Preparation of gold nanoparticles embedded in lead zirconate titanate films

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Nanocomposite films consisting of nanoscale metal particles such as gold, silver, and copper embedded in a dielectric matrix have been studied for many applications [1]. Nanocomposite films could be fabricated by sputtering, sol-gel, ion implantation, chemical vapor deposition, and aerosol deposition methods [2]. However, these methods are complex, including instrument requirements, complicated processes, and small doped amount into the matrix. In the study, we demonstrate a simple method that prepare the gold/ lead zirconate titanate (PZT) nanocomposite films by organic chemical addition method.

In our study, when AuCl₄ ions were complexed with dodecylamine, the AuCl₄ dodecylamine complexes remained stable during the period of gel drying and organic solvent prevented PZT precursor from water to form precipitation. A solution of Au³⁺ ions are added into the metalorganic decomposition (MOD) precursor of PZT, and small Au particles could form due to the decomposition of AuCl₄⁻ dodecylamine complexes in the high temperature heating process (as shown in Figure 1). Because the PZT precursor is easily hydrolysis, the [AuCl] need to hydrophobize in toluene were separated from the aqueous phase. The 1g PZT solution is mixed with 0.1, 0.5, and 1 ml of 25 mM HAuCl₄, and uniformly stirring. Each film is dried at 120 °C and heated to 200-450 °C in air. Figure 2 shows X-ray diffraction (XRD) spectra of Au/PZT composite film with contain HAuCl₄, as-deposited, and post-annealed film at $450\,^{\circ}$ c and found the composite film has been fully crystallized. Moreover, we observe the compressive residual stress and small crystalline size in the as-deposited film. No peak of gold is found since the amount in the film is too small to detect. Figure 3 shows the transmission electron microscope (TEM) images and electron energy-loss spectrometers (EDS) of Au/PZT composite films with containing HAuCl₄ of as-deposited and post-annealed samples. As shown in Figure 3(a), there aren't any Au nanoparticles in the PZT matrix and Au composition can be observed from TEM images and EDS analysis. As post-annealed at 450 $^{\circ}_{\rm C}$, the Au nanoparticles are precipitated in the PZT matrix and can be detected by EDS analysis (Figure 3(b)). The size of gold nanoparticles is about 5-10 nm in the PZT matrix. Polycrystalline structures of Au nanoparticles can be identified from weak rings and some scattered spot clearly. As shown in Figure 4 (a), there are not obviously absorption peak in the absorption spectra with increasing HAuCl₄ contents. The extinction coefficient is not any tendency with increasing HAuCl₄ contents. Figure 4(b) shows absorption properties of the postannealed composite Au/PZT thin films at 450 °C. The stronger absorption with increasing HAuCl₄ contents and the SPR absorption peak is found at about 620 nm. The extinction coefficient is increasing with increasing HAuCl₄ contents. These phenomena appear to be consistent with the TEM images and EDS analysis. In this study, we present the results of convenient chemical synthesis to fabricate nanocomposite Au/PZT films with obvious SPR phenomena.

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Fig. 1 Schematic of organic chemical addition method of nanocomposite films.



Fig.2 XRD spectra of composite film with containing 1 ml HAuCl₄, as-deposite, and post-annealed film at 450 °C for 30 min.



Fig. 3(a)



Fig.3 TEM images and EDS analysis of Au/PZT composite films. (a) as-deposited (b) post-annealed film at 450 °C for 30 min.



Fig.4 The extinction coefficient of composite films with various $HAuCl_4$ contents (a) as-deposited and (b) 450 \degree .