J Electroceram (2010) 24:20–24
DOI 10.1007/s10832-008-9453-y

Low loss flexible $SrTiO_3$/POE dielectric composites for microwave application

Feng Xiang · Hong Wang · Haibo Yang · ZiYuan Shen · Xi Yao

Received: 31 August 2007 / Accepted: 12 February 2008 /

Abstract Flexible, high dielectric constant and low dielectric loss composites for microwave application fabricated with $SrTiO_3$ (STO) ceramic filler dispersed inside a thermoplastic polyolefin elastomer (POE) polymer matrix have been studied in this paper. The dielectric property and the mechanical property of STO/POE composites filled with different volume fraction of ceramic filler were investigated. The results indicated that with the increase volume fraction of ceramic filler, both the permittivity and the dielectric loss of composites increased. Good frequency stability within a wide range was observed in all the samples. For the composites containing 40 vol% STO, the composites has a tensile strength of 2.75 MPa with an elongation of about 90% at break value. The permittivity and the dielectric loss of the composites were 11.0 and 0.01 in microwave frequency, respectively. A microstrip transmission line on the composites containing 40 vol% STO as a microwave substrate is designed and measured after bending at different angles, meanwhile the transmission coefficients of the microstrip transmission line were unchanged when bending angle is less than 60°. This indicates that the STO/POE composites have the promising characteristics for potential applications in microwave substrate, flexible dielectric waveguide and related flexible microwave devices.

Keywords Composites · Dielectric properties · Mechanical properties

In recent years, there is an increasing interest on the flexible dielectric waveguide in the microwave frequency because of the various potential applications, including communication, military and medical care [1–4]. 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 application of a flexible polymer or a kind of ceramic is greatly restricted in the core of the flexible dielectric waveguide [3, 4]. Polymer-matrix composites reinforcing by ceramic powder would be an effective approach to improve the properties of materials in order to satisfy the flexibility and high dielectric constant [5–8]. 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. So 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 $SrTiO_3$ (STO) filler into thermoplastic polyolefin elastomer (POE) matrix. The dielectric properties and the mechanical properties of the STO/POE composites were investigated and reported in detail. At the same time, a microstrip transmission line on the composites substrate was manufactured and characterized.

1 Experimental

The POE (density=0.87 g/cm^3; glass transition temperature= −55 °C; elongation exceeds 800%) used was Engage™ POE 8100 (Dupont Dow Co., USA). The pure $SrTiO_3$ filler (Guo Teng 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

F. Xiang · H. Wang (✉) · H. Yang · Z. Shen · X. Yao
Electronic Materials Research Laboratory,
Key Laboratory of the Ministry of Education,
Xi'an Jiaotong University,
Xi'an 710049, China
e-mail: hwang@mail.xjtu.edu.cn

constant and the dielectric loss of 140 and 0.008 respectively in the microwave frequency range [9].

The surface modified STO filler and the POE were mixed for 12 min in a Rheomix600p system (Rheomix600p, HAAKE Co., Germany) operated at 60 rpm and 180 °C. Then, the mixture was put into a disk mold and hot-pressed under stress of 10 MPa at a temperature of 180 °C for 5 min.

The microstructure was analyzed using a scanning electron microscope (SEM) (JSM-6460, JEOL Ltd. Japan). Stress–strain behavior of STO/POE composites was measured on a tensile test machine PT-1036PC (Perfect Instrument Co. Ltd, China Taiwan) with a deformation speed of 5 mm/min.

The broad-band dielectric response of the composite samples was studied from 1 KHz to 8 GHz by different dielectric measurement systems as described as in the followings. The low frequency dielectric response in the range of 1 KHz–1 MHz was measured using a high-precision HP4284A LCR meter. Dielectric measurement in the high frequency range of 1 MHz–1 GHz were carried out by HP4291B impedance analyzer with HP16453A dielectric material test fixture. The frequency dependant dielectric behavior of the samples in microwave range from 500 MHz to 8 GHz were obtained by using open-reflection method with a HP8270ES network analyzer and a HP85070c open-ended coaxial line probe [10]. Dielectric measurements as a function of temperature were carried out with HP4291B impedance analyzer between −75 and 75 °C in conjunction with a computer-controlled Delta 9023 temperature chamber (Delta Design, Inc., San Diego, CA) at a heating rate of 3 °C/min. The temperature coefficient (α_ε) of dielectric was calculated from the following equation:

$$\alpha_\varepsilon = \frac{\varepsilon_{T_1} - \varepsilon_{T_0}}{\varepsilon_{T_0} \times (T_1 - T_0)} \tag{1}$$

where ε_{T_1} and ε_{T_0} is the permittivity in T_1 °C and T_0 °C, respectively.

