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Wavelengths effect on mass ablation of laser decontamination on aluminum surface

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1. Introduction

Laser decontamination technology has advantages such as very high decontamination efficiency and very low secondary waste generation even though it has some disadvantages as high initial equipment cost and a little bit slow decontamination speed. Tam et al. [1] reported the laser cleaning techniques for the removal of surface particulates. The existing decontamination methods such as chemical process using an organic solvent or inorganic acid, and a physical method using blasting or brushing generate secondary wastes due to chemicals and abrasives [2]

The coolant system of the research reactor in Korea and equipment in hot cells of nuclear facilities are made of aluminum. In the present study, we investigate the effect of wavelength of Q-switched Nd:YAG laser on the removal of Cs, Co, Eu, and Ce on an aluminum surface. Correlation between alumina ablation mass and the laser fluence was evaluated to find the effect of laser fluence on the removal of contaminants on the aluminum surface.

2. Experimental

2.1 Specimen Preparation and Analysis

Aluminum specimens were cut into a rectangular form for experimental specimen. And then polished with abrasive papers, washed with ethyl alcohol, dried and photographed. They were dipped into an ultrasonic cleaner for 30 minutes and dried. For an artificial contamination, a small amount of solution containing Cs^{+},Co^{2+},Eu^{3+} and Ce^{4+} was thrown on specimen surfaces, respectively. The relative atomic molar percent of the metal surface elements before laser irradiation was analyzed by SEM (scanning electron microscopy, JEOL Ltd. Model: JSM-6300) and EPMA (Electron probe micro analyzer) as given in Table 1.

Table 1. Chemical composition of the Al surfaces before decontamination (relative atomic molar %) .

Element	N	O	Al	Ag	H	Eu	Cs	Ce	Co
$Co(NH_4)_2(SO_4)_2$	Trace	13.6	81.3	0.4	trace	-	-	-	4.7
CeO_2	-	21.1	73.4	0.3	-	-	-	5.2	-
Eu_2O_3	-	14.0	80.8	0.1	-	5.1	-	-	-
$CsNO_3$	Trace	26.8	68.7	0.2	-	-	4.3	-	-

2.2 Laser irradiation

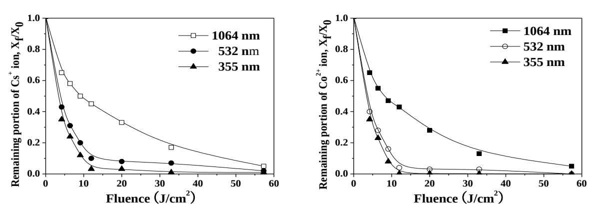
Q-switched Nd:YAG laser (Quentel Co. Model: Brilliant b) with a fundamental, second harmonic and third harmonic generation was employed. The repetition rate was 10Hz (three wavelengths) and pulse duration was 6ns (1064nm), 5ns (532nm) and 5ns (355nm). The specimen was mounted on a stage that allowed the specimen holder to move of 25mm x 25mm in the X and Y directions.

3. Results and Discussion

3.1. Effect of fluence

Removal efficiency of the four contaminants on the aluminum specimen was investigated against the laser fluence the number of laser shots at 8 and the results are given in Figs.1 and 2.

As shown in Fig. 1, the removal efficiency of the two contaminants are decreased drastically in the fluence range of 0 to 12 J/cm^2 at 532nm and 355nm, respectively. However, the remaining portion of the two contaminants is slowly decreased in the whole experimental range at 1064nm. Comparing Cs^{+} ion with Co^{2+} ion, Co^{2+} ion is removed easily.



(a) (b)

Fig. 1. Remaining portion of (a) $CsNO_3$ and (b) $Co(NH_4)_2(SO_4)_2$ against laser fluence (8 shots).

The remaining portion of (a) Eu^{3+} ion and (b) Ce^{4+} ion on aluminum specimens against the laser fluence after 8 shots under the three wavelength conditions is shown in Fig. 2. The remaining portion of the two contaminants is also decreased drastically in the fluence range of 0 to 12 J/cm^2 at 532nm and 355nm. The remaining portion of the two contaminants is slowly decreased in the whole experimental range at 1064nm. Comparing Fig. 1 with Fig. 2, Eu_2O_3 and CeO_2 are more difficult to remove than $CsNO_3$ and $Co(NH_4)_2(SO_4)_2$.

