

Effects of Content of Chopped Glass Fibers on the Properties of Silica Filled PTFE Composites

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How to cite this paper: Yuan, Y., Lin, H.D., Jiang, Z.H., Chi, Z.F., Yao, M.H. and Zhang, S.R. (2017) Effects of Content of Chopped Glass Fibers on the Properties of Silica Filled PTFE Composites. *Journal of Materials Science and Chemical Engineering*, 5, 36-44.

<https://doi.org/10.4236/msce.2017.57005>

Received: April 10, 2017

Accepted: July 4, 2017

Published: July 7, 2017

Abstract

In this work, the polytetrafluoroethylene (PTFE)-based composite substrates were manufactured by mixing, calendaring, hot-pressing sintering. The composition of all the samples was PTFE, SiO₂ and chopped E-glass fibers. The effects of content of E-glass fibers on the properties of the SiO₂ filled PTFE composites were investigated, including density, water absorption, dielectric properties (ϵ_r , $\tan\delta$), coefficient of thermal expansion (CTE) and temperature coefficient of dielectric constant (τ_ϵ). The compositions of inorganic materials mixture are (62 - x) % SiO₂ + x % E-glass fiber (x: mass ratio to composites, x = 0, 1, 1.5, 2, 2.5, 3). The results show that as the content of E-glass fibers is 2.5 wt.%, this composite obtains optimal properties, including excellent dielectric properties ($\epsilon_r \sim 2.9123$, $\tan\delta \sim 0.0011$), acceptable water absorption of 0.075%, temperature coefficient of dielectric constant of 10 ppm/°C and coefficient of thermal expansion of 15.87 ppm/°C.

Keywords

Polymers, Composite Materials, PTFE, Dielectric Properties, E-Glass Fiber

1. Introduction

Microwave composite substrate materials play a key role in global society with a wide range of applications from terrestrial and satellite communication including software radio, GPS, and DBS TV to environmental monitoring via satellites. PTFE polymer has been widely studied as high-frequency microwave substrates material because of its ideal microwave dielectric properties, temperature and chemical resistance [1] [2]. However, PTFE has some disadvantages, such as high linear coefficient of thermal expansion (CTE \sim 109 ppm/°C) and poor for-

mability [2]. In order to solve these problems, many researchers have tried to reduce its high CTE and improve the formability by adding inorganic materials into PTFE matrix, such as silica (SiO_2) [3] [4], alumina (Al_2O_3) [5], magnesium titanate (MgTiO_3) [6], TiO_2 [7], CaTiO_3 [8], and so on. Among above composites, the PTFE/ SiO_2 composites have low dielectric constant, dielectric loss and low CTE. It has been reported that 60 wt.% untreated SiO_2 filled PTFE composite has a dielectric constant of 2.9, dielectric loss of 0.0024 and CTE of 45 $\text{ppm}/^\circ\text{C}$ [3] [4]. Chopped E-glass fiber is incorporated in the PTFE matrix to control surface texture and to enhance mechanical properties. Experiments on ceramic filled PTFE composites (PTFE/ Al_2O_3 , PTFE/ SrTiO_3 and PTFE/ CaTiO_3) show that 2 wt.% E-glass chopped fiber in PTFE matrix exhibit desirable mechanical and surface properties [9]. It is well known that copper conductor layer is laminated over PTFE substrate for circuit fabrication. Therefore, the CTE of composite is a key parameter for microwave substrate, which should be close to that of copper (CTE \sim 17 $\text{ppm}/^\circ\text{C}$) [10]. As is well know, E-glass fiber has a CTE of approximately 4.8 $\text{ppm}/^\circ\text{C}$, which is far less than that of PTFE about 109 $\text{ppm}/^\circ\text{C}$. In this work, the effects of content of chopped E-glass fibers on the properties of the SiO_2 filled PTFE composites were investigated, including density, water absorption, dielectric properties, coefficient of thermal expansion and temperature coefficient of dielectric constant.

2. Experimental Procedures

The raw materials used in this study were PTFE suspension (TE-3865C, DuPont, China), E-glass fiber (Nanjing Glass Fiber Research and Design Institute, china) and fused amorphous SiO_2 powder ($\geq 99.5\%$, Huawei Powder Technology Co, Ltd, China) with an average size of 8.3 μm . The performances of raw materials are shown in **Table 1**. Surface of glass fiber and silica are treated with silane coupling agents. SiO_2 powders, short E-glass fibers and PTFE aqueous dispersion were weighed accurately and mixed by high speed dispersing machine for 1 h to prepare the composites. The obtained slurry was then filtered at 25°C to remove water. Afterwards, the dough is pressed into a sheet by a calender. Then, the obtained sheet was hot pressed and sintered into a rectangular shape at 20 MPa in 370°C in a program controlled furnace for 2 h, and then cooled with furnace.

