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Dielectric properties of $SrTiO_3$/POE flexible composites for microwave applications

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Abstract

Composites fabricated with $SrTiO_3$ ceramic filler dispersed inside a thermoplastic polyolefin elastomer (POE) polymer matrix, with excellent dielectric properties and good flexibility, have been studied in this paper. The relative permittivity of $SrTiO_3$/POE composites blended with different volume fraction of ceramic filler was investigated as a function of temperature and frequency. The results indicated that with the increase of ceramic filler, both the relative permittivity and the dissipation factor of composites increased. Good frequency stability within a wide range was observed in all the samples. The theoretical dielectric constant obtained from the effective medium theory of Bruggeman's model is in good agreement with the experimental data. For the composites containing 40 vol.% $SrTiO_3$, the dielectric constant and the loss tangent were 11.0 and 0.01 at 900 MHz, respectively, while the mechanical test results showed good flexibility on the final quality of the composites with the elongation of 90%. This indicates that the ST/POE composites have the promising characteristics for potential applications in the flexible dielectric waveguide and related flexible microwave devices.

Keywords: Dielectric properties; Composites; Flexibility

1. Introduction

In recent years, there is an increasing interest on the flexible dielectric waveguide in the microwave frequency because it has various potential applications, including communication, military and medical care.[1–3] The flexible dielectric waveguide consists of a flexible core and a flexible cladding. The core would be a flexible dielectric material with high dielectric constant and low dielectric loss in the microwave frequency range. The cladding would also be made of a low loss dielectric material with low dielectric constant, and its dielectric constant should be smaller than that of the core.[4,5] However, all flexible dielectrics with low losses in the microwave frequency, such as polystyrene, polyethylene, polypropylene, and polytetrafluoroethylene (PTFE), have small dielectric constants (about 2). On the other hand, the flexibility of the high dielectric constant ceramic material is very poor. Therefore, the application of a flexible polymer or a kind of ceramic is greatly restricted in the core of the flexible dielectric waveguide.[4] Polymer-matrix composites reinforcing by ceramic powder would be an effective approach to improve the properties of dielectric materials in order to satisfy the requirement of the flexibility and high dielectric constant.[6,7] By integrating high dielectric constant ceramic powder with superior flexibility of the polymer matrix, one can obtain composites with high dielectric constant and good flexibility. This kind of composites is highly suitable for the flexible core of the waveguide.

In this paper, high dielectric constant flexible microwave composites were achieved by introducing the ceramic filler with high dielectric constant into polymer matrix with high flexibility. Thermoplastic polyolefin elastomer (POE) is selected as the polymer matrix. $SrTiO_3$ dielectric ceramic (ST) is selected as the ceramic filler. POE is an ethylene–octene copolymer that has excellent flow characteristics and performs well in a wide range for general purpose of thermoplastic elastomer applications. ST ceramic has attracted attention of researchers in microwave dielectrics because of their low loss tangent, high relative permittivity, and wide adjustable α_ε range.[8] ST/POE flexible composites with various ST filler volume fractions were prepared by using the extrusion technology. The dielectric properties and the mechanical properties of the ST/POE composites were investigated and reported in detail.

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2. Experimental

The raw materials selected were a thermoplastic polyolefin elastomer (POE) for the matrix and a $SrTiO_3$ dielectric ceramic for the filler. The POE (density: 0.87 g/cm^3; glass transition temperature: −55 °C; elongation exceeds 600%) used was Engage^{TM} POE 8100 (Dupont Dow Co., USA). The pure $SrTiO_3$ filler (GuoTeng Co. Ltd., China) has an average grain size of about 100 nm and a density of about 5.3 g/cm^3, with the dielectric constant and the loss tangent of 160 and 0.006, respectively in the 1–5 GHz frequency range. The filler and the POE were mixed with various volume fractions for 20 min in a Rheomix600p system (HAAKE Co., Germany) operated at 60 rpm and 220 °C. Then, the mixture was put into a disk mold and hot-pressed under stress of 10 MPa at a temperature of 150 °C for 10 min. The sample was allowed to cool down and the stress was removed after that. In order to let the ST filler to distribute uniformly in POE matrix, the above mentioned technological parameters were found to be the optimum.

In the high frequency range of 1 M–1 GHz, the relative permittivity and dissipation factor were measured by HP4291B impedance analyzer with HP16453A dielectric material test fixture. For the dielectric measurement, the samples are discs with a diameter of 15 mm, about 1 mm thick.

