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Microwave Dielectric Properties of $MgTiO_3$ by Sintering MgO and TiO_2 Nanostructures

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1. Introduction

Magnesium titanate ($MgTiO_3$) is a well-known microwave dielectric ceramic material with wide applications such as in capacitors, resonators, filters, antennas for communication, radar, direct broadcasting satellite and global positioning system operating at microwave frequencies [1–4]. In order to control the combined dielectric properties of a high dielectric constant and a low dielectric loss, it is necessary to produce $MgTiO_3$ powders with the highest possible purity, well-defined particle morphology, and small particle size distribution. Therefore, much attention has been focused on the synthesis methods of $MgTiO_3$. Various synthesis methods were reported in the past decade, such as solid-state reaction method [4–6], thermal decomposition of peroxide precursors [7], chemical coprecipitation [8], mechanochemical complexation route [9], metalorganic chemical vapour deposition [10], metalorganic solution deposition technique [11] and sol–gel method [2,12,13]. Though these methods have shown successful results, a method to fabricate the magnesium titanate with high quality factor at a low sintering temperature is still desired.

2. Experimental details

The MgO and TiO_2 nanopowders were prepared separately by the chemical solution methods as described below. MgO: Magnesium acetate $(CH_3COO)_2Mg \cdot 4H_2O$ (Showa chemical, purity ~99%) was used as starting materials, and ethylene glycol (EG) (TEDIA company) was used as solvent. These materials used without further purification. Polyvinyl pyrrolidone (PVP, MW= 58000) (ACROS organic chemicals) employed as a capping agent. 1.68 grams of magnesium acetate and 9.1 grams of PVP were dissolved in 200 mL of EG. The solution was kept in a round bottom flask at 197°C and held for 4 h under ambient pressure. The cooled precipitates were collected by centrifugation at 6000 rpm, followed by washing with ethanol several times, and dried at 80°C for 8 h.

TiO_2: Five milligrams of commercial TiO_2 powders (Showa chemical, purity >99%) was added to (180 mL, 1 M) aqueous NaOH in a Teflon vessel, which was placed inside a sealed stainless vessel. The solution was heated to 130°C and held for 24 h. The cooled precipitates were collected, washed in DI water, and then washed using nitric acid solution until the value of pH was below 7, and dried at 80°C in air.

MgO: TiO_2 (1.23:1 molar ratio) nanopowders were mixed and ballmilled for 24 h with zirconia beads and distilled water. The milled mixtures were dried at 140°C for 12 h, ground, and sieved. The mixed powders were calcined at 900°C for 2 h. Polyvinyl alcohol (PVA) (2 wt %) solution was added as a binder. A disk with a diameter of 11 mm and a thickness of 5.5 mm was formed using uniaxial pressing. After debinding of PVA at 650°C for 4 h, the compacts were sintered for 4 h at elevated temperatures (1200, 1300, and 1400°C).

3. Discussions

Figure 1 shows the SEM images of the synthesized MgO and TiO_2 nanostructures. Insets in Fig. 1(a) and (b) shows the TEM images of the MgO and TiO_2, respectively. An MgO nanoparticles with a diameter of ~0.5 um was found to be consisted by a folded MgO sheet. The TiO_2 nanowires were hollow inside. Because of the fine structure, the specific surface area of the MgO and TiO_2 nanostructures measured by BET were 84.68 m^2/g and 219.733 m^2/g, respectively. Such high BET values ensure high reactivity for solid-state diffusion during subsequent heat-treatment.

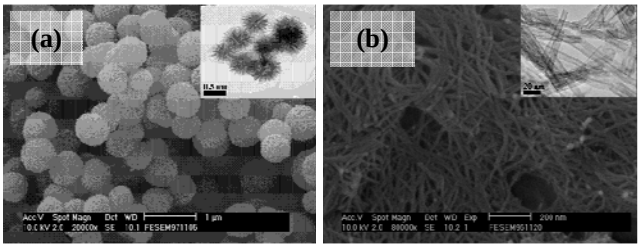

Fig. 1 SEM images of (a) MgO nanoparticles and (b) TiO_2 nanowire. Insets show the corresponding TEM images.

Figure 2 showed DTA-TGA curves for the MgO-TiO_2. Two distinct exothermal peaks were found in the vicinity of ~400 and 1200°C. The former one is attributed to the combustion of organics and the later one is to the phase transformation.

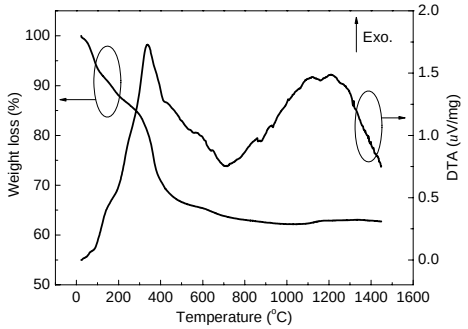

Fig.2 DTA-TGA curves for MgO-TiO_2.

