

Factors affecting surface roughness of Al_2O_3 films deposited on Cu substrates by an aerosol deposition method

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In an aerosol deposition method, factors affecting the surface roughness of Al_2O_3 thick films deposited on Cu substrates at room temperature were studied to propose guidelines for the growth of aerosol-deposited ceramic films with a flat and smooth surface morphology. The surface roughness and deposition rates of aerosol-deposited Al_2O_3 films strongly depended on the initial surface roughness of substrates. Even on flat substrates, however, their surface roughness became worse with an increase of their film thickness. Through the investigation of surface morphologies from the initial growth stages to the final, it was revealed that the deterioration of the surface roughness of Al_2O_3 films on flat substrates was substantially associated with the evolution of craters during the deposition. The fundamental cause of these results was explained by impact of hard Al_2O_3 particles above 1 μm in size by comparison with the surface morphologies of the aerosol-deposited Ag or polyimide films.

Key words: Aerosol deposition Method, Room temperature, Surface roughness, Crater, Cu substrate, Al_2O_3 Film.

Introduction

Today's electronic industries require large-scale integration and miniaturization of electronic products combined with increasing customer demands in telecommunication, computer, and automotive for rapid upgrades of functionality of products, which leads to a change of 3-dimensional integration as an alternative technology to the existing 2-dimensional integration with restrictions of integration and miniaturization. This technology based on stacking with embedded active and passive components is one of the most promising ways to integrate electronic components and to rapidly transmit signals in high frequencies [1-5].

However, in order to realize the 3-dimensional integration, there is a major problem to overcome, which is the firing temperature of different materials such as ceramics and metals. Ceramic materials give high performance in micro-electronic devices as well as high dielectric properties which cannot be expected in other materials such as metals and polymers, but the integration of ceramics and other materials such as metals and polymers always faces the problem caused by the high firing temperature of ceramics [6, 7]. To solve these problems, the process temperature of the ceramics should be reduced to below the melting point of metals or polymers so that ceramics and metals can be incorporated into the same components. For this reason, we adopted an aerosol deposition method (ADM) with which it is possible to fabricate dense ceramic thick

films at room temperature and has the advantage of creating junctions between different materials for realization of 3-dimensional integration [8-10].

Despite these advantages, however, it has been generally reported that surfaces of aerosol-deposited ceramic films have a tendency to be rough. In general, surface roughness can affect electrical properties such as electrical capacity, electronic conductivity, surface energy and sheet resistance, and roughness can determine the performance and reliability of devices, especially, when thin-film technology is applied. If the surface roughness of ceramic films is increased, the roughness of the conductor surface becomes accordingly increased during metallization, so that transmission loss at interface regions between the ceramic and metal interconnect become increased due to both signal attenuation and dispersion of the interfaces. This transmission loss results in a disturbance to the current flow in the conductor, which is especially noticeable at higher frequencies. As a result, surface roughness plays an important role in device performance, and more generally, in realization of 3-dimensional integration based on the stacking of different materials such as ceramics and metals [11, 12]. Therefore, for realization of 3-dimensional integration with ADM, it is important to fabricate flat and smooth films. And to do that, we need to understand the causes that roughen surfaces of aerosol-deposited films.

In this study, factors affecting the surface roughness of Al_2O_3 thick films on Cu substrates at room temperature using ADM were studied to propose guideline for the growth of aerosol-deposited Al_2O_3 thick films with flat and smooth surface morphologies. These studies proceed by observing the morphologies of each Al_2O_3 coating layer from the initial growth stages to the final thick film.