In order to demonstrate the flexibility of the composites and give a clearer insight into the dielectric property of the STO/POE composites as a candidate for microwave conformal devices, a microstrip transmission line on the composites containing 40 vol% STO as a microwave substrate was designed. The substrate was 1-mm thick. The microstrip transmission line was fabricated with 0.1-mm-thick copper foil used to adhere to the substrate. The designed operating frequency of the microstrip transmission line was 3 GHz, and its impedance was 50 Ω. Two 50-Ω SMA connecters were used as the input and output ports. The transmission coefficient S_{11} and S_{12} of the microstrip transmission line was measured after bending at different angles to demonstrate the flexibility of the composites in microwave range from 2 to 5 GHz by a HP8270ES network analyzer. A standard full short-open-load-thru calibration was used.

2 Results and discussion

The stress–strain curve of STO /POE composites with different volume fraction of STO ceramic filler are shows in Fig. 1, the inset show the flexibility of the composites containing 40 vol% STO. The results showed that the mechanical properties decreased with the increasing of volume fraction of filler. The interface between ceramic filler and the polymer matrix and the dispersion of the filler particles in the matrix play an important role in determining the properties of a composite. SEM micrographs of STO/POE composites with different volume fraction of filler are shown in Fig. 2. In the case of the composite with 10 vol % STO, ceramic fillers were uniformly distributed in the polymer, as shown in Fig. 2(a). The tensile strength and elongation were above 7 MPa and approximately 600%, respectively. However, with the increase of volume fraction of filler, the composite with higher volume fraction of filler revealed serious agglomeration, resulting in worse mechanical properties. The micrograph showed that the filler is less homogeneously distributed in the composite with 40 vol%, as shown in Fig. 2(d). Compared to pure polymer matrix, both tensile strength and elongation were sharply decreased when 40 vol% filler was added into polymer. The composite has a tensile strength of 2.75 MPa with an elongation at break value of about 90%. Thus, the results indicate that the homogeneous distribution of fillers and firm interaction with the polymeric matrix are very important for the mechanical properties of composites [11–13].

As seen in the Fig. 3, the permittivity and dielectric loss of STO/POE composites with different volume fraction of fillers appear nearly a constant at frequencies from 500 MHz to 5 GHz. With the increase of STO filler ratio, the permittivity of composite increases significantly. For the 40 vol% STO composites, permittivity can reach approx-

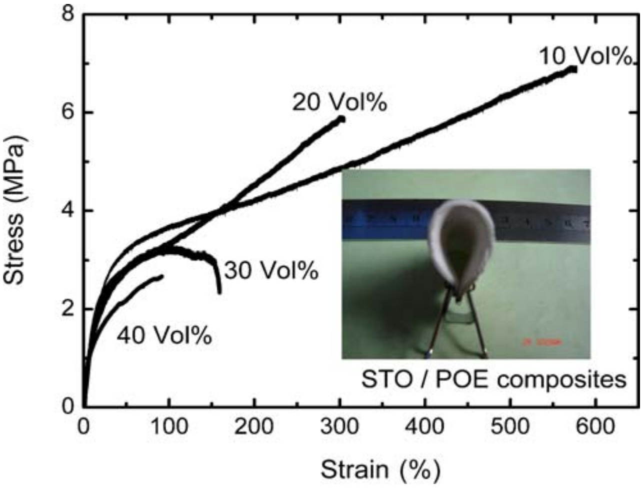

Fig. 1 The stress–strain curve of STO/POE composites with different volume fraction of STO ceramic filler. The *inset* show the flexibility of the composites containing 40 vol% STO

