

Effects of Ceramics on Dielectric Properties of Polystyrene / Ceramics Composites at Microwave Frequencies

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ABSTRACT

Dependencies of dielectric properties on MgTa₂O₆, MgNb₂O₆, and MgWO₄ (Mg-based ceramics) fillers of the polystyrene (PS) matrix composites were investigated as a function of frequency. With increasing frequency from 1 GHz to 7.3 GHz, the dielectric constant (K) of the composites was not changed significantly, while the dielectric loss ($\tan\delta$) of the composites was slightly decreased. The K , $\tan\delta$, and temperature coefficient of resonant frequency (TCF) of the composites were dependent on the type and amount of ceramics at 11 GHz. Also, several theoretical models have been employed to predict the effective dielectric constant of the composites and the results were compared with experimental data. Typically, a K value of 6.67, $\tan\delta$ of 0.56×10^{-3} , and TCF of -4.99 ppm/°C were obtained for the PS composites with 0.4 volume fraction of MgNb₂O₆ at 11 GHz.

Key words: Dielectric properties, Composites, Polystyrene, Mg-based ceramics

1. Introduction

Combining the dielectric properties of ceramics and the chemical stability, mechanical flexibility, and processing possibility of polymers, polymer / ceramic composites consisting of ceramic particles in a polymer matrix have been widely studied for microwave substrate and electronic packaging applications. For high frequency applications and resonant frequency selectivity of microwave devices, these composites should have a low dielectric constant (K) in order to reduce the signal propagation delay and low dielectric loss ($\tan\delta$) for better device performance.

Among various polymers, polystyrene (PS) was selected in this study as a polymer matrix due to its low processing temperature (160°C), good flexibility, and excellent dielectric properties ($K = 2.14$, $\tan\delta = 4 \times 10^{-4}$ at 8 GHz).¹⁾ Most studies on PS matrix composites have focused on improving the dielectric properties of the matrix through the addition of a ceramic filler with superior dielectric properties. Khastgir *et al.*²⁾ and Yu *et al.*³⁾ reported on the dielectric properties of PS / TiO₂ and PS / AlN composites, respectively. However, the microwave dielectric properties of PS / TiO₂ composites were not reported and the PS / AlN composites could not be applied to various microwave devices due to the low K of the AlN ceramics. Although the dielectric properties of PS / BaTiO₃ composites were reported recently,^{4,5)} the higher $\tan\delta$ of these composites precludes their practical application. Therefore, the dielectric properties of PS composites

filled with ceramics with various K and low $\tan\delta$ should be investigated at microwave frequencies.

In our preliminary results,⁶⁾ Mg-based ceramics including MgTa₂O₆, MgNb₂O₆, and MgWO₄ were found to have high Qf (quality factor, $Q = 1 / \tan\delta$) values (MgTa₂O₆ = 166,032 GHz, MgNb₂O₆ = 108,882 GHz, MgWO₄ = 23,068 GHz), along with different K values (MgTa₂O₆ = 25.48, MgNb₂O₆ = 20.84, MgWO₄ = 12.44) and large variation of the temperature coefficient of resonant frequency (TCF) values (MgTa₂O₆ = 24.35 ppm/°C, MgNb₂O₆ = -63.17 ppm/°C, MgWO₄ = -56.6 ppm/°C). Therefore, these Mg-based ceramics were adapted as fillers of the composites in this study in order to investigate the effects of ceramic fillers with various dielectric properties on the microwave dielectric properties of PS / ceramic composites.

In this study, the dependences of dielectric properties on type and amount of ceramic fillers of PS / Mg-based ceramic composites were investigated at microwave frequencies. The experimental K of the composites was compared with the effective K calculated by several theoretical models. The TCF of the composites was also discussed in order to assess the thermal stability of the composites.

