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Decontamination of Radioactive Material by Nd:YAG Laser†

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Metal ion removal characteristics of a Q-switched Nd:YAG laser on Type 304 stainless steel were investigated by varying the shot numbers under three wavelength conditions at 12.0 J/cm^2. It was found that Cs^+ ions and Co^{2+} ions were satisfactorily removed within 10 laser shots at 532 nm and at 355 nm. It was difficult to remove the metal ions on Type 304 stainless steel at 1064 nm. Decontamination characteristics of 4 kinds of materials artificially contaminated with Co^{60} and Cs^{137} were also investigated at 532 nm. Decontamination of the paint-coated surface was easy and the decontamination factors were over 1339 after two times' application of laser irradiation at 9.4 J/cm^2. For type 304 stainless steel, inconel 600 and concrete specimens, decontamination factors were over 110 after two times' application of laser irradiation at 14.2 J/cm^2. It was also found that Cs^{137} and Cs^{134} were easily ablated when the laser fluence is 14.2 J/cm^2 during the 20 laser shots at 532 nm.

Key Words: Decontamination, Q-switched Nd: YAG laser, Cs^{137}, Co^{60}, Ablation.

INTRODUCTION

Radioactive contaminants on material surface must be decontaminated to reduce the radiation exposure of workers during the overhaul period of the nuclear power plant. Laser decontamination is a relatively new surface decontamination technique and the technique satisfies the next requirements[1]: safety, cost-effectiveness, waste minimization, high decontamination factor and feasibility of industrialization. Furthermore, laser decontamination is applied to remove contaminants from different kinds of materials by remote operation. More than 50 techniques have been developed to achieve a high decontamination factor at various sites[2]. In nuclear facilities, contaminants usually contain radionuclide and the contaminated material is metal, polymer, slate, glass and concrete. The objective of the study is to evaluate the decontamination performance of a Q-switched Nd:YAG laser system on the specimens contaminated with a radioactive isotope solution that contains Cs^{137} and Co^{60}. Especially, the effect of laser fluence and a laser shot numbers on the decontamination of 4 kinds of materials was emphasized.

EXPERIMENTAL

Type 304 stainless steel, Inconel 600 and concrete were cut into a rectangular form for the experimental specimens. The pieces were polished with abrasive papers and washed with water and ethyl alcohol. On the other hand, rectangular form of Type 304 stainless steel was covered with epoxy paint. Four kinds of specimens have been used for laser decontamination tests. For metal ion removal tests, $Co(NH_4)_2(SO_4)_2$ and $CsNO_3$ solutions were dropped onto the Type 304 stainless steel surfaces and then dried for the tests. The relative atomic molar percent of the metal surface elements before and after laser irradiation was measured by EPMA. Relative atomic molar percent before laser irradiation is listed in Table-1. The specimens for decontamination tests were prepared as follows: an isotope solution that contains Cs^{135} and Co^{60} was slowly dropped onto the surface of specimens by injector separately and then dried in a room temperature. The radioactivity of each specimen was measured by multichannel analyzer with high purity Ge detector before laser irradiation. After laser irradiation, the radioactivity of specimen was also measured by the same process.

A Q-switched Nd: YAG laser (Brilliant b, Quantel Co.) with pulse duration of 6 ns and a maximum repetition rate of

TABLE-1
RELATIVE ATOMIC MOLAR PERCENT
BEFORE LASER IRRADIATION

Element	N	O	Ni	S	Cr	Fe	H	Cs	Co
$Co(NH_4)_2(SO_4)_2$	2.0	10.7	6.9	0.2	18.0	59.1	Trace	-	3.1
$CsNO_3$	4.5	25.4	5.9	-	13.6	45.7	-	5.2	-

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10 Hz emits a fundamental wavelength at 1064 nm and the maximum pulse energy determined from the energy meter was 870 mJ/pulse. A second harmonic generation of a Q-switched Nd:YAG laser with a pulse duration of 5 ns and a wave length of 532 nm was used. Maximum pulse energy at 532 nm was 450 mJ/pulse. Fig. 1 shows a schematic diagram of the experimental apparatus for the decontamination tests. A surrogate specimen was mounted on a X-Y stage. An articulate mirror was used for the transfer and convergence of a laser beam at the target point. Laser fluence was calculated from the measurement of the pulse energy with an energy meter and from the estimation of the beam size.

