Lead Zirconate Titanate (PZT) Thin Film Deposition in Facing Target Sputtering¹

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The deposition rate and crystallographic structure of PZT thin film deposited by facing target sputtering have been studied experimentally in this paper. The deposition rate increases with increase of sputtering power or with decrease of substrate temperature, and it has a minimum value when the thin film is deposited at the total gas pressure from 0.05 to 1.2 Pa. The perovskite phase and pyrochlore phase exist in all as-deposited samples of PZT thin films, and the relative content of various phases is varied with sputtering conditions.

1. Introduction

Lead zirconate titanate (PZT) thin films have a large amount of potential applications such as memory device, electro-optical device, sensor and actuator etc. One of the most important is used as ultrasonic transducers in medical, thick fog and under-water acoustic imaging systems. It is important that perovskite PZT thin film with excellent electric properties can be integrated onto a silicon chip with an on-board circuitry at low substrate temperature at which the circuitry can not be damaged. A variety of thin film preparation techniques such as sputtering, laser ablation, chemical vapor deposition (CVD) and sol-gel method have been used for fabrication of PZT thin films¹⁻⁵⁾. Among the sputtering processes, facing target sputtering technique is the unique one, which can be used for depositing almost all kinds of materials at a high deposition rate at low substrate temperature. It has been reported that the as-deposited thin film has good uniformity in a large area and excellent crystallographic structure with the same composition as that of the target because the substrate is outside the plasma and not attacked by the high energy particles, and that the results can be repeated well⁶⁻⁸⁾. It is very important that the sputtering technique with RF discharge is suitable for deposition of dielectric thin films under stable condition. However,

it has not yet been reported that PZT thin film had been deposited by this method. For PZT ceramics, RF discharge was very stable when RF power was changed from 400 to 800 W in the range of total gas pressure between 0.05 and 1.2 Pa in the present work. And the perovskite phase has been obtained in the as-deposited PZT thin film at such low substrate temperature as 200°C by means of facing target sputtering technique.

2. Experimental Procedure

The instrument used for sputtering was FTS-1CB system (Osaka Vacuum Co.). The distance between the two targets was fixed at 140 mm, and the distance from the surface of substrate to the central line of the two targets was changed from 125 mm to 160 mm. Two heat-pressing sintered ceramic plates ($\text{Pb}_{1.2}\text{Zr}_{0.58}\text{Ti}_{0.42}\text{O}_x$) with diameter of 100 mm and 5 mm thick were used as targets. PZT thin films were deposited on substrate Pt/Ti/SiO₂/Si, in which Pt layer was (111) oriented, and the thickness of Pt, Ti and SiO₂ layer was 900 nm, 50 nm and 600 nm, respectively. Table 1 showed the sputtering conditions in this work. The deposition rate was defined by testing the thickness of film in the sputtering time. The crystallographic structures of PZT thin film samples were determined by X-ray diffraction (XRD) analysis with Cu K α radiation of 50 kV and 150 mA (Rigaku 2500).

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Table 1 Sputtering conditions of PZT films

Input RF power (W)	400~800
Sputtering duration (min.)	120
Total gas pressure (Pa)	0.05~1.2
Sputtering gas: O ₂ /Ar	4:1
Substrate temperature (°C)	160~550
Substrate structure	Pt/Ti/SiO ₂ /Si

Fig. 1 Deposition rate of PZT films sputtered at 360°C, 120 min, different pressure.

3. Results and Discussion

3.1 Deposition rate

Fig. 1 shows the dependence of deposition rate on sputtering total gas pressure. It is found that there is a minimum value in the deposition rates of PZT thin films at 0.2 Pa when the thin films are deposited in the pressure range between 0.05 Pa and 1.2 Pa. And then the deposition rate increases slowly with increase of total gas pressure. Here all curves of the deposition rate under different sputtering power are similar to each other. It is easy to find that the deposition rate increases with increase of sputtering power. But when total gas pressure is lower than 0.2 Pa, the deposition rate under RF700W is higher than that under RF800W.

It is found from **Fig. 2** that with increase of substrate temperature, the deposition rate of PZT thin films decreases relatively. And especially when the thin film is deposited at low total gas pressure such as 0.05 Pa, it decreases quickly, because the re-evaporation rate of sputtered particles increases quickly with increase of substrate temperature in the case of low total gas pressure.