2. Experimental Procedure

Oxide powders with high-purity above 99.9% were used as starting materials. The powders were weighed according to the compositions of MgTa₂O₆, MgNb₂O₆, and MgWO₄, and then milled with ZrO₂ balls for 24 h in ethanol. To obtain the single phase of each ceramic composition and the optimal microwave dielectric properties of the composites, MgWO₄ was double calcined at 700°C and 1100°C for 3 h, while MgTa₂O₆ and MgNb₂O₆ were double calcined at

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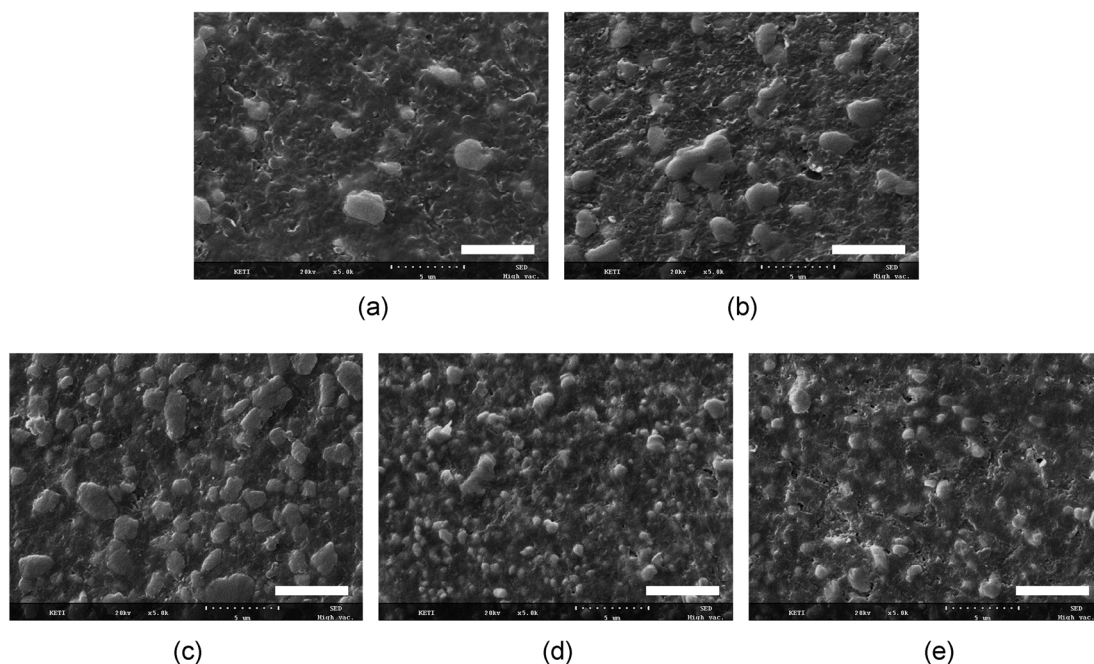


Fig. 1. SEM micrographs of polystyrene / Mg-based ceramic composites with various volume fractions (V_f) of ceramics; (a) $0.2 V_f$ MgNb_2O_6 , (b) $0.4 V_f$ MgNb_2O_6 , (c) $0.6 V_f$ MgNb_2O_6 , (d) $0.4 V_f$ MgTa_2O_6 , and (e) $0.4 V_f$ MgWO_4 (bar = 5 μm).

1100°C and 1350°C for 3 h, respectively. The calcined powders were re-milled with ZrO_2 balls for 24 h in ethanol and then dried. Polystyrene (PS) with molecular weight of 280,000 (gel permeation chromatography (GPC)) was weighed and placed into the chamber of a Torque Rheometer (Rheomix 600p, ThermoHaake, Germany) at 200°C, followed by slow addition of MgTa_2O_6 , MgNb_2O_6 , and MgWO_4 powders. After mixing at a speed of 50 rpm for 1-2 h, the PS / ceramic composites were removed from the chamber and hot-pressed at 200°C and 15 MPa for 1 h.