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Experimental procedure

The Al_2O_3 films were deposited on Cu substrates of different roughness using ADM. The details of the ADM equipment including the aerosol chamber and deposition chamber have been reported elsewhere [13, 14]. Two series of substrates were prepared. The first series of substrates was scratched by sandpaper, which had a rough morphology with a rms roughness of 297 nm. The second series of substrates was polished by a chemical process in order to generate an equivalent surface roughness, which had a flat and smooth morphology with a rms roughness of 2 nm. For the starting powder, fine Al_2O_3 powder 0.5 μm in average diameter and with a purity of 99.3% were used. The powder was dispersed by vibration into a helium carrier gas, forming an aerosol of solid particles in the helium gas. The aerosolized powders were ejected into a vacuum chamber and were targeted on the substrates 1 cm from nozzle. The aerosolized ceramic particles impact the substrate and form a dense film at room temperature. The substrate was continuously scanned back and forth by a stage. To observe the growth of aerosol-deposited Al_2O_3 films from the initial growth stages to the final thick film, the number of times that aerosol was scanned on the substrates, from 1 to 80, was increased in stages. During deposition, it took

15 seconds for one scan. Ag powders and Polyimide powders with different hardnesses and particle sizes were also used as starting powders in order to alter the surface roughness of Al_2O_3 films. The average particle sizes of Ag powders and PI powders were approximately 1 μm and 1.5 μm , respectively. Field-emission SEM was utilized for morphological observation of the deposited films. An atomic force microscope (AFM) was used to observe the surface morphology and surface roughness of substrates and films. The thickness of the coating layers was measured using a surface profilometer.

Results and Discussion

Effect of the substrate roughness on Al_2O_3 films

In this section, we present the effect of the initial Cu substrate roughness on the surface morphologies of aerosol-deposited Al_2O_3 thick films. SEM images of Cu substrates, with different rms roughness of approximately 297 nm and 2 nm, are shown in Figs. 1(a) and (c), respectively. When Al_2O_3 thick films were deposited on these two substrates with different morphologies, their surfaces became deteriorated with rms roughness of 566 nm and 116 nm and had considerably different morphologies as shown in Figs. 1(b) and (d). In Fig. 1(b), we can show that the rough surfaces

