Laser-induced Ejection of a Millimeter-sized Liquid Droplet from a Metal Surface by IR Laser Irradiation at Three Different Pulse Durations

Hiroyuki Niino*, Tetsuo Sakai**, Shinji Ookuma**, Naoto Okada**, Yoshinori Kato***, Takashi Kurita***, and Toshiyuki Kawashima***

* National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki, 305-8565 Japan E-mail: niino.hiro@aist.go.jp

** Corporate Manufacturing Engineering Center, Toshiba Corp., Yokohama, Kanagawa, 235-0017 Japan
*** Industrial Development Center, Hamamatsu Photonics K. K., Hamamatsu, Shizuoka, 431-1202 Japan

Laser-induced cleaning by the elimination of liquid contaminant droplets from the surface of a solid substrate is an attractive application of laser technology. We report the laser-induced ejection of an organic carbonate liquid droplet from an aluminum surface using three different infrared (IR) laser pulse durations (5.7 ns, 35 ns, and 90 μ s). Single-shot cleaning of a large area sample through a non-contact process was achieved by irradiation with a nanosecond-pulsed laser. The dynamic behavior of liquid desorption was observed with a high-speed CCD camera. The threshold laser fluences for liquid desorption by laser irradiations were determined for each of the lasers. The laser pulse duration increased with increasing threshold laser fluence.

DOI: 10.2961/jlmn.2019.03.0010

Keywords: laser cleaning, aluminum metal plate, organic carbonate liquid, high-speed CCD camera observation

1. Introduction

Elimination of liquid contaminants from the surface of solid substrates by laser-induced explosive vaporization is an attractive surface cleaning technique [1]. Explosive vaporization of liquid at the surface of a metal film by pulsed laser irradiation was reported by Tam and colleagues [2,3]. Their pioneering work also revealed the dynamics of the transient behavior of water and alcohols at metal-liquid interfaces by optical probing. This technique can be applied, for instance, to the cell assembly process of lithium-ion secondary batteries [4]. Electrolyte droplets are often deposited around an injection hole during electrolyte filling into an aluminum case, which may induce porosity defects when the sealing cap is laser-welded to the aluminum case; therefore, these droplets should be removed before the laser-welding process.

In our previous work, the laser-induced ejection of a millimeter-sized liquid droplet from the surface of an aluminum metal plate was observed with a 1 J pulse⁻¹ class nanosecond-pulsed laser (pulse duration: 5.7 ns, wavelength: 1053 nm) [5]. Organic carbonate, a liquid medium for the electrolyte of lithium-ion batteries, was examined as the liquid with an aim to developing an effective non-contact cleaning method for industrial application. Single-shot cleaning over a large area of a solid surface was achieved using highpower laser equipment.

In the present work, the effect of the pulse duration was investigated using two other lasers (pulse duration (exposure time): 35 ns and 90 μ s) and monitoring the transient dynamics with a high-speed CCD camera. The threshold laser

fluences for liquid desorption under laser irradiation were thus determined.

2. Experimental Method

The beam of a prototype nanosecond-pulsed laser (Hamamatsu Photonics K. K., wavelength: 1064 nm, pulse-duration: 35 ns, pulse repetition rate: 1 Hz, laser pulse energy: 1-0.4 J pulse⁻¹) was incident on the sample surface at an angle of 45° through a concave lens in the ambient air. The laser fluence on the surface was varied by changing the pulse energy of the laser equipment.

A microsecond-pulsed laser beam was prepared by scanning an IR continuous wave (cw) laser (Furukawa Electric, Yb-doped fiber laser, $\lambda = 1084$ nm, single-mode (M² < 1.1), 1 kW average power) on the sample surface with a galvanometer scanner (scanning speed on the plate: 3.3 m s^{-1}). The beam was focused with an f-theta lens (Showa Optronics Co.; Ltd., FT300/5-1080F, non-telecentric lens, f = 306 mm). The laser beam diameter on the sample surface was ca. 300 µm; therefore, the exposure time (τ) at a specific position of the cw laser was estimated to be $\tau = 90 \text{ µs}$ ($\tau = 300 \text{ µm} / 3.3 \text{ m s}^{-1}$).