The Microstructure of the specimens was observed using a scanning electron microscope (SEM, SNE-1500M, SEC, Korea). The apparent density of the composites was measured by the Archimedes method. The relative density was obtained from the theoretical values by the mixing rule.⁷⁾ Powder X-ray diffraction analysis (XRD, D/Max-3C, RIGAKU, Japan) was carried out to determine the crystalline phases of the composites. The dielectric properties were measured by the open-ended coaxial resonator probes method⁸⁾ at a frequency range from 1 GHz to 7.3 GHz. At 11 GHz, the dielectric properties of the specimens were measured by the Hakki and Coleman method.⁹⁾ The temperature coefficient of resonant frequency (TCF) was measured by the cavity method¹⁰⁾ in a temperature range from 25°C to 80°C.

3. Results and Discussion

3.1. Physical properties of the composites

SEM micrographs of polystyrene (PS) / MgTa_2O_6 , MgNb_2O_6 , and/or MgWO_4 (Mg-based ceramics) composites are shown in Fig. 1. Mg-based ceramic particles were uniformly dis-

persed in the PS matrix and the melted PS completely covered the ceramic particles, which induced low porosity of the composites. With increasing ceramic content (Figs. 1 (a)-(c)), the interparticle distance was decreased, while the connectivity among the ceramic particles and the interfacial areas between PS and ceramics were increased. For the composites with 0.4 volume fraction (V_f) of ceramics (Figs. 1(b), (d), (e)), the particle size of MgNb_2O_6 was larger than those of MgWO_4 and/or MgTa_2O_6 and the interface areas between PS and the ceramics of the PS / MgTa_2O_6 composites was larger than those of the PS / MgWO_4 and/or PS / MgNb_2O_6 composites due to the larger specific surface area by the difference of particle size.

Fig. 2 shows the apparent and relative densities of PS / Mg-based ceramic composites with various V_f of ceramics. The apparent density of the composites was increased with increasing ceramic content. For the composites with equal content of ceramics, the apparent density of the PS / MgTa_2O_6 composites was larger than those of the PS / MgWO_4 and/or PS / MgNb_2O_6 composites. These results are due to the density values of individual components ($\text{MgTa}_2\text{O}_6 = 7.816 \text{ g/cm}^3$, $\text{MgWO}_4 = 6.893 \text{ g/cm}^3$, $\text{MgNb}_2\text{O}_6 = 4.995 \text{ g/cm}^3$, PS = 1.05 g/cm^3). The relative density of the composites was slightly decreased with the ceramic content due to the increase of porosity by weak adhesion at the interface between PS and the ceramics. For the composites with the same content of ceramics, the relative density of PS / MgTa_2O_6 composites was much smaller than those of the PS / MgNb_2O_6 and/or PS / MgWO_4 composites. This reflects the larger porosity of the composites with MgTa_2O_6 relative to those of the other composites due to the larger interface

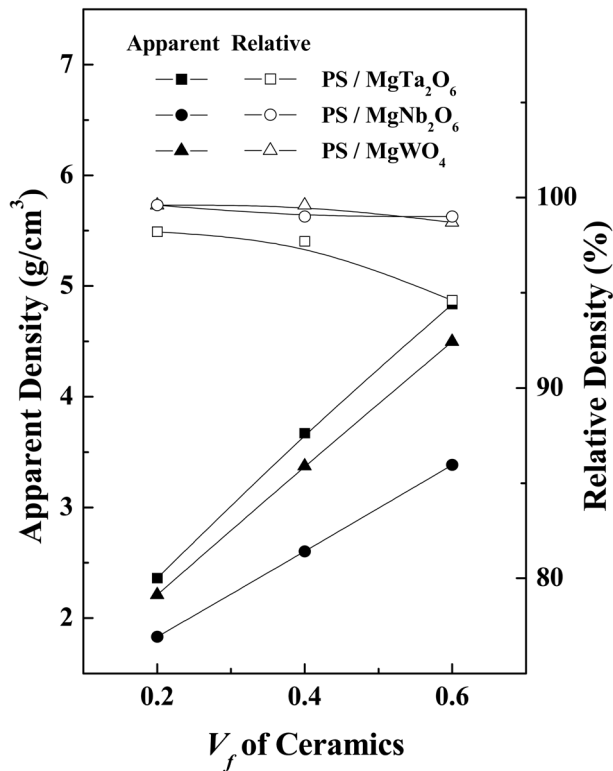


Fig. 2. Apparent and relative densities of polystyrene (PS) / Mg-based ceramic composites with various volume fractions (V_f) of ceramics.