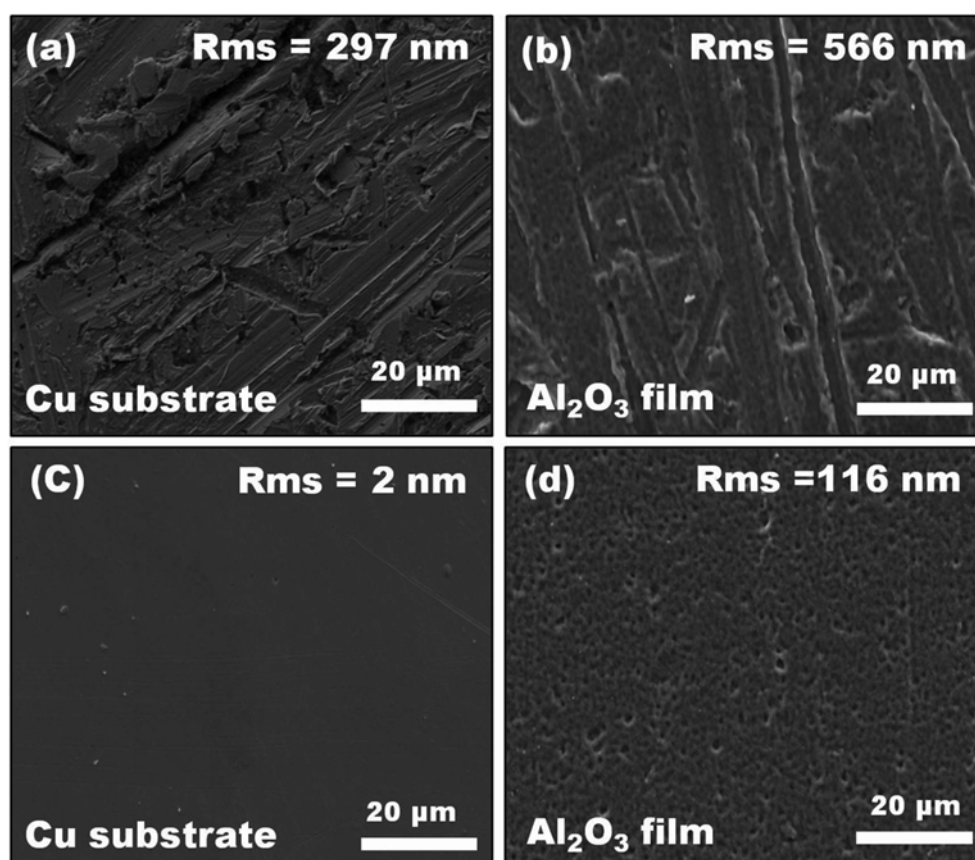


Fig. 1. SEM micrographs of Cu substrates with different surface roughnesses and Al_2O_3 films deposited on those: (a) Cu substrate with rms roughness of 297 nm, (b) Al_2O_3 film with rms roughness of 566 nm, (c) Cu substrate with rms roughness of 2 nm, and (d) Al_2O_3 film with rms roughness of 116 nm.

of the Cu substrate are transferred to surfaces of Al_2O_3 films and can observe surface relief such as craters. In Fig. 1(d), we can also show traces of obvious craters, despite the fact that the film was deposited on a flat and smooth Cu substrate.

These observations indicate that the surface roughness of Al_2O_3 films is strongly dependent on both the morphologies of the initial Cu substrates and craters that arise in the ADM process. In addition, the morphologies of these substrates considerably affect the thickness of Al_2O_3 films. Al_2O_3 film deposited on rough Cu substrate had the thickness of 1 μm , while flat Cu substrate had the thickness of 3 μm . From the above results, we confirmed that the surface features of the substrates considerably affect the surface morphology as well as film thickness of aerosol-deposited Al_2O_3 films. However, the facts which claim our attention from these results are the formation of craters regardless of the surface roughness of Cu substrates. In previous reported studies, it was reported that craters were formed by mechanical impact of particles, but the fundamental causes of craters have not been clarified yet [15, 16].

Effect of surface roughness on evolution of craters

In order to explain the causes of craters during ADM, we tried to approach growth processes of Al_2O_3 films through relations of surface roughness and film thickness from the initial growth stages to the final thick film. In these experimental processes, by using flat Cu substrates with an approximately rms roughness of 2 nm, we only considered the effect of craters and ignored the effect of the initial substrates roughness. Fig. 2(a) shows the rms roughness as a function of the scanning number. In Fig. 2(a), it is observed that the rms roughness within the scan range from 1 to 10 increased linearly with the scanning number. However, as the scanning number gradually increased up to 80, the rms roughness had a saturating trend, despite a rapid increase of the scanning number. Fig. 2(b) shows the films thickness as a function of the scanning number. In the scan range from 2 to 80, we observed that the films thickness increased linearly with the scanning number. However, in the case of one scan in which bonding between the substrate and particles is formed, the film thickness had higher values. It is considered that bonding between substrate-particles and particle-particles have a different mechanism. To confirm the change of the film thickness for every scan, its relation is represented as follows:

$$\Delta T / \Delta S_n = \frac{\Delta \text{film thickness}}{\Delta \text{scanning number}}, \quad (1)$$

where ΔT is the variation of the films thickness, ΔS_n is the variation of the scanning number, and n is the scanning number, respectively. Fig. 2(c) shows the correlation between $\Delta T / \Delta S_n$ and the scanning number. In Fig. 2(c), $\Delta T / \Delta S_n$ after one scan had about the same value in the range between 0.02 μm and 0.04 μm , whereas $\Delta T / \Delta S_n$ during the first scan was 0.15 μm .