The frequency dependant dielectric behavior of the samples in microwave range from 500 M to 5 GHz were obtained by using open-reflection method with a HP8270ES network analyzer and a HP85070c open-ended coaxial line probe.[9–11] For the error correction over the measurement frequency range, a calibration was performed by placing known standards (air, short, deionized water) on the end of the open-ended coaxial line probe.

The microwave dielectric measurement at approximately 5.011 GHz were performed on a 5.011 GHz split post-resonator[11–13] with a HP8270ES network analyzer. The samples fitted the cavity dimension were 1 mm thick and 35 mm diameter discs as used in the open-reflection method.

Dielectric measurements as a function of temperature were carried out with a HP4284 LCR meter from 1 KHz to 1 MHz between −180 °C and 65 °C, measurement were made while heating at a rate of 3 °C/min. For this measurement, silver electrodes were painted on the both side on the sample.

Density measurement was made by Archimedes method with distilled water as the liquid medium. Microstructure of the composites was observed using scanning electron microscopy (SEM). Tensile tests were made on hot-pressing samples (thin plate) using a tensile test machine PT-1036PC (Perfect Instrument Co. Ltd., China, Taiwan) with a 5 mm/min deformation speed.

3. Results and discussion

3.1. Micrographs

Fig. 1 shows SEM micrographs of the 40% ST composites. In this image few large ST filler agglomeration can be observed. There is no obvious phase separation, which means that the ST filler is compatible with the POE matrix, the ST filler can be distributed uniformly in POE matrix. It is indicated that the composites sample has excellent homogeneity.

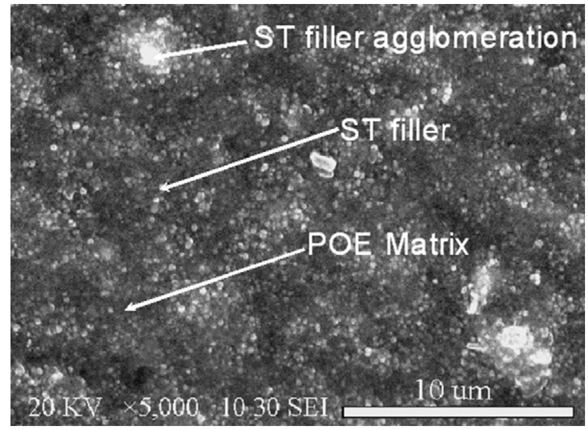

Fig. 1. SEM image of 40 vol.% ST composites.

3.2. Dielectric properties

Fig. 2(a) presents frequency dependence of relative permittivity of the composites. With the increase of ST filler amount, the dielectric constant of composite increases significantly. For the 40 vol.% ST composites, the average dielectric constant can reach approximately 11.0, which is highly improved compared with the pure polymer matrix (about 2.1). As shown in Fig. 2(b),

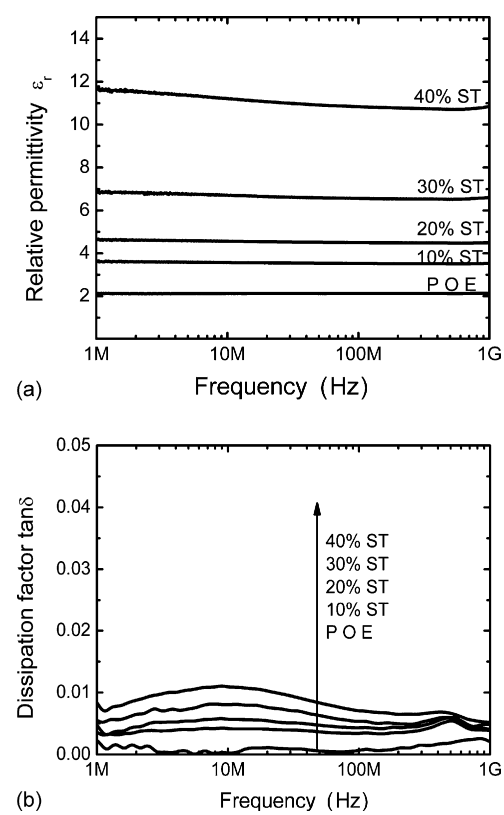

Fig. 2. The frequency dependence of the relative permittivity ε_r (a) and dissipation factor tan δ (b) for ST/POE composites blended with different volume fraction of ST ceramic filler (measured by HP4291B impedance analyzer).

