Understanding diffusion layer agitation by cavitation in electroplating based on high-speed monitoring

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A B S T R A C T

Ultra-sonic electroplating has gained attention owing to various advantages such as the promotion of mass transport to the substrate surface, improvement of the surface properties of the film, and improvement of limiting current density. However, no studies have clarified the mechanism in diffusion layer agitation caused by cavitation during ultrasonic electroplating. Here, we investigate the main factor of agitation by using a high-speed imaging technique to capture the agitation effect of shock waves and microjets generated from laser-induced cavitation on the diffusion layer of electroplating. The physical parameters of the agitation were characterized using image analysis and a micro-pressure gauge. The results revealed that only microjets affected the agitation phenomenon. The flow velocity was 21 m/s, and the water hammer pressure was low, at least below 0.05 MPa. Our results suggest that the flow velocity, and not the water hammer pressure, plays an important role in the agitation phenomenon on the substrate surface by cavitation.

1. Introduction

Electroplating is widely used because it can quickly and uniformly form a metal film on a substrate. The reactions during electroplating are subject to various restrictions. These include a decline in the limiting current density, a decline in the current efficiency, and the deterioration of the surface conditions of the plating film owing to the metal ion migration rate-limiting in the diffusion layer, which occurs at the substrate/solution interface during electroplating. To address these restrictions, the supply of metal ions by agitation should be promoted. Ultrasonic electroplating technology has been investigated as a method for achieving the agitation [1–7]. It has various advantages such as promoting mass transport to the substrate surface [2,3], improving the surface properties of the film [4,5], and improving the limiting current density [6,7]. It is widely and commonly used. In ultrasonic electroplating, these advantages are achieved by promoting the supply of metal ions through liquid flow such as shock waves and microjets, which are generated by cavitation bubbles when ultrasonic waves propagate through the liquid [8]. However, no studies have clarified the mechanism of agitation caused by cavitation during an ultrasonic electroplating. The difficulty in determining the details of agitation phenomena in an ultrasonic electroplating lies in these processes that occur on a small scale (tens of µm to hundreds of µm), are fast (tens of µs), and have poorly reproducibility [9].

Laser-induced cavitation is a technique used to improve the reproducibility of the cavitation phenomena [10–12]. Laser-induced cavitation is caused by the instantaneous vaporization of a liquid owing to the energy of a focused high-intensity laser. As laser-induced cavitation is triggered by laser irradiation, it can generate cavitation at any location and time with high reproducibility. Although the mechanisms of ultrasound-induced cavitation and laser-induced cavitation are different, the resulting shock waves and microjets are the same. Therefore, we believe that laser-induced cavitation can be used to elucidate the main factor of agitation in ultrasonic electroplating.

In this study, we attempted to elucidate the main factor of agitation by using a high-speed camera to capture the agitation effect of the shock waves and microjets generated from laser-induced cavitation on the diffusion layer of electroplating. In addition, the physical parameters of the agitation were measured using image analysis and a micro-pressure gauge.

2. Experimental details

The cavitation imaging system comprises a cavitation induction optical system and a high-speed imaging optical system (Fig. 1). In the cavitation induction optical system, a high-power Q-switched Nd:YAG laser (LOTIS Tii, pulse width: 5 ns, Q-SW laser) with a wavelength of 532 nm was used as the light source. The pulse energy of the Q-SW laser was adjusted to 15 mJ by using a half-wave plate (1/2λ) and a polarizing beam splitter (PBS) that was installed at the output side of the light beam.
source. The laser emitted from the polarizing beam splitter was focused on an electrochemical (EC) cell by a focusing lens (f = 25 mm) to induce cavitation. The high-speed imaging optical system was constructed using a semiconductor pulsed laser (Cavilux) with a wavelength of 640 nm for illumination, shadowgraph optics, and a high-speed camera (HPV-X2). The light emitted from the semiconductor laser was irradiated into the EC cell as a collimated light with a diameter of approximately 1 mm by a lens system, which comprised a lens and a pinhole. The light passing through the EC cell was detected by a high-speed camera after passing through a band-pass filter (BPF), which was installed to eliminate noise. The BPF selectively transmitted light around the wavelength of 640 nm.

