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Microstructure of PZT films deposited by Inductively Coupled Plasma Assisted Aerosol Deposition Method

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Lead zirconate titanate (PZT) films were deposited by inductively coupled plasma (ICP)-assisted aerosol deposition (AD) method. The influence of RF power was investigated for the deposition rate and microstructure of the PZT films. With our experimental deposition system, the deposition rate of PZT film was approximately 1μm/min when using the AD method without plasma assist. On the other hand, the deposition rate rose to over 10μm/min using the ICP-assisted AD method with an RF power of 800 W. The cross-sectional microstructure of the film was difference using an RF power of 800W and that without plasma assistance.

Key words: ICP-assisted AD method, PZT film, Microstructure

1. INTRODUCTION

PZT film has garnered much attention because of its excellent piezoelectric properties and applications in various devices, such as piezoelectric actuators and ultrasound transducers [1-3]. In recent years, it has become a necessary to produce dense PZT films with thickness over 1μm on Si and metal substrates. There are some reports on the fabrication of PZT thick film by sol-gel, sputtering or hydrothermal synthesis method. Unfortunately, there are several problems associated with these methods, including crack occurrence and peel-off from the substrate.

On the other hand, PZT films thicker than 10μm can be easily fabricated by an aerosol deposition (AD) method, which produces a high deposition rate and good adherence to the substrate [4-7]. However, annealing at approximately 600°C was required to recover dielectric properties of the PZT film in order to realize dielectric properties comparable to those obtained using conventional deposition methods, because structure defects were introduced inside the PZT film during the deposition. When considering of device applications, a greater reduction in the processing temperature is needed for the integration of electro-ceramic materials with metal materials, which have low melting temperature.

We have been investigating ways of improving the dielectric properties of the as-deposited PZT film and methods to reduce the processing temperature. For example, PZT film was fabricated by the AD method using direct current (DC) plasma irradiation to the aerosol to improve the dielectric properties [8]. It is considered that water (H_2O) on the surface of the PZT powder was absorbed by helium plasma irradiation. However, it is necessary to further improve the dielectric properties of the film more.

In this study, PZT film was fabricated by the ICP-assisted AD method, and the influence of RF power on the deposition rate, microstructure was thoroughly investigated.

2. EXPERIMENTAL PROCEDURE

The experimental setup for the ICP-assisted AD method is schematically shown in Fig.1. The apparatus and details of the normal AD method have been were reported elsewhere [6]. The preparation of PZT film is as follows.

Aerosol chamber was filled with PZT dry powders and these chambers were then vacuumed. Carrier gas was introduced into the aerosol chamber and mixed with the particles to introduce an aerosol flow in the chamber. The aerosol flow was transported through the tube to the nozzle and then ejected from the nozzle into the processing chamber. To investigate the influence of ICP-irradiation to the aerosol, we introduced a plasma cavity between the aerosol chamber and the processing chamber. The plasma cavity was formed using silica glass and a four-turn copper coil. One end of the coil was coupled to an RF power generator (JEOL RF-12020) at 13.56MHz via a matching network (JEOL RF-34021) while the other end was directly grounded. This formed the aerosol nozzle and acted as part of the aerosol tube. Helium gas was used as the carrier gas and the consumption 3.0 L/min. The distance between the silica glass nozzle and the substrate was 15 mm. The deposition time was 3 min for all the samples.

Figure 2 is shows that the SEM image of the powder using the deposition tests. This powder was annealed in an electric furnace at 825 °C for 4 h. The average particle size of the powder measured using this image, was 0.46μm.

The microstructure of the samples were analyzed with X-ray diffraction (XRD, Rigaku RINT-2500), and observed with scanning electron microscopy (SEM, JEOL JSM-5410).