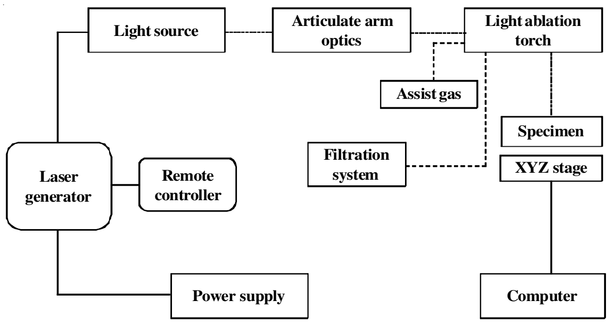


Fig. 1. Schematic diagram of the experimental apparatus

RESULTS AND DISCUSSION

Metal ion removal tests: Fig. 2 shows the remaining portion (X_f/X_o) of cesium ions on the type 304 stainless steel specimens against the number of laser shots at 12.0 J/cm^2 under three wavelength conditions. X_f and X_o are, respectively, the residual and initial relative atomic molar per cent of the contaminants. The remaining portion of Cs^+ ions is slowly decreased with the increase of the number of laser shots at 1064 nm. After 32 laser shots, the remaining portion of Cs^+ ion is higher than 0.2. The remaining portion of Cs^+ ions, however, is drastically decreased at 532 nm and at 355 nm. The figure shows that the removal efficiency on Cs^+ ion at 355 nm is higher than that at 532 nm. However, the efficiency gap is negligible within 8 laser shots. After 8 laser shots, the remaining portion of Cs^+ ion is almost 0. For the three wavelength conditions, ablation plasma is clearly visible when the surface is irradiated.

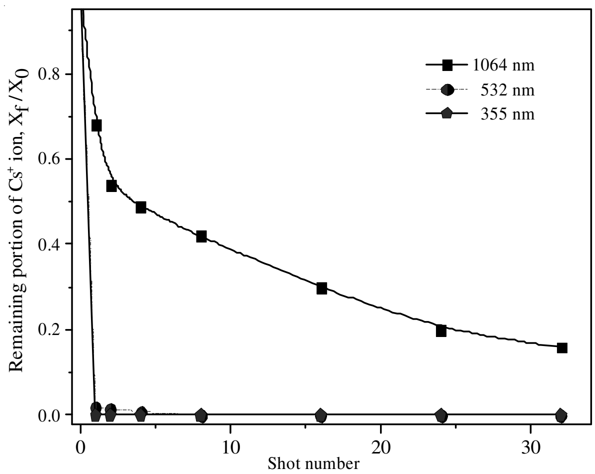


Fig. 2. Remaining portion of cesium ions against the number of laser shots under three wavelength conditions (12 J/cm^2, SUS 304 and 10 Hz)

Fig. 3 shows the remaining portion of cobalt ions on the Type 304 stainless steel specimens against the number of laser shots at 12.0 J/cm^2 under three wavelength conditions. Comparing Fig. 3 with Fig. 2, the removal tendency of laser on the metal ions is same in the experimental error range. At 1064 nm, Cs^+ ions are more easily removed than Co^{2+} ions. The remaining portion of Co^{2+} ions is also slowly decreased with the increase of the number of laser shots at 1064 nm. The remaining portion of Co^{2+}, however, is drastically decreased at 532 nm and at 355 nm. After 8 laser shots, the remaining portion of Co^{2+} ion is also almost zero. Consulting the test results of Figs. 2 and 3, it is found that laser fluence of 12.0 J/cm^2 is not sufficient to remove contaminants during the 32 laser shots at 1064 nm.

From the test results, it was determined that the laser fluence for the decontamination tests is around 12.0 J/cm^2 at 532 nm.

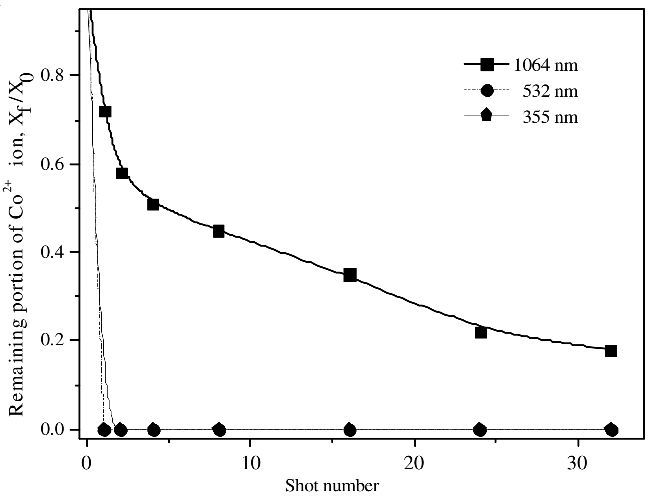


Fig. 3. Remaining portion of cobalt ions against the number of laser shots under three wavelength conditions (12 J/cm^2, SUS 304 and 10 Hz)

Decontamination tests: Fig. 4 shows the decontamination test results on the 4 kinds of materials at 9.4 J/cm^2 under 532 nm. For the 3 kinds of test specimens, decontamination factors exceed 10 after second application. However, decontamination of concrete is difficult at 9.4 J/cm^2. Concrete is hardly ablated by laser and decontamination factors do not exceed 2. In case of paint coated surface, paint is satisfactorily ablated by laser and decontamination factors are 1339.3 for Co^{60} and 1728.7 for Cs^{137}. For all the test specimens, decontamination of Cs^{137} is easier than that of Co^{60}.

