CHAPTER 4

CONCLUSIONS

4.1. CONCLUSIONS

The PZT and PLZT powders were synthesized hydrothermally from lead acetate trihydrate, zirconium n-propoxide and titanium isopropoxide. Lanthanum acetate used for preparing PLZT at 12 mol % lanthanum. The important parameters in hydrothermal synthesis are the concentration of mineralizer, pH value, temperature and time. The base mineralizer type and concentration play an important role in promoting the solubility and rearrangement of the titania and zirconia gels. The smaller the base cation radius, the more efficient it is to diffuse into the gel network. The mineralizer should be above its critical level for providing both soluble Pb species and base ions to cause disruption of the titania and zirconia gels network.

Choosing carefully the concentration to be used at a moderate synthesis temperature and time in each concentration can control the particle size, morphology and perovskite phase of the PZT powders. Increasing mineralizer concentration will lead to polynuclear growth due to the agglomeration of the cubic PZT particles and increases in the solubility of Zr^{4+}, Ti^{4+} and Pb^{2+}, which can accelerate the recrystallization process. This process may lead to the larger particle size.

The pH value is one of the important factors the critical pH value for this present work is 13. The synthesis temperatures and time are another two important factors. At low synthesis temperature without a very high mineralizer concentration, perovskite PZT cannot form even in a long synthesis time. Increasing synthesis temperature can reduce the minimum concentration required for PZT formation also

reducing the agglomerate particle size. Increasing synthesis time can lead to a narrow size distribution but increasing in the particle size of PZT powders. In the case of low mineralizer concentration, PZT powders are produced at high synthesis temperature together with long synthesis time. So, for each mineralizer concentration, they can work only with their moderate synthesis temperature and time.

The thermal decomposition and its conversion to PZT were studied by thermogravimetric analysis (TGA) and differential thermal analysis (DTA). The results confirmed that both PZT and PLZT powders from hydrothermal process lost weight at about 100 °C from the evaporation of water or hydroxyl group and the second weight loss occurred at about 900 °C due to the evaporation of lead oxide.

The moderate conditions for synthesized rhombohedral PZT are 4.0 M KOH at a synthesis temperature of 200 °C for 6 hours under autogeneous pressure, which results in PZT powders with mean particle sizes of 0.4 μm.

For tetragonal PZT, the moderate conditions are 4.0 M KOH at synthesis temperature of 100 °C for 48 hours which produced PZT powders with a mean sizes ranging from 0.3 μm and also exhibited narrow particle size distribution.

The conditions for synthesized PLZT powders are 4.0 M KOH at 200 °C for 6 hours. Under these conditions, PLZT has a cubic shape and its sizes ranged from 0.33 μm to 1.0 μm.

The optimize sintering condition for PZT ceramics is 1250 °C for 5 hours with 94.7 % theoretical density, 13.5 % linear shrinkage and the grain sized are between 3.0 μm to 5.0 μm.

Ceramics PZT from hydrothermal PZT ceramics has coexist phases between rhombohedral and tetragonal, (when these two ceramics were chosen from the rhombohedral structure powders). This is because lead losses during sintering process due to the precipitation of ZrO_2 in hydrothermal PZT.

From the impedance spectroscopy results, we can summarize as follows. With 12 mol % of La^{3+} addition bring the conductive mechanism of PZT ceramics from nearly complete grain conductivity to grain and grain boundary mixed conductivity. The PbO aggregated in the grain boundary are responsible for grain boundary conductivity and lead vacancies coming from dopants are responsible for grain conductivity. From ac and dc conductivity component were found; Jonscher power-law behavior was obtain and strong low frequency dispersion. It was observed in a wide frequency range, which supports that, the charge carriers of ether electronic or ionic nature. The charge carriers are hopping in the system and their collective interaction lead to this phenomenon. The ferroelectric bulk (grain) conductivity gives up the main contribution to dc component.

4.2. SUGGESTIONS FOR FUTURE WORK

1. For future work, the exactly phase transformation between tetragonal and rhombohedral by varying pH of the final solution before hydroyhermal treatment should be tried out.
2. Try to find out the reaction mechanism in hydrothermal processing by using of the FT-IR technique with different holding periods at fixed pH and temperature.
3. Try to find out the more efficient way to dope La^{3+} ions by the conventional method between La_2O_3 or $La(OAc)_3$ with PZT hydrothermally synthesized powders.
4. Try to find out the possible way to synthesis PZT powders with a nanometer particle size (<100 nm) by reducing the holding periods at the fixed pH and temperature.