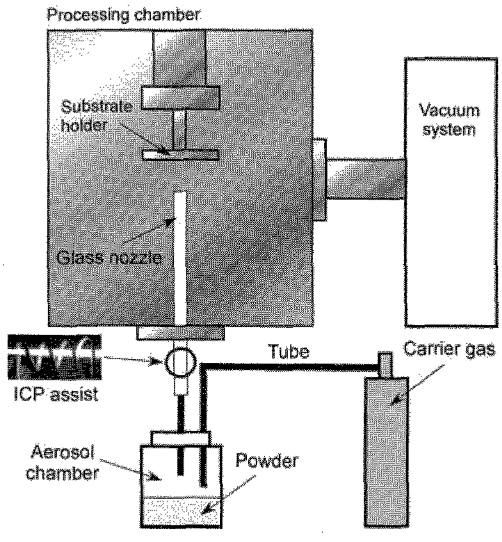

Fig.1 Illustration of ICP-assisted AD method

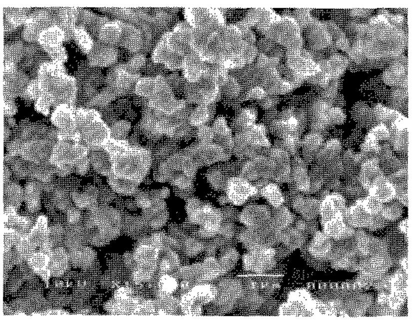

Fig.2 SEM image of the powder using deposition test

3. RESULTS AND DISCUSION

Figure 3 shows the photograph of a circular PZT film deposited on a glass substrate by the ICP-assisted AD method with an RF power of 800W. The diameter of this circular film is about 8 mm and the film thickness is about 80μm in the center. The glass nozzle shape and the flow distribution of the helium gas in the nozzle influenced the shape of the film.

Figure 4 represents the relationship between the film thickness and the RF power in the PZT films. The deposition time was 3 min for all the samples. The film thickness of these samples was measured at the center point. The film thickness was approximately 3μm when using AD method without plasma assistance, whereas the film thickness was around about 80μm when using the ICP with an RF power of 800 W. From these results, it was observed that the deposition rate tends to increase when the RF power is increased.

Figure 5 shows the cross-sectional microstructures of the PZT films deposited on the glass substrate using the AD method without ICP assist and with ICP assist. As can be seen in both the photographs, a thick and dense PZT film was formed on the substrate. However, the microstructure of Fig.5 (A) and (B) is quite different. The microstructure in Fig.5 (A) is the same as observed in other reports [9]. The difference in the microstructure in Fig.5 (B) is as the other reports. Some particles were observed in this microstructure of the film. The average particle size, which was measured from this photograph, was about 0.34μm. The average particle size of the powder using the deposition test, which was measured from this result, was 0.46μm. The average particle size of the sample and the powder was not different form this result. There is a possibility that the there is a difference in the deposition mechanism between the normal AD method and the ICP-assisted AD method.

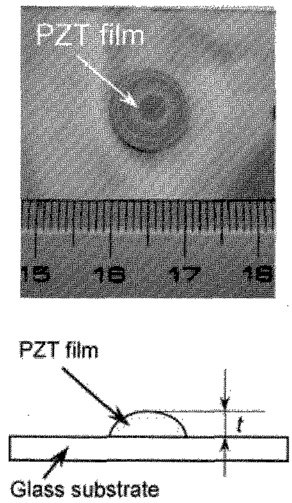

Fig.3 Photograph and illustration of a circular PZT film deposited on a glass substrate

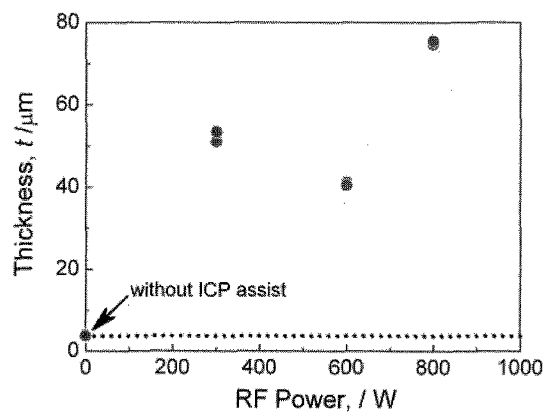

Fig.4 Relationship between RF powder and film thickness under same deposition time

Figure 6 shows the XRD patterns of the PZT film deposited by ICP-assisted AD method. It was observed that the diffraction peak angel of PZT (110) tends to increase while the RF power was increased. Half value of width in PZT (110) decreased while the RF powder increased. For example, the diffraction peak angle on PZT (110) in the film deposited by normal AD method is 30.72°. On the other hand, the diffraction peak angle on PZT (110) in the film deposited by ICP-assisted AD method with the RF power of 800W is 30.98°.

The structure defects such as residual defects and dislocations inside the film influence the peak shift on XRD profile. The dielectric properties of as-deposited PZT film by using normal AD method are low, because the structure defects would be introduced inside the PZT film during the deposition. On the other hand, the PZT film with few structure defects would be fabricated by ICP-assisted AD method. We consider that this method has the possibility of the fabrication of as-deposited PZT film with high dielectric properties.