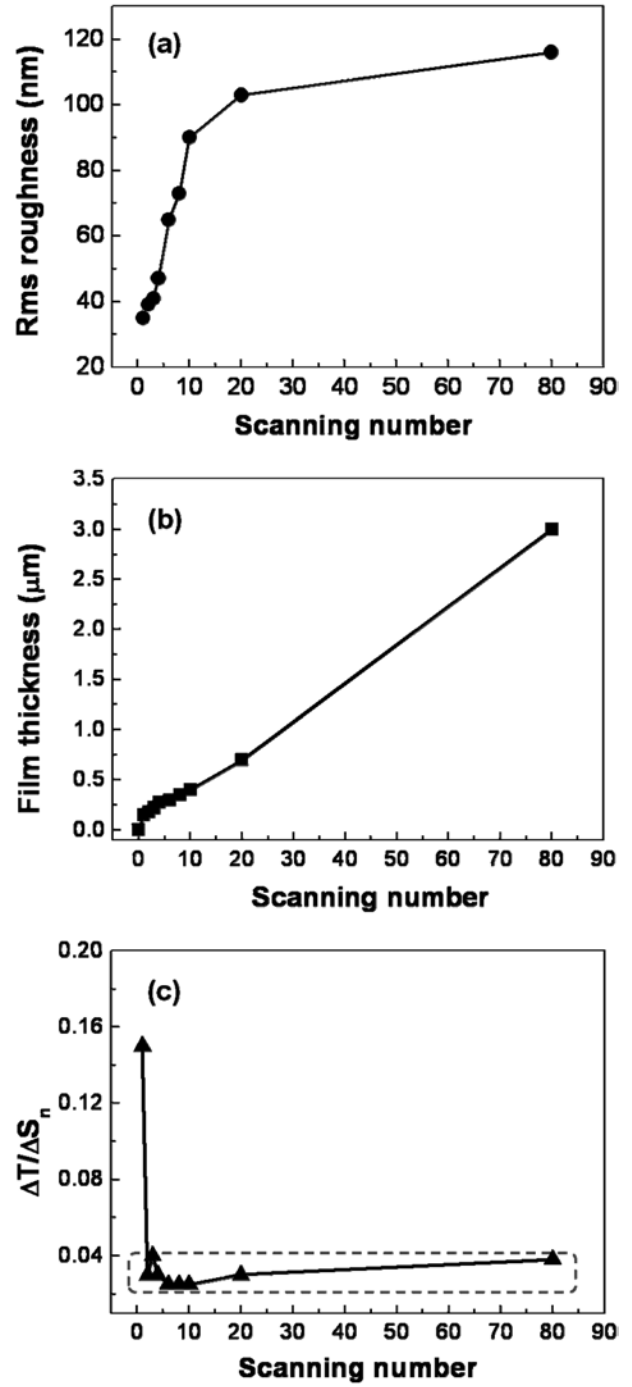


Fig. 2. (a) rms roughness of Al_2O_3 films and (b) the thickness of Al_2O_3 films as a function of the scanning number. (c) Correlation between $\Delta T / \Delta S_n$ and the scanning number.

To derive more detailed information about the causes of craters based on the results shown by Fig. 2, we observed the surface morphologies of Al_2O_3 films from the initial stages to the final thick film. Fig. 3 shows the micrographs, observed by SEM and AFM, of Al_2O_3 films deposited on Cu substrates as the scanning number increases. Fig. 3(a) shows the morphology of an Al_2O_3 film after one scan, where the remains of craters are observed, which thereby increase rms roughness. As the scanning number was

