3Pa3-2

Proceedings of Symposium on Ultrasonic Electronics, Vol. 33 (2012) pp. 345-346
13-15 November, 2012

Piezoelectric Film Fabrication by Electrospray Deposition
エレクトロスプレーデポジション法による圧電薄膜作製に関する研究

Mihoko Sato[1‡], Namihira Takao[2], and Makiko Kobayashi[3] ([1]Dep. Eng., Kumamoto Univ.; [2]Bioelectrics Research Center, Kumamoto Univ.; [3]Grad. School Sci.& Tech., Kumamoto Univ.)
佐藤美穂子 [1‡], 浪平隆男 [2], 小林牧子 [3] ([1]熊本大　工, [2]熊本大　バイオエレクトリクス研究センター, [3]熊本大院　自然科学研究科)

1. Introduction

Piezoelectric film has been widely used for ultrasonic non-destructive testing due to the simmplicity and cost effectiveness. Sol-gel techiqnue is one of the major thin film fabrication methods because of low instalation cost and mass production possibility. However, it is reralatively difficult to fabricate >1μm by one coating due to high internal stress during thermal process, and this thickness is too low for many industrial non-destructive applications. Sol-gel composite materials have been developed to overcome thickness limitation.[1)] Spray technique could be attrative because of curved surface suitablity, though spray coating process and thermal process must be still repeated in order to achieve desired thickness.[2)]

Electrospray deposition (ESD) could realise desired thickness by one coating process. ESD was developed to fabricate uniform radioactive thin film in 1950s,[3)] then this technique was used to fabricate metal oxide thin film in 1990s.[4)] In this method, a polymer solution is subjected to an electrical field, and when the electrical field reaches a critical value, a charged jet of the solution is ejected. Since it is possible to make nano- or micro- size particles or fibers by this method, it could be possible to release internal stress of sol-gel solution during thermal process and it would result in thcik film fabrication in one caoting process. Therefore, piezoelectric thick film fabrication was attempted by ESD in this contribution.

2. Experimental set-up and procedures

The schematic of experimental setup is shown in **Fig 1**. It consists of a plastic cylinder filled with sol-gel solution, a plastic tube, a metallic nozzle with ~1mm inner diameter, a high voltage source, a substrate, and a metal plate connected with ground. Ejecting speed and the amount were controlled electrically. The sol-gel solution was filled the plastic tube and the metallic nozzle as well, though there was no dripping before electrical charge. Steel substrates with dimensions of 4.2mm thickness, ~50mm length, and ~50mm width were used, since steel is common material for industrial structures.

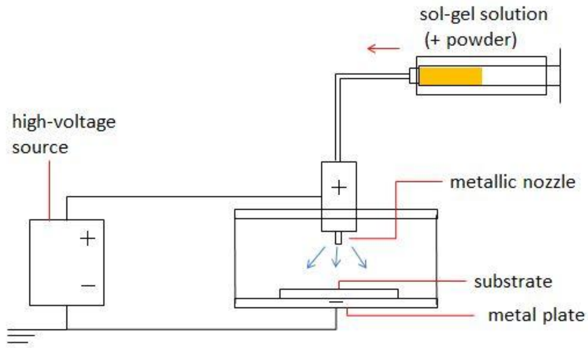

Fig.1 Experimental setup schematic of ESD.

As a sol-gel solution and a piezoelectric powder, lead zirconate titanate (PZT) was chosen, because PZT was one of the most popular piezoelectric materials for ultrasonic NDT applications due to high piezoelectricity, reasonably high Curie point for up to 100°C, and low cost. After ESD, the following process was simmilar to sol-gel process, i.e., drying process at 150°C by a hot plate, annealing process at 650°C by a furnace, then poling prcess were followed afrer ESD.

3. Results and Discussions

First, pure PZT sol-gel solution was filled into the plastic cylinder and ~15kV high voltage was applied. Thin film was successfully fabricated onto a steel substrate. An optical image of the sample after annealing process was shown in **Fig. 2**. Film thickness of PZT film was measured by a micrometer and it was ~1μm. It is noted that the film was very smooth and had good adhesion, thus ~1μm thick film was obtained by pure sol-gel by one coating, and thickness could be even higher. However, it seems that the film thickness is not uniform. Further investigation is required to improve film uniformity. After poling, ultrasonic performance was checked, but no signal was confirmed due to too high frequency.

----
kobayashi@cs.kumamoto-u.ac.jp

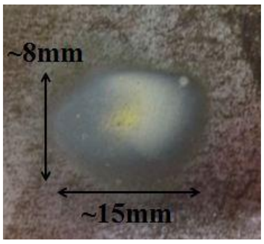

Fig.2 Optical image of PZT film made by pure PZT sol-gel ESD.

