radioactivity, decontamination of metals, CO₂ laser, absorption of light, ablation, surface cleaning, radioactive wastes.

INTRODUCTION

Contamination of an object by radioactive isotopes can often cause serious problems not only for its further deployment or use of the material it was made from, but also for radioactivity disposal, because much bigger storage volumes are usually required compared to the total volume of radioactivity. This is especially important for metal surfaces, like vessels, tubes, reservoirs and similar items from nuclear facilities, that have been in contact with radioactivity as the radionuclides are concentrated in the near surface region. Their decontamination by conventional chemical methods would require large amounts of dangerous solvents which creates large secondary waste.

* Dedicated to Professor Slobodan Ribnikar on the occasion of his 70th birthday

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Advances in laser and scanning technologies, along with improved radiation detection techniques, have led to new possibilities of surface decontamination. The ability of lasers to deposit large amounts of energy in short pulses onto surfaces and, thus, to evaporate a thin layer of material from them, without unnecessary heating or melting, offers a possibility of developing an efficient approach to the problems of surface cleaning. The mechanism of cleaning is usually ablation. Further, the evaporated material can be removed by a gas stream.

Laser decontamination can be competitive with conventional methods in spite of some inherent disadvantages. Namely, absorption of light by metals is generally low. Additionally, it is a physical principle that the absorption of a certain metal decreases as the light wavelength increases.¹ The term "laser ablation" will refer to the collective processes occurring upon the interaction of high-power laser radiation with a solid material whereby a thin layer of material is removed from the surface.

KrF excimer² (emits at 248 nm – ultraviolet region) of Nd:YAG³ (1.064 μm near infrared region) pulsed lasers of higher repetition rates have usually been used for laser decontamination up to now. We wanted to try using a pulsed carbon dioxide laser ($\lambda = 10.6 \mu\text{m}$, IR) in order to show that ablation of metal surfaces can be efficiently performed with it, and to suggest that this type of laser might also be competitive mainly due to its high energy efficiency (sometimes reaching 25 %), and the consequential reduced laser energy cost. To illustrate this possibility, we made a rough calculation of the energy that must be absorbed (Q_{abs}) by a metal surface in order to elevate the temperature of its surface by 2000 K (close to evaporation), along with the incident laser energies (Q_{inc} , fluences, J cm^{-2}) necessary for this²

$$Q_{\text{abs}} = (1 - R) Q_{\text{inc}} = S x \rho c_v \Delta T \quad (1)$$

where R is the coefficient of reflection of the polished metal surface, $S = 1 \text{ cm}^2$ is the area, ρ is the metal density, c_v – the thermal capacity of the metal ($c_v \approx 0.4 \text{ J g}^{-1} \text{ K}^{-1}$ for both metals investigated – copper and stainless steel). Table I shows the results obtained for $x = 5 \times 10^{-5} \text{ cm}$, *i.e.*, the average absorption layer thickness for both metals and both wavelengths considered.

TABLE I. Estimated energies for surface ablation (copper and stainless steel) and the necessary laser fluences (for CO_2 and KrF laser)

$\lambda/\mu\text{m}$	$Q_{\text{abs}}/\text{J cm}^{-2}$				$Q_{\text{inc}}/\text{J cm}^{-2}$	
	Cu ($\rho = 9 \text{ g cm}^{-3}$)		StSt ($\rho = 8 \text{ g cm}^{-3}$)		Cu	StSt
10 (CO_2)	R = 0.975		R = 0.90		14	3
0.248 (KrF)	R = 0.50	0.36	R = 0.58	0.32	0.7	0.8

As can be seen from Table I, both copper and stainless steel require higher laser fluences if a carbon dioxide laser is used. However, these fluences are easily achievable even with ordinary lasers of moderate fluences, for instance with the

pulsed CO₂ laser we used in this work. If the laser is of insufficient power coating the surface with a strongly absorbing material can be considered.

