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Elucidation of Droplet Development Process in Al Thin Films Prepared by Ion-Beam Evaporation

Hideki Kawahara, Hiroaki Shishido, Hideki Yanagi, Takashi Yunogami, Tsuneo Suzuki, Hisayuki Suematsu, Weihua Jiang, and Kiyoshi Yatsui
Extreme Energy-Density Research Institute, Nagaoka University of Technology, Nagaoka, Niigata 940-2188, Japan
Fax: 81-258-47-9890, e-mail: kawa@etigo.nagaokaut.ac.jp

Thin films of Al were prepared by irradiation of pulsed ion (75% proton) beam on an Al target. With the increase in the number of the irradiation, surface roughness of the target and the number of droplets on the thin films increased. The morphology of the irradiated surface target was observed with an optical microscope. As a result of analysis, many spherical projections were found and considered as the origin of the droplets. It was inferred that the spherical projections were made by quenching of a portion of the melted Al target.

Key words: ion-beam evaporation, thin films, surface roughness, surface morphology, droplet

1. INTRODUCTION

When intense pulsed light ion beam (LIB) irradiates on a solid target, high-temperature and high-density ablation plasma is generated. Utilizing the plasma, thin films can be deposited on a substrate which is facing the target. This method was termed pulsed ion -beam evaporation (IBE)[1-3]. Various thin films including $B_{12+x}C_{3-x}$[4], $SrAl_2O_4$: Eu,Dy[5] and TiFe were successfully prepared by IBE.

However, some thin films prepared by IBE method have high surface roughness, since there are many droplets on the surface. In a pulsed laser deposition (PLD) method, whose process is similar to that of IBE, surface roughness of the thin film is dependent on the grain size[6] and the relative density[7] of the target. This phenomenon was explained by the uneven energy deposition by a laser on the rough surface of the target. Fundamentally, the PLD method and the IBE method are the similar preparation technology of thin film. In the PLD method, only the surface of the target is heated by a laser beam. On the other hand, in the IBE method, the target is heated by a LIB within the *range*. Specifically, the *range* of the LIB for the Al target is estimated to be 14μm for a proton beam with the energy of 1MeV. It is easily expected that the molten layer can be waved by external force, i.e. the reaction by ablation plasma. Thus, in the IBE method, it is necessary to take the waved molten layer into consideration.

In the IBE method, the conditions to decrease droplets have not been made clear. Furthermore, systematic measurements and its explanation have not been reported at all. Therefore, these need to be elucidated, particularly, for the preparation of the flat thin films. In our previous results, surface roughness of Al and Ag targets increased with the increase in the number of ion beam irradiation[8]. By measuring the surface roughness of the thin films deposited after each irradiations. Systematic results which show the relation between the surface roughness of the target and that of the thin films can be obtained.

In this study, experimental results on the surface roughness of Al thin films and the Al target are described. A model is proposed to explain the relationship between the surface roughness of the thin film and the target in IBE process.

2. EXPERIMENTAL PROCEDURE

2.1 Experimental setup

Figure 1 shows the experimental setup of the IBE process. Experiments were carried out using an intense, pulsed power generator, "ETIGO-II." The LIB is produced by a magnetically insulated ion beam diode, where the motion of electrons was insulated by the transverse magnetic field. Using a polyethylene sheet as a flashboard anode, the ion species were mostly protons more than 75 %, which was diagnosed with an energy spectrometer. The operating conditions for the ion beam evaporation were accelerating voltage of 1MV (peak), current of 80 kA and pulse width of 70 ns (FWHM: full width at half-maximum). An Al target was inclined by 30 degrees to the ion beam axis. The distance from anode surface to the target was defined as d_{AT}, and the distance from the target to the substrate was defined as d_{TS}. The thin film preparation chamber were evacuated to $\sim 10^{-4}$ Torr.

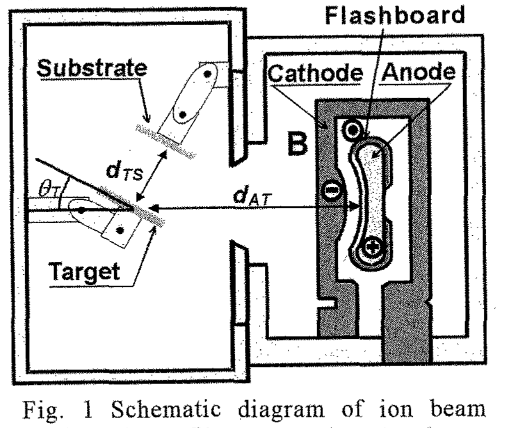


Fig. 1 Schematic diagram of ion beam diode and thin film preparation chambers.

