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Electrical Properties of PLZT Thin Films Prepared by a Sol-Gel Method

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The electric properties of sol-gel derived PLZT thin films was investigated. With less than 10 mol% lanthanum contents, this film obtains the single phase of perovskite structure and its dielectric constant is more than 870. The leakage current of PLZT thin film are one order of magnitude less than that of PZT and slightly larger than that of acceptable value for 64(M)DRAM. The equivalent SiO_2 thickness of PLZT decrease with film thickness and 0.67nm with 100nm thickness.

1. Introduction

Recently, the bit density of DRAM's is increasing four times per three years and the capacitor area becomes smaller. However, keeping off the soft-error, the storage capacitors maintain a definite capacity. As the result, we need the materials with high dielectric constants for retaining the capacity. From this viewpoint, ferroelectric materials with extreme high dielectric constants are expected to apply to the insulator of DRAM capacitor.

PLZT is a well known ferroelectric material with high dielectric constant and many studies of the electric properties of bulk PLZT were reported in the literature[1-4]. However, a few attempt have been reported to evaluate the electric properties of PLZT thin films[5-7]. In this study, we report the crystal structure and the electric properties of PLZT thin films prepared by a sol-gel method.

2. Experimentals

$Pb_{(1-x)}La_xZr_{0.52}Ti_{0.48}O_3$ films with various lanthanum contents have been fabricated by a sol-gel method. The precursor chemicals we used were lead acetate [$Pb(CH_3COO)_2\cdot 3H_2O$], lanthanum acetate [$La(CH_3COO)_2\cdot 1.5H_2O$], zirconium tetra-n-buthoxide [$Zr(OC_4H_9)_4$] and titanium tetra-i-propoxide [$Ti(OC_3H_7)_4$].

The precursor solution was spin coated onto platinum coated silicon wafers. In order to achieve the required thickness, several layers were applied and a thermal bake cycle was introduced after each spin-on coating. A final furnace anneal at 650-700°C in air was performed in order to obtain the perovskite structure. Top electrodes of platinum were then deposited with metal mask. The area of electrodes were 0.01cm^2.

3. Results

The crystalline properties of PLZT depend upon lanthanum concentration. Fig.1 shows the X-ray diffraction patterns of the PLZT thin films with various lanthanum concentrations. With less than 10 mol% of the lanthanum content, the PLZT films show the single phase of the perovskite structure. With more than 10 mol% of it, they show mixture of two phases of the perovskite and the pyrochlore structure.

Fig.2 shows the relationship between the lanthanum concentration and the relative dielectric constant of 300nm thick PLZT films. In a PZT without lanthanum, the relative dielectric constant and the equivalent SiO_2 thick-

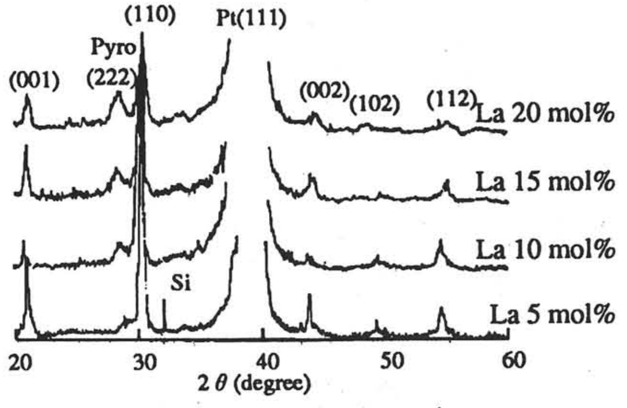

Fig.1 The X-ray diffraction pattern of PLZT films with various lanthanum concentration.

ness are 2300 and 0.67nm, respectively. This relative dielectric constant is larger than that of the bulk values of 800[1]. Increasing the lanthanum concentration, the relative dielectric constant was decrease. With less than 10 mol% lanthanum contents, i.e. single phase of the perovskite structure, the relative dielectric constant of 300nm thick PLZT films are larger than 1040.