Fig. 5 shows the decontamination test results on the 3 kinds of materials at 14.2 J/cm^2 under 532 nm. For all the test specimens, decontamination factors exceed 110. In case of concrete, decontamination factors are 110.7 for Co^{60} and 127.3 for Cs^{137}. For the Type 304 stainless steel and Inconel specimens, decontamination performance is improved by the increase of the laser fluence to 14.2 J/cm^2.

Conclusion

Q-switched Nd:YAG laser system was employed to perform a comparative study on the laser decontamination of type 304 stainless steel, Inconel 600, concrete and paint coated surface. To obtain the high decontamination factors, the laser shot numbers

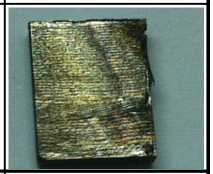
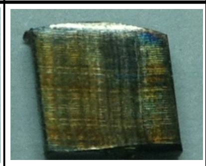
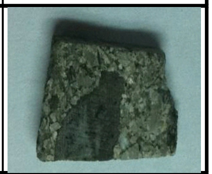
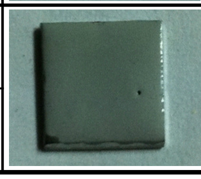
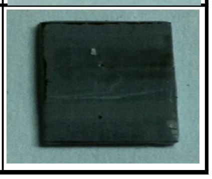
Specimen type		Before Deconta-mination (Bq)	1st application		2nd application		Photographs	
			(Bq)	Efficiency / DF	(Bq)	Efficiency / DF	Before	After
Inconel	Co-60	80.49	22.46	72.10 / 3.6	9.05	88.76 / 8.9		
	Cs-137	136.29	32.84	75.90 / 4.2	11.76	91.37 / 11.6		
SUS 304	Co-60	137.09	11.22	91.82 / 12.2	1.13	99.18 / 121.3		
	Cs-137	179.95	11.80	93.44 / 15.3	1.26	99.30 / 142.8		
Concrete	Co-60	215.54	127.99	40.62 / 1.7	120.22	44.22 / 1.8		
	Cs-137	330.83	199.08	39.82 / 1.7	191.22	42.20 / 1.7		
Paint + SUS 304	Co-60	455.35	1.01	99.78 / 450.8	0.34	99.93 / 1339.3		
	Cs-137	656.90	1.89	99.71 / 650.4	0.38	99.94 / 1728.7		

Fig. 4. Decontamination test results at 9.4 J/cm^2 (532 nm, string type lens, stepwise, N_2 gas flow and 10 shots/spot)

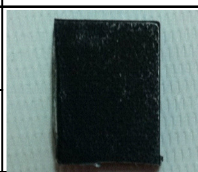
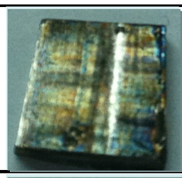
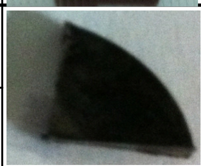
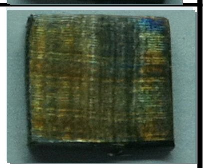
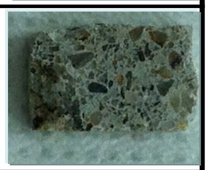
Specimen type		Before Deconta-mination (Bq)	1st application		2nd application		Photographs	
			(Bq)	Efficiency / DF	(Bq)	Efficiency / DF	Before	After
Inconel	Co-60	37.39	7.54	79.83 / 5.0	0.33	99.12 / 113.3		
	Cs-137	58.66	10.22	82.57 / 5.7	0.42	99.28 / 139.7		
SUS 304	Co-60	28.17	1.81	93.57 / 15.6	0.15	99.47 / 187.8		
	Cs-137	43.92	2.40	94.54 / 18.3	0.26	99.41 / 168.9		
Concrete	Co-60	70.86	23.13	67.36 / 3.1	0.64	99.10 / 110.7		
	Cs-137	110.75	31.76	71.32 / 3.5	0.87	99.21 / 127.3		

Fig. 5. Decontamination test results at 14.2 J/cm^2 (532 nm, string type kens, stepwise, N_2 gas flow and 10 shots/spot)

and laser fluence should be increased or decreased according to the property of the contaminated surface. During the laser irradiation, nitrogen gas was flown to avoid the deposition of the ablated dusts. The decontamination of the paint was easiest among the test specimens. This is attributed to the low melting point and low boiling point of paint. Decontamination of concrete was more difficult. This can be also attributed to the physical property of concrete. It is assumed that some portion of liquid phase contaminants was penetrated into the pores during the artificial contamination and decontamination of concrete was more difficult at lower laser fluence. A study on the quantitative interpretation of the laser decontamination is necessary.

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