2.2 Experimental method

Table I shows experimental conditions. For paying attention to the increase in surface roughness of the target by the increase in the number of LIB irradiation, the following experiments were carried out. (1) the Al target before irradiation was observed with an optical microscope and the surface roughness and the target mass were also measured. (2) an Al thin film was prepared on a Si substrate by irradiation of the LIB on the Al target. (3) the irradiated Al target was observed again with the optical microscope. The surface roughness and the target mass were also measured. Ablated mass was calculated from a mass difference between the target masses before and after the LIB irradiation. (4) the prepared Al thin film was observed with the optical microscope. (5) a new Si substrate was placed instead of the former substrate. The same Al target was used. The process of (2)-(5) was repeated for 10 times.

Table I Experimental condition.

Accelerating voltage (peak)	1 MV
Pulse width (FWHM)	70 ns
Energy density of ion beam	30 J/cm^2
Ion species	H^+ (75%)
Number of shots	1～10
Pressure	2×10^{-4} Torr
Target	Al
Angle of target (θ_T)	30 deg.
Substarate	Si (100)
Substarate temperature	R.T.
d_{AT}	200 mm
d_{TS}	100 mm

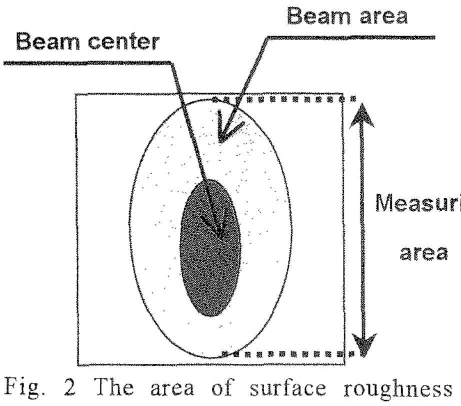


Fig. 2 The area of surface roughness measurements on irradiated the target.

The ablated mass was measured using an electronic balance. Figure 2 shows the schematic damage pattern of target irradiated by an LIB. The target roughness was measured using the surface profiler. Furthermore, the surface morphology of the ten LIB irradiated target observed by a scanning electron microscope (SEM) operated at acceleration voltage of 5.0 kV.

3. RESLUTS

3.1 Surface roughness of target

Target surface roughness (R_a) after LIB irradiations is shown in Fig. 3. The target before LIB irradiations is mirror finished surface, and surface roughness (R_a) is ~ 0.1 μm. In proportion to the number of LIB irradiations, the target surface roughness increased.

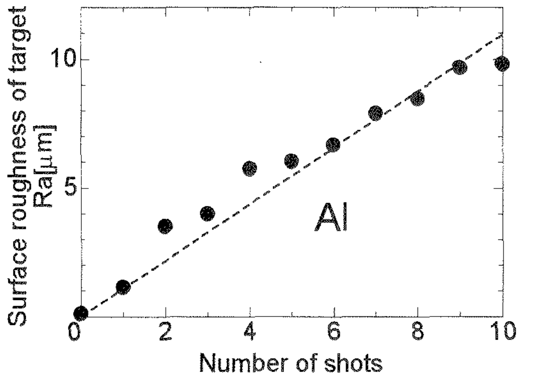


Fig. 3 Target surface roughness change due to the increase in the number of LIB irradiations.

3.2 Surface morphology of thin films

Figures 4 (a), (b) and (c) show optical micrographs of the surface of the Al thin films prepared by the first, the fifth, and the tenth LIB irradiation, respectively. White parts on the Al thin film prepared by first LIB irradiation and black parts on the other films are droplets. Occupied areas of droplets on the Al thin films are shown in Fig. 5. From Figs. 4 and 5, the amount of droplets was increased with increasing the number of irradiation. Moreover, droplets with sizes of 10-50μm are seen.

As LIB irradiation was repeated, the target surface roughness increased (Fig. 3). The number of droplets was increased with the increase in the target surface roughness.

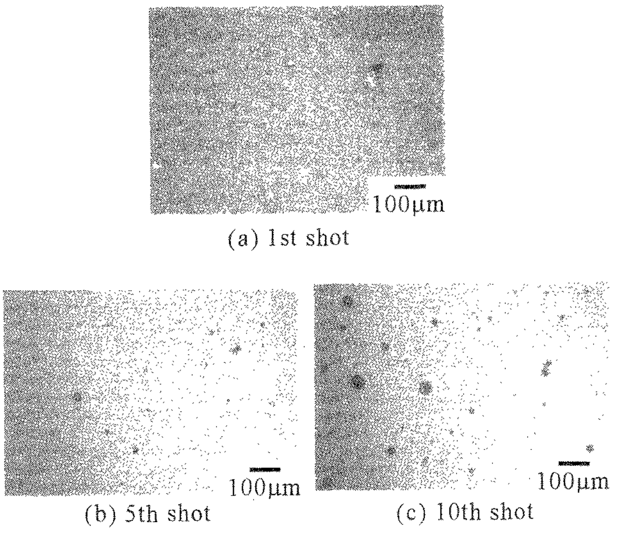


Fig. 4 Optical micrographs of the surface of Al thin films prepared by (a) the first, (b) the fifth and (c) the tenth LIB irradiation.