Then, the mixture of PZT sol-gel solution and PZT powders were filled into the plastic cylinder and ~15kV high voltage was applied again. The sol-gel composite film was successfully deposited onto a steel substrate. An optical image of the sample after annealing, poling, and top electrode fabrication process was shown in **Fig. 3**. Film thickness of PZT/PZT film was measured by a micrometer and it was ~30-40μm. It means that thickness limitation was successfully enlarged.

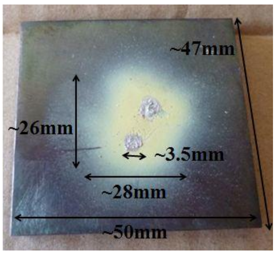

Fig.3 Optical image of PZT film made by pure PZT sol-gel ESD.

Microstructure of sol-gel composite was observed by Scanning Electron Microscope (SEM) and it was shown in **Fig. 4**. It is very similar to sol-gel composite made by air-spray technique. It is hard to distinguish sol-gel origin and powder origin.

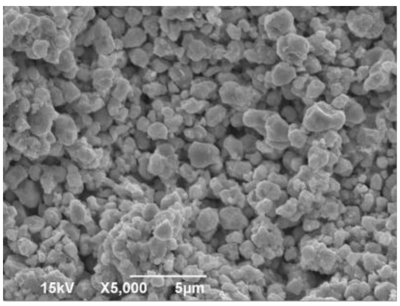

Fig.4 SEM image of PZT/PZT film made by ESD.

Ultrasonic response of PZT/PZT made by air-spray and ESD were shown in **Fig. 5** and **Fig. 6**, respectively. Clear reflected echoes from bottom surface of the substrates were confirmed and even ESD samples showed broadband characteristic and high SNR. The ESD sample seems to have higher frequency component than air-spray, and it resulted from lower film thickness and high porosity. Currently signal strength of ESD sample was much lower (by 20-30dB) than air-spray sample. Further investigation is required to improve signal strength.

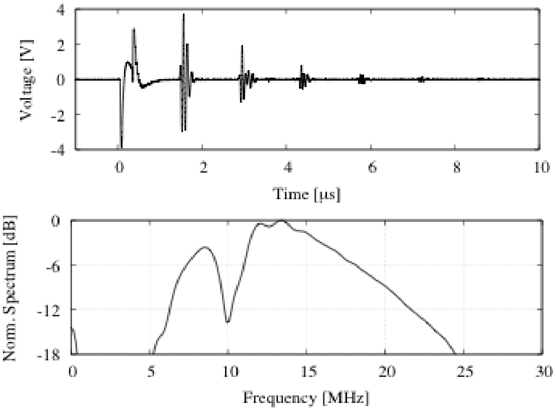

Fig.5 Typical ultrasonic response of PZT/PZT by air-spray technique: upper) in time domain, bottom) in frequency domain.

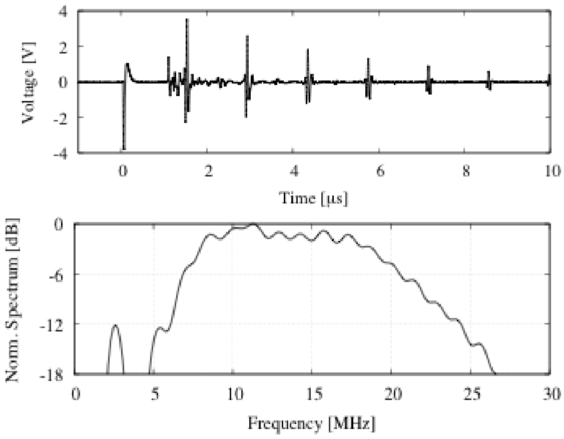

Fig.6 Typical ultrasonic response of PZT/PZT by ESD: upper) in time domain, bottom) in frequency domain.

4. Conclusions

Pure sol-gel and sol-gel composite films were successfully fabricated by ESD, and ~1μm and ~30-40μm thick films were instantly achieved without crack. SEM image of sol-gel composite made by SEM was very similar to the one made by air-spray. Ultrasonic response was confirmed from sol-gel composite film made by ESD.

References

1. D.A. Barrow, T.E. Petroff, R.P. Tandon and M. Sayer: J. Appl. Phys. **81** (1997) 876.
2. M. Kobayashi, C.-K. Jen, Y. Ono, K.-T. Wu and I. Shih: Jpn. J. Appl. Phys. **46** (2007) 4688.
3. D. J. Carswell, J. Milsted: J. Nucl. Ener. **4** (1954) 51.
4. C. Chen, E. M. Kelder, P. J. J. M. van der Put and J. Schoonman: J. Mater. Chem. **6** (1996) 765.