Microwave Processing of Electronic Ceramics

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Abstract

PZT ceramics were rapidly sintered in a 2.45GHz microwave oven without no addition of lead to the atmosphere and afterwards the piezoelectric properties were investigated. The specimens were sintered between 1050 and 1130℃ with the sintering time totaling less than 20 minutes, reducing processing times and costs from those of established methods. The relative dielectric constant, mechanical quality factor and electro-mechanical coupling factor of the PZT specimens at 1090℃ were 1900, 80, 0.53 respectively, which is comparable to conventionally sintered values.

1. Introduction

The microwave sintering method has several advantages when compared to conventional sintering method. These include a shorter processing time, energy efficient processing, uniform heating of products and a small grain size[1]. These advantages in microwave processing are from fundamentally different heating mechanism from conventional process. Generally heat is originating from external heating sources. In microwave sintering process, however, heat is generated internally and rapidly by microwave. It heats the entire specimen which enables the unique heating of even large shapes.

Many researches have been applied using microwave energy to sinter carbides, borides, and ceramic composites, as well as oxide ceramics[2-4]. But very few researches are attempting to sinter ferroelectric materials. In the following experiment, microwave processing was applied to sinter PZT ceramics which are most widely used piezoelectric materials[5]. Previously, the sintering of lead perovskite materials was not always easy due to the volatilization of PbO. Many past efforts concentrated on the reduction of the amount of PbO volatilization, which was usually accomplished through atmosphere control with lead rich powder in doubly sealed ceramic vessles[6].

In this study, PZT was sintered by a microwave sintering method, without the addition of Pb to the atmosphere, and afterwards the electrical properties were investigated.

2. Experimental Procedure

The microwave hybrid heating system was constructed through the use of a modified 1 kW, 2.45GHz microwave oven(Goldstar Co.). The schematic of the microwave sintering apparatus

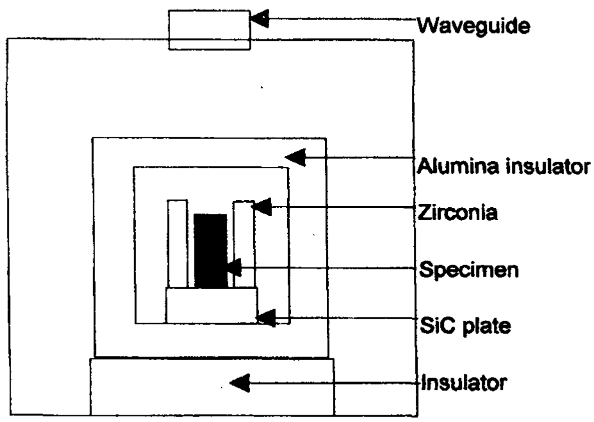

Fig. 1. Schematic drawing of microwave sintering apparatus with 1kW, 2.45GHz magnetron

is shown in Fig. 1. Insulation brick was placed on the bottom of microwave cavity and SiC plate and zirconia fiber brick (Zirca Products Inc., ZYZ-3) was used as a susceptor, creating to hold the specimens during processing. The SiC and zirconia fiber brick are capable of heating specimen from room temperature to high temperature through absorbtion of 2.45GHz microwave frequency due to their high loss[1)]. The microwave hybrid heating system was used in this heating system. The heat created by microwave-material interaction as well as through radiant and conductive heat from susceptors. The arbitrary temperature was measured using an optical pyrometer(Minolta Camera Co., TR-600A) positioned in a 8mm diameter hole in the center front brick. The size of the hole was sufficiently small as to not prohibit black body radiation.

The composition of the materials investigated in this series of experiments was $(Pb_{0.95}Sr_{0.05})(Zr_{0.53}Ti_{0.47})O_3+0.8wt\%Nb_2O_5$. Two millimeter thick by twelve millimeter diameter pellets were formed under 1ton/cm² pressure from a powder with an average particle size of 3.1 microns. The PZT specimens were sintered in the microwave hybrid heating system for 5 minutes at several sintering temperatures.

After sintering, electrodes were screen printed on both side of specimens, and the specimens were fired at 800℃ for 10 minutes. Proceeding electroding, the specimen were poled with 2.5KV/mm applied field for 30 minutes before measuring the electrical properties. The relative dielectric constant, mechanical quality factor(Qm), and electro-mechanical coupling factor(kp) of the specimens were measured by using Network Analyzer(HP 3577A). The bulk density of specimens were measured by archimedes method. The microstructure and composition of specimens were characterized by SEM and XRD.

3. Results and discussion

A plot of density of sintering temperature is shown in Fig. 2. The density of sintered pellets increased as the temperature of the specimens rose to a maximum of 1090℃. The maximum sintered density achieved was 7.71g/cm³. This sintered density is a comparable value to the conventional sintering method. Above this specified temperature, the density of the pellets begin to decrease due to the volatilization of PbO.