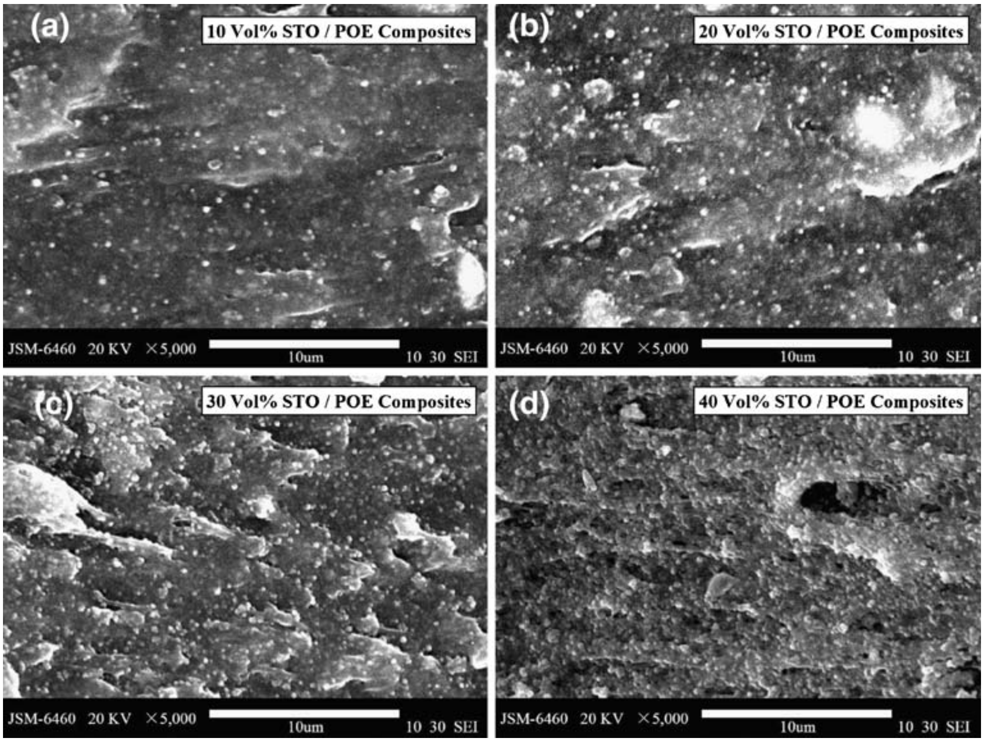

Fig. 2 SEM micrographs of STO/POE composites with different volume fraction of STO ceramic filler. (**a**) 10 vol%, (**b**) 20 vol%, (**c**) 30 vol%, (**d**) 40 vol%

imately 11.0, which is highly improved compared with the pure POE matrix (about 2.1). Dielectric loss of the composites slightly increases with the amount of STO filler increasing.

The core of the flexible dielectric waveguide would be a flexible dielectric material with high dielectric constant and low dielectric loss in microwave range. Thus, the composites containing 40 vol% filler were investigated as a function of temperature and frequency. The frequency dependence of the permittivity and dielectric loss tanδ of composites at frequency between 1 kHz and 5 GHz are shows in Fig. 4. The permittivity of composites shows good stability in a wide range of frequency. This indicates that the flexible, high dielectric constant and low dielectric loss ST/POE composites have the promising characteristics for potential microwave applications in a wide range of operating frequencies.

Temperature dependence of the permittivity and dielectric for composites in the temperature range from −75 to 75 °C at 50 MHz is shown in Fig. 5. In the composites, the permittivity decreases with temperature increasing. The dielectric loss is low, with only a slight increase as temperature increasing. The α_ε of composites was approximately −1,480 ppm/°C in 50 MHz.

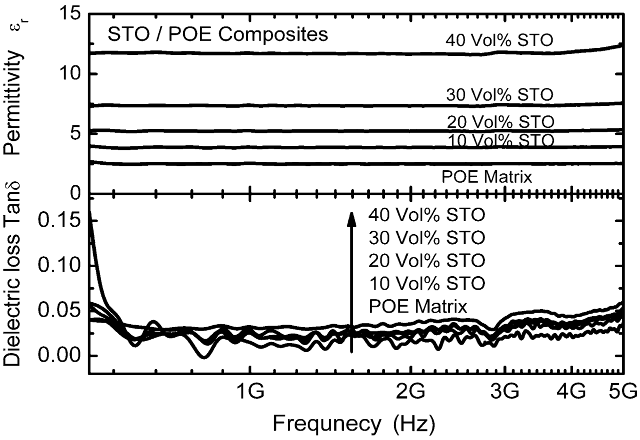

Fig. 3 The frequency dependence of the permittivity ε_r (*top*) and dielectric loss tanδ (*bottom*) of STO/POE composites with different volume fraction of STO ceramic filler at microwave frequencies (measured by open-reflection method)

