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Characteristics of sol–gel derived PZT thin films with lead oxide cover layers and lead titanate interlayers

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The sol–gel derived $PbZr_{0.53}Ti_{0.47}O_3$ (PZT) films were fabricated on the bare $Pt/Ti/SiO_2/Si$ substrates or the same substrates coated by the $PbTiO_3$ (PT) interlayers. The post-deposition annealing temperature and time were optimized when the PbO cover layers and PbO vapour-containing atmosphere were compared with each other and adopted as the method to diminish the lead-loss problem during the high-temperature post-deposition annealing. The X-ray diffraction patterns, microstructures, and electrical properties such as relative permittivity, ε_r, remanent polarization, P_r, and coercive electrical field, E_c, were investigated in relation to the annealing conditions. The PZT films deposited on the bare $Pt/Ti/SiO_2/Si$ substrates under the PbO vapour-containing atmosphere showed better electrical properties. This indicates that the PbO vapour-containing atmosphere may be the better method of lead-loss-prevention to process the lead-containing films rather than the PbO cover layer method. The electrical characteristics of the PZT films, $\varepsilon_r = 1150$, a dissipation factor of 0.039, $P_r = 26\ \mu C\ cm^{-2}$, and $E_c = 40.5\ kV\ cm^{-1}$, were measured at 1 kHz. When PZT films were deposited on substrates coated by the PT layers, PZT-PT films with single perovskite phase were derived by post-deposition annealing at 500 °C for 1 h. However, the relative electrical properties are very poor, i.e. $E_r = 160$, $P_r = 2.0\ \mu C\ cm^{-2}$ and $E_c = 75\ kV\ cm^{-1}$. The optimum combination for preparing PZT–PT films is a 40 nm $PbTiO_3$ interlayer and annealing conditions of 6 h at 550 °C in a PbO vapour-containing atmosphere; the derived films exhibit electrical properties of $E_r = 885$, $P_r = 21.5\ \mu C\ cm^{-2}$ and $E_c = 64\ kV\ cm^{-1}$. The combination of inserting a PT interlayer and annealing in a PbO vapour-containing atmosphere can prevent the formation of electrical short paths. In this case, nearly pin-hole-free PZT films can be grown on the PT (interlayer) $/Pt/Ti/SiO_2/Si$ substrates. It is believed that it is possible to prepare the PZT films with nano-scale uniformity, reproducible quality, which may be worth considering for commercial applications.

1. Introduction

Lead zirconate titanate (PZT) is known to be a promising material for integrated memory, optical and microelectromechanical devices such as acoustic pressure sensors, micro-positioning actuators, piezoelectric micromotors, piezoelectric force sensors, and pyroelectric infrared detectors, etc., because of its excellent ferroelectric, optical, piezoelectric, and pyroelectric properties [1–7]. The recent progress in thin-film deposition technology has made it possible to deposit a homogeneous PZT thin film on silicon-based substrates, and also realized the possibility of use for integrated devices. Among the many silicon semiconductor compatible thin-film deposition processes, the sol–gel process provides an inexpensive and convenient route to prepare PZT thin films on diverse substrates. All the starting chemicals are mixed on the molecular level in the solution, so that the composition of the multicomponents system is easily controlled and a high degree of homogeneity of the film can be realized. However, the average percentage of electrical short circuits of the capacitors, formed between the bottom platinum electrode and the array of individual gold top electrodes, of PZT thin films derived from the sol–gel process, is slightly higher than that from other processing methods. This could be a barrier to sol–gel derived PZT integrated devices. How to diminish this problem is a challenging issue when the commercialization of these devices is considered.

The PZT 53/47 (Zr/Ti ratio) composition, which is near the centre of the morphotropic phase boundary (MPB, from Zr/Ti ratios of 40/60 to 65/35) region and separates the rhombohedral and tetragonal phases, possesses excellent dielectric, piezoelectric and

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ferroelectric properties. In the sol–gel process, the formation temperature of the perovskite phase of PZT 53/47 thin films has been reported to be higher than 600 °C [8]. However, the necessary high-temperature post-deposition annealing may cause broadened electrical properties of the PZT film owing to lead loss by evaporation in local areas of the film surface during high-temperature annealing. Moreover, the locally lead-deficient phase may offer a path for electrical short circuits of the PZT film capacitor. Some ways have been proposed to reduce this problem. PbO coating on sol–gel derived PLZT thin films has recently been studied by Tani and Payne [9]. The dielectric constants of PbO-coated PLZT films were reported to be twice as large as those of uncoated films, and only the coated films showed the well-defined ferroelectric hysteresis loop. This seems to be one approach to solve the lead-loss problem and even a way to promote the electrical properties. Tani and Payne also pointed out that PbO is capable of diffusing into the nearby substrate because the consumption rate of the PbO coating layer depends on the kind of electrodes and substrates. The PbO(*s*) of the coating layer may be reduced to Pb(*l*) in the area because of the locally low oxygen partial pressure. The diffusion of Pb(1) atoms into PLZT film then becomes possible. It has been reported that the reactions of lead with the platinum electrodes or titanium adhesion layer are common in the integrated structures [10, 11]. The effect of the PbO cover layer on PZT thin films has also been studied by same group. A similar trend in the influence of the PbO cover layer on the electrical properties has been reported [12]. On the other hand, using PbO vapour-containing atmospheres in the post-deposition annealing process of PZT thin films is a potential solution to preparing thin films of high quality and performance. This technique is easily utilized in processing lead-containing bulk ceramics via the conventional mixing-oxides approach [13] and may offer the same merits as the PbO cover layer method. This has the advantage that problems with the possible diffusion of PbO cover layer do not occur in this case. To the best of our knowledge, the effect of the PbO vapour-containing annealing atmosphere on the preparation of PZT thin films has not yet been reported. Consequently, the effects of the PbO cover layer, and a PbO vapour-containing atmosphere were compared in relation to the annealing conditions of PZT 53/47 thin films in this investigation.