3. Characterization Studies

Scanning electron microscopy (SEM, model JEOL JSM-6490) was used to observe the morphology of E-glass fibers and microstructure of the composites.

Table 1. Properties of PTFE, SiO_2 and E-glass fiber.

	Dielectric constant (ϵ_r)	Dielectric loss $\tan\delta$	Density/ (g/cm^3)	Coefficient of thermal expansion (CTE)/($\text{ppm}/^\circ\text{C}$)
PTFE	2.1	0.0003	2.2	109
SiO_2	3.83	0.0025	2.3	0.5
E-glass fiber	6.11	0.006	2.53	4.8

The coefficients of thermal expansion of the composites were measured by TMA 2940 according to IPC-TM-650 2.4.41 [11]. The densities of the composites were measured by the Archimedes method. Moisture absorption of the composites was figured out as reported earlier by Murali *et al.* according to IPC-TM-650 2.6.2 [11]. The dielectric constant, dielectric loss and temperature coefficient of dielectric constant of the composites were measured by Agilent E8363A microwave network analyzer using stripline resonator method according to IPC-TM-650 2.5.5.5 specification [12]. The testing frequencies covered the region from 7.0 GHz to 13.0 GHz. In this article, ϵ_r and $\tan\delta$ of the composites reported was at a frequency around 10 GHz.

4. Results and Discussion

Figure 1 shows the SEM micrographs of the non-treated E-glass fibers and treated E-glass fibers. It could be observed that E-glass fibers become smoother and no agglomerate after being treated by silane coupling agent. **Figure 2** shows the brittle fractured cross sectional SEM micrographs of composites with different contents of E-glass fiber. In this work, as the glass fiber content is relatively small, it is difficult to find fiberglass by SEM, further indicating that the glass fibers are evenly distributed in the composite materials.

The variation in experimental and theoretical density with different contents of E-glass fiber in the composites is shown in **Figure 3(a)**. The theoretical density of the composites ρ_c is calculated using the rule of mixtures (Equation (1)) [6].

$$\rho_c = \rho_f V_f + \rho_m V_m \quad (1)$$

where ρ_f and ρ_m are the densities and V_f and V_m are the volume fractions of filler and matrix, respectively. From **Figure 3(a)**, it is obvious that the experimental density increases with E-glass fiber loading up to 2 wt.% and then keep unchanged with E-glass fiber loading up to 3 wt.%. This result is due to the density of the glass fiber is greater than that of silica. So the density of composites increases with an increase of content of E-glass fiber. It can be seen that the experimental density increases slightly with the loading of the filler. The difference between the experimental density and the theoretical density is due to the higher

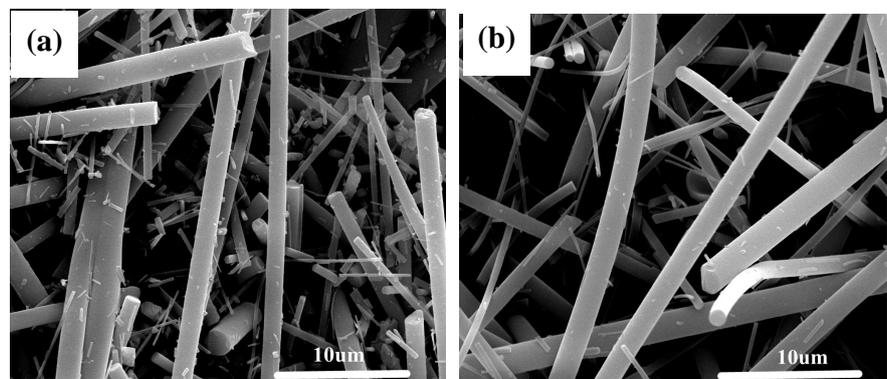


Figure 1. SEM micrographs of (a) non-treated fused E-glass fiber (b) treated E-glass fiber.