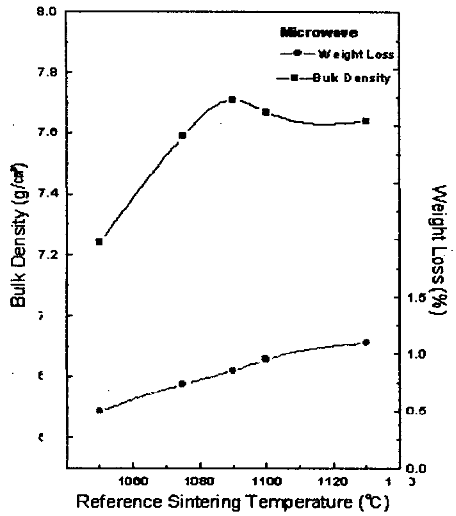

Fig. 2. Bulk density and weight loss of the microwave sintered specimen for 5 min vs reference sintering temperature.

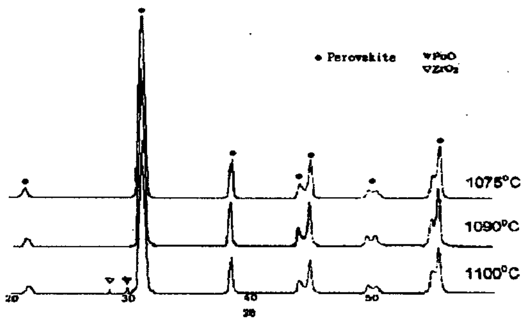

Fig. 3. XRD patterns of the microwave sintered specimen as a funtion of reference sintering temperature

An XRD study was used to investigate the surface composition of the specimen and to realize if a phase change had occured during the sintering process. A summary of the XRD pattern results from microwave sintered specimens is given in Fig. 3. Below 1075℃ only the PZT phase was present, but above 1090℃ small clusts ZrO_2 phase formed due to the volatilization of PbO from the pellets surface. At temperature of 1100℃, small region of the PbO phase could be detected on the surface, in addition to the small cluster of ZrO_2. As the sintering the PbO phase is thought to form as PbO liquifies in the sample bulk and migrated to the surface as the sintering temperature is increased. This low density formation of ZrO_2 and the volatilization of PbO at the tempreature above 1090℃ coincides with the bulk density decrease and the weight loss of the specimen as previously shown in Fig. 2. Fig. 4 shows the microstructure of sintered specimen. Specimen sintered at 1050℃ was not sintered with small size grains inside it. At 1090℃, further grain growth occured and maximum density was achieved. But further grain growth did not seem to occur at higher sintering temperature.

Fig. 4. SEM fracture photographs of the microwave sintered specimen for 5 min

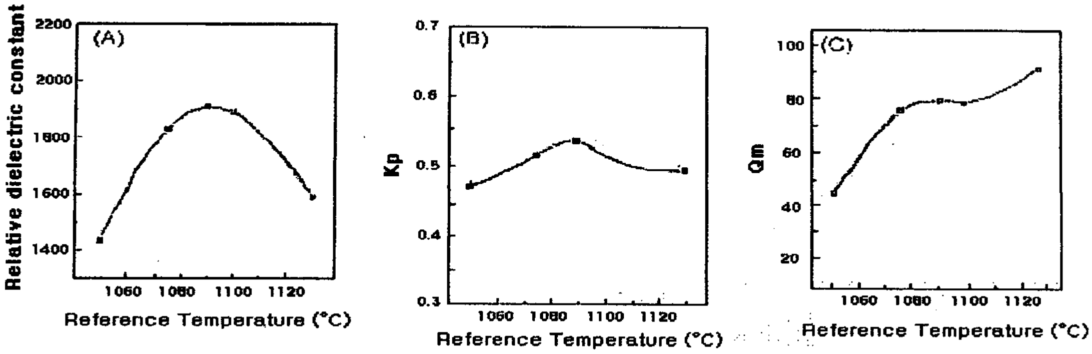

Fig. 5. Electrical properties of the microwave sintered specimen for 5 min vs reference sintering temperature (a) Relative dielectric constant, (b) Electro-mechanical coupling factor and (c) Mechanical quality factor

In order to achieve a high dielectric constant and electro-mechanical coupling factor, it is crucial that the sintered body should be dense and that the material composition be uniquely composed of the ferroelectric perovskite phase. In this study, the specimen sintered above 1090℃ corresponded to the highest dielectric constant. The specimen sintered at 1050℃ exhibited a porous microstructure which is most likely contributed their low dielectric constant. Likewise, the specimen sintered above 1090℃ showed a decrease in the dielectric constant. This phenomenon was attributed to the PbO volatilization and the formation of second phase, as discussed previously. The relative dielectric constant, mechanical quality factor and electro-mechanical coupling factor of the PZT specimens microwave sintered at 1090℃ were 1900, 80, 0.53 respectively, respectively, as shown in Fig. 5, which is comparable to conventionally sintered values.

4. Conclusions

The microwave hybrid sintering method was successfully applied to PZT ceramic sintering without implementation a PbO atmosphere control. The microwave hybrid heating was adequately rapid to sinter the specimens with little PbO loss. Thus, PZT dielectrics with high density and comparably good electrical properties were obtained within a 20 minutes processing time.

References

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