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Effects of microwave heating on dielectric and piezoelectric properties of PZT ceramic tapes

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Abstract

Commercial lead zirconate titanate (PZT) perovskite powders were used to fabricate ceramic tape and then sintered by microwave and conventional methods. Both dielectric and piezoelectric properties of PZT ceramic tapes were studied in terms of sintering process. X-ray diffraction analysis (XRD) and scanning electron microscopy (SEM) show the PZT perovskite phase with smaller grain size and dense microstructure can be obtained at a lower sintering temperature by microwave process. It was also observed that shrinkage ratio and bulk density of the tapes sintered at 800 °C were obtained about 19% and 7.46 g/cm^3 by the microwave heating method, respectively, that is corresponding to those values of sintered PZT tapes at 950 °C by conventional process. Moreover, the dielectric constant and maximum permittivity are increased about 30% as compared with conventional processing method. The experimental results demonstrated that the characteristics of the PZT tapes could be significantly improved by microwave heating method. These results demonstrate that such a simple approach can upswing the piezoelectric and dielectric properties of these tapes by using microwave process with a short heating time.

Keywords: C. Dielectric properties; C. Piezoelectric properties; D. PZT; Microwave heating

1. Introduction

PZT ferroelectric materials have been widely used in various electronic devices such as sound element, transducer, actuator, memory and light modification because of excellent functional properties, such as piezoelectricity, dielectricity, pyroelectricity and electroopticity, etc. [1–3]. In general, PZT powders have been prepared by conventional sintering method but such reactions often lead to compositional and structural inhomogeneities in the ceramic produced [2]. Moreover, the ceramics prepared at high temperatures have drawbacks, such as very large particle size, higher impurity content due to repetitive calcination and grinding steps, and lower chemical activity, and therefore are not suitable for enhancing the dielectric properties for high performance uses. In recent years, many scientists have focused on the investigation of microwave sintering for the feasibility of various materials [4,5]. The primary advantages of using microwaves sintering are (a) rapid kinetics leading to savings of time and energy, (b) rapid internal heating, and (c) lower temperature synthesis [6].

As a result, the PZT bulk devices showed enhanced dielectric and piezoelectric properties have been reported as compared with conventional process method [7].

Therefore, the effects of microwave heating on dielectric and piezoelectric properties of PZT ceramic tapes fabricated by tape casting method will be focused in the present study. In addition, both X-ray and microstructure will be analyzed to correlate with dielectric and piezoelectric properties of PZT ceramic tapes sintered by microwave heat treatment. The PZT tapes prepared by conventionally heating process are also measured for comparison.

2. Experimental procedure

Commercial lead zirconate titanate (PZT) perovskite powders [HIZIRCO-750] were used as start materials. According to the powder catalog, the shrinkage ratio, dielectric constant and electromechanical coupling factor of the

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commercial PZT samples sintered by the conventional process were reported to be about 18.5%, 3000 and 58.8%, respectively. In order to obtain green tape of the PZT material, a 66% solid content of the plasticizers and binder were added and mixed for 24 h to obtain the slurry. Subsequently, the tape casting was performed using a laboratory caster (Model-2104 AEM Inc.) and the green tape was punched to be as round green sheet with a diameter of 10 mm and thickness of 125 μm. After that, these samples were stacked using six pieces of PZT green sheet between two dense ZrO_2 plates as a sandwich configuration with the ZrO_2 powder uniformly sprinkled on upper and lower surface of these samples in order to prevent adhesion of the samples. Then, the samples were heated at a slow heating rate 1 °C/min up to a temperature of 550 °C and held for 6 h to burnout the organic additives.

The microwave heating was performed in a single mode microwave furnace, the microwave heating system delivers approximately 500 W of microwave energy at a 2.45 GHz frequency, and the power can be controlled using an electric tuner. The samples were microwave sintered at 800, 850, 900, 950 °C for 20 min and the sintering temperature was detected from microwave cavity by IR thermometer (RAY-MX4+, USA) from cavity window. In order to enhance pumping microwave energy absorption of PZT green tapes, SiC plate was used as a susceptor. In contrast, the PZT tapes were also conventionally sintered at 800, 850, 900, 950 °C using a resistance furnace with a heating rate of 3 °C/min and held time for 2 h, respectively. The sintered samples with a diameter of about 8 mm and thickness of about 100 μm were pasted with Ag electrode with a diameter of 6 mm on both upper and lower surface of the samples, and then fired at 500 °C for 20 min. After that, the samples were pooled with a dc electric field of 2.5 kV/mm for 5 min at room temperature.