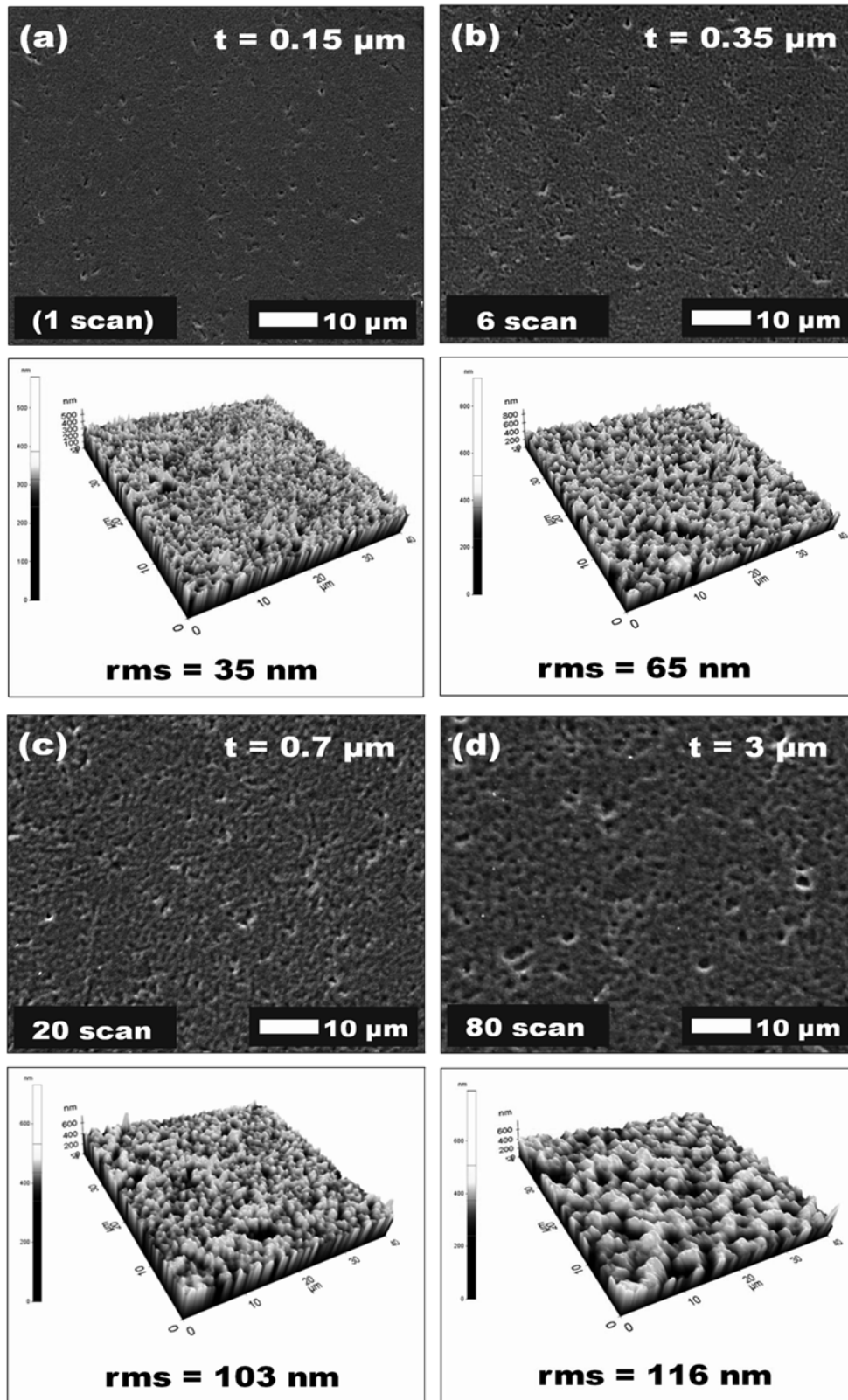


Fig. 3. SEM images and 3-dimensional AFM images of Al_2O_3 films deposited on Cu substrates with rms roughness of 2 nm with increasing the scanning number: (a) 1 scan, (b) 6 scan, (c) 20 scan, and (d) 80 scan.

increased up to 6 as depicted in Fig. 3(b), the number of craters was increased and some of them increased in size. The evolution of these craters deteriorated the morphology

of the Al_2O_3 film, which resulted in an increase of the rms roughness. When the scanning number reached 20 as shown in Fig. 3(c), both the sizes and the number of craters

