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Low-temperature sintering and microwave dielectric properties of Al_2TeO_6–TeO_2 ceramics

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ABSTRACT

We propose Al_2TeO_6–TeO_2 ceramics as a candidate for use as low-temperature co-fired ceramics (LTCC). We investigated microwave dielectric properties and low-temperature sintering conditions for Al_2TeO_6–TeO_2 ceramics. The calcined Al_2TeO_6 powders were sintered at 900 °C for 2–10 h with 30–50 wt% additive TeO_2. X-ray powder diffraction patterns showed that the sintered samples were Al_2TeO_6–TeO_2 composite with no other phase. The apparent density was improved with the additive TeO_2 content of up to 45 wt%. The dielectric constant (ε_r) increased by adding TeO_2 content from 35 to 45 wt%, although the quality factor ($Q \cdot f$) decreased. During sintering at 900 °C, the ε_r of the Al_2TeO_6–TeO_2 decreased slightly, whereas the $Q \cdot f$ increased gradually. The observed microstructures showed that the longer sintering time makes fewer pores in Al_2TeO_6–TeO_2 ceramics. Sintering at 900 °C for a long time contributes to densification, but it simultaneously causes TeO_2 evaporation. To prevent TeO_2 evaporation, we investigated the effects of annealing at 750 °C after sintering at 900 °C. Apparent densities or ε_r for the annealed samples were higher than those of the non-annealed samples. The $Q \cdot f$ improved with increasing annealing duration time, suggesting that sintering proceeded well during annealing with slower TeO_2 evaporation at 750 °C. The results show that annealing at 750 °C is effective to facilitate sintering and to control TeO_2 evaporation.

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1. Introduction

Recently, low-temperature co-fired ceramics (LTCC) have attracted attention for the development of next-generation miniaturized microwave/millimeterwave communication devices [1–10]. Using the LTCC, co-firing of dielectric ceramics with internal electrodes is possible, where silver is usually used for the electrode. Because the melting point of silver is approximately 961 °C in air [11], the sintering temperature for the dielectric ceramics must be lower than 961 °C. However, until now, almost all micro/millimeter-wave dielectric ceramics require sintering temperatures higher than 1000 °C to obtain a higher quality factor ($Q \cdot f$), resulting from good densification and crystallinity [2,12–16]. Higher $Q \cdot f$ is an important factor for higher-frequency telecommuting applications such as filters and resonators [2]. This study was conducted to develop new dielectric ceramics with a high quality factor, even though the sintering temperature of the ceramics is lower than 961 °C of the silver melting point.

We have proposed that ceramics based on Al_2TeO_6 are candidate compounds for the LTCC because they contain Te oxide, which is characterized by its low melting point. For example, the melting point of TeO_2 is approximately 730 °C [17]. Some studies have been reported that the Te based binaries such as TiO_2–TeO_2, Bi_2O_3–TeO_2, and CuO–TeO_2 can be densified by sintering at temperatures lower than 900 °C [3–6,10]. Additionally, the Al based oxides tend to show lower dielectric constant [2,13,15,16]. Lower dielectric constant in addition to lower dielectric loss is also important for miniaturization of the higher-frequency telecommuting devises [2]. Su et al. have described that the Al_2TeO_6 possesses both of low-dielectric constant (ε_r) and dielectric loss ($\tan\delta$) in the frequency range of 1 kHz–1 MHz [18]. The ε_r and the $\tan\delta$ at 1 MHz have been reported as 9.4 and 0.03, respectively. That fact implies that the Al_2TeO_6 is also characterized by low-dielectric constant and dielectric loss in the microwave frequency range. However, no reports in the relevant literature describe studies of microwave dielectric properties in the Al_2TeO_6.

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Given that perspective, we tried earlier to prepare Al_2TeO_6 ceramic samples as a pilot study, but it was difficult to obtain dense ceramics, when the sample was sintered at temperatures below 960 °C. Not sintered sample, but the pressed sample was used to investigate the dielectric properties in the frequency range of 1 kHz–1 MHz in the literature [18]. It also presumes that preparing the sintered Al_2TeO_6 is difficult, because dielectric properties are usually investigated by using a sintered sample. Particularly, densification is more significant for the quality factor in the microwave frequency range. To overcome the difficulty, this study was undertaken to prepare dense Al_2TeO_6 ceramics by adding TeO_2 with the lower melting point. We investigated the low-sintering condition and crystallinity of Al_2TeO_6–TeO_2 ceramics. Then we investigated the microwave dielectric properties of the prepared samples and discussed relation between the sintering conditions and the microwave dielectric properties.

2. Experimental procedures

Al_2O_3 (purity: 99.99%) and TeO_2 (purity: 99.9%) were weighed as the mole ratio of 1:1. They were ball-milled for 24 h in ethanol solvent using alumina balls with 5 mmϕ diameter. After drying, the mixed powders were calcined in air at 550–650 °C for 10 h. The calcined Al_2O_3–TeO_2 powders were mixed with additional TeO_2 of 30–50 wt% and were again ball-milled for 24 h. The powders were molded into pellets with uniaxial pressure of approximately 7.8 MPa. Then cold isostatic pressing (CIP) of the molded samples was conducted under pressure of 200 MPa. The obtained pellets were sintered at 900 °C for 2–10 h in air. Some pellets were annealed at 750 °C for 24 h after sintering at 900 °C for 2 h.