The phase structures of sintered tapes were identified with X-ray diffraction (XRD). The microstructure of the samples tapes was examined by the field emission scanning electron microscopy (FE-SEM, JEOL JSM-6500F). The dielectric properties were measured using precision L.C.R. meter (HP4284A) at an applied voltage of 1 V rms with 1 kHz frequency and oven (Delta-9023). The piezoelectric properties of small signal were examined using impedance analyzer (HP4194A) and piezoelectric parameters were calculated with reference to the IRE standard [8,9].

3. Results and discussion

The phase characterization of the PZT samples sintered by microwave and conventional methods was measured by XRD and shown in Fig. 1. As the PZT tapes were sintered below 950 and 1050 °C in microwave process and conventional process, respectively, only pure perovskite phase was observed. However, above those temperatures, the secondary phase could be detected as shown in the $2\theta \sim 28.2^\circ$ of XRD patterns. The secondary phase is probably pyrochlore phase that can be attributed to evaporation and dissipation of PbO at a higher sintering temperature. The bulk density was also measured for the PZT tapes sintered in both microwave and conventional process. It was observed that the bulk density increases from 7.46 to 7.6 g/cm^3 for the samples sintered in microwave process from 800 to 900 °C, respectively, above that, a slight decrease in bulk density (7.51 g/cm^3) was observed, which is consistent with the observation in XRD patterns shown in Fig. 1. In contrast, as the samples were sintered at 800 °C in conventional process, a lower bulk density of about 5.8 g/cm^3 was measured, indicating that the sintering densification can be enhanced via a microwave sintering process.

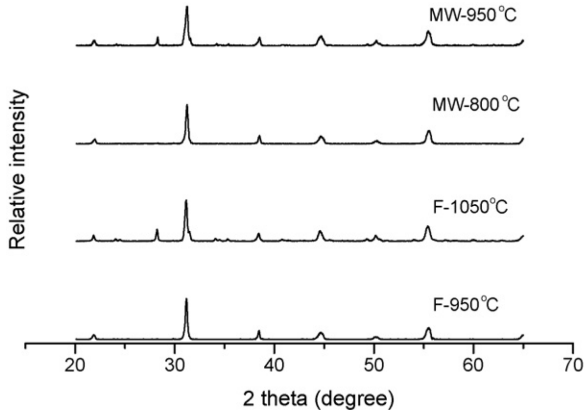

Fig. 1. XRD pattern of microwave (MW) and conventional process (F).

The shrinkage ratio dependence of sintering temperature is shown in Fig. 2. It was observed that shrinkage ratio increased with increasing sintering temperature until 900 °C, above that, it changed little. Furthermore, it indicated that a much faster shrinkage ratio could be obtained in microwave heating than that in conventional heating. The changes of the shrinkage properties of sintered tapes show enhanced dense properties of the sintered tapes from the microwave process. The sintering shrinkage in diameter and thickness is about 19% for the PZT tape sintered at 800 °C for 20 min by microwave process. In contrast, in conventional process, a shrinkage ratio of about 19% was obtained at sintering temperature of 950 °C for 2 h. If based on the same sintering shrinkage, the PZT tapes were

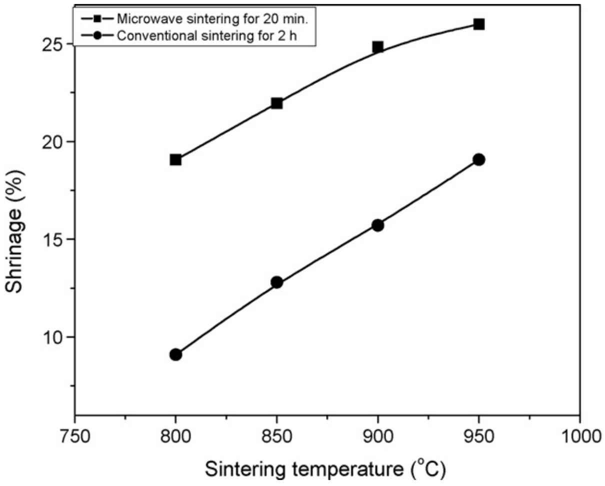

Fig. 2. Shrinkage ratio dependence of sintering temperature for PZT tapes sintered by microwave and conventional processes.