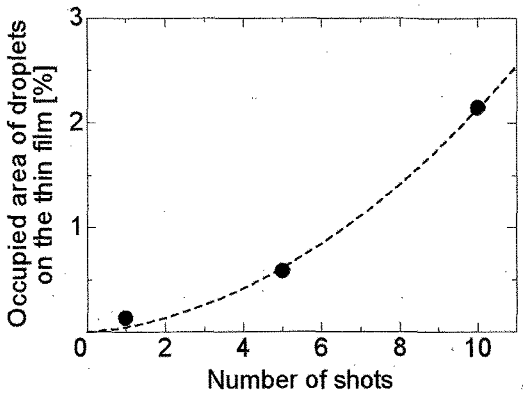
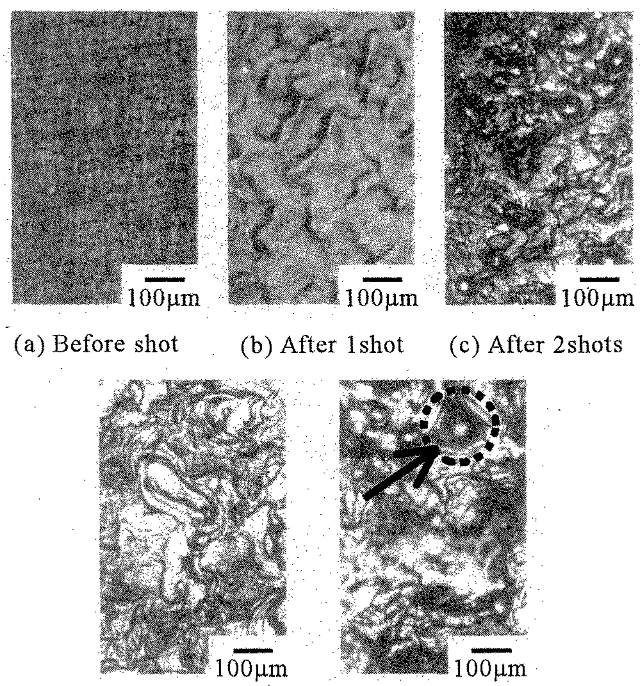


Fig. 5 Occupied areas of droplets on the Al thin films prepared by the first, the fifth and the tenth LIB irradiation.

3.3 Surface morphology of the target

Figure 6 shows optical micrographs of Al target surface (a) before, (b) after one, (c) after two, (d) after five and (e) after ten LIB irradiations. From these micrographs, surface morphology changes as the number of LIB irradiations increases. Moreover, many spherical objects, whose example was shown by an arrow in Fig. 6(e), were observed. An enlarged optical micrograph of this spherical object is shown in Fig. 7. This micrograph showed that the spherical object existed on the top of a projection.

A scanning electron micrograph of the spherical object is shown in Fig. 8. Many spherical objects exist. Enlarged scanning electron micrograph of this spherical object is shown in Fig. 9. The diameter of the spherical object is about 70μm and there are horizontal stripes in the lower half of the spherical object.



(a) Before shot (b) After 1shot (c) After 2shots

(d) After 5shots (e) After 10shots

Fig. 6 The surface morphology of Al target (a) before, (b) after one, (c)after two, (d)after five and (e) after ten LIB irradiations.

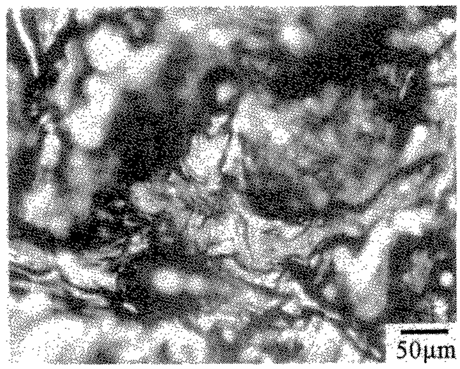


Fig. 7 The spherical projection on Al target after ten LIB irradiations.

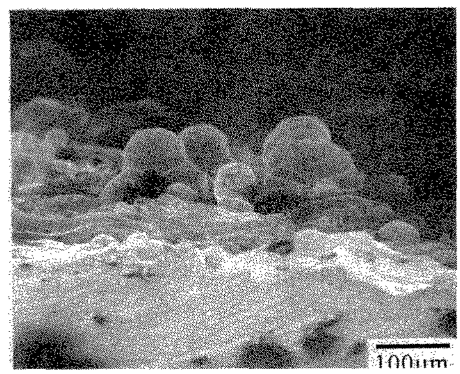


Fig. 8 A scanning electron micrograph of many spherical objects on Al target after ten LIB irradiations.

