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MRB 9113 No. of Pages 6

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Materials Research Bulletin xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Materials Research Bulletin

journal homepage: www.elsevier.com/locate/matresbu

# Dependence of dielectric properties on multilayered structures of $MgTa_2O_6$ and $MgMoO_4$/PTFE composites

Chang Jun Jeon[a], Eung Soo Kim[b], Jeong Ho Cho[a,*]

[a] Electronic Materials & Component Center, Korea Institute of Ceramic Engineering and Technology, Jinju 52851, South Korea
[b] Department of Materials Engineering, Kyonggi University, Suwon 16227, South Korea

ARTICLE INFO

*Article history:*
Received 31 August 2016
Received in revised form 13 December 2016
Accepted 15 January 2017
Available online xxx

*Keywords:*
A. Ceramics
A. Composites
D. Dielectric properties
D. Dielectrics
Temperature coefficient

ABSTRACT

Effects of multilayered structures on the microwave dielectric properties of composites of polytetrafluoroethylene (PTFE) with ceramic fillers of $MgTa_2O_6$ and $MgMoO_4$ were investigated as a function of ceramic content. Dielectric constants (*K*), dielectric losses (tan$\delta$), and temperature coefficients of resonant frequency (*TCF*) of the 0–3 type composites were found to depend on the type and amount of ceramic material. Multilayered 2–2 type composites with 0.2 ceramic volume fraction ($V_f$) showed strong dependence of tan$\delta$ on multilayer structures. This was due to strain differences between different layers in the multilayered 2–2 type composites. Several theoretical models were employed to predict the effective *K* of the composites and were compared with experimental data. Typical results of $K = 3.53$, tan$\delta = 1.75 \times 10^{-3}$, and $TCF = -3.49$ ppm/°C were obtained for a double-interfaced (middle $MgTa_2O_6$) 2–2 type composite with 0.2 $V_f$ of ceramic.



## 1. Introduction

Polymer-based composites have been widely investigated for use as high-performance materials suitable for microwave substrates and electronic packaging. Their suitability is due to their mechanical flexibility, chemical stability, ease of processing, and tuneable properties that allow tailoring of dielectric properties. For stable and selective resonant frequencies and high-frequency use in microwave devices, these composites require a low dielectric constant (*K*) for fast signal speed, low dielectric loss (tan$\delta$) for better device functionality, and a near-zero temperature coefficient of resonant frequency (*TCF*) for thermal stability.

The microwave dielectric properties of composite materials depend not only on the type of filler and matrix, but also on their coupling behavior [1]. This coupling concept is called connectivity and was first introduced by Newnham et al. [2]. Composites with 0–3 connection type (three-dimensionally connected polymer phase loaded with isolated ceramic particles) are especially attractive for various applications because they can be easily prepared, which enables the creation of the best possible design structures. It has been reported that the layered 2–2 type composite structures with low tan$\delta$ can be obtained from 0–3 type dielectric and magnetic composites with homogeneously distributed ceramic fillers [3]. Teirikangas et al. [4] also reported that composites with vertical 2–2 type multilayered structures showed lower tan$\delta$ than composites with horizontal 1–3 type multilayered structures.

Layer-structured materials show considerable stresses as the layers have different elastic or thermal properties [5]. The dielectric properties of thick/thin film structures are significantly affected by internal stresses at the interfaces between the film and the substrate, with tan$\delta$ increasing due to the strains ($\varepsilon$) of the thick/thin films [6]. Ceramic-based composite layers have shrinkage mismatch and diffusion between their phases or layers during sintering which greatly influences the dielectric properties of the final product. Polymer-based composite layers do not suffer from these effects and show improved dielectric properties. Therefore, the homogeneous dispersion of the ceramic filler, the type of multilayer structure, and $\varepsilon$ of layers can affect tan$\delta$ of composite.