A shorter annealing time is another way to reduce lead loss, i.e. rapid thermal processing [14]. Because of the shorter high-temperature treatment period, the possibility of lead loss is reduced. A lower annealing temperature is also a way by inserting an interlayer into the substrate and sol–gel derived PZT film [8]. The presence of a $PbTiO_3$ interlayer has been found to be effective in reducing the heat-treatment temperature in the preparation of PZT 53/47 thin film with single perovskite phase from 600 °C down to 500 °C. The PZT perovskite phase formation process is a nucleation-controlled process [15–17] rather than a diffusion-controlled process and the $PbTiO_3$ interlayer is able to offer plentiful nucleation sites and decrease the nucleation activation energy as well, that is, it behaves as a seeding layer. The equilibrium vapour pressure of PbO(*g*) over PbO(*s*) is reduced by two orders as the temperature falls from 600 °C down to 500 °C [18]. This may repress the lead loss to the atmosphere during the post-deposition annealing when the formation temperature of PZT films with single perovskite phase is decreased via the insertion of a $PbTiO_3$ interlayer. The possibility of forming local electrical short circuits can be reduced because the lead loss is suppressed. Although many studies of interlayer effects have been reported, they all basically emphasized the use of thin films for DRAM applications (i.e. with about 0.3 μm or less thickness). When the PZT film is considered as a material for microelectromechanical devices, a slightly thicker film, i.e. about 1 μm, may be necessary. From these points of view, the present work studied the effect of $PbTiO_3$ interlayer on the PZT film for microelectromechanical applications.

The influence of annealing atmosphere (i.e. P_{O_2}) on perovskite crystallization from an amorphous phase has been reported. The oxygen concentration in the amorphous films prior to annealing has been shown to be one of the most critical parameters when films of reproducible texture and microstructure are prepared by the sol–gel process with rapid thermal annealing [19]. The oxygen content of as-sputtered PLZT films was found to have a profound effect on the transformation to perovskite during subsequent annealing [20]. To avoid the effects of P_{O_2} on the perovskite phase formation, in the present study, high-temperature firing was carried out for every layer of film, instead of low-temperature pyrolysis, in order to form perovskite phase for each deposition. This may simplify the situation with regard to the annealing atmosphere, because emphasis may be placed on comparing the post-deposition annealing effect on PZT films of the PbO cover layer method and the PbO vapour-containing atmosphere.

In the present study, an attempt was made to compare the benefits of the PbO cover layer method with the PbO vapour-containing atmosphere, in the optimization of annealing conditions for PZT 53/47 films. As a result, annealing temperature and time were varied to achieve the best electrical properties. The influence of the $PbTiO_3$ interlayer on PZT 53/47 films were also studied. It was found that the PbO vapour-containing annealing atmosphere method is an effective approach to prepare lead-containing films of high quality. Nearly pin-hole-free PZT films can be prepared on the $PbTiO_3$ interlayer coated substrates and the $PbTiO_3$ interlayer may contribute to the uniformity of the electrical properties of PZT films. It is believed it may be possible to prepare PZT films with nanoscale uniformity and of reproducible quality, which are worth considering for commercial microelectromechanical applications.

2. Experimental procedure

2.1. Precursors solution

The sol–gel precursors solution of $PbZr_{0.53}Ti_{0.47}O_3$ (0.4 M) and $PbTiO_3$(0.15 M) were prepared from lead acetate, $Pb(CH_3COO)_2$, titanium isopropoxide,

$Ti[(CH_3)_2CHO]_4$, and zirconium *n*-butoxide, $Zr(C_4H_9O)_4$, dissolved in glacial acetic acid, 2-propanol, and 1-butanol. Lead acetate was dissolved in glacial acetic acid; zirconium *n*-butoxide and then titanium isopropoxide were added to the lead acetate solution. During solution preparation, 20 mol% extra $Pb(CH_3COO)_2$ was added. This excess amount of lead was used to compensate for lead loss during the 600 °C firing. Many studies report that this may not only enhance the crystallinity, and promote the perovskite phase percentage, but also increase the relative permittivity [21, 22]. These solutions were hydrolysed with an appropriate amount of water when the ethylene glycol was added as cross-linking agent; this may reduce the possibility of cracking of the gel films during the drying process. The solution was then further diluted with 2-propanol, 1-butanol and acetic acid to form the solution used for deposition. The details of the precursor solution preparation are similar to the process suggested by Yi and Sayer [23]. For the PbO cover layer, the 0.7 M PbO precursor solution was prepared by dissolving lead acetate in the glacial acetic acid.

2.2. PbO cover layers and PbO vapour-containing atmosphere for the annealing process

As shown in Fig. 1, solution of 0.4 M PZT was deposited on to $Pt/Ti/SiO_2/Si$ wafers by spin-coating at 4000 r.p.m. for 20 s. After deposition, samples were dried on a hot plate at 110 °C for 5 min, and then samples were heat treated directly at 600 °C for 20 min after each layer was deposited. Because the perovskite phase was expected to form directly from the amorphous phase, the pyrochlore phase formation was eliminated completely. The final PZT films consisted of eight layers and were approximately 1.2 μm thick. In the post-deposition annealing, the samples were either placed into a crucible containing PbO powder, or coated with a PbO cover layer, then placed directly into a preheated box furnace at 600 °C for *x* h ($x = 0, 2, 4$ or 6). The PZT film samples with a PbO cover layer were denoted P-PZT-*x*, while samples of PZT films obtained from the PbO vapour-containing atmosphere annealing for *x* h, were denoted as PZT-*x*.)

2.3. $PbTiO_3$ interlayers

Meanwhile, the $PbTiO_3$ interlayers were prepared on the $Pt/Ti/SiO_2/Si$ wafers by spin-coating at 4000 or 6500 r.p.m. for 20 s to produce films 40 and 70 nm thick, respectively, after firing at 500 °C for 20 min. Then the PZT films were deposited on to the surface of the $PbTiO_3$ interlayer, using conditions similar to those of the preceding experiment, Section 2.2. However, as depicted in Fig. 1, the firing temperatures for $PbTiO_3$ and PZT multilayers were 500, 550, and 600 °C. The temperatures for post-deposition annealing were the same as the firing temperatures of the respective samples. The final annealing for $PbTiO_3$-PZT multilayers was only carried out in the PbO vapour-containing atmosphere. The samples were