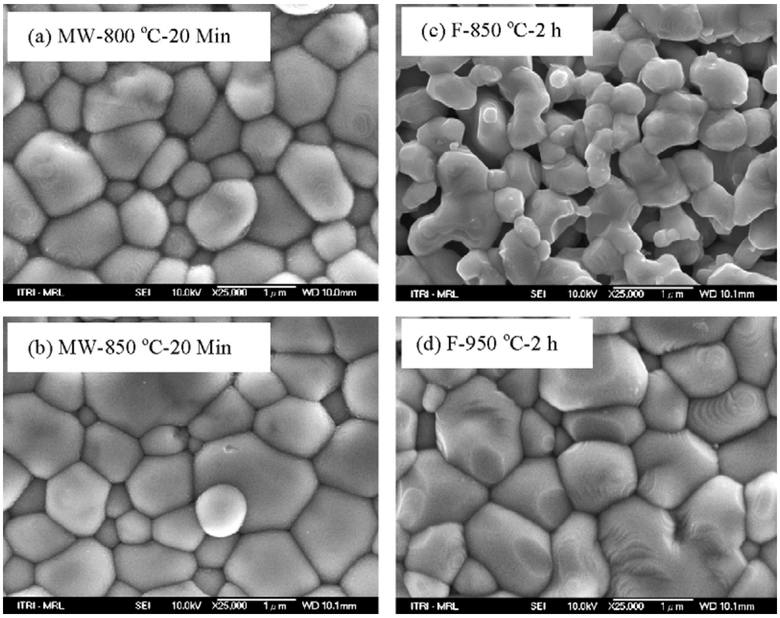

Fig. 3. Microstructure of microwave and conventional sintering PZT tape.

obtained by sintering at 800 °C for 20 min in MW process while in the case of the conventional sintering process, the PZT tapes was required to sinter up to 950 °C for 2 h. This indicated that the sintering temperature to reach a similar shrinkage can be decreased by about 150 °C and the sintering time can be reduced in microwave process as compared to those in conventional process. Fig. 3 shows the microstructures of the sintered tapes in terms of sintering temperature and sintering process. The PZT tape with smaller gain size and denser microstructure can be obtained by microwave process at 800 °C as illustrated in Fig. 3(a). However, many pores are clearly observed on surface of the samples sintered at 850 °C by conventional process as shown in Fig. 3(c). A higher sintering temperature such as 950 °C was required for the PZT tapes to reach a dense microstructure of Fig. 3(d) by conventional process.

The dielectric properties of the sintered PZT tapes by microwave and conventional process are shown in Fig. 4. It was found that the relative dielectric constant in microwave process shows an abrupt increasing change from 1340 to 2876 in the temperature range of 800 to 950 °C, whereas in conventional process, the dielectric constant was increased from 520 to 2100 with increasing sintering temperature. On the other hand, the dielectric loss was decreased from 3 to 1.1% and from 26 to 6.9% for the PZT tapes sintered by microwave and conventional process with increasing sintered temperature, respectively. It was also reported in the literature that the dielectric constant is strongly dependent on the shrinkage and densification of PZT ceramic. Moreover, higher temperatures usually guide to higher dielectric constant [10].

