Available online at www.sciencedirect.com

SciVerse ScienceDirect

Ceramics International 38 (2012) 5621–5627

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Effects of interlayer roughness on deposition rate and morphology of aerosol-deposited Al_2O_3 thick films

Chang-Wan Kim [a], Joo-Hyun Choi [b], Hyung-Jun Kim [a], Dong-Won Lee [a], Chang-Yong Hyun [b], Song-Min Nam [a,*]

[a] *Department of Electronic Materials Engineering, Kwangwoon University, 447-1, Wolgye-dong, Nowon-gu, Seoul 139-701, Republic of Korea*

[b] *Department of Materials Science & Engineering, Seoul National University of Science & Technology, 232, Gongneung-ro, Nowon-gu, Seoul 139-743, Republic of Korea*

Received 2 February 2012; received in revised form 2 April 2012; accepted 2 April 2012
Available online 9 April 2012

Abstract

The influence of the surface roughness of Al_2O_3 interlayers on the growth of Al_2O_3 thick films fabricated by an aerosol deposition (AD) process was investigated as an approach to improving the plasma resistance of the films. The Al_2O_3 interlayer was fabricated by a plasma electrolytic oxide (PEO) method. This method is capable of fabricating films on the entire surface area of 3-dimensional substrates, whereas the AD process has difficulties with depositing films on complex shapes, such as on edges and corners, and inside holes. To prevent degradation of the plasma resistance with increasing working time, the thickness of the Al_2O_3 interlayer was increased by the PEO method. The surface roughness of the Al_2O_3 interlayer was increased linearly by increasing the thickness of the Al_2O_3 interlayer. On Al_2O_3 interlayers with surface roughness values of more than 1.5 μm (R_a), Al_2O_3 films were not grown by the AD process. To investigate the effect of the surface roughness of the Al_2O_3 interlayer on the growth of Al_2O_3 films on the Al_2O_3 interlayer, we attempted to deposit Al_2O_3 films on an Al_2O_3 interlayer whose surface roughness was decreased from 1.5 μm to 0.8 μm by polishing. As a result, an Al_2O_3 film of 2.0 μm in thickness was grown by the AD process. These study results support the conclusion that controlling of the surface roughness is the most important factor in aerosol-deposited film growth.

Keywords: D. Al_2O_3; Aerosol deposition process; Plasma electrolytic oxide coating; Deposition rate; Roughness of substrate

1. Introduction

The dry etching process commonly uses a gas plasma that is capable of high-speed etching of a wafer [1]. The components of the chamber, such as the window, tubes, and plates in the chamber, are easily damaged by exposure to plasma during the process [2,3]. Contaminating particles introduced by the damage lead to a decrease in the production yield. On the other hand, a high plasma density is required to achieve high aspect ratios and good feature size control [4]. For these reasons, the components used in the etching process are protected against exposure to plasma [2,3,5].

Until now, plasma resistant ceramic bulks have been applied as components of the chamber, such as monitor windows, tubes, plates, etc. [6]. However, it was expensive to sinter the plasma resistant ceramic bulks that were used as components. Therefore, plasma resistant ceramic coating technologies, such as the thermal spray and sol–gel processes, have been widely developed [7–9]. However, these processes have a limited impact in increasing the lifetime of components due to the high porosity and rough surfaces of the coatings, which leads to contaminating particles being introduced due to exposure to the plasma. There are also difficulties involved in increasing the thickness of the coating [10]. Consequently, these problems lead to decreased production yields [11].

In order to overcome these problems, our research group used an aerosol deposition (AD) process to increase the plasma resistance properties of components. The AD process [8,12] has merits such as a high deposition rate and the ability to produce a high-density films without voids which has excellent electrical and mechanical properties [13–15]. However, it is hard to deposit films on complex shapes such as inclined edges,

* Corresponding author. Tel.: +82 2 940 5764; fax: +82 2 942 5764.
E-mail address: smnam@kw.ac.kr (S.-M. Nam).

0272-8842/$36.00
http://dx.doi.org/10.1016/j.ceramint.2012.04.003

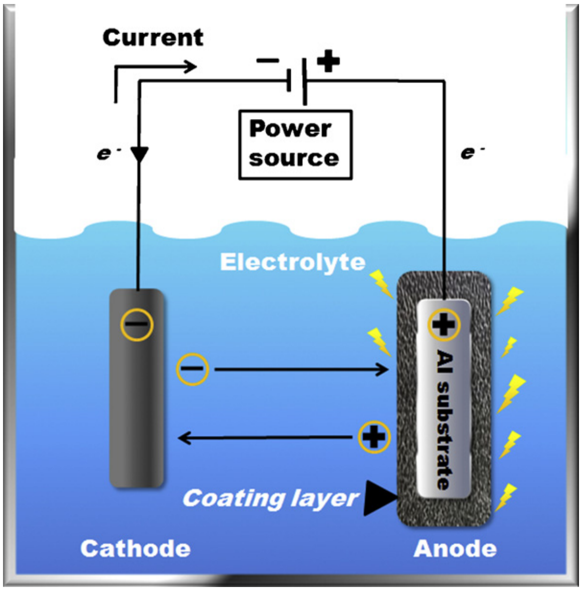

Fig. 1. Schematic diagram of the PEO apparatus.