areas between PS and the ceramics, as shown in Figs. 1(b), (d), and (e). However, all of the composites showed higher relative densities than 95% of the theoretical values, and therefore the effects of density on the microwave dielectric properties of the composites can be ignored.¹¹⁾

Fig. 3 shows the XRD patterns of the PS / Mg-based ceramic composites with the V_f of the ceramics. The XRD pattern of PS showed a typical amorphous halo (Fig. 3(a)). With increasing MgNb_2O_6 content (Figs. 3(b)-(d)), the amorphous halo of PS around $2\theta = 20^\circ$ was decreased, while the intensity of the MgNb_2O_6 crystalline phase was increased. Although the broadness of the XRD peak with MgNb_2O_6 content was largely unchanged, the XRD peak intensities of amorphous PS were remarkably decreased with increasing MgNb_2O_6 content. This is attributed to the higher integral intensity of MgNb_2O_6 relative to that of PS. A similar tendency of the XRD patterns was confirmed for the PS / MgTa_2O_6 and PS / MgWO_4 composites. For the composites of PS with the same volume fraction ($0.4 V_f$) of MgNb_2O_6 , MgTa_2O_6 , and/or MgWO_4 (Figs. 3(d)-(f)), single crystalline phases of orthorhombic columbite (MgNb_2O_6), tetragonal tri-rutile (MgTa_2O_6), and/or monoclinic wolframite (MgWO_4) structures were confirmed, respectively. Based on the XRD patterns of the specimens, reaction compounds between PS and the Mg-based ceramics were not detected, and, in turn, chemical reactions between PS and the Mg-based ceramics were not observed.

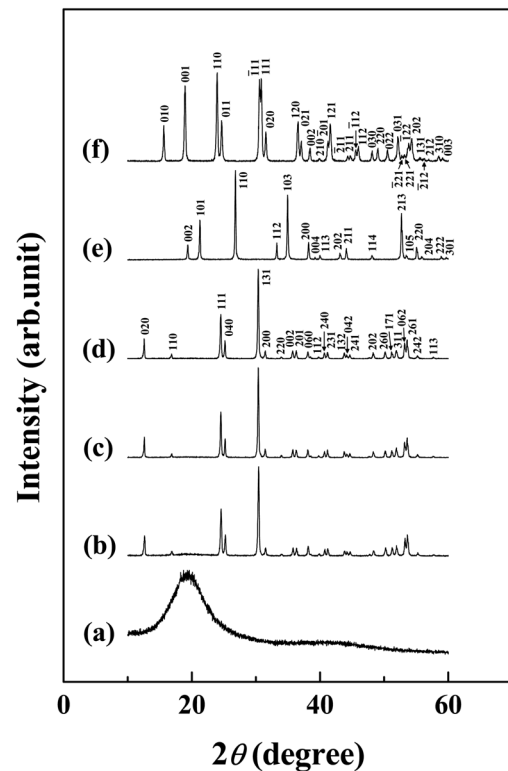


Fig. 3. X-ray diffraction patterns of (a) polystyrene (PS) and PS / Mg-based ceramic composites with various volume fractions (V_f) of ceramics ((b) $0.2 V_f \text{MgNb}_2\text{O}_6$, (c) $0.4 V_f \text{MgNb}_2\text{O}_6$, (d) $0.6 V_f \text{MgNb}_2\text{O}_6$, (e) $0.6 V_f \text{MgTa}_2\text{O}_6$, and (f) $0.6 V_f \text{MgWO}_4$).