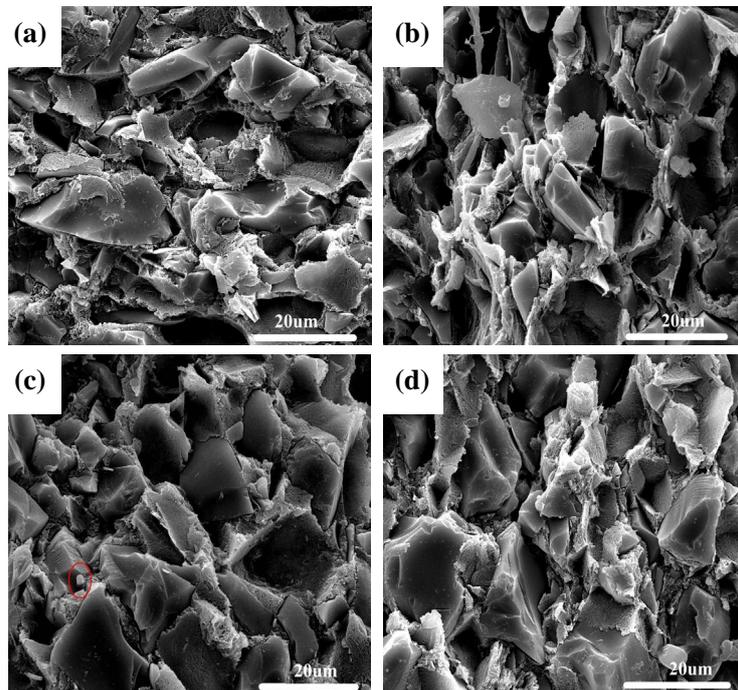


Figure 2. Brittle fractured cross sectional SEM micrographs of composite at different contents of E-glass fiber (a) 0 wt.%; (b) 1.5 wt.%; (c) 2.5 wt.%; (d) 3 wt.%.

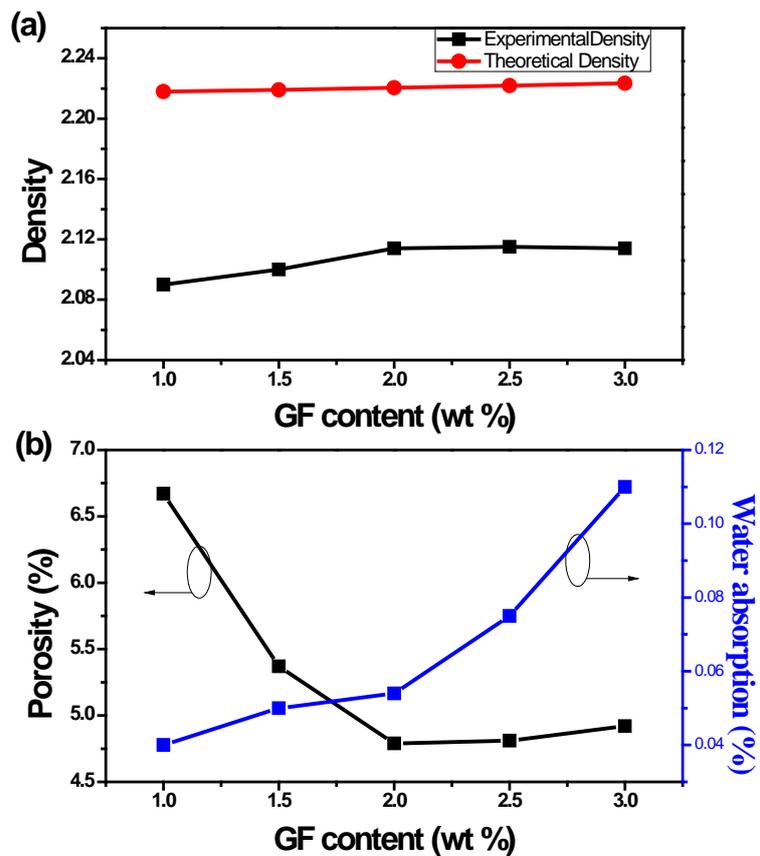


Figure 3. (a) The variation in experimental density and theoretical density with respect to E-glass fiber loading; (b) The variation in porosity and water absorption with respect to E-glass fiber loading.

packing load. The formation of pores in the composite material may be the main reason for this deviation, which is not considered in the theoretical calculation. The variation in porosity and water absorption with different contents of E-glass fiber in the composites is shown in **Figure 3(b)**. It can be seen that porosity decreases with E-glass fiber loading up to 2 wt.% and then slowly increases with E-glass fiber loading up to 3 wt.%. However, water absorption of the composites increases with different contents of E-glass fiber. This is maybe attributed to the fact that the capillary action of the E-glass fibers causes the water to spread along the interface.