corners, and inside deep holes. For this reason, in order to improve the coating of the weak coating regions when using the AD process, we attempted to employ Al_2O_3 interlayer films by using a plasma electrolytic oxide (PEO) coating method, which is able to fabricate Al_2O_3 film on complex shapes [9]. However, the Al_2O_3 interlayers deposited by the PEO method became the main sources of contaminating particles during the plasma etching process due to the surface morphology of the Al_2O_3 interlayer having high porosity and roughness and a low film density.

In this study, we attempted to fabricate Al_2O_3 thick films on an Al_2O_3 interlayer by using an AD process which is capable of fabricating dense and uniform films. However, the Al_2O_3 films deposited by the AD process were not grown on the Al_2O_3 interlayer. Through referring to the literature [16], we concluded that these results were caused by the surface roughness of the substrates.

Therefore, we compared the Al_2O_3 interlayer with a glass substrate, on which generally successful depositions of Al_2O_3 films have been reported. As a result, we confirmed that the difference in the roughness values between the Al_2O_3 interlayer and the glass substrate is largest difference between the two substrates. Also, it has been reported that the substrate roughness affects the surface morphologies [16].

Therefore, the main purpose of this paper is to investigate the relationship between the deposition rates of Al_2O_3 films formed by the AD process and the surface roughnesses of Al_2O_3 interlayers fabricated by the PEO method, with the aim of identifying the optimal conditions for successful fabrication of a dense and thick Al_2O_3 film.

2. Experimental

Fig. 1 shows a schematic illustration of the PEO apparatus. The Al_2O_3 interlayers were fabricated by the PEO method, which was applied to a square-shaped Al substrate (Al 6061) with a size of 20 mm × 20 mm × 10 mm as illustrated in Fig. 2(b). The applied voltage was 430 V in the anodic electrode from a power supply unit and the pulsed power supply allows control of the current density (about 200 A/cm^2). The electrolyte consists of an aqueous solution of potassium hydroxide and sodium silicate. The thickness of the Al_2O_3 interlayer was influenced by the working time. We varied the working time in the range from 30 min to 60 min. The deposition conditions for the Al_2O_3 interlayer are summarized in Table 1.

The Al_2O_3 films were deposited on the Al_2O_3 interlayer by an AD process as shown in Fig. 2(b). An Al_2O_3 powder with an average diameter of 0.5 μm and a purity of 99.8% (ALM-43,

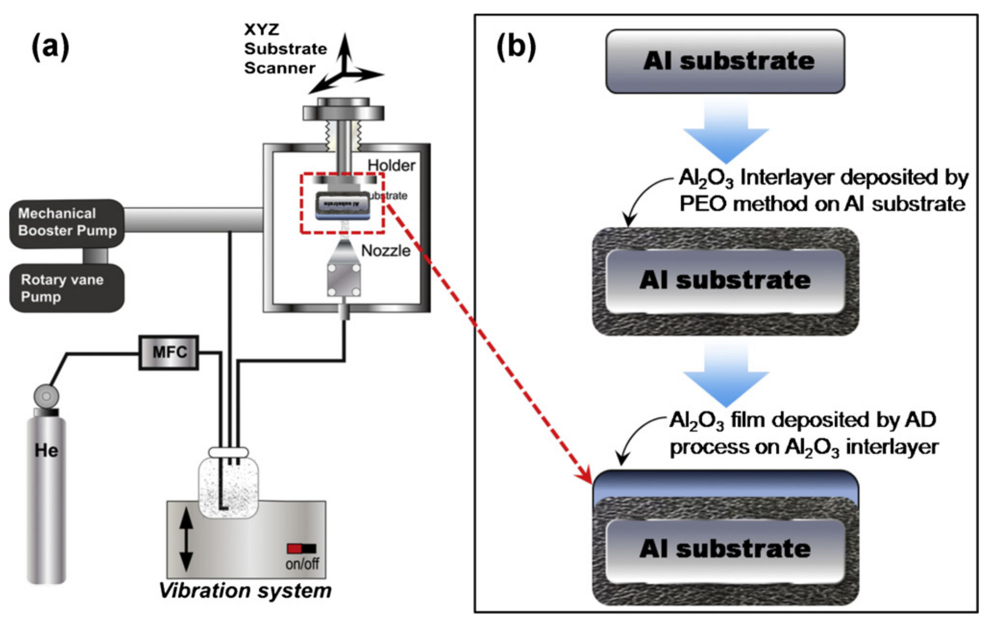

Fig. 2. Schematic diagram of (a) the AD apparatus, (b) an Al substrate, an Al_2O_3 interlayer deposited by the PEO method on an Al substrate, and an Al_2O_3 film deposited by the AD process on an Al_2O_3 interlayer.