The crystalline phase of the prepared sample was characterized at room temperature using X-ray powder diffraction (XRPD; X'pert MPD Pro; PANalytical B.V.) with Cu Kα radiation, where the conditions of XRPD voltage and current were respectively 45 kV and 40 mA. The apparent densities of the prepared samples were measured using Archimedes' method. The microstructure images of the prepared samples were investigated using a scanning electron microscope (SEM; JSM-6330F; JEOL Inc.). Differential thermal analysis (DTA) and thermogravimetry (TG) were conducted at temperatures of 20–900 °C with a rate of 10 °C/min to investigate the weight change and the melting point of TeO_2 in the samples.

The microwave dielectric properties of the prepared samples were investigated using the Hakki and Coleman method [2,19], for which a pellet sample was positioned between two copper plates. The microwave signal from the sample was investigated using a network analyzer (8720ES; Agilent Technologies Inc.). The dielectric constant (ε_r) and the quality factor ($Q \cdot f$) were calculated using the TE_{011} resonant mode investigated at room temperature [2,19]. The temperature coefficient of the resonator frequency (τ_f) was investigated at temperatures of 20–80 °C.

3. Results and discussion

3.1. Calcination condition to synthesize Al_2TeO_6

XRPD patterns of the Al_2O_3–TeO_2 powders calcined at 550–650 °C for 10 h are presented in Fig. 1. When the calcination temperature was 550 °C, reflections corresponding to Al_2O_3 and tetragonal TeO_2 were observed. At the calcination temperature of 600 °C, reflections corresponding to Al_2TeO_6 appeared in addition to the Al_2O_3 and the TeO_2 reflections. With higher calcination temperatures, the reflection intensities of the TeO_2 and the Al_2O_3 decreased, whereas the Al_2TeO_6 reflection intensities increased. When the calcination temperature was 620 °C, only Al_2TeO_6 reflections were observed, indicating that single phase of Al_2TeO_6 is obtainable at this calcination temperature. The reported TGA has showed an endothermal peak related to the formation of Al_2TeO_6 at 620 °C [18], which is good agreement with this XRPD result. We confirmed that a broad XRPD peak was observed around $2\theta = 25°$, coming from the presence of TeO_2 glass, when the Al_2O_3–TeO_2 powders were sintered at 650 °C. Consequently, we concluded that 620 °C is an appropriate condition for the calcination temperature for Al_2TeO_6.

3.2. Sintering condition to prepare dense Al_2TeO_6–TeO_2 ceramic samples

XRPD patterns of the sintered Al_2TeO_6–TeO_2 samples are portrayed in Fig. 2, where the samples were prepared by sintering the calcined Al_2TeO_6 powders with additive 30–50 wt% TeO_2 at 900 °C for 2 h. The calcined temperature was 620 °C, as described above. The observed reflections of the sintered samples can be indexed by two phases: Al_2TeO_6 and tetragonal TeO_2. With increasing additive TeO_2 contents, the TeO_2 reflection intensities increased slightly.

XRPD patterns of the Al_2TeO_6 with additive 40 wt% TeO_2 sintered at 900 °C for 2–10 h are depicted in Fig. 3. Reflections corresponding to Al_2TeO_6 and tetragonal TeO_2 were observed. With longer sintering duration time, the reflection intensities of the TeO_2 decreased gradually, suggesting that the TeO_2 is evaporated during sintering at 900 °C.

TG and DTA of the Al_2TeO_6 with additive 40 wt% TeO_2 are presented in Fig. 4. In the DTA, the endothermic peak was observed at 707 °C, at which point the sample weight began to decrease. The results demonstrate that TeO_2 contained in the sample begins to melt at 707 °C.

The relations between apparent densities and the sample preparation conditions ((1) additive TeO_2 amount and (2) sintering duration time) are presented in Fig. 5. It is difficult to estimate the relative densities in the Al_2TeO_6–TeO_2 composite series because TeO_2 is evaporated during sintering. In the case of the additive 30 wt% TeO_2, the apparent density was the lowest among the samples in the case of additive TeO_2 of 30–50 wt%, when the sintering duration time was 2 h. With increasing the additive TeO_2 from 30 wt% to 45 wt%, the apparent density increased. The

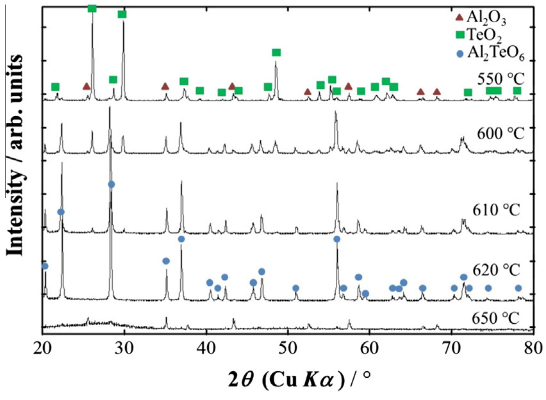

Fig. 1. XRPD patterns of the Al_2O_3–TeO_2 calcined for 10 h at 550–650 °C.

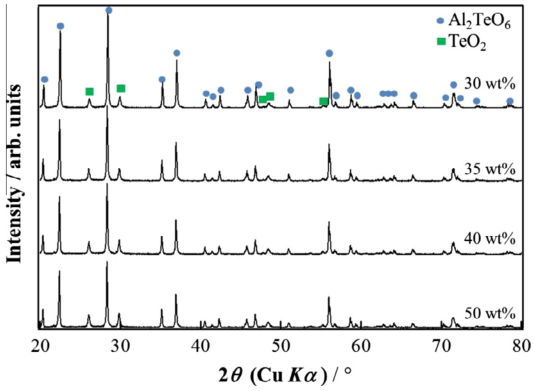

Fig. 2. XRPD patterns of the Al_2TeO_6 sintered for 2 h with additive TeO_2 of 30–50 wt% at 900 °C.

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