3.2. Dielectric properties of the composites

Fig. 4 shows the dependences of the dielectric constant (K) on the frequency (1-7.3 GHz, at $0.2-0.6 V_f$) and various V_f of the ceramics (at 11 GHz) of the PS / Mg-based ceramic composites, respectively. For all of the composites with various V_f values of the ceramics, K showed a nearly constant value in the frequency range from 1 GHz to 7.3 GHz, as shown in Fig. 4(a). However, the K of the composites was increased with the ceramic content at the same frequency, due to the higher K of the ceramics than that of PS. As shown in Fig. 4(b), the K of the composites was also increased with the ceramic content at 11 GHz. For the composites with the same content of ceramics, the K of the PS / MgTa_2O_6 composites was higher than those of the PS / MgNb_2O_6 and/or PS / MgWO_4 composites due to the difference of the K values of the Mg-based ceramics ($\text{MgTa}_2\text{O}_6 = 25.48$, $\text{MgNb}_2\text{O}_6 = 20.84$, $\text{MgWO}_4 = 12.44$). Comparing the K values of the composites at 1-7.3 GHz (Fig. 4(a)), good frequency stability as a function of the K of the PS / Mg-based ceramic composites was confirmed over a wide frequency range, although the K values of the composites with the same content of ceramics were slightly decreased at 11 GHz (Fig. 4(b)).

For the effective design of polymer / ceramic composites from the view point of electronic packaging and substrate applications, the precise prediction of the effective K of the

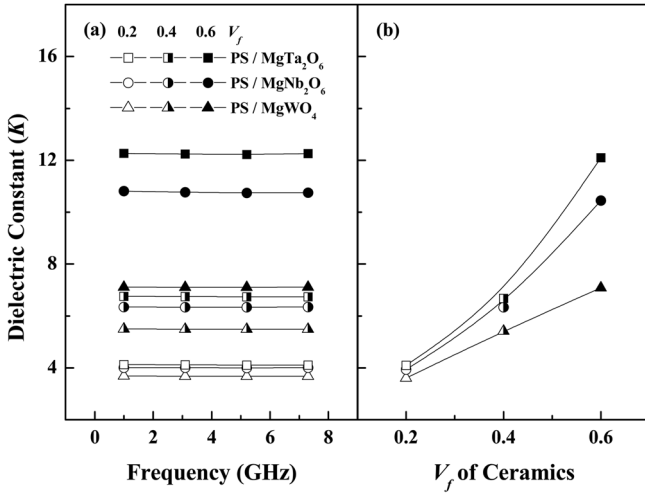


Fig. 4. Dependences of dielectric constant (K) on (a) frequency (1-7.3 GHz, at 0.2-0.6 volume fraction (V_f)) and (b) various V_f of ceramics (at 11 GHz) of polystyrene (PS) / Mg-based ceramic composites.

composites from the K and V_f (volume fraction) of each component is very important. Generally, the effective K of the composites is dependent on the K and the amount of ceramics, because the K of the ceramics is higher than that of the polymer. Also, the shape, size, and spatial arrangement of ceramic fillers have decisive effects on the effective K of the composite. Many theoretical models have been developed to describe the effective K of composites in which fillers with various shapes and sizes are distributed randomly. The effective K of the composites was calculated by conventional theoretical models, such as Maxwell-Garnett¹²⁾ (Eq. (1)), Lichtenecker¹³⁾ (Eq. (2)), Bruggeman¹⁴⁾ (Eq. (3)), Jayasundere-Smith¹⁵⁾ (Eq. (4)), Effective Medium Theory (EMT)¹³⁾ (Eq. (5)), and Poon-Shin¹⁶⁾ (Eq. (6)).