Fig.3 shows the dependence of the leakage current on the lanthanum concentration which is measured at the bias condition of 1.65V. For 0 mol% lanthanum contents (i.e. PZT), the leakage current is $\approx 7\mu A/cm^2$. However, for PLZT films which contain more than 3 mol% lanthanum, the leakage current are one order of magnitude less than that of PZT. This result is brought by adding lanthanum which compensates the enhanced conductivity arising from oxygen and lead vacancies [1]. For the DRAM application, it needs small leakage current to retain the electric charge in definite time. For example the capacitor insulator of 64(M)DRAM is required the leakage current less than $0.1\mu A/cm^2$. The leakage current of PLZT films are slightly larger than this required value.

Fig.4 shows the X-ray diffraction patterns of PLZT thin films with various film thickness which contain 5 mol% lanthanum. Though the X-ray diffraction intensity of perovskite structure increases with film thickness, the peaks of that structure do not disappear at the

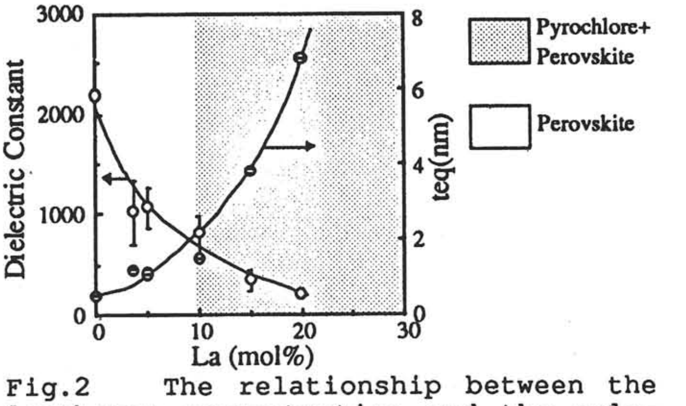

Fig.2 The relationship between the lanthanum concentration and the relative dielectric constant of 300nm thick

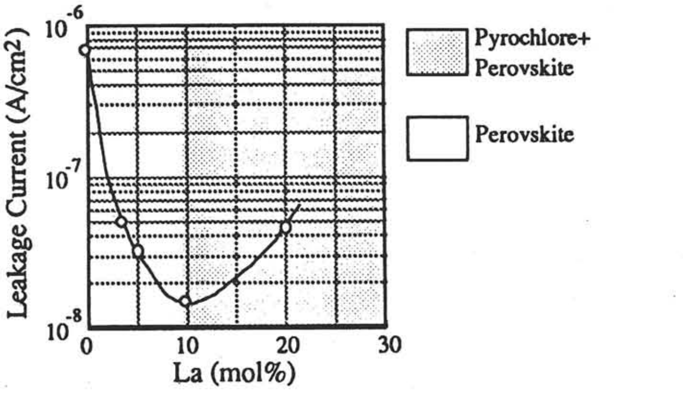

Fig.3 The dependence of the leakage current of 300nm thick PLZT film on the lanthanum concentration which is measured at the bias condition of 1.65V.

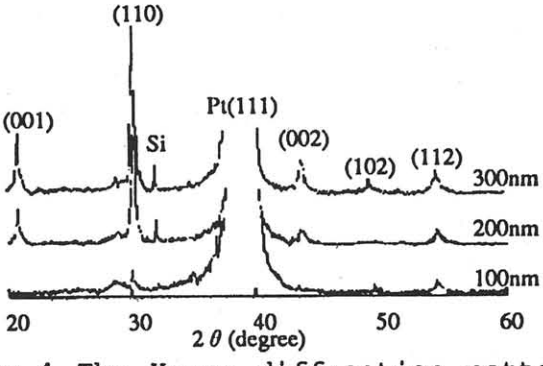

Fig.4 The X-ray diffraction pattern of PLZT films with various film thickness which content 5 mol% lanthanum.