Table 1
Experimental parameters of PEO method.

Substrate	Al 6061
Composition	KOH, Na_2SiO_3
Voltage	430 V
Current	200 A/cm^2
Working time	3600 s
Thickness	5–30 μm

Table 2
Experimental parameters of AD process.

Powder	α-Al_2O_3 (ALM-43)
Substrate	Al 6061
Carrier gas	He
Size of nozzle orifice	10 mm × 0.4 mm
Scanning rate	2 mm/s
Working pressure	10–40 Torr
Consumption of carrier gas	5 L/min
Distance between substrate and nozzle	10 mm
Deposition temperature	Room temperature
Deposition time	50 min
Deposition area	10 mm × 10 mm
Vibration speed	200–400 rpm

Showa Denko Co. Ltd., Japan) was used as the starting powder. The Al_2O_3 particles were aerosolized in an aerosol chamber by means of a vibration and mixing system; a schematic illustration of this apparatus is shown in Fig. 2(a). The particles were transported into the deposition chamber by He gas and accelerated through a nozzle. The gas flow rate was 5 L/min. The distance between the substrates and the nozzle was 1 cm and the deposition time was 5 min. The deposition conditions for the aerosol-deposited Al_2O_3 films (AD-films) are summarized in Table 2.

The surface morphologies of the AD-films were observed using an optical microscope. The microstructure of the Al_2O_3 interlayer and the AD-films were observed by field-emission scanning electron microscopy (FE-SEM, S-4700, HITACHI). The thickness of each film was measured by a surface profilometer (XP-100, AMBIOS Technology Co., USA). Finally, the Al_2O_3 interlayer and Al substrates were polished by a ceramic and metal polisher (SYSTEAM 2000, LECO) to control their surface roughness.

3. Results and discussion

The Al_2O_3 interlayers were formed by the PEO method on Al substrates, following the optimized experimental procedure shown in Table 2. Fig. 3 shows a plan-view SEM image of an Al_2O_3 interlayer formed by the PEO method. It shows a microstructure consistent with those found in previous works [6]. Compared to the Al substrates, the Al_2O_3 interlayer had a much rougher surface with holes, including deep central shrinkage holes which were unfavorable in terms of the plasma resistance.

Next, to overcome the disadvantages of the PEO method, we attempted the deposition of Al_2O_3 films by the AD process on Al_2O_3 interlayers. In order to find the optimized deposition

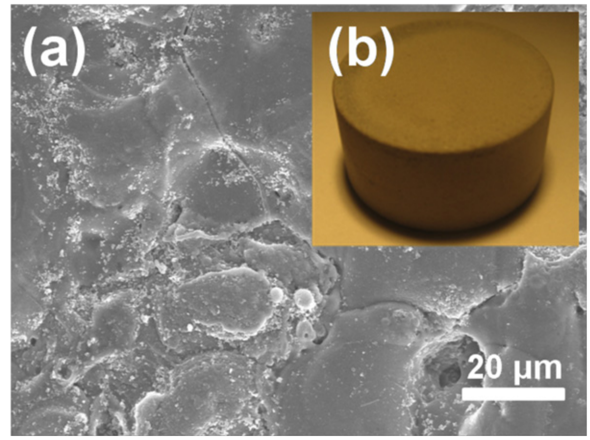

Fig. 3. Plan-view SEM image of (a) the Al_2O_3 interlayer deposited by the PEO method on an Al substrate and (b) a photograph of the same.

conditions as shown in Table 1, the Al_2O_3 films were deposited by an AD process onto glass substrates, since it has been reported that ceramic thick films have been successfully deposited on glass substrates using the AD process [15]. Fig. 4(a) shows an optical image of an aerosol-deposited Al_2O_3 film (AD-film) on glass with a hardness of 600 ± 5 HV and a surface roughness (R_a) of 0.004 μm; a dense and flat film was clearly confirmed to have been formed by the AD process. The thickness of the Al_2O_3 film was approximately 5 μm and it exhibited a hardness of 1200 ± 5 HV and a surface roughness (R_a) of 0.085 μm, respectively. Based on the above optimized experimental conditions, Al_2O_3 thick films were deposited on an Al_2O_3 interlayer with a hardness of 270 ± 5 HV and a surface roughness (R_a) of 1.5 μm, as shown in Fig. 4(b). The AD-films had very rough surface morphologies and their thicknesses were not confirmed, unlike those formed on the glass. Also, the deposition of AD-films was not successful in any of the attempts. Therefore, it is considered that this result was caused by the difference between the surface properties of