$$K_{eff} = K_m + 3V_f K_m \frac{K_f - K_m}{K_f + 2K_m - V_f(K_f - K_m)} \quad (1)$$

$$\ln K_{eff} = V_f \ln K_f + V_m \ln K_m \quad (2)$$

$$V_f \frac{K_f - K_{eff}}{K_f + 2K_{eff}} + V_m \frac{K_m - K_{eff}}{K_m + 2K_{eff}} = 0 \quad (3)$$

$$K_{eff} = \frac{K_m V_m + K_f V_f [3K_m / (K_f + 2K_m)] [1 + 3V_f (K_f - K_m) / (K_f + 2K_m)]}{V_m + V_f [3K_m / (K_f + 2K_m)] [1 + 3V_f (K_f - K_m) / (K_f + 2K_m)]} \quad (4)$$

$$K_{eff} = K_m \left[1 + \frac{V_f (K_f - K_m)}{K_m + n V_m (K_f - K_m)} \right] \quad (5)$$

$$K_{eff} = K_m \left[1 + \frac{V_f (K_f / K_m - 1)}{V_f + (V_m / 3) (K_f V_m / K_m + V_f + 2)} \right] \quad (6)$$

where K_m , K_f , V_m , and V_f are the dielectric constants and the volume fractions of the polymer matrix and ceramic filler, respectively, K_{eff} is the effective dielectric constant of the composites, and n is the fitting parameter or the morphol-

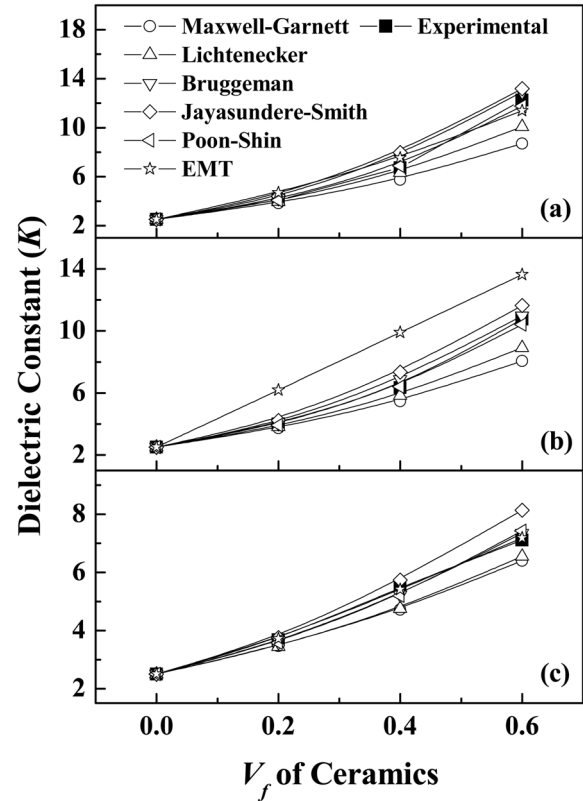


Fig. 5. Comparison of theoretical and experimental dielectric constant (K) of polystyrene / (a) MgTa₂O₆, (b) MgNb₂O₆ and/or (c) MgWO₄ composites with various volume fractions (V_f) of ceramics at 7.3 GHz.

ogy factor. The K_{eff} values predicted by Eqs. (1)-(6) were compared with the experimental results at 7.3 GHz and are shown in Fig. 5. The Maxwell-Garnett, Lichtenecker, Bruggeman, and Jayasundere-Smith models were only valid for low filler content (up to 0.2 V_f), because these models do not consider the interfacial interactions (adhesion, porosity, and interfacial polarization) between the polymer matrix and ceramic fillers with various shapes and sizes. Therefore, the shape and size of the fillers as well as the interfacial interactions between the polymer matrix and ceramic fillers should be considered along with the V_f and K of the ceramic fillers. For the composites with MgTa₂O₆ and/or MgNb₂O₆, the experimental K was in good agreement with the K_{eff} predicted by the Poon-Shin model (Figs. 5(a), (b)). This stems from the fact that the Poon-Shin model takes into account the interaction between the particulates consisting of a pure medium and inclusions embedded in the medium. The Poon-Shin model was applicable throughout the range of the V_f of the inclusions and was reasonably good for all values of the K of the composites.¹⁶⁾ Meanwhile, the experimental K of the PS / MgWO₄ composites showed a good fit with the K_{eff} predicted by the EMT model (Fig. 5(c)). The n value of the EMT model was determined empirically and was found to be 0.05 for the PS / MgWO₄ composites. A small value of n indicates spherical shape of the filler parti-