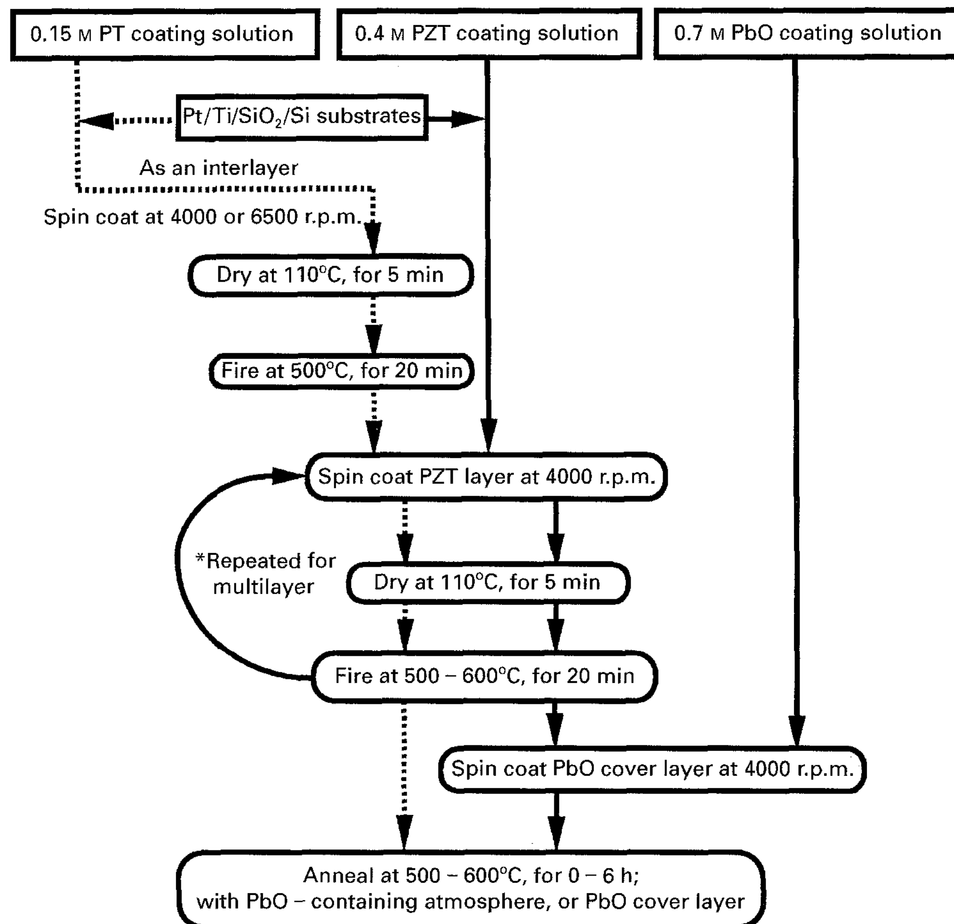

Figure 1 Flow chart of a processing route for sol–gel derived PZT films.

denoted PZT-4PT or PZT-7PT for the samples with 40 or 70 nm thick $PbTiO_3$ interlayers.

2.4. Characterization

Structural characterization of the films was carried out by X-ray diffraction. Film thicknesses were measured using a profilometer after acid etching a portion of the film. Surface microstructures were examined by commercial atomic force microscopy (AFM; Nanoscope III-AFM, Digital Instruments). The top electrodes of Au/Cr layers with 1 mm diameter were deposited on the PZT film surfaces through a shadow mask. Dielectric properties were measured at room temperature using an LCR meter operated at 1 kHz. A Sawyer–Tower circuit was used to characterize the P–E hysteresis loop measured at 1 kHz and 2, 4, or 8 V (V_{p-p}) driving voltages.

3. Results and discussion

3.1. PbO cover layers and PbO vapour-containing atmosphere for the annealing process

The X-ray diffraction (XRD) patterns of PZT and P–PZT samples for different annealing times, i.e. different x values, are shown in Fig. 2. Obviously, the well-developed single perovskite phase was already formed in the sample of PZT-0 (the as-fired PZT film). All the samples show the presence of perovskite phase, but the pattern of the P–PZT-6 sample indicates existence of unknown phases which are neither perovskite nor pyrochlore phase. For the PZT 53/47 films, the pyrochlore phase has very broad major peaks at 2θ of 29.5°, 34.2° and 49.2° [8]. This may possibly be ascribed to the diffusion of the lead atoms into the PZT layer from the PbO cover layer. Further studies are necessary to elucidate this. Histograms are used to illustrate the distribution of relative permittivities for the samples, and the related data are shown in Fig. 3. The relative permittivities for as-fired PZT films, i.e. PZT-0 samples, are 550–700. The maximum relative permittivity for P-PZT-2 was about 850 indicating that the PbO cover would appear to be able to enhance the electrical properties in the case of a relatively shorter annealing time. However, the 4 h annealed samples exhibit the narrowest range of the distribution of all samples studied. Moreover, the PZT-4 sample shows the highest value of relative permittivity, ε_r, which is about 1150. Both types of sample exhibit a broadened distribution range for 6 h annealing, which is perhaps due to the problems of lead diffu sion into and lead loss from P-PZT-6 and PZT-6 samples,

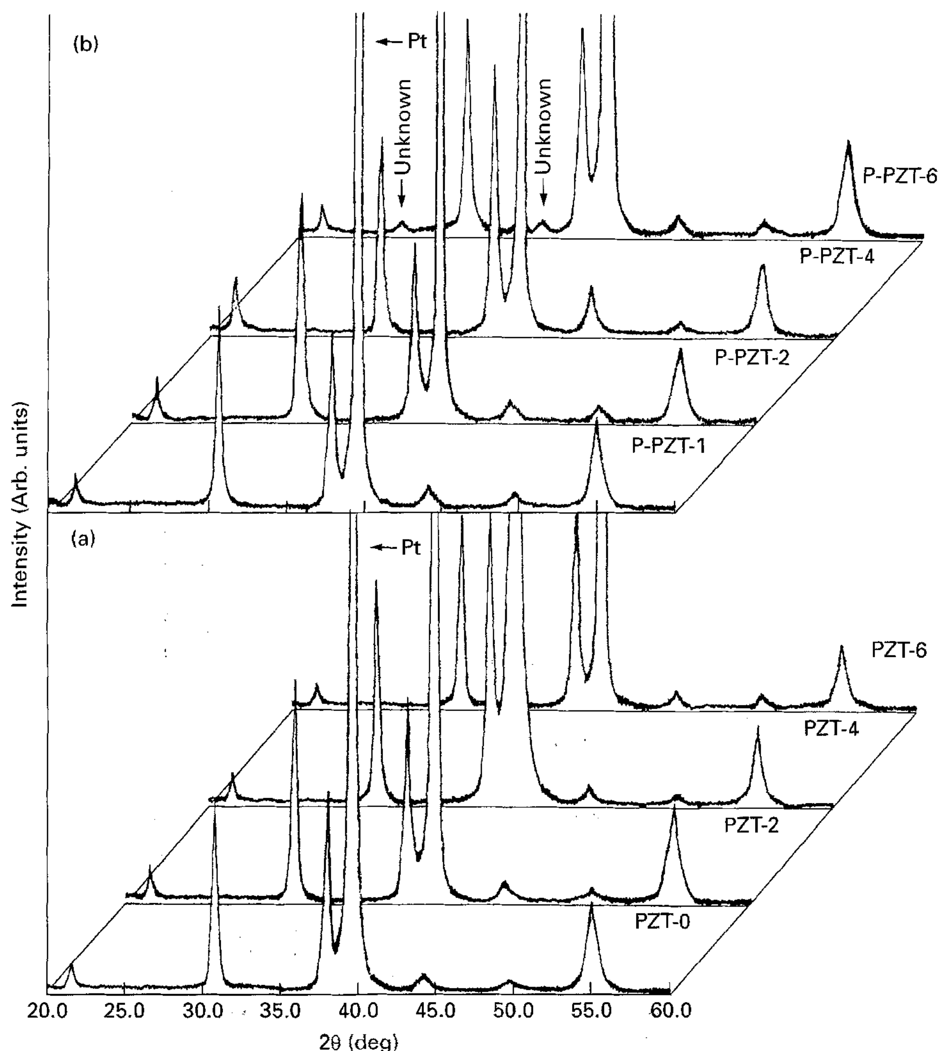

Figure 2 XRD patterns of PZT 53/47 films annealed at 600 °C for various times in (a) a PbO vapour-containing atmosphere or (b) covered by a PbO layer.

