Millimeter-wave Dielectric Ceramics of Alumina and Forsterite with High Quality Factor and Low Dielectric Constant

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ABSTRACT

Millimeter-wave dielectric ceramics have been used like applications for ultrahigh speed wireless LAN because it reduces the resources of electromagnetic wave, and Intelligent Transport System (ITS) because of straight propagation wave. For millimeter-wave, the dielectric ceramics with high quality factor (Q·f), low dielectric constant (εr), and nearly zero temperature coefficient of resonant frequency (τf) are needed. No microwave dielectric ceramics with these three properties exist except Ba(Mg1/3Ta2/3)O3 (BMT), which has a little high εr. In this paper, alumina (Al2O3) and forsterite (MgSiO3), candidates for millimeter-wave applications, were studied with an objective to get high Q·f and nearly zero τf. For alumina ceramics, Q·f more than 680,000 GHz was obtained but it was difficult to obtain nearly zero τf. On the other hand, for forsterite ceramics, Q·f was achieved from 10,000 GHz of commercial forsterite to 240,000 GHz of highly purified MgO and SiO2 raw materials, and τf was reduced a few by adding TiO2 with high positive τf.

Key words: Millimeter-wave dielectric materials, Microwave materials, Alumina, Forsterite, Quality factor

1. Introduction

Recently, microwave telecommunication has been developed for wide applications, such as mobile phone, wireless LAN and Intelligent Transport System (ITS). Microwave dielectric materials are expected to be developed for variety of application including miniaturization for mobile phone, transmitter and receiver with high performance for base station, and ultrahigh speed wireless LAN and ITS for millimeter wave range. The development trend of microwave dielectric materials is schematically shown in Fig. 1. In this figure, Quality factors (Q·f) measured in variety of microwave dielectric materials are shown as a function of their dielectric constants (εr). Both Q·f and εr are two of three key factors describing dielectric property. Q is inverse of the dielectric loss (tan δ) and large εr shortens wavelength, as followed by the relation: \( \lambda = \frac{c}{f \sqrt{\varepsilon_r}} \). The temperature coefficient of resonant frequency (τf) is the other key factor describing microwave dielectric property. τf is expected to be close to zero.

In particular, millimeter-wave dielectric ceramics have been recently attracted much attention. They are planned to be applied in ultrahigh speed wireless LAN, because they can reduce the resource of electromagnetic wave. Millimeter-wave dielectric ceramics are also planned to be used in the ITS including car anti-collision system etc., by using excellent wave property of straight propagation. Further, there are much needs to reduce cross-coupling between microstrip lines on circuit boards, and to minimize signal delay times. The signal propagation speed is inversely proportional to εr of IC dielectric substrate, so that performance is enhanced as εr decreases. For these applications, dielectric ceramics with high Q·f and low εr are strongly needed, and nearly zero τf is also needed for more high quality applications. None of the presently available microwave dielectric ceramics can satisfy all of these three factors.

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except for Ba(Mg_{0.5}Ta_{0.5})O_3 (BMT) which shows a little higher $\varepsilon_r$ than expected for millimeter-wave application.

Alumina ($\text{Al}_2\text{O}_3$) and forsterite ($\text{Mg}_3\text{Si}_2\text{O}_7$) ceramics, which show much lower $\varepsilon_r$ than BMT, are also candidates for millimeter-wave applications. Especially, alumina ceramics has ultrahigh Q-f of 360,000 GHz and raw $\varepsilon_r$ of 9.8. Moreover, alumina has been used as insulator such as substrate and package materials because of its mechanical hardness. On the other hand, forsterite has lower $\varepsilon_r$ than alumina. Forsterite ceramics have been used as electronic parts such as resonator core, electron tube stem, base parts supporting dielectric resonator etc., because of the low $\varepsilon_r$ and high insulation resistance even in the microwave frequency range. Therefore, it is also expected as a candidate for the application to IC dielectric substrate.

In this study, alumina and forsterite ceramics were synthesized by using high-purity and fine-grain-size raw powders to obtain excellent materials with high Q-f and nearly zero $\tau_t$. On the other hand new designed forsterite ceramics were synthesized by using highly purified MgO and SiO_2 raw materials. Moreover, reduction in $\tau_t$ was attempted by adding a few amounts of TiO_2 with high positive $\tau_t$ into forsterite ceramics.

2. Experimental Procedures

High purity (99.99%) and fine grain (170 mm) powder, Tasmicron TM-DAR (Taiimei Chemical Co., Ltd.) was used for synthesis of alumina ceramics. Pellets with diameter of 14 mm were formed by uni-axial pressure of 98 MPa after wet ball-milling of the powder and sintered at temperature from 1350 to 1600°C for 5 h in air.

To prepare forsterite ceramics, high purity (99.99%) MgO powder manufactured by gas-phase oxidation process of magnesium was used. The powder showed single crystal grains with fine particle size distribution in the range of 0.08 to 0.1 mm and large specific surface area of 2,600 m^2kg^-1.

High purity SiO_2 powder was used for synthesis of forsterite ceramics. The powder has 99.8% purity and particle size of 1.6 mm and specific surface area of 4,000 m^2kg^-1.