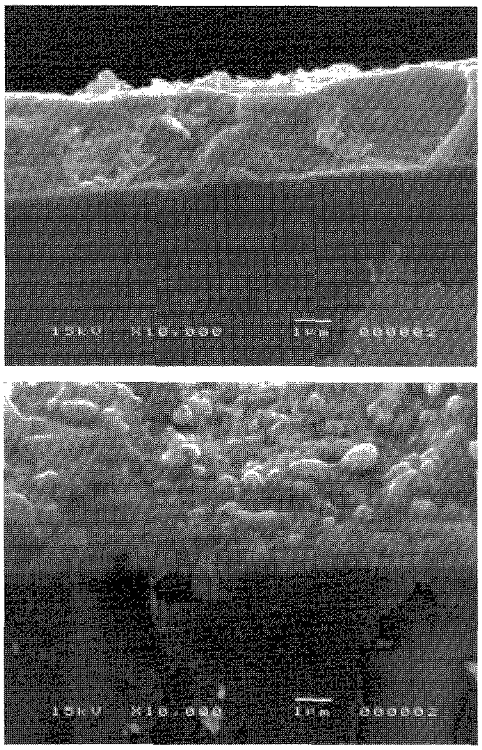

Fig.5 Cross-sectional microstructure of the PZT film (A) without ICP assist, (B) with ICP assist (800W)

4 CONCLUSIONS

PZT films were fabricated by the ICP-assisted AD method, and the influence of RF power was investigated on the deposition rate and microstructure of the films. The deposition rate with our experimental system was about 1μm/min, while using the AD method without plasma assistance. On the other hand, the deposition rate increased to over 10μm/min when by using the ICP assisted AD method. Additionally, the cross-sectional microstructure of the film was changed by the increasing the RF power. The cross-sectional microstructure was very different in films without plasma assistance and with an RF power of 800W. The observed results indicate a possibility that the deposition mechanism differs between the normal AD method and the ICP-assisted AD method.

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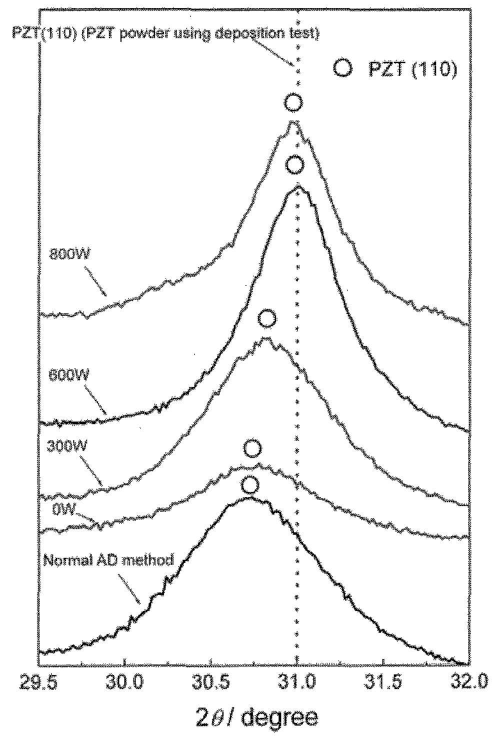

Fig.6 XRD patterns of PZT film fabricated by AD method without ICP assist and with ICP assist

References

[1] A. Wu, M. Vilarinho, M. Isabel, Miranda and J. Baptista, *Journal of the American Ceramic Society*, **83**, 1379-1385(2000).

[2] H. D. Chen, K. R. Udayakumar, C. J. Gaskey, L. E. Cross, J. J. Bemstein and C. Niles, *Journal of the American Ceramic Society*, **79**, 2189(1996).

[3] S .Watanabe and T. Fujii, *Applied Physics Letters*, **66**, 1481(1995).

[4] J. Akedo and M. Lebedev, *Japanese Journal of Applied Physics*, **38**, 5397-5401(1999).

[5] J. Akedo and M. Lebedev, *Applied Physics Letters*, **77**, 1710-1712(2000).

[6] J. Akedo, *Material Science Form*, **449-452**, 43-48, (2004).

[7] J. Akedo, *Journal of the American Ceramic Society*, **89**, 1834-1839(2006).

[8] J. Akedo, M. Lebedev and S. Baba, *Japanese Journal of Applied Physics*, **42**, 5931-5935(2003).

[9] Y. Kawakami, S. Aisawa and J. Akedo, *Japanese Journal of Applied Physics*, **44**, 6934-6937,(2005).

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