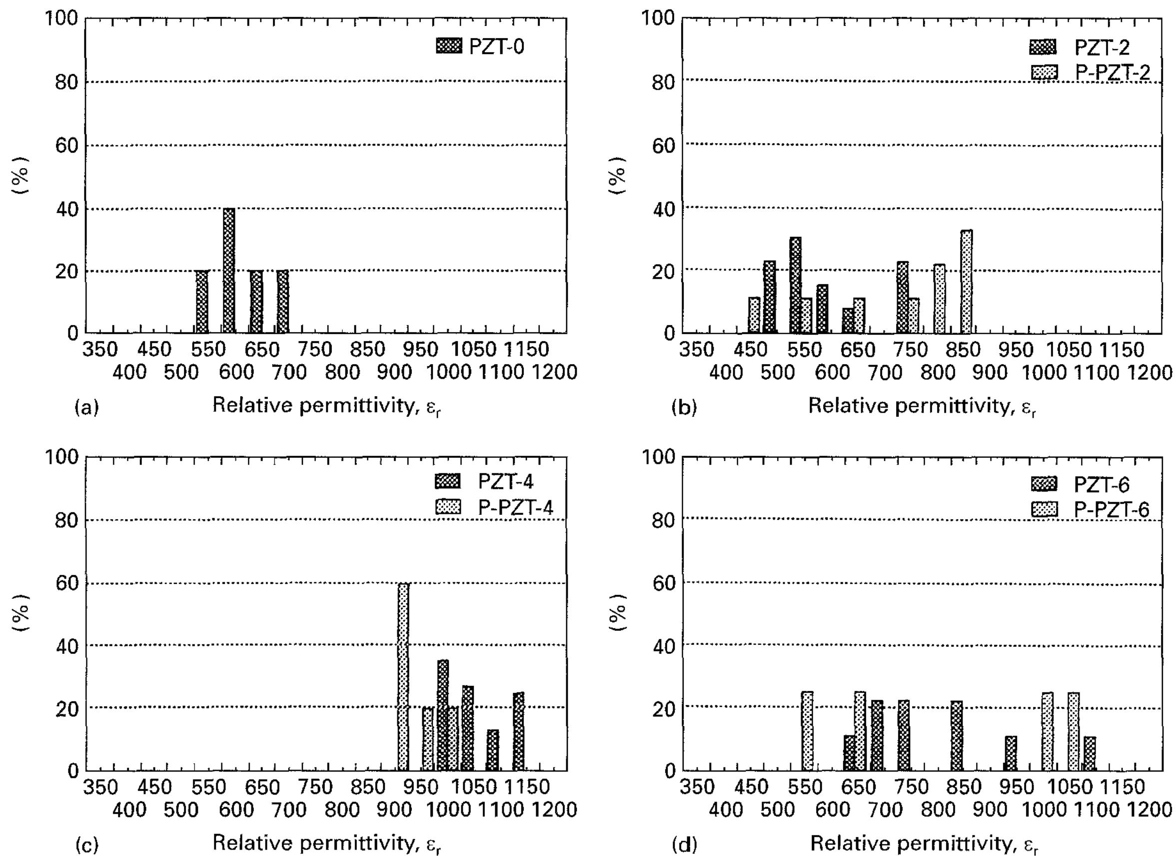

Figure 3 Histograms of relative permittivity, ε_r, versus annealing time for PZT and P–PZT films.

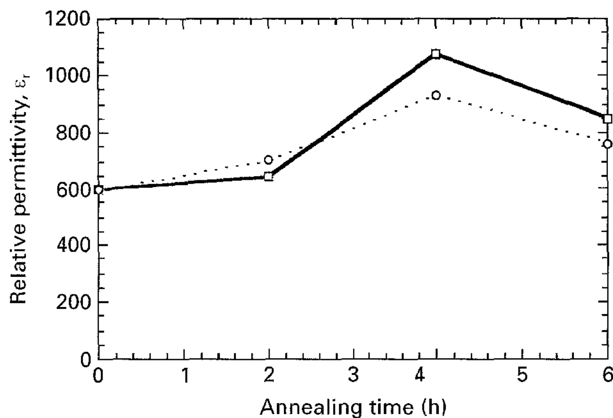

Figure 4 The average relative permittivity, ε_r, as a function of annealing time for (–□–) PZT and (- -○- -) P–PZT films.

respectively. Fig. 4 shows that the average value of relative permittivity increases as the annealing time increases up to 4 h, and then declines.

The histograms of Fig. 5 show the distributions of remanent polarization (i.e. P_r) for those samples. The P_r for the as-fired sample, i.e. PZT-0, is around 16–22 $\mu C\,cm^{-2}$. For P-PZT samples, the P_r values increase slightly when the annealing time is increased to 4 h. Nevertheless, the P_r values suddenly fall for the P-PZT-6 samples. This is possibly related to the existence of an unknown non-ferroelectric phase in the P-PZT-6 samples. The same annealing time dependent trend is observed in the PZT-x samples, but the P_r values for PZT-6 samples do not fall, rather they remain at the same level as in PZT-4 samples and even have the highest P_r value, i.e. 28 $\mu C\,cm^{-2}$. In general, the PZT-4 samples possess the narrowest distribution range of P_r values which are 21–25 $\mu C\,cm^{-2}$. The values of coercive electrical field (i.e. E_c) show a similar distribution to that of remanent polarization. The E_c values for PZT-4 samples are about 40–60 $kV\,cm^{-1}$, while the E_c values for P-PZT-4 samples are about 40–65 $kV\,cm^{-1}$. The annealing time-dependent average value of P_r for both types of sample are depicted in Fig. 6. PZT-4 exhibits not only better average electrical properties, but also good uniformity. Fig. 7. shows the hysteresis loops of PZT-4 samples taken under different applied voltages, i.e. 2, 4, 8 V (V_{p-p}). The well-saturated hysteresis loop can be obtained by increasing the driving voltage to only 8 V.