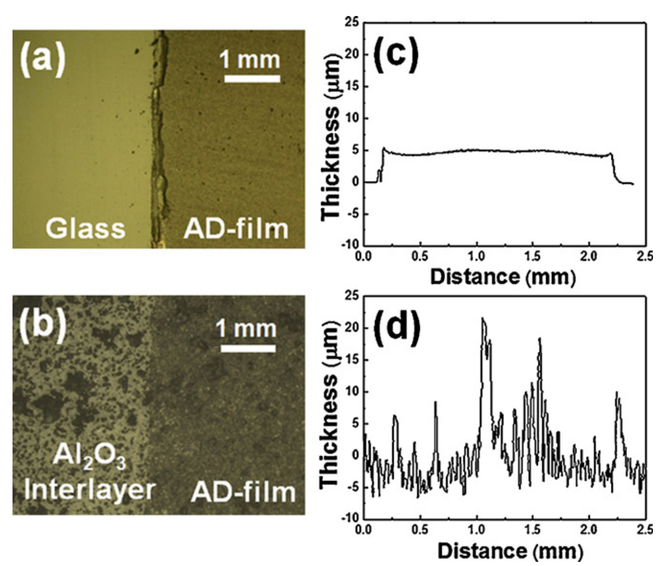

Fig. 4. Optical images of (a) an AD-film on a glass substrate and (b) an AD-film on an Al_2O_3 interlayer, and (c and d) the thickness profiles of the films in (a) and (b), respectively.

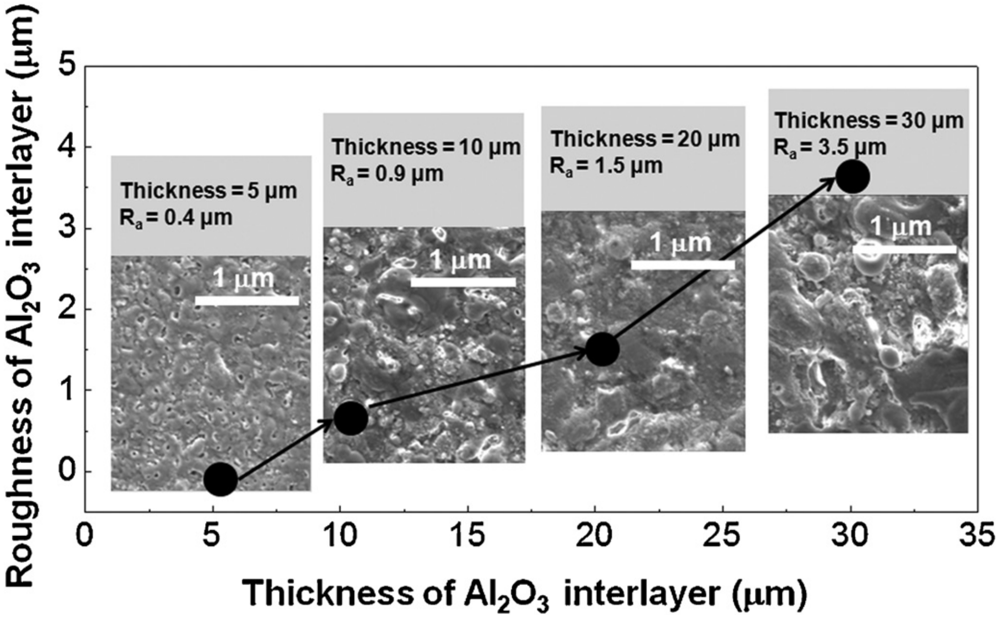

Fig. 5. Plan-view SEM images of the surfaces of Al_2O_3 interlayers with different thicknesses deposited by the PEO method on Al substrates.

the glass substrate and those of the Al_2O_3 interlayer. The surface roughness of the glass substrate ($R_a = 0.004$ µm) was very much lower than that of the Al_2O_3 interlayer ($R_a = 1.5$ µm). With these results, it is presumed that the main factor governing the fabrication of the AD-films on the Al_2O_3 interlayer was the surface roughness, because the difference between the surface roughness values of the glass substrate and the Al_2O_3 interlayer was much larger than the difference between the hardness values. Therefore, Al_2O_3 interlayers with different surface roughnesses were prepared in order to investigate the effect of surface roughness on the AD process.

The Al_2O_3 interlayers with different thicknesses were fabricated on the Al substrate because changing the thickness of the Al_2O_3 interlayer affects the surface roughness of the Al_2O_3 interlayer [6]. Fig. 5 shows the surface roughness of the Al_2O_3 interlayer as a function of its thickness. As the thickness of the Al_2O_3 interlayer was increased from 5 µm to 30 µm, the surface roughness of the Al_2O_3 interlayer was accordingly increased from 0.4 µm to 3.5 µm, respectively.

To explain the relationship between the surface roughness of the Al_2O_3 interlayer and the growth of AD-films, the deposition results of AD-films in relation to Al_2O_3 interlayer roughness were analyzed. Fig. 6 shows the results for the AD-films on Al_2O_3 interlayers with different surface roughnesses. When the surface roughness (R_a) values of Al_2O_3 interlayers were 0.4 µm and 0.9 µm, respectively, the thicknesses of the deposited AD-films were approximately 4 µm and 2 µm, respectively. However, in the case where the surface roughness (R_a) was larger than 1.5 µm, AD-films were not grown. As a result, it is considered that the decrease in thickness of the AD-film is associated with an increase of the surface roughness of the Al_2O_3 interlayer.