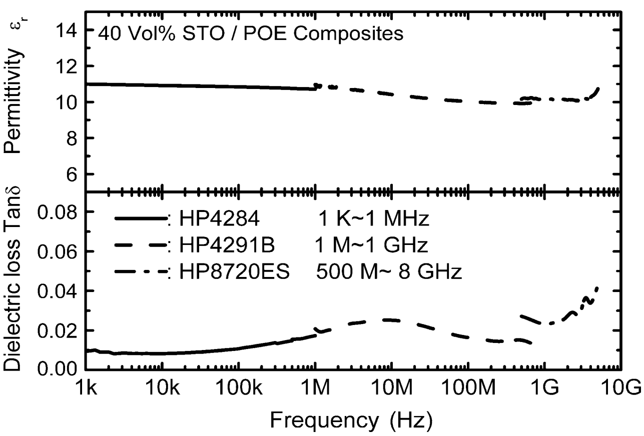

Fig. 4 The frequency dependence of the permittivity ε_r (*top*) and dielectric loss tanδ (*bottom*) of the STO/POE composites containing 40 vol% STO filler

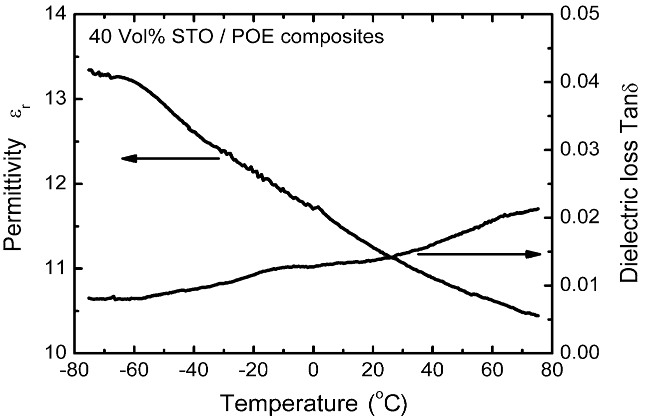

Fig. 5 The permittivity ε_r and the dielectric loss tanδ as a function of temperature for a composites containing 40 vol% STO filler, the measuring frequency was 50 MHz

A microstrip transmission line on the composites containing 40 vol% STO as a microwave substrate (length= 50 mm, width=10 mm, thickness=1 mm) is designed and measured after being bended at different angles to demonstrate the flexibility of the composites. The permittivity and the dielectric loss of the composites were 11.0 and 0.01 in microwave frequency, respectively. The microstrip transmission line had a thickness of 0.1 mm, a line-width of 0.8 mm, and a line-to-ground plate spacing of 1 mm, as shown in Fig. 6(a).

One of the major questions concerning the microstrip transmission line on flexible composites substrate is how the performance of composites substrate is changing as the bending angle is increasing. Figure 6(b) and (c) shows the transmission coefficient S_{11} and S_{12} of the microstrip transmission line dependence on different bending angles. The simulations of the microstrip transmission line were carried out with Ansoft HFSS for comparison. The insets in (b) and (c) show the simulated transmission coefficient at 0° bending angle.

As shown in Fig. 6(b) and (c), a minor disagreement is observed in the measured and simulated the transmission coefficient S_{11} and S_{12} of the microstrip transmission line at 0° bending angle, and this is likely due to the fact that the parameters between simulation and measurement might be slightly different. When the flexible substrate was bent, the transmission coefficients of the microstrip transmission line showed no significant change at 0°, 30° and 60° bending angle. However, the transmission coefficient at 90° bending angle was obvious shifted, which is probably due to the

