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Hybrid bilayer structure $PbTiO_3$/PVDF-TrFE prepared by spin coating method for capacitor applications

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ABSTRACT – Performance of high capacitance density device is the requirement to fabricate the films based capacitor. Thus, the need to have high dielectric ceramic material is quite major factor instead conventional metal oxide material. This study presents the preparation of nano-films lead titanate in various annealing temperature for the fabrication of bilayer structure lead titanate/poly(vinylidene fluoride) trifluoroethylene material combination.

1. INTRODUCTION

Development of high performance capacitor is reaching towards new generation where the ferroelectric ceramic material take places as the active dielectric layer. The motivation of this study is to produce high capacitance device that can stand the nanoscale films thickness. Variation in annealing treatment give significant impact on ceramic thin films preparation [1–3]. However, the ceramic films like lead titanate (PbTiO3) are well known by its brittle structure where the part of loss dissipation getting very high compared to the polymer films. Thus, the combination of PbTiO3 and poly(vinylidene fluoride) trifluoroethylene (PVDF-TrFE) where both poses ferroelectric behavioral becomes the promising material for layered films capacitor.

The fabrication was done by simple and cost effective sol-gel spin coating method that being varied at spinning speed property. Sol-gel spin coating method is the most preferable method to be used in fabricating such nanoscaled thin films device. This is due to the fact that homogeneous multilayered films can be achieved that give effect on reduced leakage current density. Furthermore, the method is in line with industrial mass production complimentary with large and complex area covering system yet effective manufacturing cost indeed.

2. METHODOLOGY

2.1 $PbTiO_3$ Layer

The starting materials, lead acetate trihydrate [PbAc; 99% purity], titanium isoproproxide [TTIP; 97% purity], and solvent 2-methoxyethanol. The solutions were prepared at 0.4mol.L^{-1} with presence of glacial acetic acid (GAA) as a stabilizer and with additional of 10wt% of PbAc powder. The deposition process was maintained at 1500 rpm spin speed for 30 seconds. The prepared films were dried at 150˚C and then involved various annealing temperature (400 to 600˚C).

2.2 PVDF-TrFE Layer

The PVDF-TrFE [70:30mol% composite] powders were dissolved in methyl ethyl ketone (MEK) solvent at concentration of 30g.L^{-1}. The spinning process involved at 3000 rpm spin speed for 90 seconds. The samples were annealed at 120˚C about an hour then cooled at 50˚C for 2 hours. The bilayer structure samples were characterized for dielectric and ferroelectric property with structural observation by atomic force microscopy.

3. RESULTS AND DISCUSSION

The dielectric measurement based on metal-insulator-metal configuration operated at 0.5mVac signals is as shown in Figure 1. Particularly, the measurement of dielectric films at low annealing temperature concerned on 1 kHz operating frequency where crucially is made for low voltage application. This is the substantial property owing to the polarization occurred at interfacial layer of dielectric-metal layer [4].

Dielectric permittivity of entire films at increase annealing temperature showed exponential increase. Thus, it is predicted that by increasing the temperature more than 600˚C, the result may yield to the saturation region. Therefore, a film prepared by 500˚C annealing temperature is presumed as the transition prior to the saturation value and the dielectric value is about 150, the highest obtained in this study. On the other hand, such crystalline substrate with platinum coated and particle seeded as reported in previous study ensured better property of microstructural $PbTiO_3$ films as well as increase of dielectric permittivity [5–7]. Inverse observation showed tangent loss for entire films where it behaves polynomial curvature and stated the optimum sample (anneal 500˚C) obtained minimum tangent loss about 0.024.

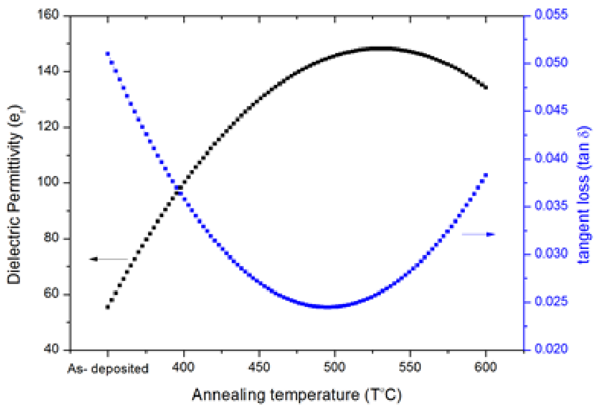

Figure 1 Plot of dielectric permittivity and tangent loss of $PbTiO_3$ films prepared with variation of annealing temperature.

Capacitance value of each sample is calculated using Equation (1). Where εr and εo is dielectric and air permittivity respectively, A is the area of plate-to-plate metal contact, and d presents the dielectric films thickness. Thus, the capacitance value of optimum sample prepared by 500˚C annealing is about 90.96 nF.

$$C = \frac{\varepsilon_r \varepsilon_o A}{d} \quad (1)$$

Table 1 Dielectric property of $PbTiO_3$ films prepared with different annealing temperature.

Samples (T˚C)	**Dielectric permittivity (ε_r)**	**Tangent loss (tan (δ))**
As-deposited	50	0.051
400	112	0.036
450	122	0.027
500	150	0.024
550	140	0.029
600	138	0.038

4. CONCLUSIONS

$PbTiO_3$ films were successfully prepared by simple spin coating method at different annealing temperature. This study emphasizes the optimum $PbTiO_3$ films with 500˚C annealing treatment managed to obtain high dielectric constant and adequate low tangent loss as well. In future discussion, this deposition parameter will be then implied for the fabrication of bilayer structured $PbTiO_3$/PVDF-TrFE for MIM capacitor device.

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