In addition, the deposition rate of PZT thin film

Fig. 2 Deposition rate of samples sputtered at: 700 W, various substrate temperature. **Fig. 3** XRD patterns of PZT thin films deposited at different temperature.

increases with shortening the distance between the surface of substrate and the central line of the two targets.

3.2 Crystallographic structure

Fig. 3 shows the XRD patterns of PZT thin films deposited at 0.8 Pa, RF600W and different substrate temperature. It is found that there are perovskite phase, pyrochlore phase, and lead or titanium oxide phases in all of the as-deposited samples, and the relative contents of various phases are varied with deposition temperature. When the substrate temperature increases from 160°C to 437°C, the peaks of (110) planar PbO₂ and (222) planar pyrochlore phase increase too; and when the substrate is heated to 550°C, the peak of PbO₂ disappears and the peak of (222) planar pyrochlore phase becomes very weak, while the peak of (100)

Fig. 4 XRD patterns of PZT thin films deposited at RF700W, 360°C, various total gas pressure.

Fig. 5 The relative contents of the phases in PZT films deposited at 360°C, FR700W, 120 min., different total gas pressure.

planar perovskite phase appears, and the peak of (400) planar pyrochlore phase becomes very strong. And the peak of (111) planar perovskite phase becomes strong and sharp too. It is very important that the perovskite phase can be obtained in the PZT thin film deposited at low substrate temperature as 200°C in this work.

Fig. 4 shows the XRD patterns of PZT thin films deposited at different total gas pressure. It is found that with decrease of total gas pressure, the peaks of (110) planar PbO₂ phase and (222) planar pyrochlore phase in the XRD patterns decrease obviously, and meanwhile the peaks of (110) and (111) planar PZT perovskite phase increase relatively. It indicates that there are more lead oxide reacted and more perovskite phase formed when the total gas pressure decreases, which means that

Fig. 6 XRD patterns of PZT thin films deposited at 360°C, 120 min., 0.5 Pa, different sputtering power.

lower total gas pressure is advantageous to formation of perovskite phase of PZT thin films in this case.

Fig. 5 summarizes the total gas pressure dependence of relative contents of different phases in the as-deposited PZT thin films. It is shown clearly that the relative contents of lead oxide and perovskite phase depend strongly on the total gas pressure at which the PZT thin films are deposited. With decrease of total gas pressure, the relative content of (111) oriented perovskite phase increases obviously. And there is a maximum value in the relative content of (110) oriented perovskite phase when the sample is deposited at 0.2 Pa of total gas pressure. So perovskite phase is the main phase in PZT thin film deposited at low total gas pressure.

Fig. 6 shows the XRD patterns of PZT thin films deposited under different sputtering power. It is found that with decrease of sputtering power, the peak of (111) planar perovskite phase increase obviously, and the peaks of (110) planar PbO₂ phase and (222) planar pyrochlore phase decreases relatively. It indicates that lower sputtering power is advantageous to formation of (111) oriented perovskite phase of PZT thin film.

It has been reported that the (111) orientation of PZT perovskite phase may be attributed to the intermetallic compound (Pt₃Ti) formed on Pt/Ti/SiO₂/Si substrate, because the lattice parameter of Pt₃Ti matched closely that of (111) PZT and the interfacial energy was quite low⁹. The lattice matching may give nucleation sites for PZT thin films to grow preferentially along the (111) direction at low temperature. However, the correlation among the rates of deposition, re-evaporation and migration

is very important for growth of (111) PZT thin film during sputtering process⁹⁾. When deposition rate is low, the migration is enhanced, so the (111) PZT perovskite phase is formed preferentially in the case of low total gas pressure or low sputtering power.

4. Conclusions

The deposition rate of PZT thin films increases with increase of sputtering power or with decrease of substrate temperature, and it has a minimum value at 0.2 Pa when the thin films are deposited at the total gas pressure from 0.05 to 1.2 Pa.

The perovskite phase can be obtained at low substrate temperature such as 200°C. Low total gas pressure, or low sputtering power is advantageous to formation of (111) PZT perovskite phase in our sputtering condition.

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