100nm film thickness. Fig.5 gives the relative dielectric constant versus thickness of PLZT films having 5 mol% lanthanum content. The relative dielectric constant increases with the thickness and has the tendency to flatten out at greater thicknesses. In the case of PZT films, the equivalent SiO_2 thickness is nearly constant with the range of less than 300nm film thickness [8]. However, the equivalent SiO_2 thickness of PLZT film decrease with the film thickness. Therefore PLZT is more suitable to apply to DRAM capacitor than PZT. Fig.6 shows the dependence of the leakage current on the film thickness which is measured at the bias condition of 1.65V. The leakage current of 100nm thick PLZT film is 2μA/cm^2 and this value smaller than that of 300nm PZT films.

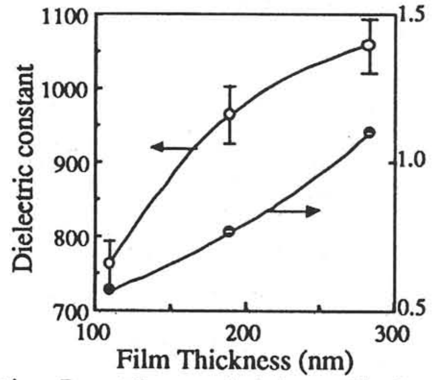

Fig.5 The relative dielectric constant versus thickness of PLZT films having lanthanum 5 mol% contents.

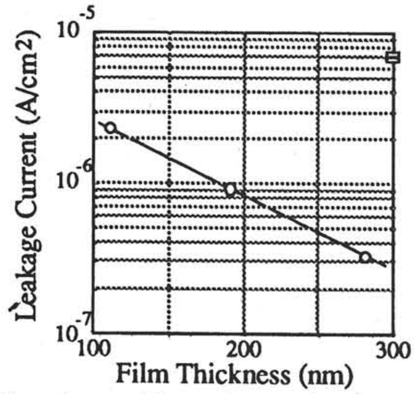

Fig.6 The dependence of the leakage current on the film thickness measured at the bias condition of 1.65V; O :the PLZT films having lanthanum 5 mol%, ▤ :the PZT film having no lanthanum.

Fig.7 shows the time dependent dielectric breakdown of PLZT film under the various stress conditions. The films are 5 mol% lanthanum content, and relative dielectric constant and equivalent SiO_2 thickness are 870 and 1.35nm, respectively. The cumulative failure percent of PLZT thin film show the Weibull distribution and the gradient of Weibull distribution of life time versus log t_{BD} increase with the stress voltage. Because of this change of the gradient, further improvement of defect properties of PLZT film is required to determine the acceleration factor of electric field.

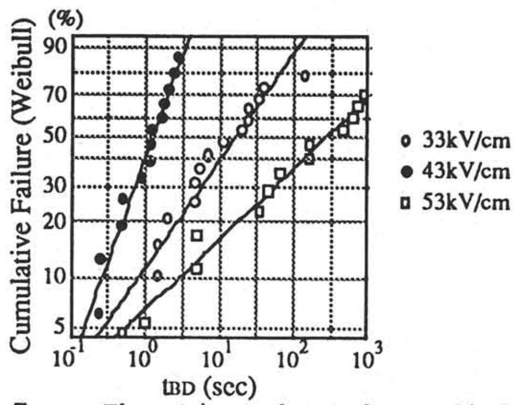

Fig.7 The time dependent dielectric beakdown of PLZT film having lanthanum 5 mol% content under the various stress conditions.

4. Conclusion

The electric properties of sol-gel derived PLZT thin films was investigated. With less than 10 mol% lanthanum contents, it obtains the single phase of perovskite structure and its dielectric constant is more than 870. The leakage current of PLZT thin film are one order of magnitude less than that of PZT and slightly larger than acceptable value for 64(M)DRAM. The equivalent SiO_2 thickness of PLZT film decrease with film thickness and become 0.67nm with 100nm thickness. The time dependent dielectric breakdown properties of PLZT film depend upon the stress voltage and acceleration factor has not yet been determined.

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