The related surface microstructures of PZT films are shown in the Fig. 8. Obvious grain growth is observed when the comparisons are made between the PZT-0 and PZT-4 or P-PZT-4 samples. The grain size of P-PZT-4 is slightly larger than that of PZT-4. In addition, the grain size for PZT-6 differs very little from PZT-4, but blurred grain boundaries are observed in some areas. This may be attributed to the lead loss of the samples, which takes place during the post-deposition annealing process, from the grain-boundary region. The pressure difference in the vicinity of the grain boundary becomes higher than in other areas owing to the smaller surface curvature radius [24]. As a result, lead loss tends to occur in the grain-boundary region rather than in other areas. The broadened distribution range of electrical properties for PZT-6 samples is expected, due to the lead loss. The surface microstructures of P-PZT-6 are similar to those of PZT-6. From the above results, the improvement of electrical properties between the as-fired films and the 4 h annealed films can rationally be attributed

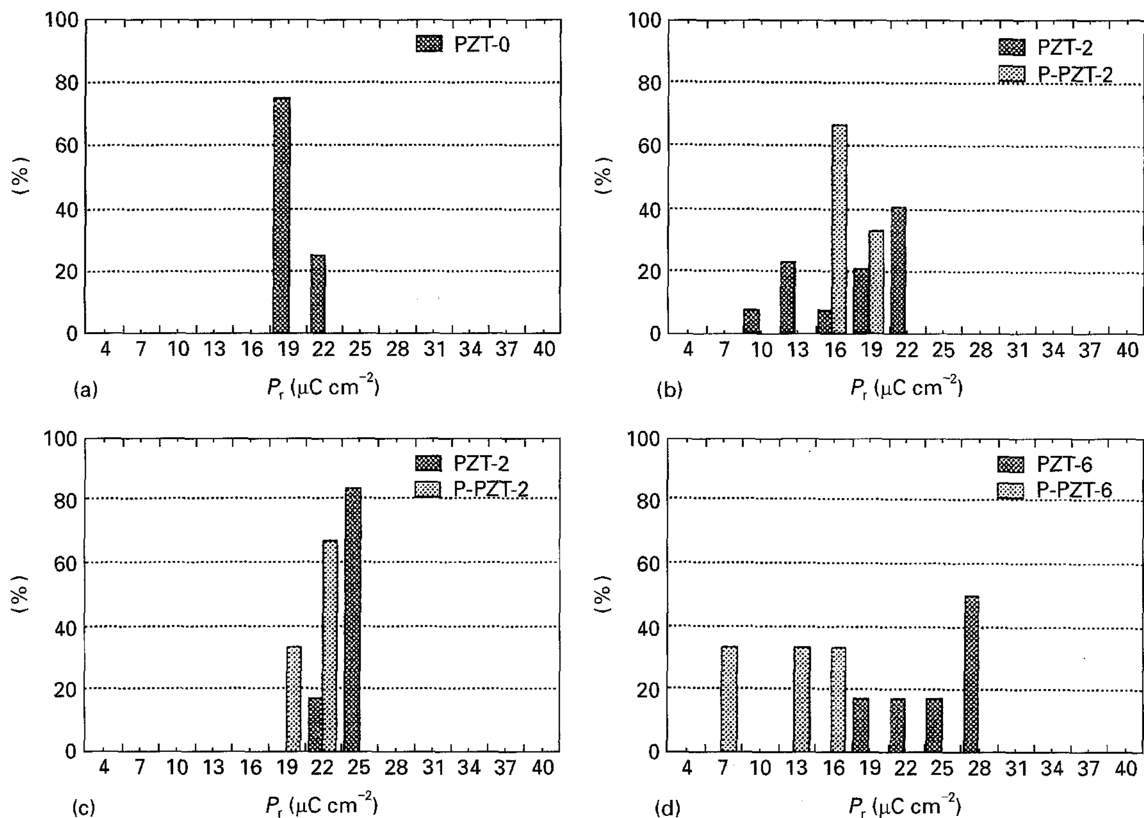

Figure 5 Histograms of remanent polarization, P_r, versus annealing time for PZT and P–PZT films.

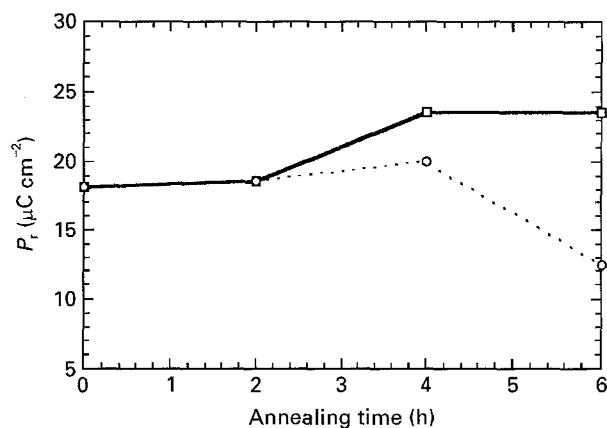

Figure 6 The average remanent polarization, P_r, as a function of annealing time for (–□–) PZT and (- -○- -) P–PZT films.

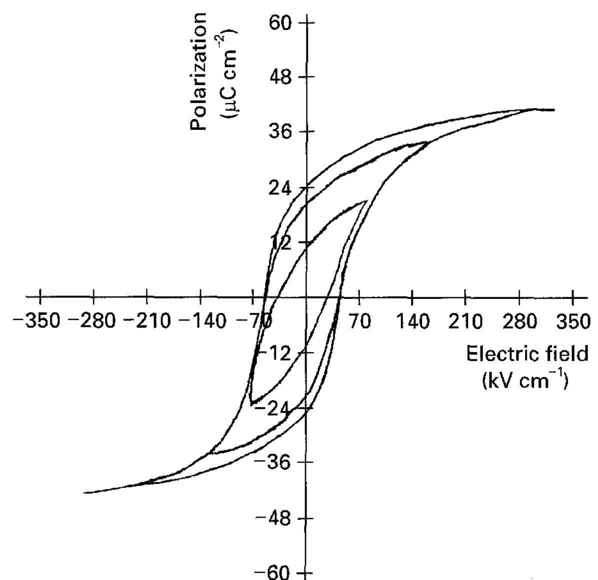

Figure 7 Hysteresis loops of the PZT-4 films measured at driving voltages of 2, 4, and 8 V (V_{p-p}) at 1 kHz.

to grain-growth phenomena from the as-fired films to the 4 h annealed films. In contrast, the maximum values of relative permittivity and remnant polarization do not increase when the annealing time is increased from 4 h to 6 h. It is also logical that there is no further grain growth in that range. The 4 h annealing in the PbO vapour-containing atmosphere appears to be the optimum process to prepare PZT films with good electrical properties and uniformity for the current experimental set up. The longer period of annealing, i.e. over 4 h, may broaden the distribution range of physical properties and deteriorate the electrical properties.