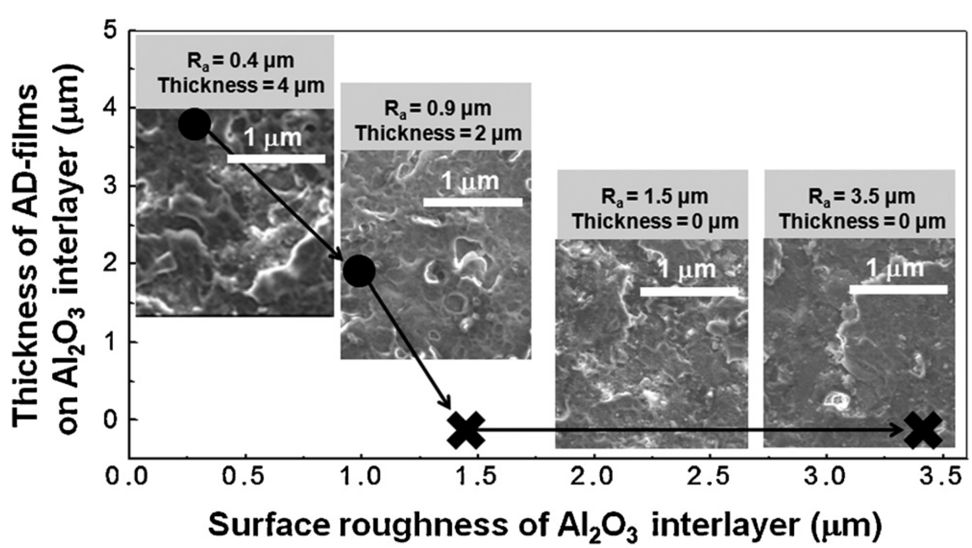

Fig. 6. Plan-view SEM images of the surfaces of AD-films on Al_2O_3 interlayers with different thicknesses deposited by the PEO method on Al substrates.

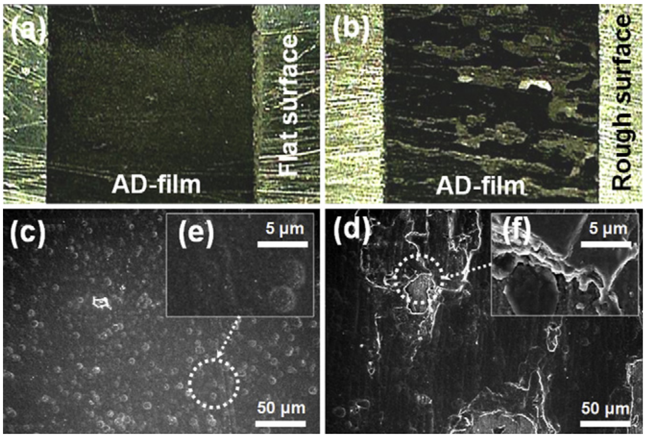

Fig. 7. Plane-view optical image of AD-films fabricated on Al substrates with (a) flat (R_a = 1 μm) and (b) rough (R_a = 20 μm) surfaces. Plan-view SEM images of the surfaces of AD-films on Al substrates with (c) flat surfaces, (d) rough surfaces and (e and f) high magnification images of (c and d), respectively.

In order to confirm the above results, two types of Al substrates with flat and rough surfaces were prepared. Fig. 7(a) and (b) shows plan-view optical images of the AD-films that were deposited on these two types of Al substrates, respectively. When using flat Al substrates (R_a = 1 μm), dense AD-films were formed over an unbroken area as shown in Fig. 7(c). In contrast, when using a rough Al substrate (R_a = 20 μm), the surfaces of AD-films were deteriorated with broken areas as shown in Fig. 7(d). In addition, the deposition rate of AD-films on rough Al substrates was decreased compared to that on flat Al substrates. These results confirm that the surface roughness of the substrate had an influence on the properties of the deposited AD-films. Rough surfaces of substrates bring about a low deposition rate and unfavorable film morphologies such as broken areas.

The causes of the above results were considered, as shown in Fig. 8. Although the deposition mechanism of the AD process is not yet understood, it is considered that bonding between particles results from the plastic deformation and fracture of particles during AD processing [17]. The results for the two cases described above suggest that the deposition rate and morphologies were determined by the procedure that is used. Fig. 8(a) and (b) depicts the growth processes of AD films on Al_2O_3 interlayers with flat and rough surfaces, respectively.

In the case of the flat substrate, where have slight indentation in the surface, as shown in Fig. 8(a). First, the starting particles collided with the flat substrate. Upon impact, the primary particles were pulverized due to their large kinetic energy. The kinetic energy of each particle was converted into bonding energy between the substrate and the primary particle. Also, particle-to-particle bonding was caused by plastic deformation or fractures of pulverized particles. Consequentially, the starting particles deformed into a dense, uniform, and hard ceramic layer and the slightly indented surface fill with hard ceramic layer.