Variation of dielectric constant and dielectric loss with respect to weight fraction of E-glass fiber in the PTFE matrix is shown in **Figure 4**. Dielectric constant increases with E-glass fiber loading up to 3 wt.%. Dielectric loss increases with E-glass fiber loading up to 1.5 wt.% and then decreases with E-glass fiber loading up to 2.5 wt.%. Then dielectric loss increases with E-glass fiber loading up to 3 wt.%. This result is due to the dielectric constant of the glass fiber is greater than the dielectric constant of the silica. At the same time, density increases with E-glass fiber loading as shown in **Figure 3**. Thus, the number of polarized particles increases in per unit volume and ϵ_r of the composites increases. Furthermore, when the composite has fewer pore, the air content decreases, resulting in a decrease of dielectric loss. At the same time, the dielectric loss of E-glass fiber is higher than that of silica and PTFE. So a minimum dielectric loss is obtained with 2.5 wt.% content of E-glass fiber.

The variation of CTE at different directions (X/Y/Z) is shown in **Figure 5**. The value of CTE at Z direction increases firstly with 1.5 wt.% content of E-glass fiber, and then decreases up to 2.5 wt.% content of E-glass fiber, and then shows a little increase at 3.0 wt.% fiber. However, the thermal expansion changes at X and Y directions are not significant. A minimum CTE of composites is obtained when content of E-glass fiber is 2.5 wt.%. E-glass fiber is tightly connected with PTFE and embedded in PTFE homogeneously. E-glass fiber could restrain the

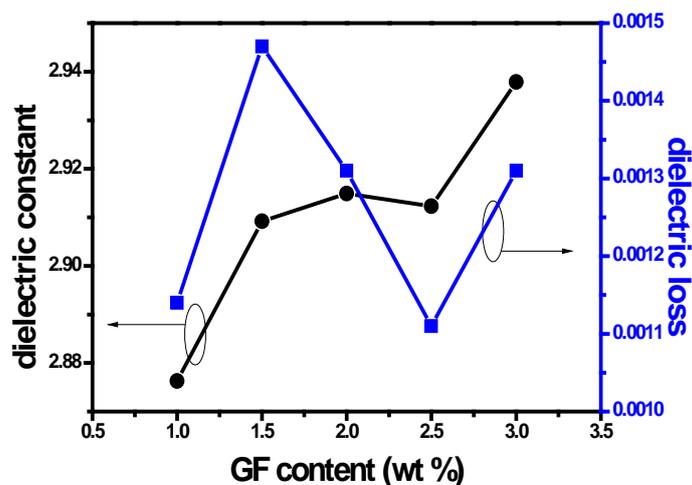


Figure 4. The variation of dielectric constant and dielectric loss at 10 GHz of PTFE/SiO₂ composites with respect to E-glass fiber loading.

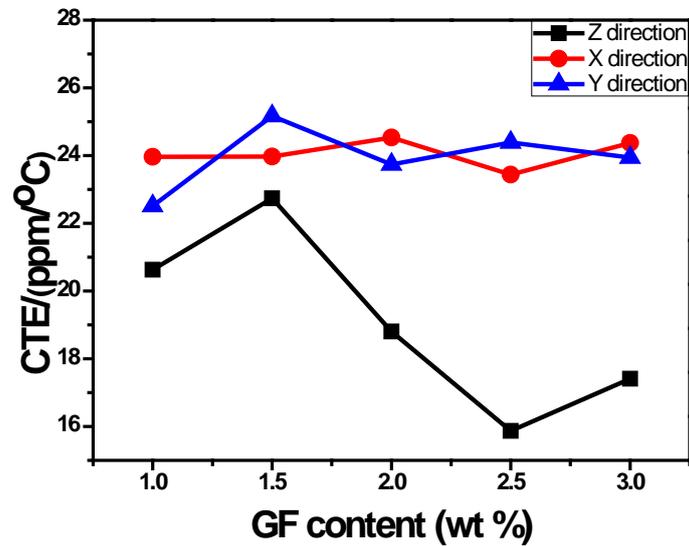


Figure 5. The variation of CTE with respect to E-glass fiber loading.