Figure 3 (a) and (b) shows the XRD patterns of the MgO-TiO_2 calcined at 900°C for 2h and sintered at 1200-1400°C for 2h, respectively. The XRD of the calcined MgO-TiO_2 indicated the phases were composed of $MgTiO_3$, Mg_2TiO_4, and $MgTi_2O_5$. After sintering at 1200-1400°C, the lossy $MgTi_2O_5$ phase was absent.

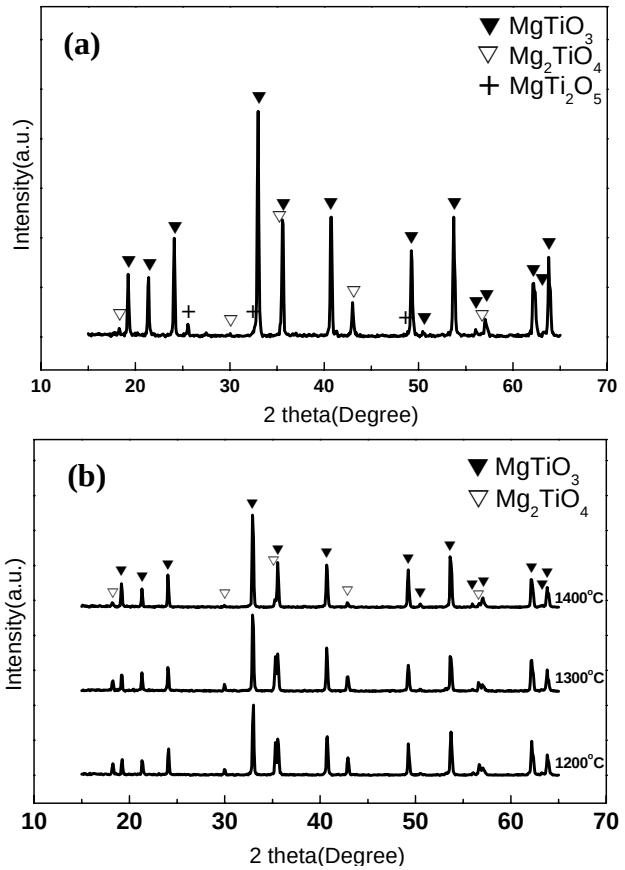

Fig.3 XRD of (a) calcined powders and (b) sintered ceramic compacts.

SEM images of the sintered magnesium titanate were shown in Fig. 4. The grain size increased with sintering temperature. As a result, the dielectric constant increased with the sintering temperature. The pores between inside the grain boundaries decreased with the sintering temperature. Because of the high sinter ability of the nano-size starting materials, the dielectric constant achieved a maximum at ~1300°C that is much lower than that ever been reported in literature. Most of all, 1 very large value of quality factor multiples its resonant frequency (~320000 GHz) was obtained at 1300°C. The Q*f of the $MgTiO_3$ or Mg_2TiO_4 is known to be around 16000 GHz. The obtained high Q*f value is associated with uniformly distributed grain size and dense compact. Furthermore, the sintering temperature needed to achieve such a high value is as low as ~1300°C, which is about 150°C lower than that prepared from the conventional solid-state sintering.

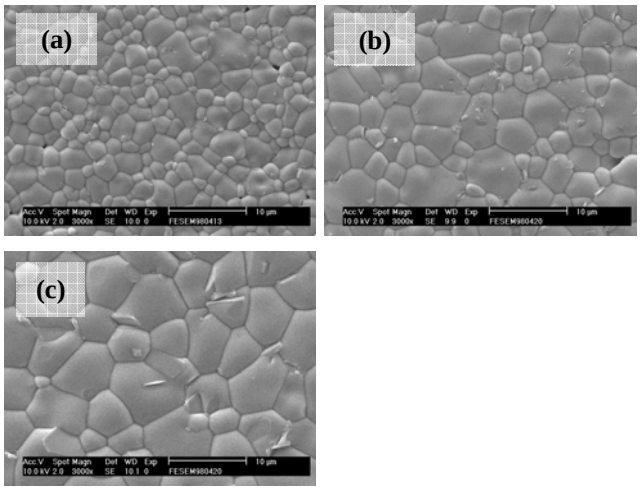

Fig.4 SEM images of magnesium titanate sintered at (a) 1200°C (b) 1300°C and (c) 1400°C.

4. Conclusion

We reported the solid-state sintering process by using the MgO nanopowders and TiO_2 nanowires as starting materials. The MgO nanopowders and TiO_2 nanowires were synthesized separately using chemical solution methods. Both of the MgO nanopowders and TiO_2 powders showed a high specific surface area by the BET measurement. The magnesium titanate sintered by MgO nanopowders and TiO_2 nanowires sample showed excellent microwave properties at a low sintering temperature. A very large value of quality factor multiples its resonant frequency (~320000 GHz) was obtained at a temperature as low as 1300°C. This study exploited the hybrid application of the MgO and TiO_2 nanostructures, which has great potential for application in ceramic processes in which a low sintering temperature is desired.

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