In the case of a rough substrate as shown in Fig. 8(b) and (c), this substrate can be subdivided in three sections. First, when

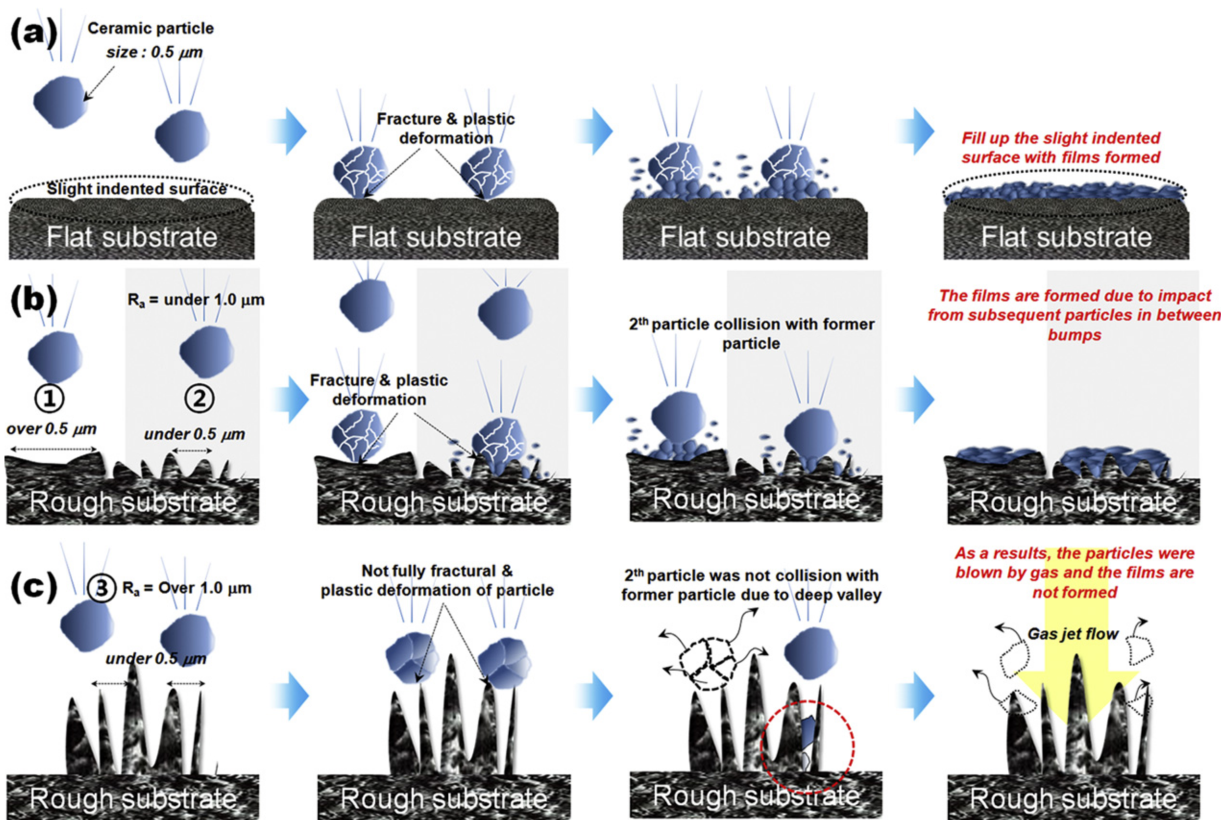

Fig. 8. Schematic diagrams showing Al_2O_3 films being formed by mechanical impacts of Al_2O particles on flat substrate (a), rough surface with under 1.0 μm roughness (R_a) (b), and rough surface with over 1.0 μm roughness (R_a) (c).

the interval between bump and bump is larger than a particle size, the particles were broken and successfully converted to form a film. This is similar to the flat substrate situation. Second, when the interval between bump and bump is smaller than a particle size and roughness of surface is smaller than 1.0 μm, the valley between bumps on the substrate are filled with fracture and plastic deformation of Al_2O_3 particles, and then Al_2O_3 films are formed due to impact from subsequent particles. Third, In the case of substrate roughness is above 1.0 μm, the particles were not fully broken due to dispersion of their kinetic energy. The valleys are not filled with 0.5 μm Al_2O_3 particles due to the deep valley and the former Al_2O_3 particles were blown by gas get flow. So, subsequent particles were not collision with former particle. Consequentially, The Al_2O_3 particles do not convert to form films.

Namely, the distance of between bumps and roughness were effect to film growth. So, the deposition rate of films on a rough substrate was lower than the deposition rate of films on a flat substrate and broken areas were observed in the films.