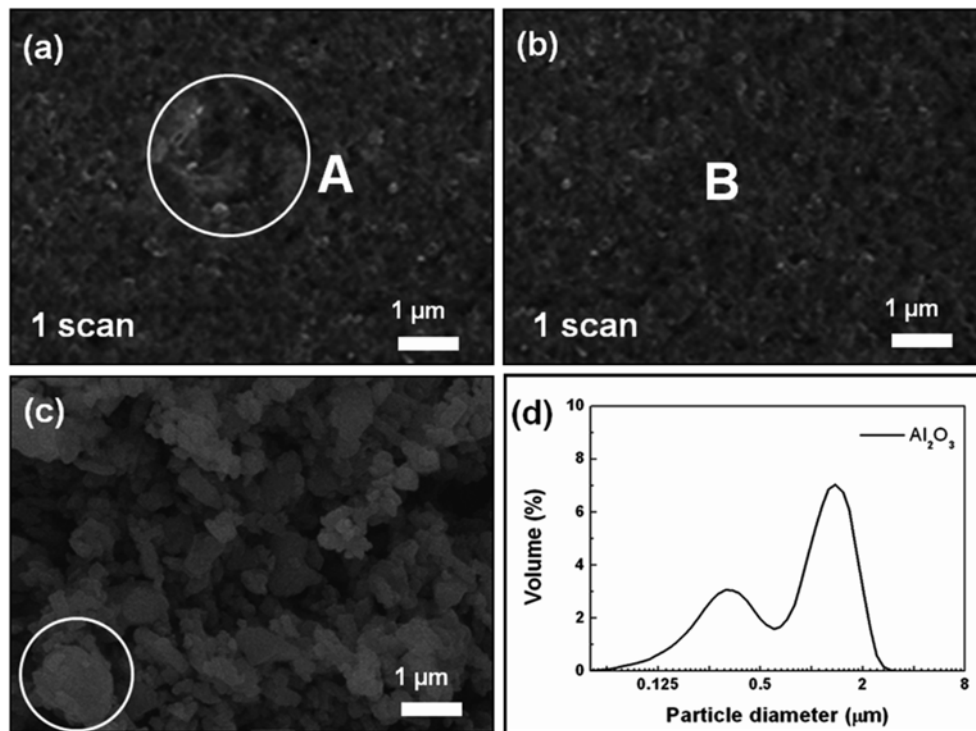


Fig. 4. SEM micrographs of (a) Al₂O₃ films with craters after one scan, (b) dense Al₂O₃ films after one scan, and (c) the as-received Al₂O₃ powders. (d) The particle size distribution of the starting Al₂O₃ powders.

increased more and more, so that their the morphology of the film was further deteriorated with an increase of the film thickness. Fig. 3(d) shows the morphology of an Al₂O₃ film in the final stage. In Fig. 3(d), the sizes of craters were larger than in the case of Fig. 3(c), but the number of existing craters did not increase. This result explains a saturating trend of rms roughness as the scanning number increases, which corresponds with the results of Fig. 2(a).

From the results obtained in Figs. 2 and 3, the evolution of craters due to an increase in the size and the number of craters with the growth of Al₂O₃ films was revealed by analysis of rms roughness and by observation of the surface microstructures, which resulted in the deterioration of the surface roughness of the Al₂O₃ films.

Consideration of crater evolution

Through the results in Figs. 2 and 3, we confirmed that the evolution of craters was a leading cause of surface roughness in ADM. Based on this a fact, we considered the fundamental cause of craters. We focused on a correlation between the initial stage of Al₂O₃ film formation and how the starting powder acted as source of film formation in ADM to know the main cause of craters formation. Figs. 4(a) and (b) show SEM images of Al₂O₃ films deposited by scanning only once. In a A region of Fig. 4(a), a crater with diameter of approximately 1 μm was observed. In the B region without craters as shown in Fig. 4(b) are can observe dense Al₂O₃ films with a fine grain size. Figs. 4(c) and (d) show an SEM image of the Al₂O₃ powder and the

particle size distribution of the Al₂O₃ starting powder, respectively. In Fig. 4(d), the particle size is broadly distributed in the range of 0.1 μm to 3 μm, and there are many Al₂O₃ particles above 1 μm included. Similarly, Al₂O₃ particles above 1 μm and Al₂O₃ particles with a range around approximately 0.5 μm were observed in Fig. 4(c).