3.2. $PbTiO_3$ interlayers

For this procedure the gel powder of $PbTiO_3$ was derived by baking the solution on a hot plate at 300 °C for 10 min, and then the gel powder was heat treated at 450 and 500 °C for 20 min. The related XRD patterns are shown in Fig. 9. The apparent peaks of $PbTiO_3$ perovskite phase are observed in the pattern of 500 °C, 20 min heat-treated gel powder. This heat-treatment process was adopted as the firing conditions for preparing the $PbTiO_3$ interlayers, so that the crystallized $PbTiO_3$ interlayer with the single perovskite phase can be used as a seeding layer. The necessary formation temperature of single perovskite phase of PZT (53/47) film can therefore be reduced. Moreover, in this section of the experiment, the PbO vapour-containing atmosphere was applied simultaneously to

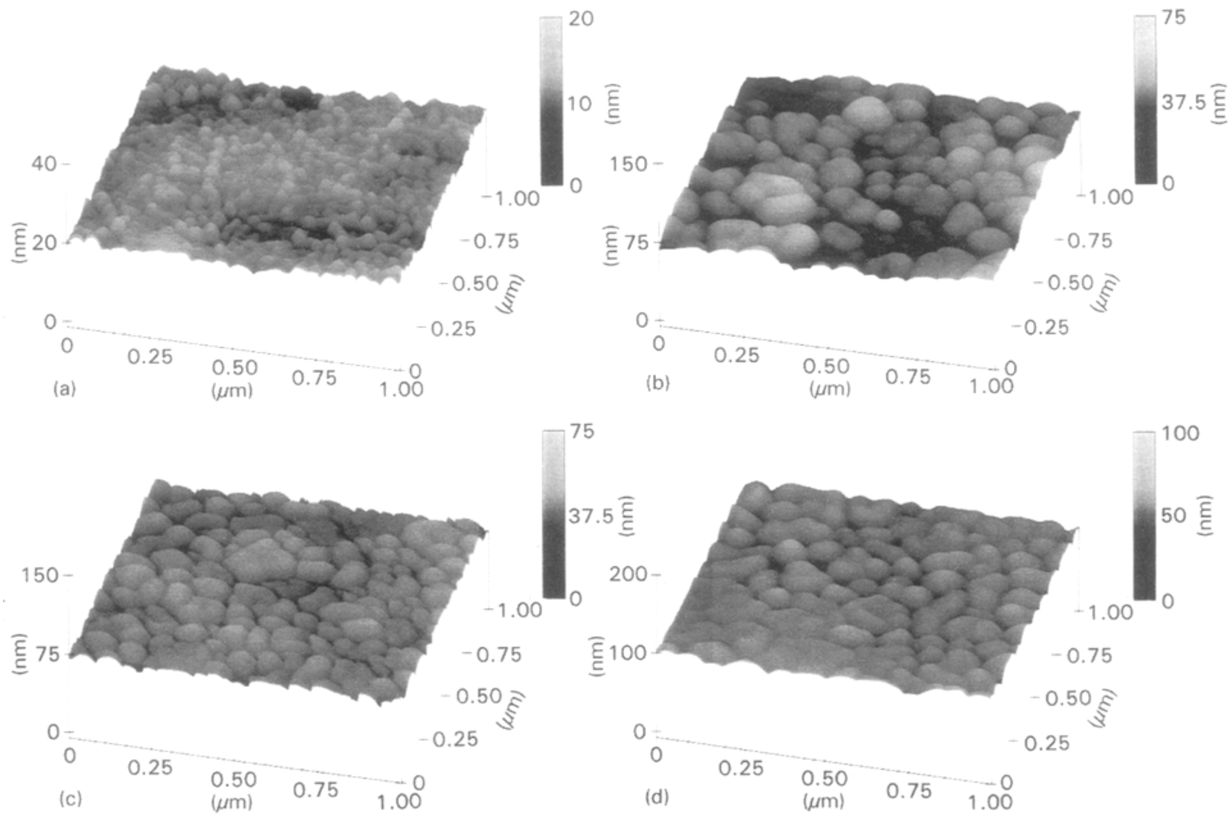

Figure 8 AFM images of surface morphology for (a) PZT-0, (b) P-PZT-4, (c) PZT-4, and (d) PZT-6 samples.

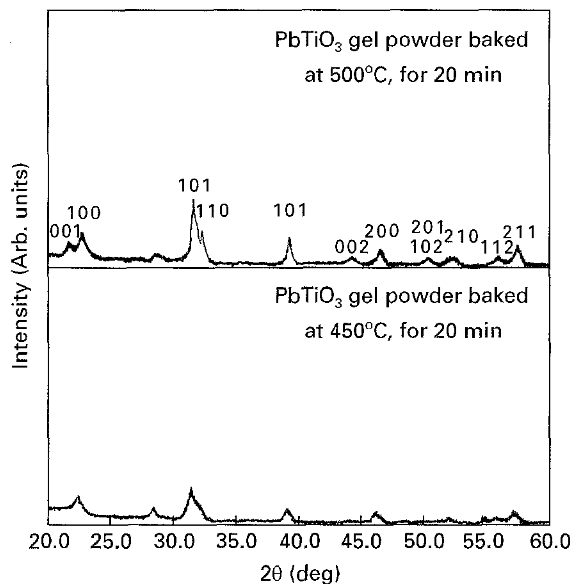

Figure 9 XRD patterns of derived gel powder of $PbTiO_3$.

prevent lead loss during the post-deposition annealing. The PbO vapour-containing atmosphere method is seen to be superior to the PbO cover layer method, from the results of the preceding experiment. The XRD patterns of the PZT-4PT samples with different annealing temperatures and times are shown in Fig. 10. For the 500 °C annealing temperature, pyrochlore phase peaks are observed for as-fired and 30 min annealed samples; however, samples of pure perovskite phase can be obtained by annealing for 1 h. These results coincide with the data reported by Kwok and Desu [8]. Additionally, the peaks of pyrochlore phase are seen for the samples annealed for 4 or 6 h, probably due to the perovskite and pyrochlore phase co-existing at 500 °C for the PZT (53/47) composition. The perovskite to pyrochlore phase transformation may occur during the long period of annealing. However, the phase transformation route, which is two-phase mixture (pyrochlore/perovskite)–single phase (perovskite)–two-phase mixture (perovskite/pyrochlore), was first observed when samples underwent annealing treatment with increasing time. A more detailed study of the microstructure is necessary to elucidate this point. For the 550 or 600 °C annealed samples, the single perovskite phase was observed for all samples. Representative XRD patterns of the 550 °C annealed samples are shown in Fig. 10.