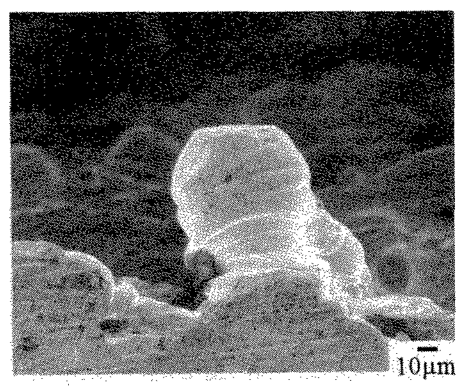


Fig. 9 A scanning electron micrograph of the spherical object on Al target after ten LIB irradiations.

4. DISCUSSION

From Fig. 3, as the number of LIB irradiations increased, the surface roughness of Al target is increased. Furthermore, the spherical objects with sizes of 20-100 μm were formed on top of projections on the target surface. From the above results, those phenomena are presumed as follow. Surface of Al target is molten by LIB irradiations, and the target receives some kind of force, i.e. the pressure by evaporating the surface of target[9]. Accordingly, the target surface is waved, and spheres at the tips will be formed by surface tension. The spherical objects shown in Figs. 7 and 8 will be solidified spheres. In Fig. 9, the spherical object on

Al target has horizontal stripes in the lower half of it. This will be due to the collapse of the solidifying spherical objects.

On the other hand, the number of droplets with sizes of 10-50 μm on the thin films increased with the increase in the surface roughness of the Al target. Therefore, it is thought that these spherical objects are the origin of droplets on the thin films.

Another hypothesis to explain the formation of droplets on the thin films prepared using the irradiated target is oxidation of target surface. As the surface roughness of the target increased, spherical objects might have been oxidized. The oxidized spherical objects on the target are hard to melt due to the high melting point of Al_2O_3. Therefore, it seems to suggest us that the number of droplets on the thin films increased as the target surface is roughened and oxidized. However, the thickness of oxide film on Al will be 0.1 μm at the most. It seems unlikely that the number of droplets increased by oxygenation of surface. The quantity of oxygen on the target surface has been measured by an energy dispersive X-ray analyzer (EDX) equipped on a SEM operated at acceleration voltage of 15kV. Experimentally, the oxygen contents of the irradiated and unirradiated target were found to be comparable. A model, as shown in Fig. 10, is proposed to explain the cause of the above-mentioned phenomenon as follows. (1) the target surface is heated by LIB irradiation and a melting layer is formed on the target surface. (2) since the heat is spread to the inside of the target, melting layer is expanded. (3) the melting layer receives the reaction force by evaporating the target. (4) the surface of melting layer is waved and splashed by the reaction force. From the splash, spherical drops are formed by surface tension of liquid Al. (5) some of those spherical drops fly toward the substrate. (6) the rest are solidified and remained on the target surface as the spherical objects on top of the projections.

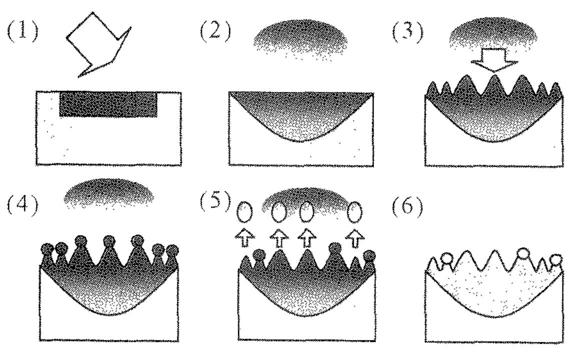


Fig. 10 A model of the droplet generating process on the thin film prepared by IBE. (1) LIB irradiation on a target, (2) melting and evaporating the target, (3) application of recoil from the ablation plasma, (4) splashing of the liquid surface, (5) formation of liquid drops and (6) cooling of the target to form spherical objects on the target.

4. CONCLUSION

After repeated LIB irradiations on an Al target, the surface roughness of Al target and the surface morphology of the target and prepared Al thin films were observed. From the experimental results, the following conclusions were obtained.

1) As the number of LIB irradiations increased, the surface roughness of the target increased, and the number of droplets with sizes of 10-50μm on the thin films increased.
2) Many spherical objects with sizes of 20-100 μm exist on top of the projections on Al target surface.

Furthermore, the following hypotheses were proposed.

1) It appears that the spherical objects formed on Al target surface are the origin of droplets on the deposited thin films.
2) Melting layer on the surface of the Al target waves due to external forces, when ablation plasma is generated. It is inferred that the spherical objects are formed by quenching the splash of melting target surface.

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