Preliminary results showed that $MgTa_2O_6$ and $MgMoO_4$ have low tan$\delta$ values above 10 GHz ($6.02 \times 10^{-5}$ and $3.03 \times 10^{-4}$, respectively). In addition, they showed different *K* (25.48 and 7.05, respectively) and *TCF* (24.35 ppm/°C and -45.29 ppm/°C, respectively). Therefore, these two ceramics, with different dielectric properties, were tested here as fillers for composites to investigate their effects on the microwave dielectric properties of the resulting composites. Polytetrafluoroethylene (PTFE) was selected as the polymer matrix due to its excellent dielectric properties ($K = 2.1$, stable over a wide frequency range up to 20

* Corresponding author.
*E-mail address:* goedc@kicet.re.kr (J.H. Cho).

http://dx.doi.org/10.1016/j.materresbull.2017.01.020
0025-5408/

Please cite this article in press as: C.J. Jeon, et al., Dependence of dielectric properties on multilayered structures of $MgTa_2O_6$ and $MgMoO_4$/PTFE composites, Mater. Res. Bull. (2017), http://dx.doi.org/10.1016/j.materresbull.2017.01.020

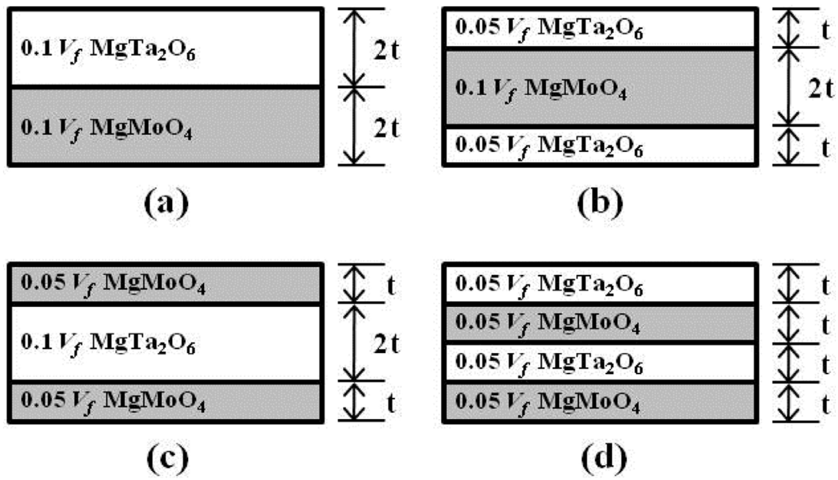


**Fig. 1.** Schematic diagrams of multilayered 2–2 type PTFE composites with 0.2 volume fraction ($V_f$) of ceramics: (a) one interface, (b) two interfaces with $MgMoO_4$ middle layer, (c) two interfaces with $MgTa_2O_6$ middle layer, and (d) three interfaces between $MgTa_2O_6$/PTFE and $MgMoO_4$/PTFE layers.

GHz; tan$\delta = 10^{-4}$ at 800 MHz), high thermal stability (applicable up to 250 °C), low moisture absorption, and good corrosion resistance [7].

This work reports the effects of ceramic content and multilayer structure on the dielectric properties of various composites of PTFE with $MgTa_2O_6$ and $MgMoO_4$, at microwave frequencies. Further, the effects of strain differences ($\Delta\varepsilon$) between the $MgTa_2O_6$/PTFE and the $MgMoO_4$/PTFE layers on tan$\delta$ of the multilayered 2–2 type composites are discussed. In addition, effective $K$ values were calculated by several theoretical models and were compared with experimentally determined values. *TCF* values of composites were also studied in relation to their thermal stability.