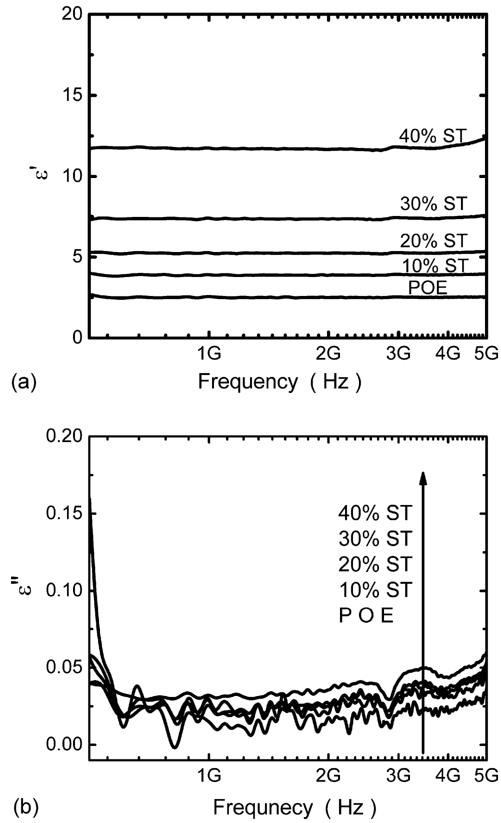

Fig. 3. The frequency dependence of the real (a) and imaginary (b) part of the relative permittivity of ST/POE composites with different volume fraction of ST ceramic filler at microwave frequencies (measured by open-reflection method).

the dielectric loss of the composites slightly increases with the amount of ST filler increasing. At 900 MHz, the loss tangent is 0.01 for the composites containing 40 vol.% $SrTiO_3$.

AS seen in Fig. 3(a) and (b), real and imaginary part of the relative permittivity of ST/POE composites with different volume fraction of ST ceramic filler at microwave frequencies from 500 MHz to 5 GHz appear nearly a constant in the whole measurement range. The plot in Fig. 3 show similar features as that in Fig. 2. It reveals that the good frequency stability of the ST/POE composites has been obtained over a wide range from 1 MHz to 5 GHz.

The microwave dielectric properties of the ST/POE composites were measured by a 5.011 GHz split post-resonator and the results are shown in Table 1. The dielectric constant and the loss tangent at about 5 GHz correspond well with the results of Figs. 2 and 3, showing good stability within a wide range of frequency. The results indicate that the 40 vol.% ST composites can satisfy the requirement of high dielectric constant for the core of the flexible dielectric waveguide.

There are some defects such as air gap and the interface phase between ST and POE in the composites materials, which can influence the relative permittivity and the dielectric loss of the composites.[14] Assuming that there are only three phases in the composites, the volume fraction of the air in the composites can be calculated by equation:

$$\rho_{\text{composites}} \times (1 - V) + \rho_{\text{air}} \times V = \rho_{\text{measured}} \quad (1)$$

where $\rho_{\text{composites}}$, ρ_{air} and ρ_{measured} is the theoretical density of composites, the density of air and the measurement density of the composites, respectively. V is the volume fraction of air in the composites. The ρ_{measured} was measured using Archimedes method. The $\rho_{\text{composites}}$ was determined using mixing rule[15]:

$$\rho_{\text{composites}} = \sum_i \rho_i V_i \quad (2)$$

where ρ_i is the density of inclusion; V_i is the theoretical volume fraction of inclusion. These results are shown in Table 2.

Considering the influence of air gap, there are three phases in the composites. The theoretical dielectric constant can be

Table 1
Microwave dielectric properties of ST/POE composites (measured by 5.011 GHz split post-resonator, 25 °C)

Sample	Resonant frequency (GHz)	Q	Loss (dB)	ε_r	$\tan\delta$ ($\times 10^{-3}$)
Empty cavity	5.011848084	14,028	41.386		
Polymer matrix	4.963332779	12,245	41.500	2.1	0.2
10% ST					
	4.940197478	3866.6	41.244	3.4	4.5
20% ST	4.896465809	2,793	41.549	4.6	4.7
30% ST	4.878503448	2094.1	41.203	6.5	6.3
40% ST	4.749000814	899.6	41.746	10.5	8.5

Table 2
The density of ST/POE composites and the virtually volume fraction of inclusion

ST filler volume fraction (%)	$\rho_{\text{composites}}$ (g/cm^3)	ρ_{measured} (g/cm^3)	V_{ST} (vol.%)	V_{POE} (vol.%)	V_{air} (vol.%)
0	0.87	0.87	0	1	0
10	1.313	1.239	9.45	85.05	5.5
20	1.756	1.567	17.85	71.38	10.77
30	2.199	1.883	25.1	58.56	16.34
40	2.642	2.219	32.52	47.78	18.7