By observing the SEM images of Figs. 4(a) and (c), we confirmed that the diameter of the crater and some Al₂O₃ powder particles were consistent at approximately 1 μm. Therefore, we concluded that the main cause of craters is caused by the impact of aerosolized Al₂O₃ particles above 1 μm in size. To gain further insight about the cause of craters, we also observed the microstructures of Al₂O₃, Ag and PI films deposited on Cu substrates under the same experimental conditions, where the Ag and PI powders used as starting powders had a mean particle size above 1 μm and a relatively low hardness in comparison with the Al₂O₃ powders. In Fig. 5, the morphologies of Al₂O₃, Ag and PI films are seen to be considerably different. The surface morphology of the Al₂O₃ film has a rough formation due to craters. However, Ag films and PI films have surface morphologies without traces of craters, despite the fact that the films were formed by particles with a size above 1 μm. These facts mean that craters are formed by mechanical impact of hard ceramic particles which have a high hardness in comparison with other materials such as polymers and metals. In addition, we can infer different mechanisms for Al₂O₃, Ag and PI films from these different morphologies. In the case of Al₂O₃ films, the aerosolized Al₂O₃ particles form dense films with

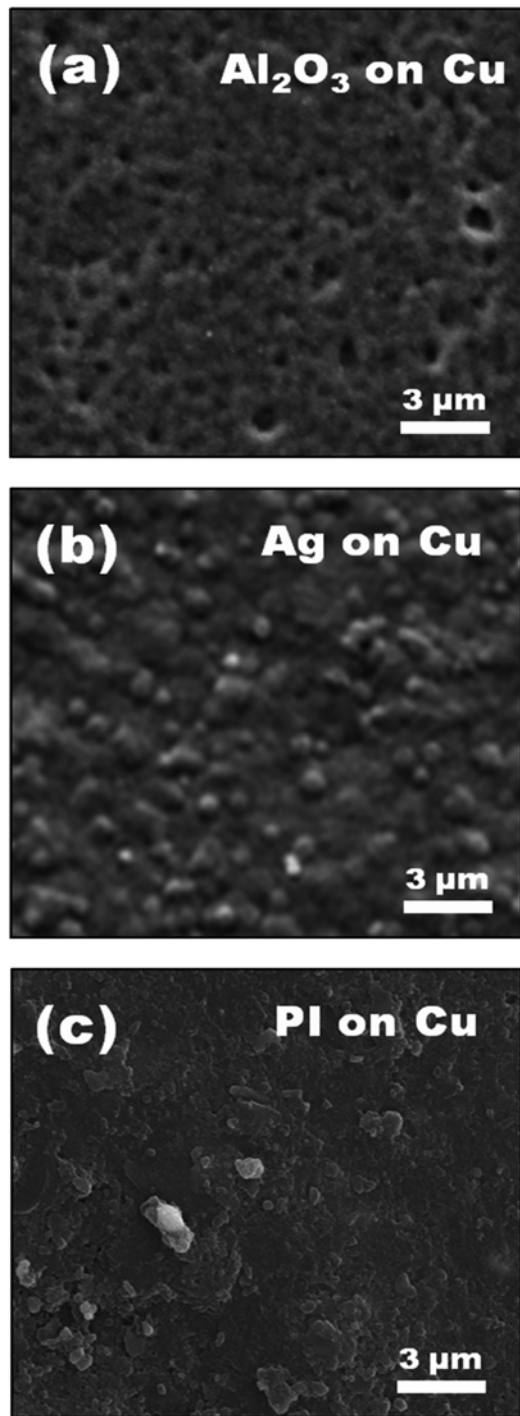


Fig. 5. SEM images of (a) Al_2O_3 film, (b) Ag film and (c) PI film with different surface morphologies.

a fine grain size due to fracture or plastic deformation. In the case of Ag films, the aerosolized ductile Ag particles form Ag films due to plastic deformation without fracture. In the case of PI films, the aerosolized PI particles form PI films due to partial fracture or plastic deformation.

From all the results so far achieved, we can explain that the surface morphologies of aerosol-deposited Al_2O_3 films were considerably associated with the surface roughness of the initial Cu substrates and that their roughness profiles