## 2. Experimental

Ceramic filler powders of $MgTa_2O_6$ and $MgMoO_4$ were separately prepared by conventional solid-state reactions from high-purity (99.9%) oxide powders. Single phases of each ceramic composition and optimal microwave dielectric properties of the composites were obtained by double calcination of $MgTa_2O_6$ at 1100 °C and 1350 °C for 3 h and $MgMoO_4$ at 650 °C and 850 °C for 3 h. The calcined powders were ground with $ZrO_2$ balls for 24 h in ethanol and then dried. 1 $\mu$m polytetrafluoroethylene (PTFE) powder was used as the polymer matrix. Its very high melt viscosity (>$10^{11}$ Pa s) [8] precludes the use of common processing techniques such as melt extrusion and injection moulding. Therefore, the composites were prepared by powder processing. The mixed ceramic/polymer powders were pressed isostatically into 15-mm-diameter discs as 0–3 type and multilayered 2–2 type composites under a pressure of 20 MPa for 1 min. The resulting pellets were heat treated at 300 °C for 1 h. The multilayered 2–2 type composites are shown in Fig. 1. All samples were prepared with the same total thickness to eliminate the unequal effects of different thicknesses on the microwave dielectric properties. Each specimen with 0.2 volume fraction ($V_f$) of ceramic had the same net compositional ratio. The number of interfaces between $MgTa_2O_6$/PTFE and $MgMoO_4$/PTFE layers was different with the lamination patterns.

Powder X-ray diffraction (XRD, D/Max–2500 V/PC, RIGAKU, Japan) was used to determine the crystalline phases of composites. The microstructures of composites were observed by scanning electron microscopy (SEM, JSM–6500F, JEOL, Japan). Apparent densities of the composites were measured by the Archimedes' method. Relative densities were obtained from the theoretical values by the mixing rule [9]. Dielectric properties ($K$ and tan$\delta$) were measured by the Hakki and Coleman method [10] at 10–14 GHz. *TCF* values were measured by the cavity method [11] from 20 °C to 80 °C.

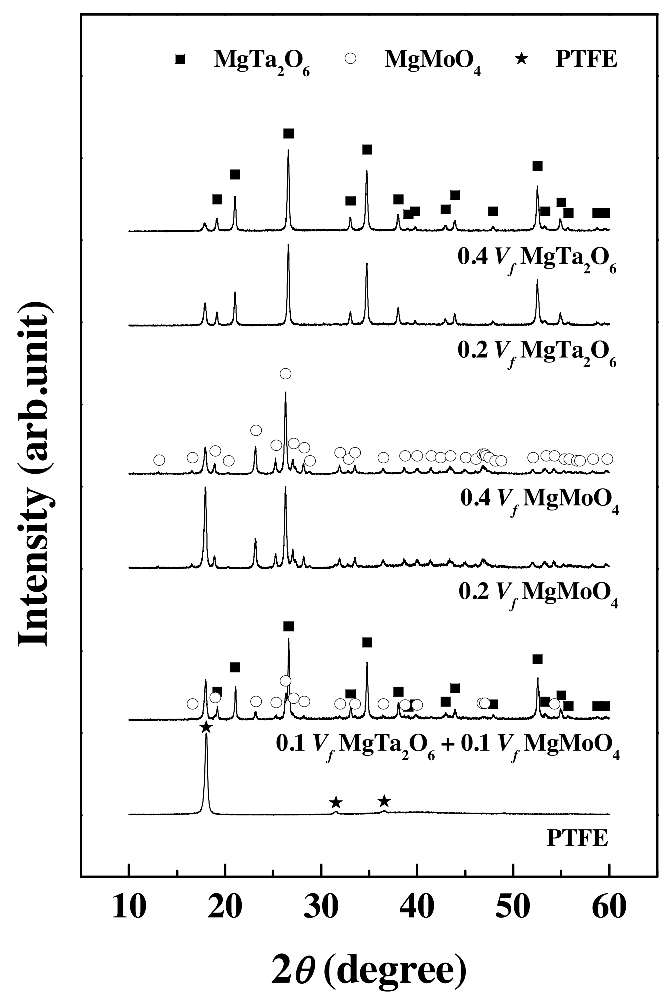


**Fig. 2.** X-ray diffraction patterns of PTFE and 0–3 type PTFE composites with various volume fractions ($V_f$) of ceramics.

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