A mixture of equal amounts of ethyl methyl carbonate and ethylene carbonate was selected as a liquid contaminant. This liquid is transparent at the wavelengths of the lasers used in this work (the absorption spectrum of the liquid is shown in Fig. 5 of Ref. [5]); therefore, the laser beam reaches the surface of the aluminum plate through the liquid.

An aluminum plate (A3003, 2 mm thick) was used as the solid substrate. To increase the wettability of the surface of the aluminum plate, an array of circular micro-structures

was produced by step-by-step laser ablation of the surface (Fig. 1). 4×4 mm² liquid droplets were prepared on the metal surface using a 0.8 μ L dropper (repeatability error: < 0.6 %). The thickness of the liquid on the sample surface was ca. 50 μ m (= 0.8 μ L / (4×4 mm²)). The pre-treatment of the sample to produce microroughness on the surface was effective for reproducible preparation of the droplets with constant thickness.

Laser-induced ejection of liquid droplets was observed using a high-speed CCD camera (Photron Inc., FASTCAM SA5) with an imaging lens (Nikon Imaging Japan Inc.; PC-E Micro NIKKOR 85 mm f/2.8D) and an optical filter (Edmund Optics; Heat Absorbing Glass KG-5). The optical filter was used for the elimination of scattered light from the incident laser beam. The frame rate of the CCD camera was set to either 150×10^3 fps or at 100×10^3 fps. The velocity of the liquid ejection was estimated from the frame-by-frame photo-images of the CCD camera equipped with an external light for illumination (Schott; Halogen light source, Mega-Light100).

The surface morphology of the sample plate was observed with a 3D morphological measurement system (Keyence Co.; VR-3000).

3. Results and Discussion

3.1 Laser-induced ejection of liquid droplets from an aluminum surface with 35 ns-pulsed laser irradiation

Figure 2 shows a side-view CCD camera image of the aluminum surface before and after 35 ns-pulsed laser irradiation at a fluence of 6.6 J cm⁻² (CCD frame rate: 150 kfps). A 0.8 μ L liquid droplet was first placed on the aluminum plate (Fig. 2(a)). After a single-shot of irradiation (Figs. 2(b) and (c)), the liquid droplet was explosively evaporated from the surface. The ejection velocity at the front of the liquid desorption jet was ca. 330 m s⁻¹. At a laser fluence of 3.3 J cm⁻², the ejection velocity at the front of the liquid jet was ca. 67 m s⁻¹, as shown in Fig. 3 (CCD frame rate: 100 kfps). An asymmetric ablation plume shown in Figs. 2 and 3 was probably due to inhomogenized fluence of the incident laser beam.



Fig. 1 Microroughness on the sample surface produced by laser ablation; (a) top-view and (b) 3D side-view images.

The ejection velocity was dependent on the laser fluence. Figure 4 shows the ejection velocity at the front of the liquid desorption plume as a function of the laser fluence. A linear relation between the ejection velocity and laser fluence in the range of 3 to 6 J cm⁻² was observed, together with a threshold laser fluence for the liquid ejection of 2.6 J cm⁻².

It is presumed that a rapid temperature increase of the metal surface caused by the laser irradiation may induce explosive evaporation of the liquid at the interface between the metal plate and the droplet; the boiling point of the liquid (ethyl methyl carbonate, b.p.= 107° C; ethylene carbonate, b.p.= 244° C) was much lower than the melting point of aluminum (m.p. = 660° C).



Fig. 2 Side-view CCD images of the laser-induced ejection of a liquid droplet (liquid thickness: 50 μ m) from an aluminum metal surface with a nanosecond laser ($\tau = 35$ ns) at a fluence of 6.6 J cm⁻². The CCD frame rate was 150 kfps. (a) Before laser irradiation, (b) 6.7 μ s after 1-pulse laser irradiation, and (c) 13.3 μ s after 1-pulse laser irradiation.





Fig. 3 Side-view CCD images of the laser-induced ejection of a liquid droplet (liquid thickness: 50 μ m) from an aluminum metal surface with a nanosecond laser ($\tau = 35$ ns) at a fluence of 3.3 J cm⁻². The CCD frame rate was 100 kfps. (a) Before laser irradiation, (b) 10 μ s after 1-pulse laser irradiation, (c) 20 μ s after 1-pulse laser irradiation, (d) 30 μ s after 1-pulse laser irradiation, (e) 40 μ s after 1-pulse laser irradiation, and (f) 50 μ s after 1-pulse laser irradiation.