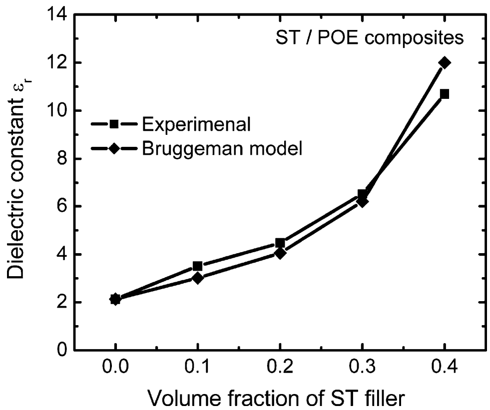

Fig. 4. The experiment and calculated dielectric constant of ST/POE composites.

calculated with the Bruggeman's model,[16,17]

$$\sum_i V_i \left(\frac{\varepsilon_i - \varepsilon_{\text{composites}}}{\varepsilon_i + 2\varepsilon_{\text{composites}}} \right) = 0 \tag{3}$$

where $\varepsilon_{\text{composites}}$ and ε_i is the dielectric constant of composites and inclusion, respectively. According to Eq. (3) and Table 2, the dielectric constant of ST/POE composites was calculated. The result is shown in Fig. 4, which is in good agreement with the experimental result.

The relative permittivity and dissipation factor as a function of temperature for composites fill with 40 vol.% of ST are shown in Fig. 5(a) and (b), respectively. The peak-like changes of the relative permittivity and the dissipation factor at −75 °C

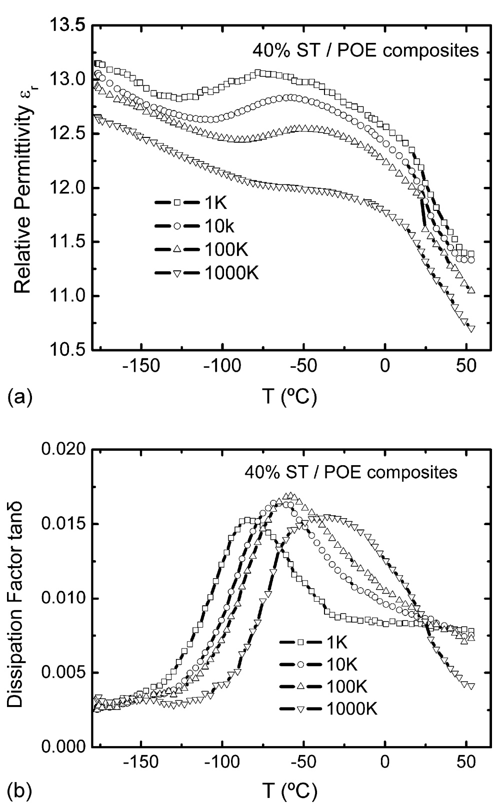

Fig. 5. The relative permittivity ε_r (a) and the dissipation factor tan δ (b) as a function of temperature for a composites containing 40 vol.% ST composites.

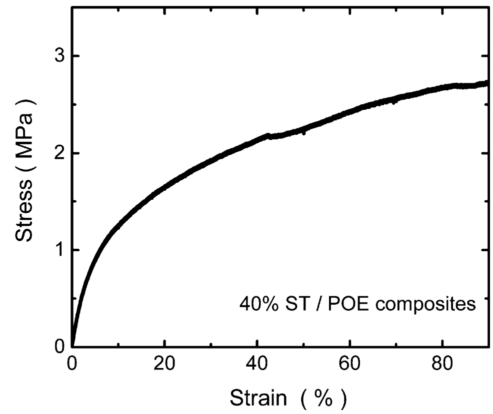

Fig. 6. The stress–strain curve of the 40 vol.% ST composites.

could be observed. Both transitions are native characteristics of POE, which have been inherited by the composites. In the ST/POE composites, the relative permittivity shows decrease with temperature increases in the range from −180 °C to 65 °C.

3.3. Mechanical test

As the matrix of composites, the good flexibility of POE polymer (more than 600% elongation) makes the flexibility of the composites also good. As shown in Fig. 6, the stress–strain curve of the 40% ST composite is presented. The composite has a critical stress of 2.75 MPa with an elongation at break value of about 90%. It is flexible enough to reach the requirement of flexible dielectric waveguide applications.