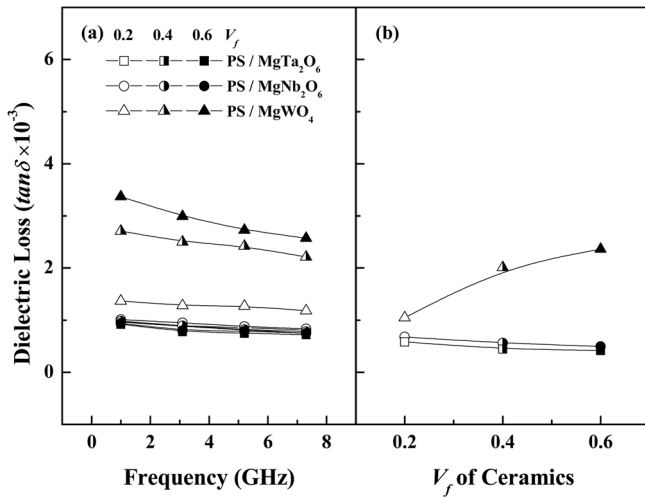


Fig. 6. Dependences of dielectric loss ($\tan \delta$) on (a) frequency (1-7.3 GHz, at 0.2-0.6 volume fraction (V_f)) and (b) various V_f of ceramics (at 11 GHz) of polystyrene (PS) / Mg-based ceramic composites.

cles.¹⁾ George *et al.* reported¹⁷⁾ that the n value of PS composites filled with $\text{Ca}[(\text{Li}_{1/3}\text{Nb}_{2/3})_{0.8}\text{Ti}_{0.2}]\text{O}_{3-\delta}$ ceramics with a spherical shape was 0.09, similar to our results. In the EMT model, the random unit cell is defined as the core of the filler K_f surrounded by a shell of host matrix K_m to describe the composite microstructure. Therefore, the random unit cell can be replaced with a material characterized by K_{eff} which considers the shape and size of the fillers, and the interfacial interactions between the ceramic fillers and the polymer matrix. The Poon-Shin model could be applied for the composites with MgTa₂O₆ and/or MgNb₂O₆ above $K = 20$,¹⁶⁾ while the EMT model could be employed for the composites with MgWO₄ below $K = 20$.¹⁾

Fig. 6 shows the dependences of the dielectric loss ($\tan \delta$) on the frequency (1-7.3 GHz, at 0.2-0.6 V_f) and various V_f of the ceramics (at 11 GHz) of PS / Mg-based ceramic composites, respectively. With increasing frequency from 1 GHz to 7.3 GHz (Fig. 6(a)), the $\tan \delta$ of the composites was slightly decreased. These results could be attributed to the relaxation frequency being below 1 GHz, which is lower than the frequency range of this study, because relaxation loss by dipolar orientation is generally generated at 10^8 Hz.¹⁸⁾ At the same frequency, the $\tan \delta$ of the PS / MgWO₄ composites was higher than those of the PS / MgNb₂O₆ and/or PS / MgTa₂O₆ composites for all V_f . In general, the $\tan \delta$ of polymer / ceramic composites increases with the ceramic content due to interfacial polarization,¹⁷⁾ which is associated with the entrapment of free charges generated in the ceramic fillers at the interfaces between the polymer matrix and ceramic fillers. However, the $\tan \delta$ of the composites with low loss ceramics was decreased with increased ceramic content due to the very low $\tan \delta$ of the ceramics.¹⁹⁾ Therefore, the $\tan \delta$ of composites with ceramic content could be affected by the $\tan \delta$ values of the ceramics. With increasing ceramic content, the $\tan \delta$ of the PS / MgWO₄ composites