EXPERIMENTAL

The experimental apparatus used in this work is shown in Fig. 1. The laser source is a pulsed Transversely Excited Atmospheric (TEA) carbon dioxide laser, operating with a non-typical gas mixture CO₂/N₂/H₂. The presence of hydrogen increases the efficiency of the system.⁴ The average energy per pulse is up to 145 mJ. This energy is irradiated in a short initial spike, with duration (FWHM - Full Width at Half Maximum) of about 120 ns ($\approx 40\%$), followed by a tail of up to 1 μ s ($\approx 60\%$). Thus, the average value of light power over the entire laser pulse is about 0.6 MW. The wavelength of emitted radiation is 10.6 μ m. The pulse rate repetition was 1 Hz. The beam was focused by a KBr-lens with a focal length of 65 mm and the area in focus was between 0.01 and 0.02 cm², depending on the aim of the experiment. The target-metal was placed near the focus to get enough power per square centimeter (irradiance, W cm⁻²).

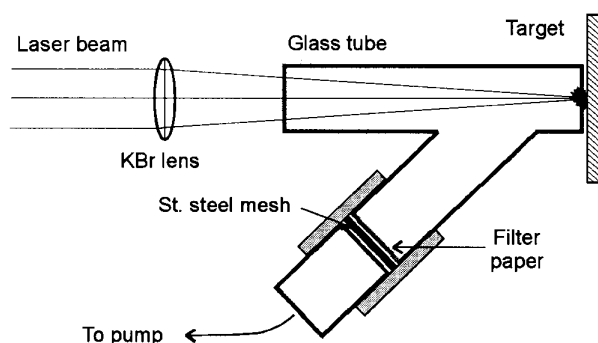


Fig. 1. Schematic representation of the experimental setup used for ablation of metal surfaces.

All targets were 1 mm thick, 25 mm long and 10 mm wide unpolished metal pieces. A restricted area (10 mm \times 1 mm) on one side was contaminated with an aqueous solution of caesium chloride labeled with the ¹³⁷Cs isotope. The water was evaporated using an IR lamp. This procedure was repeated until the relative activity of the sample exceeded the value of 10 count s⁻¹ on our GM detection system. This count rate is high enough for accurate measurements but low enough to be safe for the experimentalists. The laser beam was passed through a 7 mm internal diameter glass tube open at both sides. Another tube of the same diameter, which served as a connection with a vacuum pump, was welded to this one at an angle of 45°. The pump sucks in surrounding air at ambient pressure transferring the evaporated material with the radiocativity onto a paper filter. The filter was fixed in position by placing it on a stainless steel mesh. All experiments were performed at room temperature. The activated area was manually scanned point-by-point with a step interval of 0.5 mm using an x-y stage. Every point received 60 laser pulses. The detection system consisted of a γ -spectrometer (Canberra S100) with a pure germanium (HPGe) detector (relative efficiency $\approx 20\%$), and a GM counter with a relatively high efficiency for γ -radiation. The spectrometer was engaged only to prove the presence of ¹³⁷Cs on the samples or filters in several experiments at the beginning of the investigations. The counter was used to measure values of the relative activities in all experiments because it is an easy and sufficiently accurate way. It was adopted to have the same geometry in all measurements.

RESULTS AND DISCUSSION

The efficiency of decontamination of a sample is expressed through the decontamination factor (F_d), while the efficiency of radioactivity transfer and its collection onto the filter is described through the collection factor (S_f)

$$F_d = \frac{A_0}{A}; \quad S_f = \frac{A_f}{A_0} \quad (2)$$

where A_0 , A and A_f are the activity of the sample before ablation, the activity of the sample after ablation and the activity collected on the filter after ablation, respectively.

Following the procedure described in the previous section, the data shown in Table II was obtained. In some way it summarizes the efficiencies of decontamination achieved in our experiments.

TABLE II. Decontamination efficiencies expressed as F_d and S_f for some metals at 293 K obtained with a focused carbon dioxide pulsed TEA laser beam*

Material	F_d	S_f
Stainless steel	4.3	0.51
Copper	3.3	0.40
Aluminium	3.2	0.60

*Focal area = 0.0105 cm², pulse energy = 145 mJ, thus energy density (fluence) was about 14 J cm⁻² and the irradiance level at the exposed point was \approx 55 MW cm⁻²

Table II shows that 40 % to 60 % of the initial sample activity is transferred to the filter, while the activity remaining on the sample is roughly between one fourth and one third of the initial value, depending on the material. The F_d values for copper and aluminium are almost the same. They are generally lower than those for stainless steel, which is to be expected due to their lower absorption (higher reflectivity) of the radiation at the applied wavelength. The remaining activity can also be to some degree attributed to the scanning method.