Fig. 5 reveals relative permittivity and dissipation loss dependence of measuring temperature for the samples sintered in different processes. An apparent change in relative permittivity was found when the microwave-treated samples were measured in temperature range from 25 to 250 °C. It can be seen that the PZT tape treated in microwave process at 850 °C showed a maximum permittivity of 14,000, whereas the samples treated in conventional process at 950 °C only have

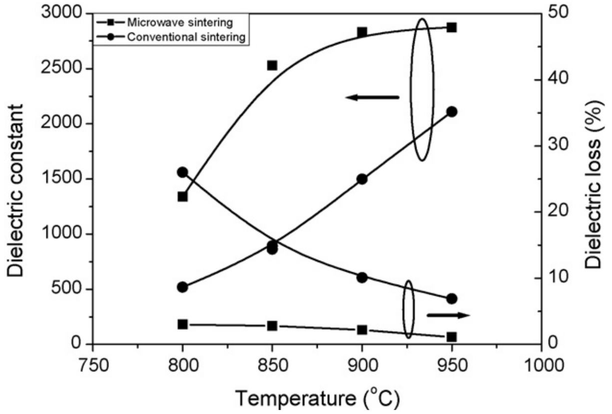

Fig. 4. Sintering temperature dependence of dielectric constant and dielectric loss of PZT tapes sintered by microwave and conventional sintering.

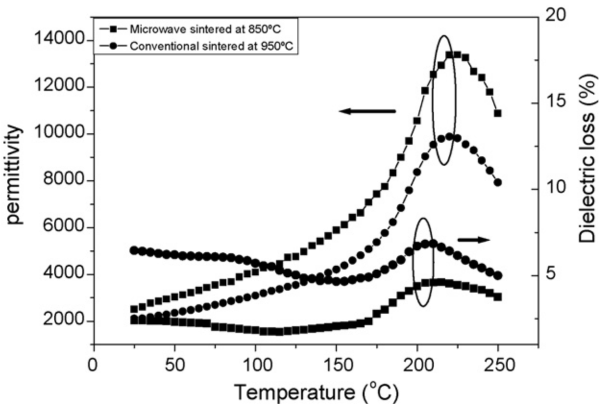

Fig. 5. Temperature dependence of dielectric constant and dielectric loss of PZT tapes sintered by microwave and conventional sintering.

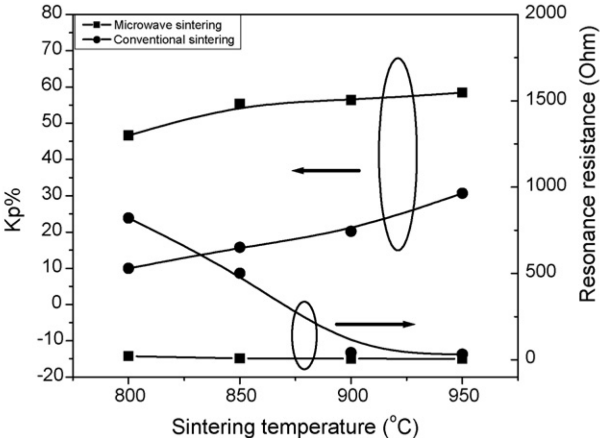

Fig. 6. K_p and resonance resistance of sintered PZT tapes as a function of sintering temperatures.

9900. Furthermore, a lower dissipation loss was obtained for the microwave-treated sample as compared with that in conventional process.

Both electromechanical coupling factor (K_p) and resonance resistance of PZT tapes as a function of temperature were also measured and shown in Fig. 6. From this figure, the K_p value is raised with the sintering temperature from 800 to 950 °C in both of microwave and conventional processes. However, it was observed that with the increase of sintering temperature, a much larger K_p value (46–58%) was obtained from microwave process compared to that (10–30%) from convention process. Furthermore, a lower resonance resistance of sintered tapes was also detected for microwave heating compared to that for conventional process. This demonstrates that PZT tape with excellent piezoelectric properties and lower sintering temperature could be obtained by microwave than that by conventional sintering process.

4. Conclusions

In summary, dielectric and piezoelectric properties of the PZT ceramic tapes sintered by microwave heating in microwave and conventional sintering method have been characterized and evaluated. It has been also demonstrated that greater shrinkage and rapid densification could be achieved in microwave process. The higher dielectric constant and lower dielectric loss of 2530 and 2.8% were obtained for the PZT tapes sintered at 850 °C by microwave sintering. Moreover, the PZT tapes sintered at 850 °C by microwave process show much higher K_p and lower resonance resistance and those values were found to be about 55.4% and $7.7\ \Omega$, respectively. It revealed that microwave treatment shows an effectively and powerful sintering process in reducing sintering temperature and enhancing the dielectric and piezoelectric properties of the PZT ceramic tapes.

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