From these considerations and the above-mentioned facts, we conclude that the roughness of the Al_2O_3 interlayer affected the deposition rate and morphology. For the fabrication of AD-films on an Al_2O_3 interlayer with a surface roughness of 3.5 μm (R_a), we used an Al_2O_3 interlayer with a thickness of 30 μm and a surface roughness of 3.5 μm (R_a), which we polished to decrease the surface roughness, as shown in Fig. 9(b). This

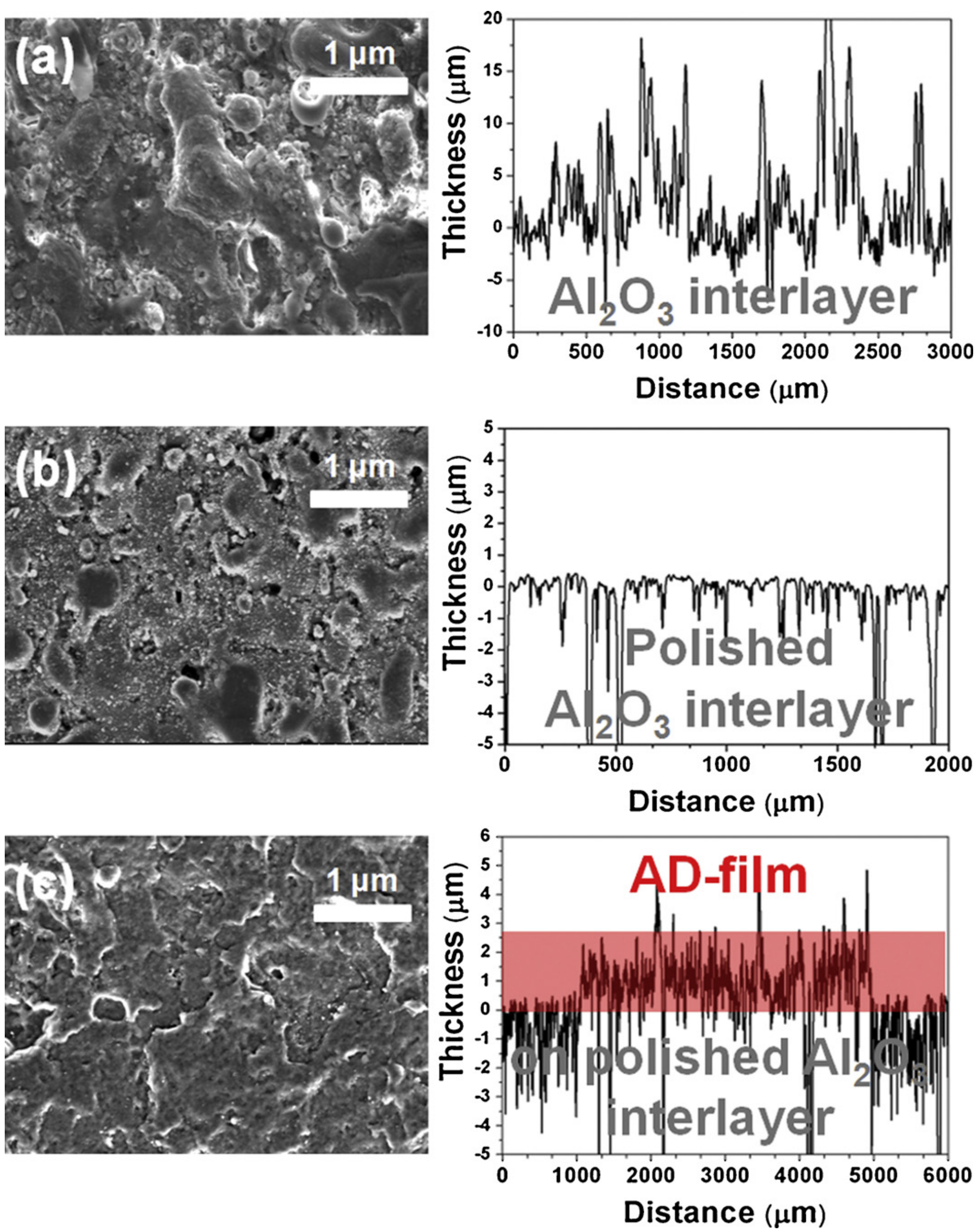

Fig. 9. Plan-view SEM images of the surface of an Al_2O_3 interlayer (a) before and (b) after polishing, and (c) AD-films on the polished Al_2O_3 interlayer and their thickness profiles, respectively.

surface roughness of the Al_2O_3 interlayer was decreased from 3.5 μm to 0.8 μm by the polishing process and then an Al_2O_3 thick film was deposited by an AD process on the polished Al_2O_3 interlayer. As a result, an Al_2O_3 thick film of 2 μm in thickness was grown by the AD process on the polished Al_2O_3 interlayer.

In this paper, we reported the effect of surface roughness on the fabrication of dense Al_2O_3 thick films by the AD process. Through this study, we have revealed that a low roughness of the substrate is one of dominant factors promoting deposition during the AD process. However, in spite of reducing the surface roughness of the Al_2O_3 interlayer to give improvements of the deposition rate and morphology, the AD-film had a limited thickness of 2 μm. In the future, to overcome this thickness limit, other factors should be investigated for the improvement of the properties of AD-films.