These powders mixed in an urethane ball mill for 20 h in distilled water, then freeze-dried. The powders were calcined at 1200°C for 3 h. The calcined powders were again ball-milled for 24 h to produce forsteritic powders with an average particle diameter of about 1 mm. The pellets with diameter of 15 mm were formed by CIP at 300 MPa. The pellets were sintered at temperature from 1300 to 1450°C for 2 h.

The crystalline phases of sintered pellets are investigated by X-ray powder diffraction. The relative density was measured by the Archimedes' method. The microstructure was observed by a Scanning Electron Microscope (SEM). The microwave dielectric properties ($\varepsilon_r$, Q-f, and $\tau_t$) of alumina ceramics were estimated using a pair of parallel conducting Ag plates in the $\text{TE}_{01}$ mode, using Hakki and Coleman's method, and those of forsterite also at 23 GHz using JIS R 1627-1996, by network analyzer.

Fig. 2. Microwave dielectric properties of alumina ceramics obtained.

3. Results and Discussion

3.1. Alumina Ceramics

Microwave dielectric property of alumina ceramics as a function of sintering temperature is shown in Fig. 2. Q-f value becomes higher as the temperature increases. In the temperature range higher than 1500°C, the large Q-f value larger than 600,000 GHz is observed. The highest Q-f value of 680,000 GHz was obtained in the temperature range of 1550 to 1650°C and in the case of 5 h sintering. This value is the highest Q-f in the world as far as we know for single-phase alumina ceramics, although the Q-f value of 1,000,000 GHz was reported in alumina single crystal. High Q in the alumina ceramics is considered to result from high purity and high density. Relative density and SEM micrographs as a function of sintering temperature are shown in Figs. 3 and 4, respectively. High relative density more than 99.5% was obtained for each sample, but grain growth was observed in the sample sintered at 1600°C, compared with the sample sintered at 1350°C. On the other hand, $\varepsilon_r$ of 10.05 and $\tau_t$ of -60 ppm/°C are not affected by different sintering temperature.

3.2. Forsterite Ceramics

Q-f values of forsterite ceramics as a function of sintering temperature are shown in Fig. 5. The highest Q-f of 240,000 GHz was obtained at sintering temperature of 1360°C. This
Fig. 3. Relative densities and grain sizes of alumina ceramics obtained.

![Graph showing the relationship between sintering temperature and grain size.]

Fig. 4. SEM photographs of alumina ceramics obtained with different sintering temperature in which the grain sizes increase according to sintering temperature.

![SEM photographs at 1350°C, 1400°C, 1500°C, and 1600°C.]

The Q-f value is much higher than that of commercially available forsterite ceramics as shown in Fig. 6. The density of the sintered sample showed 96–98% of theoretical density. The sample was single crystalline phase of forsterite with no glassy phase at the grain boundaries, which is believed to reduce dielectric loss significantly into the low level measured in the single crystal. Its SEM micrograph is shown in Fig. 7. The grain growth improved the Q-f in fine forsterite ceramics. For comparison, a micrograph of commercially available forsterite is also shown in this figure. The grain boundary observed in the commercially available forsterite is thick and includes many impurities and glassy phases, which resulted in large dielectric loss. The dielectric constants are smaller than 6.90 which is expected to bring high speed electromagnetic-wave propagation, protecting delay of signal propagation.

The temperature coefficient of resonant frequency $\tau_f$ of forsterite was estimated to be $-67$ ppm/°C which was calculated using following equation:

$$\tau_f = -(\alpha + \tau_f/2)$$

(1)
where, $\tau_0$ is temperature coefficient of dielectric constant ($\varepsilon$) and $\alpha$ is thermal expansion coefficient. They were 116 ppm/$^\circ$C and $9.4 \times 10^{-6}$ in the range of 25~700$^\circ$C, respectively. The $\tau_0$ value was attempted to be reduced close to zero by means of adding rutile (TiO$_2$) having positive $\tau_0$ of 450 ppm/$^\circ$C. However, $\tau_0$ could be reduced only a little amount from ~67 ppm/$^\circ$C to ~63 ppm/$^\circ$C, even at a large amount (30 wt%) of TiO$_2$ addition. This result comes from phase equilibrium in the MgO-SiO$_2$-TiO$_2$ ternary system. The composition with 30 wt%TiO$_2$ locates in a Mg$_2$SiO$_4$-MgTi$_2$O$_5$-ternary subsystem, then TiO$_2$ disappeared. For zero $\tau_0$ resonator, the sintering processing should be considered.

4. Conclusions

Alumina and forsterite ceramics for candidates millimeter-wave dielectrics with high Q and low $\varepsilon$, were presented in this paper. Q-factors of alumina ceramics were improved to 680,000 GHz by using high purity and fine grain size alumina raw materials. Forsterite ceramics were also improved to 240,000 GHz by using MgO raw material. Dielectric constant of forsterite is 7.8 smaller than alumina ceramics. Low dielectric constant reduces delay of signal propagation. Moreover, we tried temperature coefficient of resonant frequency ($\tau_0$) to near zero $\tau_0$ by means of adding TiO$_2$ with high positive $\tau_0$. But, $\tau_0$ did not improved because of TiO$_2$ disappeared during reaction based on phase equilibrium. Ceramics with near zero $\tau_0$ should be created near future.

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