The influence of different annealing temperatures versus annealing time in the preparation of PZT–4PT samples on the electrical properties is shown in Fig. 11. Obviously the annealing temperature of 500 °C is too low to obtain well-developed crystallinity. The relative permittivities range from 100–400 and lossy characteristic of hysteresis loop can be observed. The remanent polarization, P_r, and coercive field, E_c, are around 2.0–3.5 $\mu C\,cm^{-2}$ and 60–75 $kV\,cm^{-1}$, respectively. Although the 1 h annealed PZT–4PT samples possess the single perovskite phase, the relative permittivities and remanent polarizations for 550 °C annealed samples are higher than, or similar to, the values for 600 °C annealed samples. The best data can be obtained from the 550 °C, 6 h annealed samples, which are $\varepsilon_r = 885$, a

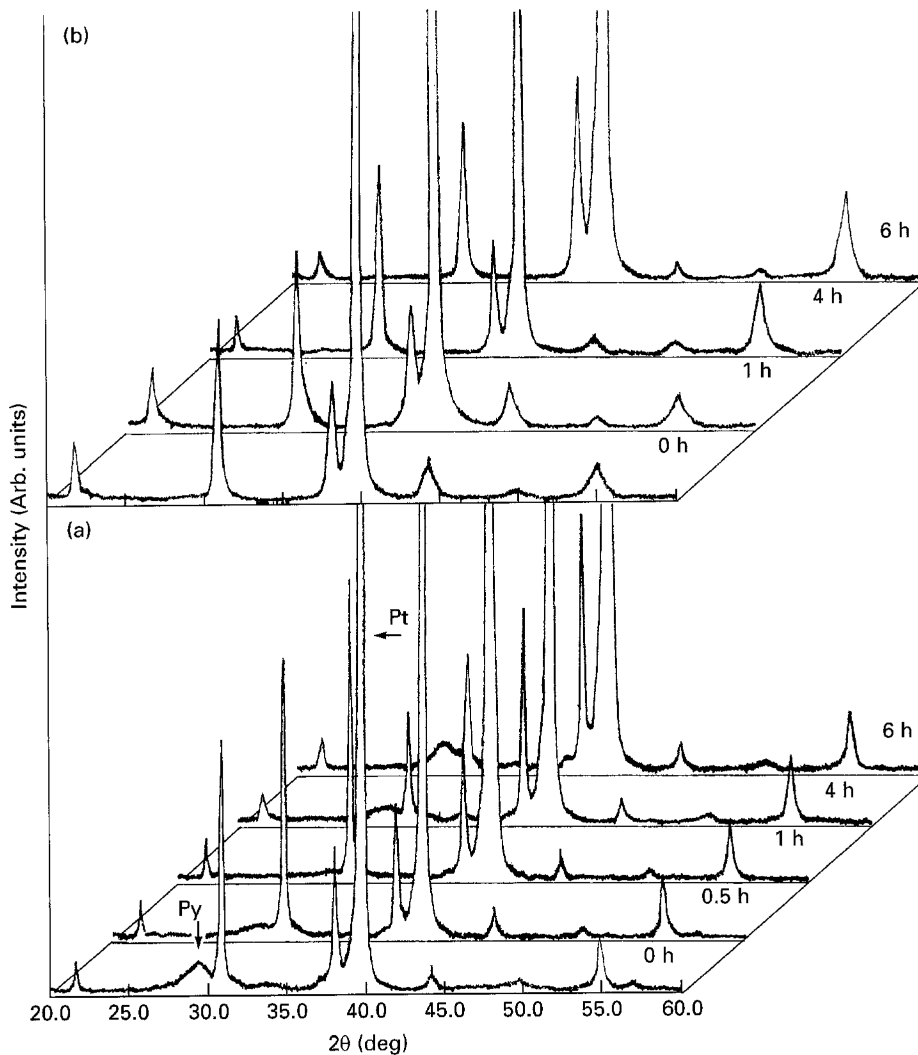

Figure 10 XRD patterns of PZT–4PT films annealed at (a) 500° C, (b) 550 °C for various times.

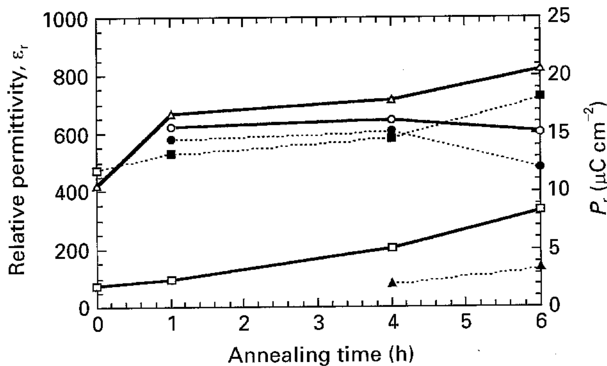

Figure 11 The average (□, △, ○) relative permittivity, ε_r, and (■, ▲, ●) remanent polarization, P_r, as functions of annealing temperature and time for PZT–4PT films (□, ■) 500 °C, (△, ▲) 550 °C, (○, ●) 600 °C.

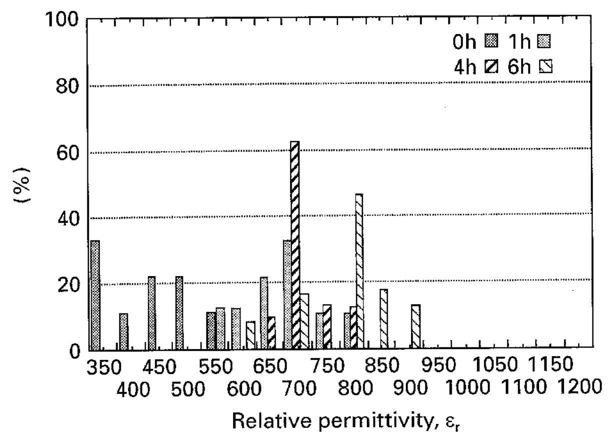

Figure 12 Histograms of relative permittivity, ε_r, versus annealing time for PZT–4PT films at 550 °C.

dissipation factor of 0.047, $P_r = 21.5\ \mu C\,cm^{-2}$ and $E_c = 64.0\ kV\,cm^{-1}$. Histograms of relative permittivities versus annealing time for 550 °C annealed samples are shown in Fig. 12. The 4 h anneal produces the narrowest distribution range in the samples, whilst samples subjected to the 6 h annealing possess the best electrical properties.