To obtain high-speed images of laser-induced cavitation using this optical system, the Q-SW laser oscillation, the semiconductor laser for illumination, and the high-speed camera must be controlled with submillisecond accuracy. To achieve such precise control, we constructed a control system triggered by a delay generator (DG), and we sent trigger signals from the DG to the three devices. The pulse oscillation of the semiconductor laser was synchronized with the framing of the high-speed camera by using the synchronization signal from the high-speed camera. For the detailed analysis, we performed pressure measurement of shock waves and microjets. An EC cell (made of quartz) with an optical path length of 10 mm was filled with water, and a micro-pressure gauge (Mueller-Platte Needleprobe) was placed in the cell. The Q-SW laser was irradiated at 400 μm from the micro-pressure gauge, and the pressure was measured.

In this experiment, it is difficult to compare the results using the cavitation radius because the cavitation size can easily change due to small differences in the laser energy absorbed by the water. Therefore, when comparing some of the results, we used the non-dimensional distance γ, which is derived by:

\[ \gamma = \frac{h}{r} \]  

where h is the distance from the substrate and r is the bubble radius.

3. Results and discussions

The cavitation generated at different distances from the substrate (1,000 μm and 250 μm) are shown in Fig. 2 (a) and (b). In both cases, shock waves were generated during cavitation generation (Fig. 2 (a)-1, (b)-1) and cavitation collapse (Fig. 2 (a)-3, (b)-3). In contrast, microjets were not generated when the Q-SW laser was irradiated at 1,000 μm from the substrate (Fig. 2 (a)-4); however, they were generated in all three trials when the Q-SW laser was irradiated at 250 μm from the substrate (Fig. 2 (b)-4). The microjets were generated toward the substrate owing to the pressure difference caused by the restriction of liquid supply from the substrate side during the contraction process of the cavitation as it entraps the surrounding solvent [13]. Therefore, microjets can be generated by moving the cavitation generation position closer to the substrate. The cavitation radius produced in this experimental system was approximately 150 μm, which is larger than the

![Fig. 1. Schematic of imaging system (a)Overall view of imaging system (b)Side view of focusing lane (red box of (a)) (c)Inside the EC cell (blue box of (a)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)](image-url)
cavitation size produced by low frequencies below 100 kHz used in
conventional ultrasonic electroplating (approximately 110 µm at 30
kHz, 34 µm at 100 kHz) [14]. Different cavitation radius may affect
the generation of microjet due to differences in surface tension. A common
method to check whether or not ultrasonic waves produce microjet is to
irradiate aluminum foil with ultrasonic and check for erosion. Erosion
tests were conducted at 3 frequencies of 28, 45, and 100 kHz (theoretical
bubble radii of 120, 75, and 34 µm, respectively). It was confirmed that
erosion occurred at all frequencies and that the lower the frequency (the
larger the theoretical bubble radius), the more severe the erosion.
Therefore, we believe that the agitation phenomenon in this system is
more powerful than that in conventional ultrasonic electroplating.
The effects of shock waves and microjets on the agitation of
the diffusion layer were captured (Fig. 3 (a) and (b)). As mentioned above,
microjets can only be generated when cavitation occurs in the vicinity of
the substrate. Under the present conditions, only shock waves were

![Fig. 2. (a) (b) Difference in behavior depending on cavitation generation location (a) 1,000 µm from substrate surface (γ = 6.25) (b) 250 µm from substrate surface (γ = 2.11) (c) Position of the microjet tip.](image)