As the liquid is transparent at the wavelengths of the lasers used in this work, the explosive evaporation of liquid was induced by photo-thermal energy transfer at the interface between metal and liquid. Aluminum plate was not ablated in this experimental condition. The ejection velocity was not significantly varied in the range of $20 - 100 \ \mu m$ of the thickness of the liquid layer.



Fig. 4 Ejection velocity at the front of the liquid desorption jet as a function of the laser fluence ($\tau = 35$ ns).

3.2 Mechanism of the laser ejection behavior for three IR lasers

Table 1 shows the threshold fluences of laser-induced ejection for the three different IR lasers examined in this work. The threshold fluence of the liquid ejection for the 90 μ s-exposure laser was observed at 130 J cm⁻². Optical microscope observations showed no photothermally-induced damage on the aluminum surface at this condition.

The threshold fluence was strongly dependent on the pulse duration of the lasers, which suggests the dispersion of photothermal energy into the bulk side of the aluminum plate under laser irradiation.

The heat penetration depth (L) is defined by the pulse duration (t) and the thermal diffusivity constant D. [1; p. 279].

$$L \approx 2\sqrt{Dt}$$
 ⁽¹⁾

 Table 1
 Threshold
 fluences
 of
 laser-induced
 ejection

 for three different IR lasers in the ambient air atmosphere.
 in the ambient air atmosphere.

IR laser (pulse duration, wavelength)	Threshold laser fluence F _{th}
¹ ns-pulsed laser (5.7 ns, 1053 nm)	¹ 0.6 J cm ⁻²
ns-pulsed laser (35 ns, 1064 nm)	2.6 J cm ⁻²
μs-exposure laser (90 μs, 1084 nm)	130 J cm ⁻²

1: Ref [5]



Fig. 5 Threshold fluence of liquid ejection as a function of the square root of laser irradiation time (t) for three IR lasers; (a) microsecond and nanosecond regions, (b) nanosecond region.

Assuming the rapid temperature increase at the surface is caused by the laser irradiation in competition with the diffusion of thermal energy into the bulk side of the aluminum plate, the square root of the laser irradiation time (t) should be proportional to the threshold fluence (F_{th}) of the liquid ejection on the basis of a simple one-dimensional energy dispersion model using Eq. (1), which gives:

$$F_{th} \propto \sqrt{t}$$
 (2)

Figure 5 shows that Eq. (2) is in agreement with the experimentally obtained values of F_{th} and τ , which suggests that the thermal energy distribution is a key for the ejection mechanism induced by the three lasers.

4. Summary

The laser-induced ejection of an organic liquid droplet from the aluminum metal surface was investigated. With a high-power ns-laser, non-contact cleaning of a large area of the solid surface is plausible in an ambient atmosphere. The threshold fluence of the liquid ejection was linearly dependent on the root of laser pulse duration, which suggests that the thermal energy distribution is a key for the ejection mechanism.

Acknowledgments

This work was supported by the Impact Program "Ubiquitous Power Laser for Achieving a Safe, Secure and Longevity Society (Program Manager: Dr. Yuji Sano)" of the Council for Science, Technology, and Innovation (Cabinet Office, Government of Japan).

References

- D. Bäuerle: "Laser Processing and Chemistry (4th Ed.)", (Springer-Verlag, Berlin & Heidelberg, 2011) Chapter: 23.7, pp.549-559.
- [2] A. C. Tam, W. P. Leung, W. Zapka, and W. Ziemlich: J. Appl. Phys., 71, (1992) 3515.
- [3] O.Yavas, P. Leiderer, H. K. Park, C. P. Grigoropoulos, C. C. Poon, W. P. Leung, N. Do, and A. C. Tam: Phys. Rev. Lett., 70, (1993) 1830.
- [4] K. Tagawa and R. J. Brodd: "Production Processes for Fabrication of Lithium-Ion Batteries (The Chapter 8 of "Lithium-Ion Batteries"), eds. by M. Yoshio, R. J. Brodd, and A. Kozawa", (Springer-Verlag, New York, 2009) p.189.
- [5] H. Niino, Y. Kato, T. Kurita, and T. Kawashima: J. Laser Micro/Nanoeng., 13, (2018) 189.

(Received: June 16, 2019, Accepted: September 29, 2019)