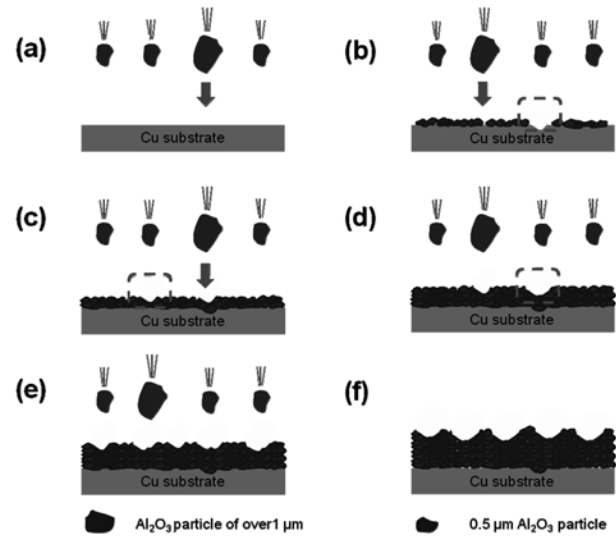


Fig. 6. Schematic drawing of the evolution of craters with an increase in the scanning number: (a) Al_2O_3 particles move to the Cu substrate, (b) $0.5\ \mu\text{m}$ Al_2O_3 particles form films, while Al_2O_3 particles above $1\ \mu\text{m}$ form craters in the Cu substrate, Al_2O_3 particles above $1\ \mu\text{m}$ (c) form new craters in the Al_2O_3 film, or (d) increase the diameter of craters, (e) The number and the diameters of craters increase with the growth of Al_2O_3 films, and (f) The sizes of craters substantially increase as the thickness of Al_2O_3 films is thicker.

deposited on flat Cu substrates were associated with the evolution of craters formed by Al_2O_3 particles with a size above $1\ \mu\text{m}$. These results are illustrated schematically in Fig. 6. The sub-micron Al_2O_3 particles move to the Cu substrate as shown in Fig. 6(a). In Fig. 6(b), Al_2O_3 particles with a size range of approximately $0.5\ \mu\text{m}$ form Al_2O_3 films, while Al_2O_3 particles above $1\ \mu\text{m}$ form craters in the Cu substrate. The Al_2O_3 particles above $1\ \mu\text{m}$ form new craters in the Al_2O_3 films previously deposited by Al_2O_3 particles of approximately $0.5\ \mu\text{m}$ as shown in Fig. 6(c) or make larger craters than the existing craters as depicted in Fig. 6(d). On the other hand, Al_2O_3 particles of approximately $0.5\ \mu\text{m}$ form dense films and retain the morphology of the substrate or film with craters previously formed. By repeating these steps of Figs. 6(c) and (d), the number and the diameters of craters are gradually increased with the growth of the Al_2O_3 films as shown in Fig. 6(e). The increase in diameters of these craters could be caused by the mechanical impact of aerosolized Al_2O_3 particles above $1\ \mu\text{m}$ in the region around craters previously formed. If the density of craters is high as shown in Fig. 6(e), new craters are hardly formed and the diameters of craters are gradually increased with increase in film thickness. In Fig. 6(f), as the thickness of Al_2O_3 films is thicker, the sizes of craters are accordingly increased, but the effect on the evolution of craters is significantly decreased. This has the relation to the saturating trend of rms roughness.

Consequently, the evolution of craters due to their generation and expansion considerably affect morphologies of Al_2O_3 films in the final stages with an increase in the film thickness. For the formation of flat Al_2O_3 films, the

generation of craters combined with their expansion should be reduced throughout the growth of Al_2O_3 films, which is closely associated with the sizes of the starting Al_2O_3 powder. The above results are expected to suggest guidelines for the growth of aerosol-deposited ceramic films with flat and smooth surface morphologies in order to apply to 3-dimensional integration of electronic components.

Conclusions

In summary, the factors affecting surface roughness of Al_2O_3 films deposited on Cu substrates at room temperature using ADM was systematically investigated in terms of the initial substrate roughness and as a function of scanning number. Our results revealed that the surface morphologies of Al_2O_3 films tended to depend strongly on the surface roughness of the initial Cu substrates and the film thickness. In addition, it was revealed that the main cause that Al_2O_3 films roughened during deposition is the evolution of craters with the growth of Al_2O_3 films. Finally, we explained that these craters are caused by impact of hard Al_2O_3 particles above $1\text{ }\mu\text{m}$ by a comparison with the surface morphologies of Ag and polyimide films.

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