4. Conclusion

The effects of the surface roughness of the substrate on the deposition rate and morphology were investigated in order to fabricate dense and thick AD-films. Using the PEO method, Al_2O_3 interlayers were prepared on Al substrates with different roughness values and then we attempted to deposit Al_2O_3 thick films on these Al_2O_3 interlayers by using an AD process. The deposition rate and morphology of aerosol-deposited Al_2O_3 thick film were dependent on the surface roughness of the Al_2O_3 interlayer. The thickness of aerosol-deposited Al_2O_3 thick films decreased with increasing surface roughness of the Al_2O_3 interlayer. When the surface roughness of the Al_2O_3 interlayer was over 1.0 μm, aerosol-deposited Al_2O_3 thick films were not grown. On the other hand, when the surface roughness was below 1.0 μm, an aerosol-deposited Al_2O_3 thick film was grown with a thickness of 2.0 μm. This results support the conclusion that a rough surface of the Al_2O_3 interface adversely affected the film growth.

Acknowledgement

The present research has been conducted using the research grant of Kwangwoon University in 2012.

References

[1] C.D.W. Wlkinson, L. Deng, M. Rahman, Issues in etching compound and Si-based devices, Japanese Journal of Applied Physics 41 (2002) 4261.
[2] J. Iwasawa, R. Nishimizu, M. Tokita, M. Kiyohara, K. Uematsu, Plasma-resistant dense yttrium oxide film prepared by aerosol deposition process, Journal of the American Ceramic Society 90 (8) (2007) 2327.
[3] J. Iwasawa, R. Nishimizu, M. Tokita, M. Kiyohara, K. Uematsu, Dense yttrium oxide film prepared by aerosol deposition process at room temperature, Journal of the Ceramic Society of Japan 114 (2006) 272.
[4] S.A. McAuley, H. Ashraf, A. Chambers, S. Hall, J. Hopkins, G. Nicholls, Silicon micromachining using a high-density plasma source, Journal of Physics D: Applied Physics 34 (2001) 2769.
[5] B.Y. Kim, D.Y. Lee, Y.N. Kim, M.S. Jeon, W.S. You, K.Y. Kim, Analysis of oxide coatings formed on Al1050 alloy by plasma electrolytic oxidation, Journal of the Korean Ceramic Society 46 (2009) 295.
[6] M. Kodo, K. Soga, H. Yosida, T. Yamamoto, Low temperature sintering of polycrystalline yttria by transition metal ion depoing, Journal of the Ceramic Society of Japan 117 (6) (2009) 765.
[7] J. Singh, D.E. Wolfe, Nano and macro-structured component fabrication by electron beam-physical vapor deposition (EB-PVD), Journal of Materials Science 40 (1) (2005).
[8] J. Akedo, Room temperature impact consolidation (RITC) of fine ceramic powder by aerosol deposition method and applications to microdevices, Journal of Thermal Spray Technology 17 (2) (2008) 181.
[9] J.A. Curran, T.W. Clyne, Thermo-physical properties of plasma electrolytic oxide coatings on aluminium, Surface and Coatings Technology 199 (2005) 168.
[10] D.H. Riu, S.W. Lee, Y.K. Jeong, S.C. Choi, Microstructural control of Y_2O_3 coating on quartz glass for the reduced thermal mismatch stress, Key Engineering Materials 601 (2004) 264–268.
[11] G.S. May, C.J. Spanos, Fundamentals of Semiconductor Manufacturing and Process Control, IEEE, New Jersey, 2006, p. 98.
[12] J. Akedo, Aerosol deposition of ceramic thick films at room temperature: densification mechanism of ceramic layers, Journal of the American Ceramic Society 89 (6) (2006) 1834.
[13] M. Lebedev, S. Krumdieck, Optically transparent, dense α-Al_2O_3 thick films deposited on glass at room temperature, Current Applied Physics 8 (2004) 233.
[14] M. Lebedev, J. Akedo, T. Ito, Substrate heating effects on hardness of an α-Al_2O_3 thick films formed by aerosol deposition method, Journal of Crystal Growth 275 (1–2) (2004) e1301.
[15] D.W. Lee, H.J. KIM, S.M. Nam, Effects of starting powder on the growth of Al_2O_3 films on Cu substrates using the aerosol deposition method, Journal of the Korean Physical Society 57 (4) (2010) 1115.
[16] D.W. Lee, S.M. Nam, Factors affecting surface roughness of Al_2O_3 films deposited on Cu substrates by an aerosol deposition method, Journal of Ceramic Processing Research 11 (1) (2010) 100.
[17] D.W. Lee, H.J. Kim, Y.H. Kim, Y.H. Yun, S.M. Nam, Growth process of α-Al_2O_3 ceramic films on metal substrates fabricated at room temperature by aerosol deposition, Journal of the American Ceramic Society 94 (9) (2011) 3131.