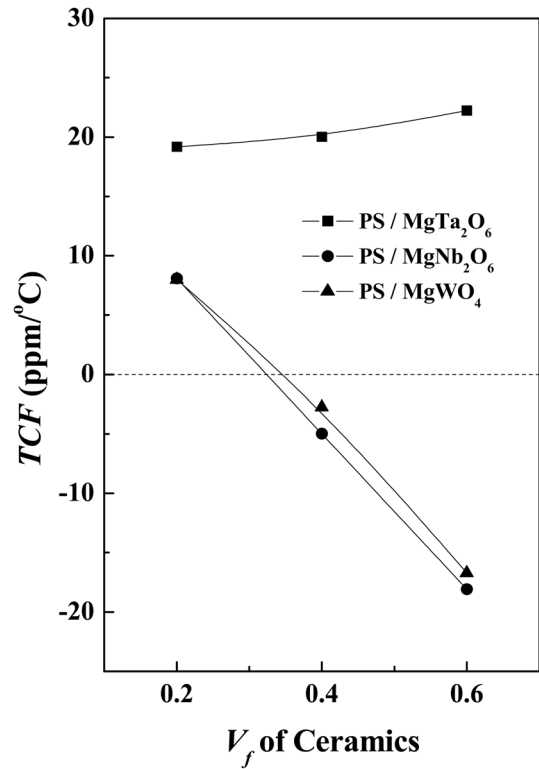


Fig. 7. Temperature coefficient of resonant frequency (TCF) of polystyrene (PS) / Mg-based ceramic composites with various volume fractions (V_f) of ceramics.

was increased, while those of the PS / MgTa₂O₆ and PS / MgNb₂O₆ composites were slightly decreased (Fig. 6(b)). These results are due to the much higher $\tan \delta$ ($Q = 1 / \tan \delta$) of MgWO₄ ($Q_f = 23,068$ GHz) compared to those of MgNb₂O₆ ($Q_f = 108,882$ GHz) and MgTa₂O₆ ($Q_f = 166,032$ GHz). For the composites with the same content of ceramics, the $\tan \delta$ values of the PS / MgNb₂O₆ and/or PS / MgTa₂O₆ composites were lower than those of the PS / MgWO₄ composites. At high frequency of 11 GHz, the $\tan \delta$ of the PS / Mg-based ceramic composites with various amounts of ceramics showed very low values below 2.4×10^{-3} , which were slightly lower than those of the composites in the range of 1-7.3 GHz (Fig. 6(a)).

Fig. 7 shows the temperature coefficient of resonant frequency (TCF) of the PS / Mg-based ceramic composites with various V_f values of the ceramics. With increasing ceramic content, the TCF of the PS / MgTa₂O₆ composites was increased, while those of the PS / MgWO₄ and/or PS / MgNb₂O₆ composites were decreased. For the composites with the same content of ceramics, the TCF of the PS / MgTa₂O₆ composites was higher than those of the PS / MgWO₄ and/or PS / MgNb₂O₆ composites. These results are due to the TCF values of the individual components (MgTa₂O₆ = 24.35 ppm/°C, PS = 18.74 ppm/°C, MgWO₄ = -56.6 ppm/°C, MgNb₂O₆ = -63.17 ppm/°C). Good thermal stability of resonant frequency (zero TCF) was obtained for the composites with a 0.4 V_f of MgWO₄.

4. Conclusions

At a frequency range from 1 GHz to 7.3 GHz, the dielectric constant (K) of polystyrene (PS) / MgTa_2O_6 , MgNb_2O_6 , and MgWO_4 (Mg-based ceramics) composites showed good frequency stability, while the dielectric loss ($\tan\delta$) of the composites was slightly decreased with the frequency. The experimental K values of the PS / MgTa_2O_6 and PS / MgNb_2O_6 composites were in good agreement with those predicted using the Poon-Shin model. However, the experimental K values of the PS / MgWO_4 composites fitted well with those predicted using the EMT model due to the low K value (< 20) of the ceramics. The K , $\tan\delta$, and temperature coefficient of resonant frequency (TCF) of the composites were affected by the type and amount of ceramics with various dielectric properties at 11 GHz.

Acknowledgments

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