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Preparation of microwave dielectric, $Sn_{0.2}Zr_{0.8}TiO_4$

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Abstract. A simple coprecipitation technique is described for the preparation of tin substituted zirconium titanate ceramic powders.

Keywords. Zirconium titanate; microwave dielectric; ZrO_2; TiO_2; SnO_2.

1. Introduction

Communication at microwave frequencies has lead to the proliferation of commercial wireless technologies such as cellular phones and global positioning systems (Hirano *et al* 1991; Huang and Weng 2000). Many kinds of dielectric materials have been developed for microwave applications (Heiao *et al* 1988; Christofferson *et al* 1994). Among them, zirconium titanate–stannate solid solutions, $Sn_xZr_{1-x}TiO_4$, are known to have a high dielectric constant, a high q value and a low temperature coefficient of resonant frequency (Wakino *et al* 1984; Huang and Weng 2000). Because of the difficulty in sintering this compound without additives, it has been sintered with additives such as ZnO and NiO. These additives, however, lead to the degradation of its dielectric properties. The three methods that are employed to reduce the sintering temperature of this compound are low melting glass additions, chemical processing and smaller particle sizes of starting materials. Hirano *et al* (1991) produced $Sn_xZr_{1-x}TiO_4$ ceramics and sintered at 1873 K for 3 h. Here we report a simple coprecipitation method to prepare fine powders of $Sn_xZr_{1-x}TiO_4$. To the best of our knowledge this method has not been described in the literature.

2. Experimental

Required quantity of $ZrOCl_2$ was weighed and dissolved in a solution containing stoichiometric amount of $TiOCl_2$ and $SnOCl_2$, which were prepared by diluting $TiCl_4$ and $SnCl_4$ solutions with ice cold distilled water. To the above mixture a standard ammonia solution was added dropwise, on which precipitation began. The addition of ammonia solution was continued until pH was 10. This ensured the completion of precipitation. The precipitate was washed free of chloride ion, filtered and dried at 373 K for 12 h. The oven dried powder was subjected to differential thermal analysis (DTA)/thermogravimetric analysis (TGA) studies at a rate of 10 K per min in air using Rheometric Scientific STA 1500+ machine. Surface area measurements were done with a machine Nova 1200 using BET analysis. The powders were calcined at 1073 K and X-ray diffractograms were recorded using Philip 1730 X-ray diffractometer. Transmission electron microscopic (TEM) pictures were recorded with JEOL model 1200 EX instrument at the accelerating voltage of 100 kV. The powder samples were dispersed using amyl acetate on a carbon coated TEM copper grid.

3. Results and discussion

Figure 1 shows the X-ray diffractogram recorded for the samples calcined at 1073 K, which is the same as that

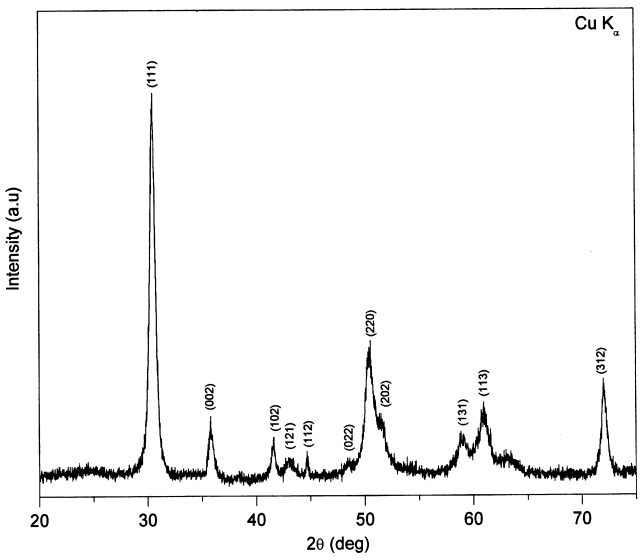

Figure 1. XRD of polycrystalline $Sn_{0.2}Zr_{0.8}TiO_4$.

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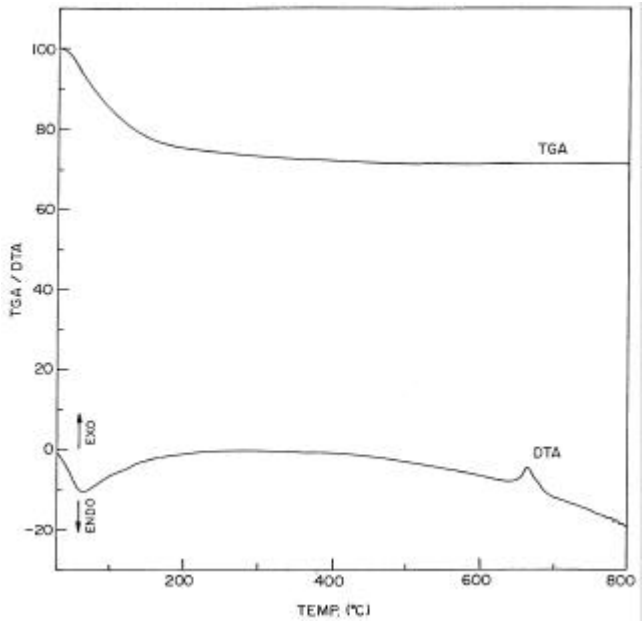

Figure 2. DTA/TGA of the oven-dried precipitate.

reported for $ZrTiO_4$ ceramics. No impurity lines were observed and well crystallized pattern was observed for samples annealed at 1073 K. The orthorhombic unit cell parameters calculated for this sample are $a = 4{\cdot}805$ Å, $b = 5{\cdot}031$ Å and $c = 5{\cdot}461$ Å. The ionic radius of Sn^{4+} being less than the Zr^{4+}, the unit cell parameters decreased with stannate substitution. It is stated that (Christofferson *et al* 1994) the role of Sn is to stabilize the interface between Zr-rich and Ti-rich domains, which form during the cation ordering transformation. The DTA/TGA curves recorded for the sample are depicted in figure 2. The weight loss up to 473 K corresponds to loss of water from the precipitate and further the weight remains constant as observed from figure 2. The exothermic peak at 675°C may indicate a possible recrystallization process occurring in the ceramics. The surface area of calcined

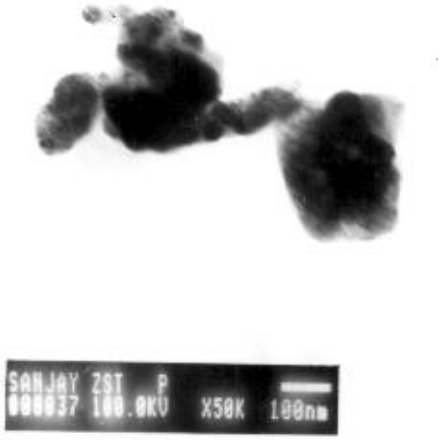

Figure 3. TEM photograph of powder sample, $Sn_{0{\cdot}2}Zr_{0{\cdot}8}TiO_4$.

powders is found to be 50 m^2/g. The average particle size is found to be 100 nm as illustrated in TEM photograph (figure 3) taken for the powder sample.

4. Conclusion

Fine polycrystalline single-phase microwave dielectric, $Sn_{0{\cdot}2}Zr_{0{\cdot}8}TiO_4$, was prepared by a simple coprecipitation method.

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