The effects of interlayer thicknesses for preparing the PZT–PT multilayer films on the electrical properties are shown in Fig. 13. In general, the influence of interlayer thicknesses is not as great as expected because the same trend can be observed for the samples with both 40 and 70 nm thick $PbTiO_3$ interlayers. However, the electrical properties for as-fired and 6 h annealed samples exhibit differences between the two samples with different thicknesses. On annealing at 550 °C, the thicker interlayer is seen to enhance the crystallinity of as-fired samples. However, the

crystallinity is improved for samples with a thinner $PbTiO_3$ interlayer after 1–4 h annealing. As a result, the value of relative permittivity of PZT–4PT is the same as that of PZT–7PT during that period. For the 6 h anneal, both PZT–4PT and PZT–7PT samples possess well-developed crystallinity and the electrical properties are simply dependent on the thickness of the interlayer, i.e. the $PbTiO_3$ layer, which has a lower relative permittivity. The multilayer of PZT–PT can be considered to be a capacitor made up of two thin-layer capacitors in series. In the fixed thickness of the PZT layer, samples with thicker $PbTiO_3$ layers, i.e. PZT–7PT, obviously possess lower relative permittivities.

3.3. The electrical short circuits

Fig. 14 shows that the PbO vapour-containing atmosphere method can offer a better means of annealing the samples than the PbO cover layer method. This is demonstrated by the dramatic reduction in the percentage of electrical short circuits. However, a long annealing time (longer than 4 h) may induce an increase in the percentage of electrical short circuits. On the other hand, the insertion of an interlayer of $PbTiO_3$ is an effective way to reduce this. Thus, possibly, the more dense PZT films with fewer electrical short circuits, such as lead-deficient phases, grain-boundary phase, oxygen vacancies etc., can be prepared by inserting a $PbTiO_3$ interlayer. On the other hand, as expected, the reduced annealing temperature for PZT–PT films, i.e. 550 °C, can decrease the percentage of electrical short circuits owing to the reduced lead loss.

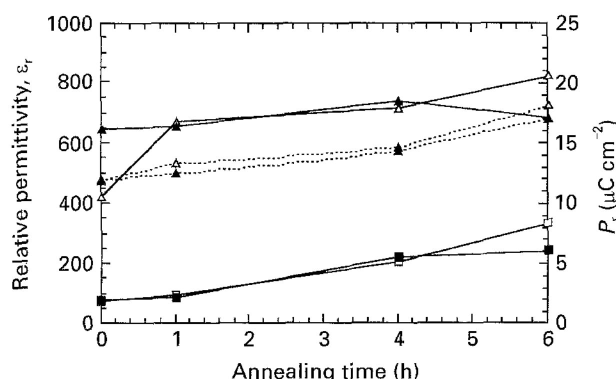

Figure 13 The effects of $PbTiO_3$ interlayer thickness for preparing the PZT–PT multilayer films on the electrical properties: (——) ε_r, (– – –) P_r: Annealing temperautres (□, ■) 500 °C, (△, ▲) 550 °C, with (□, △) PZT–4PT, and (■, ▲) PZT–7PT.

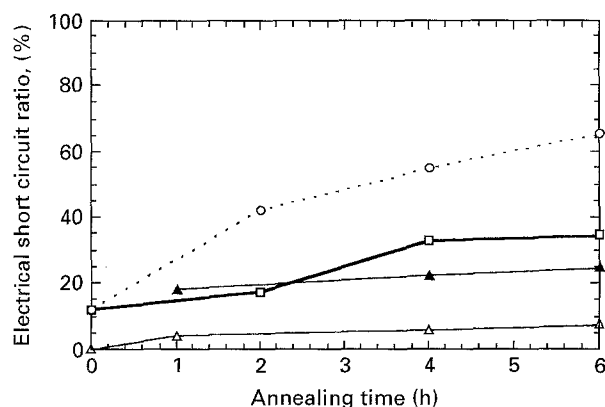

Figure 14 The effects of annealing conditions on the electircal short circuit ratios of PZT and PZT–4PT films. (-□-) PZT, 600 °C; (--○--) P-PZT, 600 °C; (—△—) PZT–4PT, 550 °C; (—▲—) PZT–4PT, 600 °C.

4. Conclusions

The optimization of post-deposition annealing parameters, such as temperature and time, for preparing sol–gel derived PZT (53/47) thin films were scrutinized in this study by comparing the different annealing environments: the PbO cover layer and the PbO vapour-containing atmosphere. Insertion of an interlayer of $PbTiO_3$ proved to be a way to prevent lead loss and to improve the crystallinity by forming a perovskite seeding layer and then reducing the perovskite phase formation temperature of PZT (53/47). The most important findings from this investigation are as follows.

1. From the results of electrical properties and the percentage of electrical short circuits, the PbO vapour-containing atmosphere method is proved to be better than the PbO cover layer method as an effective way to prepare the PZT films of high quality and uniformity via the sol–gel process. The optimized annealing conditions for the preparation of PZT (53/47) films are 4 h at 600 °C in the PbO vapour-containing atmosphere. The derived films possess electrical properties as follows: $\varepsilon_r = 1150$, a dissipation factor of 0.043, $P_r = 25\ \mu C\,cm^{-2}$, and $E_c = 43.3\ kV\,cm^{-1}$.

2. PZT–PT films with single perovskite phase can be obtained from the post-deposition annealing of 500 °C and 1 h. The derived PZT–PT films exhibit poor electrical properties with $\varepsilon_r = 160$, $P_r = 2.0\ \mu C\,cm^{-2}$, and $E_c = 75\ kV\,cm^{-1}$. The lossy characteristics of the hysteresis loop is observed.

3. The optimized combination for preparing PZT–PT films is a 40 nm $PbTiO_3$ interlayer and annealing conditions of 6 h at 550° C in the PbO vapour-containing atmosphere, and the derived films possess electrical properties which are comparable with the data of pure PZT films: $\varepsilon_r = 885$, a dissipation factor of 0.047, $P_r = 21.5\ \mu C\,cm^{-2}$, and $E_c = 64\ kV\,cm^{-1}$.

4. The combination of inserting a $PbTiO_3$ interlayer and a PbO vapour-containing annealing atmosphere can prevent the formation of electrical short circuits with nearly pin-hole-free PZT (53/47) films which can be grown on the substrates covered by a $PbTiO_3$ interlayer.

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