![Fig. 3. Effect of shock wave and microjet on agitation of diffuse layer (a) 700 µm from substrate surface (γ = 2.75) (b) 230 µm from substrate surface (γ = 1.18). (c) Images of shock wave and microjet acting on the probe tip and the pressure measured during the process. (γ = 1.29).](image)
generated when the Q-SW laser was irradiated at 700 µm from the substrate. Both shock waves and microjets were generated when the Q-SW laser was irradiated at 230 µm from the substrate. The thickness of the formed diffusion layer (shown as a white line on the substrate surface in Fig. 3 (a) and (b)) is 20–30 µm, which is similar to that of a diffusion layer (several µm to several tens of µm thick) produced during conventional electroplating. Under the condition in which only shock waves were generated, the diffusion layer on the substrate surface did not change before and after cavitation generation, and there was no agitating effect (Fig. 3 (a)). In contrast, under the condition in which both shock waves and microjets were generated, the diffusion layer on the substrate surface was locally agitated after cavitation generation, and a vortex was observed (Fig. 3 (b)). As the same phenomenon is observed in the ultrasonic electroplating, it is suggested that only microjets effect the agitating phenomenon in ultrasonic electroplating and not shock waves.

Furthermore, the physical parameters of agitation by the microjets were measured. First, the velocity of the microjets was calculated from the image of the microjets taken at a speed of 1 Mfps. Fig. 2 (c) shows a plot of the change in the tip position of the microjet over time, with the time of microjet generation being 0 µs. By obtaining an approximate line in this graph, it was found that the average velocity of the microjet is 21 m/s. This value is lower than the value measured in a previous study (about 100 m/s) [12,15]. This difference may have been caused by cavitation size. Both cavitations in the previous study had a radius of several millimeters, which is about 10 times larger than the cavitation in this study. Therefore, the cavitation energy was higher and a high-speed microjet was produced. A micro-pressure gauge was used to measure the water hammer pressure of the microjets generated by the cavitation at 400 µm from the tip (γ = 1.29), and to observe the cavitation phenomenon near the gauge (Fig. 3 (c)). In this case, the shock wave reached the tip of the micro-pressure gauge in a very short time, approximately 0.1 µs, and then, the microjet reached the tip in a few microseconds. The water hammer pressure due to the microjet was low, at least less than 0.05 MPa, although the micro-pressure gauge showed a water hammer pressure of approximately 0.2 MPa due to the shock wave. The water hammer pressure due to the shock wave may be smaller than the original water hammer pressure, considering that the shock wave hit the probe as a spherical wave, and the probe size is small (500 µm) and easily affected by the cage. However, it was shown to be at least 0.2 MPa. The small water hammer pressure of the microjet may be due to the large gamma of this condition. Dular et al. have studied the effect of microjet on aluminum substrates at various gammas, and found that the microjet damaged the aluminum substrate only at γ ≤ 0.90, where the cavitation and the substrate touch, and that no direct effect of the microjet on the substrate (i.e., microjet water hammer pressure) occurred at γ ≥ 1.90. Since the gamma in Fig. 3 is 1.29, it is considered that the microjet pressure did not occur as in the previous study. Fig. 3 (a) and (b) also show that only the microjets effect the agitation of the diffusion layer. This suggests that the flow velocity, and not the water hammer pressure, plays an important role in cavitation-induced agitation on the substrate surface.

From the above results, it is clear that volume movement by flow is necessary for the agitation effect on the diffusion layer in ultrasonic electroplating, and it is due to the microjets, and not shock waves. It is also suggested that further improvement of the ultrasonic electroplating capability could be achieved by the efficient generation of microjets.

4. Conclusion

Herein, the agitating effect of shock waves and microjets, generated by laser-induced cavitation, on the diffusion layer of electroplating was observed. It was found that the diffusion layer in the area where the microjets acted was locally agitated, suggesting that shock waves do not affect the agitating phenomenon in the ultrasonic electroplating, and only microjets do. The physical parameters of the microjets were also measured. The flow velocity was 21 m/s. However, the water hammer pressure was low, at least below 0.05 MPa. Our results suggest that the flow velocity, and not the water hammer pressure, plays an important role in the agitation phenomenon on the substrate surface by cavitation.

CRediT authorship contribution statement

Taisuke Nozawa: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration. Keiichi Nakagawa: Conceptualization, Methodology, Formal analysis, Resources, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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