4. Conclusions

By using the extrusion technology, the $SrTiO_3$ ceramic filler were dispersed inside of the POE polymer matrix and the ST/POE composites with various volume fractions have been obtained successfully. The measurement results indicate that the composites have high relative permittivity, the low dissipation factor, and good stability within a wide range of frequency from 1 MHz to 5 GHz. At high frequency, the dielectric constant and the loss tangent of 40 vol.% ST composites are 11.0 and 0.01, respectively. It also shows a very good flexibility with elongation at a break value of 90%. This indicates that the ST/POE composites have the promising characteristics for potential use as the flexible dielectric waveguide and related flexible microwave devices.

Acknowledgements

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References

1. Hofmann, A., Horster, E., Wenizierl, J., Schmidt, L. P. and Brand, H., Flexible low-loss dielectric waveguide for THz frequencies with transitions to

metal waveguides. In *33rd European Microwave Conference Proceedings, Vol 3*, 2003, pp. 955–958.

2. Kehagias, N., Zankovych, S., Goldschmidt, A. *et al.*, Embedded polymer waveguides: design and fabrication approaches. *Superlattices and Microstructures*, 2004, **36**, 201–210.
3. Kim, K. Y., Tae, H.-S. and Lee, J.-H., Measurement of dielectric and radiation losses for flexible circular dielectric waveguides in Q-band. *Microwave and Optical technology Letters*, 2002, **35**(2), 102–106.
4. Obrzut, J. and Goldsmith, P. F., Flexible circular waveguides at millimeter wavelengths from metallized teflon tubing. *IEEE Transactions on Microwave Theory and Techniques*, 1990, **38**(3), 324–327.
5. Bruno, W. M. and Bridges, W. B., Flexible dielectric waveguides with powder cores. *IEEE Transactions on Microwave Theory and Techniques*, 1988, **36**(5), 882–890.
6. Moulart, A., Marrett, C. and Volton, J., Polymeric composites for use in electronic and microwave devices. *Polymer Engineering and Science*, 2004, **44**(3), 588–597.
7. Marrett, C., Moulart, A. and Volton, J., Flexible polymer composite electromagnetic crystals. *Polymer Engineering and Science*, 2003, **43**(4), 822–830.
8. Tagantsev, A. K., Sherman, V. O., Astafiev, K. F., Venkatesh, J. and Setter, N., Ferroelectric materials for microwave tunable application. *Journal of Electroceramics*, 2003, **11**, 5–66.
9. Misra, D., Chabbra, M., Epstein, B. R., Mirotznik, M. and Foster, K., Noninvasive electrical characterization of materials at microwave frequencies using an open-ended coaxial line: test of an improved calibration technique. *IEEE Transactions on Microwave Theory and Techniques*, 1990, **39**(1), 8–14.
10. Nyshadham, A., Sibbald, C. L. and Stuchly, S. S., Permittivity measurements using open-ended sensors and reference liquid calibration-an uncertainty analysis. *IEEE Transactions on Microwave Theory and Techniques*, 1992, **40**(2), 305–314.
11. Chen, L. F., Ong, C. K., Neo, C. P., Varadan, V. V. and Varadan, V. K., *Microwave Electronics Measurement and Materials Characterization*. John Wiley & Sons Ltd., 2004.
12. Jerzy, K., Developments in techniques to measure dielectric properties of low-loss materials at frequencies of 1–50 GHz. *Journal of the European Ceramic Society*, 2003, **23**(14), 2607–2610.
13. Jerzy, K., Precise measurements of the comples permittivity of dielectric materials at microwave frequencies. *Materials Chemistry and Physics*, 2003, **79**, 195–198.
14. Xiang, F., Wang, H. and Yao, X., Preparation and dielectric properties of bismuth-based dielectric/PTFE microwave composites. *Journal of the European Ceramic Society*, 2006, **26**, 1999–2002.
15. Grewe, M. G., Gururaja, T. R., Shrout, T. R. and Newnham, R. E., *IEEE Transaction on the Ultrasonics, Ferroelectrics, and Frequency Control*, 1990,3, **7**(6), 506–509.
16. Choy, T. C., *Effective Medium Theory*. Clarendon Press, Oxford, 1990.
17. Brosseau, C. and Talbot, P., Effective permittivity of nanocomposite powder compacts. *IEEE Transactions on Dielectric and Electrical Insulation*, 2004, **11**(5), 819–832.