On the other hand, there is a significant difference in the S_f values for copper and aluminium. This can probably be attributed to differences in geometry and the pump efficiency from day to day. As evidence of such a possibility it can be stated that the entire activity after an experiment, obtained as the summation over all parts of the experimental apparatus coming in contact with the radioactivity (glass tube, filter holder) is roughly equal to the initial activity.

To prove the transfer of radioactive caesium to the filter once more spectrometric measurements of the same filter paper before and after the ablation were performed. No activity at all was found without ablation, while a spectral peak typical of ¹³⁷Cs always appeared at about 661.6 keV when the surfaces were ablated.

THRESHOLD FLUENCE

To estimate the threshold fluence for ablation, the activity of the filter after different laser pulse energies at a constant focal area were measured. These experiments were performed using a stainless steel sample the activated area of which was

about 5 mm long and 1 mm wide. The ablation was carried out in the same manner as above applying 15 laser pulses to each point. The results of these measurements, expressed through the S_f values, are given in Table III. No decontamination at all was seen for fluences of 2.1 and 3.5 J cm⁻², while the S_f values is 0.5 for a fluence of 6.0 J cm⁻². This is of the same order of magnitude as shown in Table II, where much higher fluences were applied. This means that a fluence of 6 J cm⁻² is much over the threshold value. From these results a fluence threshold value of about 4 J cm⁻² can be estimated. This value is very close to the estimated threshold shown in Table I and can easily be achieved with the laser employed.

TABLE III. Threshold conditions, expressed through S_f factors for stainless steel at 293 K, measured with the pulsed carbon dioxide focused laser beam*

Pulse energy/mJ	Energy density/J cm ⁻²	Irradiance/MW cm ⁻²	S_f
42	2.1	8.3	0
70	3.5	13.8	0
121	6.0	23.6	0.5

*Focal area = 0.02 cm²

CONCLUDING REMARKS

Considering all the above results, it can be said that the application of a carbon dioxide laser for decontamination looks quite promising. The ablation is efficient. We applied 60 pulses per scanned point to ensure that most of the activity is evaporated, but later measurements showed almost the same results with only 15 pulses. Besides, it is necessary to say that the employed laser is sufficiently convenient for basic research of this phenomenon. In practice a higher repetition rate laser combined with an improved scanning system (based on the x - y table or a computer controlled optical system⁵) would be necessary.

The investigations carried out in this work clearly show that a carbon dioxide laser can be a serious candidate for surface decontamination for several important reasons: (i) As a machine it is much cheaper than an excimer or a Nd:YAG laser of similar performance; (ii) It is energetically more efficient than any other laser, thus it can deliver the cheapest energy per quantum. However, it should be noted here that its (IR) quantum is of lower energy than that of the excimer or Nd:YAG laser; (iii) CO₂ lasers are available in many more versions than the other types; (iv) Surface coating with a strongly absorbing material can offer an additional improvement.

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ИЗВОД

ПОКУШАЈ ДЕКОНТАМИНАЦИЈЕ РАДИОАКТИВНИХ МЕТАЛНИХ ПОВРШИНА
ПОМОЋУ ИМПУЛСНОГ СО₂ ЛАСЕРА

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Деконтаминација радиоактивних металних површина путем ласера значајна је јер обећава одређене предности над конвенционалним методама: смањење секундарног отпада, смањење запремине отпада, прихватљиве цене итд. У овом раду је коришћен импулсни СО₂ ласер за чишћење металних површина путем аблације да би се показало да је то могуће чак и при ниским енергијама импулса, те да овај ласер има изгледе да буде коришћен у ове сврхе, због тога што је енергетски знатно ефикаснији од других. Ласерски сноп је фокусиран помоћу КВг сочива на површину контаминирану изотопом ¹³⁷Cs. Коришћена су три метала: нерђајући челик, бакар и алуминијум. Испарени материјал је одношен ваздушном струјом на атмосферском притиску до филтера. Присуство радиоактивности на филтеру је констатовано γ-спектрометријом и GM-бојачем. Израчунати фактори деконтаминације и фактори сакупљања на филтеру показују да долази до аблације уз релативно велику ефикасност деконтаминације.

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