thermal expansion of PTFE polymer when temperature is increased. The randomly distributed mixture of PTFE and E-glass fibers is initially compression moulded. The E-glass fibers tend to align perpendicular to the direction of compression during pressing [13]. Since the ceramic filling amount is high, the glass fiber is substantially inclined in the substrate. Therefore, the thermal expansion coefficient has more obvious change in the Z direction. The calendaring process leaves air entrapped in the structure. In the form of small micro air bubbles are possibly entrapped between the E-glass fibers. When heated, the air tends to expand much more than the polymers and glass fibers, forcing the glass fibers to expand more in X and Y direction [13]. Therefore, the thermal expansion coefficient in the X and Y direction are larger than the thermal expansion coefficient in the Z direction.

The dielectric constant temperature coefficient has a similar trend with CTE. In this work, the τ_ϵ of composites is studied within the temperature range from 0°C to 100°C. The τ_ϵ values of the composites are calculated using Equation 2 [14].

$$\tau_\epsilon = \frac{1}{\epsilon_\gamma} \times \frac{\Delta\epsilon_\gamma}{\Delta T} \quad (2)$$

where ϵ_γ is the dielectric constant at 25°C and $\frac{\Delta\epsilon_\gamma}{\Delta T}$ is the change of dielectric constant with respect to temperature. The variation of τ_ϵ with respect to E-glass fiber loading is shown in Figure 6. The most important factors that control the temperature coefficient of dielectric constant are the change in the polarization of the material with respect to its temperature and its linear coefficient of thermal expansion (Equation (3)) [14]:

$$\tau_\epsilon = \frac{\epsilon'_\gamma}{3} \left(\frac{1}{\alpha} \times \frac{\partial \alpha}{\partial T} - 3\alpha_L \right) \quad (3)$$

where the first term represents the change in polarization of the system, and the

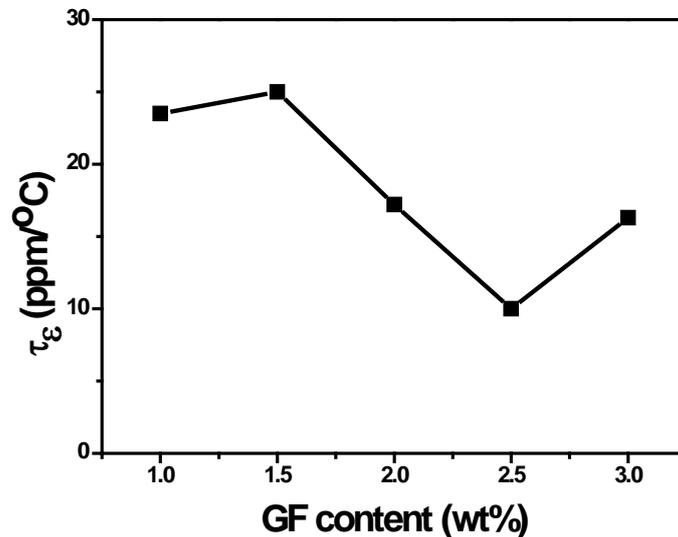


Figure 6. The variation of τ_ϵ with respect to E-glass fiber loading.

second term represents the linear coefficient of thermal expansion. In this composite system, PTFE molecules don't have polarity due to its symmetric structure. Therefore, the polarization type of PTFE is only electronic displacement polarization so that PTFE shows a negative τ_ϵ of -400 ppm/°C [10]. On the contrary, SiO₂ ceramic and E-glass fibers present a positive τ_ϵ resulted from its dominant ionic displacement polarization. In this work, τ_ϵ is dominantly controlled by the polarization variation of PTFE polymer, E-glass fibers and SiO₂ ceramic with respect to temperature, and the linear coefficient of thermal expansion of composites.

5. Conclusion

In this work, the effects of glass fiber content on the properties of the composites were investigated, such as dielectric properties, density, water absorption, coefficient of thermal expansion, and dielectric constant temperature coefficients. The microstructure of the composites was observed by scanning electron microscopy, which further proved the effect of the glass fibers in the composites. The optimum dielectric properties and the dielectric constant temperature coefficient values were obtained when the glass fiber content was 2.5%. E-glass fiber could restrain the thermal expansion of PTFE polymer when temperature is increased. The smaller thermal expansion coefficient values were obtained.

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