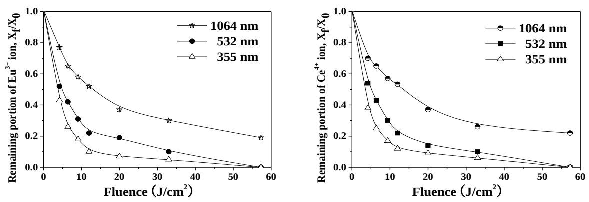


Fig.2. Remaining portion of (a) Eu_2O_3 and (b) CeO_2 against the laser fluence (8 shots).

This is might be related with their different physical properties such as the boiling point. The boiling point of $CsNo_3$(849℃) is lower than those of cobalt ammonium sulfate (2,927℃) europium oxide (4,118℃) and cerium oxide (3,500℃). When irradiated by a laser, is occurred sequently by boiling, those lower boiling points might affect the easy evaporation of cesium and cobalt ions compared with the europium and cerium oxides.

3.2. Correlation between the mass ablation and fluence

With an increase of the number of laser shots, the remaining portion of contaminants is also decreased. To investigate the effect of the ablation of base metal on the removal of the contaminants, laser ablation tests on aluminum was performed. Because the formation of an oxide during the laser irradiation decreased the laser ablation performance, the effect of laser fluence on the ablation of aluminum was difficult to investigate. Therefore the effect of laser fluence on the ablation of alumina (Al_2O_3) during 1000 laser shots at 532 nm was investigated. Fig.3 shows the mass ablation of alumina against the (fluence)$^{1/3}$.

As shown in Fig.3, mass ablation is directly proportional to the (fluence)$^{1/3}$ in the laser fluence range from 3.58 J/cm^2 to 25.48 J/cm^2. Some deviation from the linearity, however, occurs above 25.48 J/cm^2. Two explanations are possible on the deviation from linearity. 1) Plasma formed during the laser irradiation shields the laser light at higher fluence. 2) Alumina has a high thermal conductivity. Therefore, it dissipates the heat effectively around the affected area.

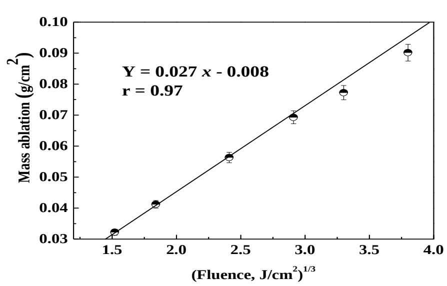


Fig. 3. Mass ablation of alumina against the (Fluence)$^{1/3}$ at 532nm.

Fabbro et al. [3] reported that mass ablation rate (*kg/s.cm^2*) is proportional to the laser fluence and inversely proportional to the laser wavelength. They suggested the empirical relation among mass ablation rate, fluence, and laser wavelength is as written in Equation (1).

$$m \approx 110\left(\frac{\phi_a}{10^{14}}\right)^{\frac{1}{3}}\lambda^{-\frac{4}{3}} \qquad (1)$$

Here, m is a mass ablation rate, ϕ_a is a laser fluence, $\frac{W}{cm^2}$, and λ is the laser wavelength, μm . Mass ablation of alumina against $\phi_a^{\ 1/3}$ at 532 nm wavelength in Fig. 3 is well coincided with the Fabbro relation in the laser fluence range of 3.58 J/cm^2 to 25.48 J/cm^2. The slope value of 0.027 is related to the physicochemical property of alumina. The intercept value of – 0.008 is related to the threshold fluence of alumina at 532 nm. Deviation from linearity above 25.48 J/cm^2 explains the removal tendency of contaminants at 532 nm well, as shown in Figs. 1 and 2 We realize that the mass ablation of alumina formed during the laser irradiation also affects the removal of contaminants.

4. Conclusion

The effects of wavelength and laser energy fluence on the laser ablation decontamination of the surrogate specimen were investigated using the three wavelength of Q-switched Nd:YAG .

For each contaminant, the order of removal efficiency at 12 J/cm^2 of a laser fluence condition was 355 nm > 532 nm > 1064 nm. It was assumed that the photochemical reaction at 355 nm enhanced the removal of the contaminants. Correlation between alumina ablation mass and the laser fluence was evaluated to find the effect of laser fluence on the removal of contaminants on the aluminum surface.

Acknowledgement

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