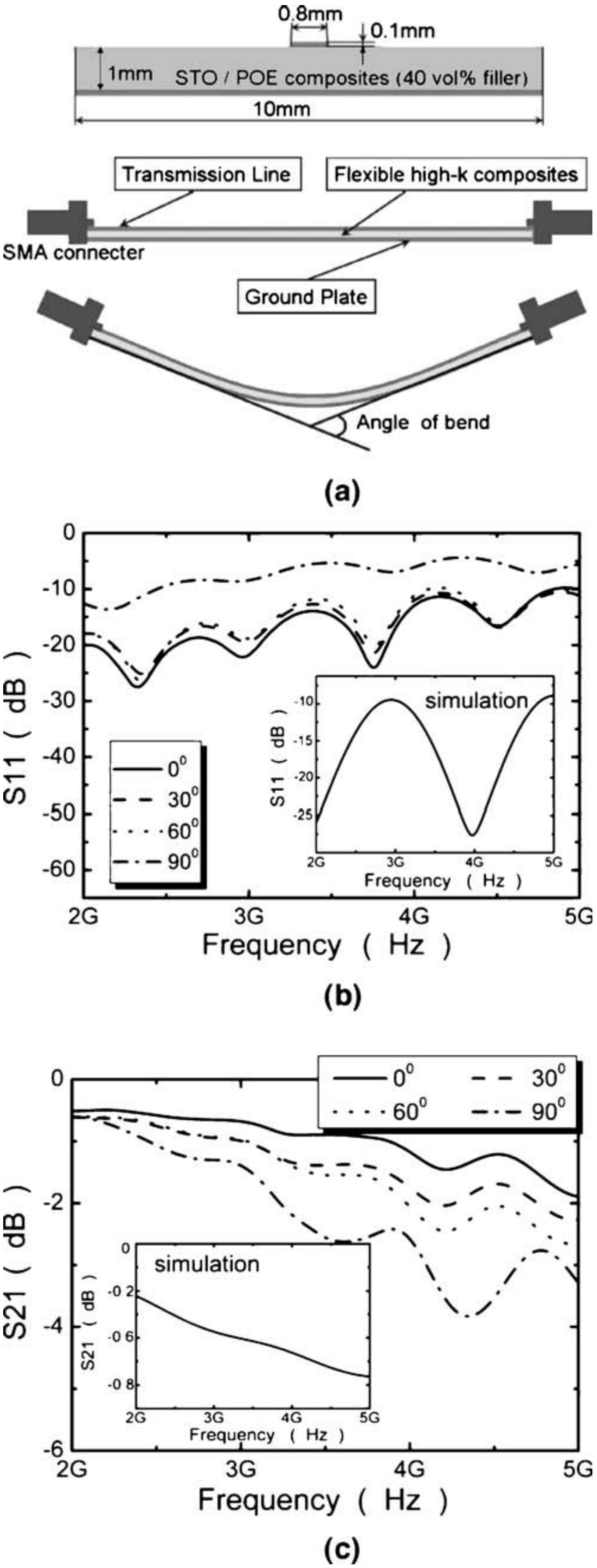

Fig. 6 Geometrical details of the microstrip transmission line on the composites containing 40 vol% STO as a microwave substrate (**a**), the transmission coefficient S_{11} (**b**) and S_{12} (**c**) of the microstrip transmission line at different angles, The *insets* in (**b**) and (**c**) show the simulated transmission coefficient at 0° bending angles

splitting the copper foil transmission line off the flexible substrate. After repeated bending of the flexible substrate, no cracks in the composites were seen. The results show that the STO/POE composites do have good flexibility and dielectric properties for the use in microwave conformal devices.

3 Conclusions

STO/POE flexible composites with various volume fractions of STO filler were prepared by using the extrusion technology. The mechanical properties of composites decreased with increasing volume fraction of filler. For the composites with 40 vol% STO filler, the composite has a tensile strength of 2.75 MPa with an elongation at break value of about 90%. The permittivity of composites is 11 and show good stability in a wide range of frequency. Dielectric loss remained below 2% in a wide range of temperature and frequency. The α_ε of composites was approximately −1,480 ppm/°C in 50 MHz. A microstrip transmission line on the composites containing 40 vol% STO as a microwave substrate was designed and measured after bending at different angles to demonstrate the flexibility and dielectric properties of the composites. The results indicate that the STO/POE composites can satisfy high dielectric constant for the flexible microwave conformal devices.

Acknowledgement This work was supported by the National 973-project of China (grant 2002CB613302), the Ph.D. Program Fund from the Ministry of Education of China (grant 20060698007) and NSFC project of China (grant 50572085).

References

1. A. Hofmann, E. Horster, J. Wenizierl, L.P. Schmidt, H. Brand, 33rd European Microwave Conference Proceedings **3**, 955 (2003)
2. K.Y. Kim, H-S. Tae, J-H. Lee, Microw. Opt. Technol. Lett **35**(2), 102 (2002)
3. J. Obrzut, P.F. Goldsmith, IEEE Trans. Microwave Theor. Tech **38** (3), 324 (1990)
4. W.M. Bruno, W.B. Bridges, IEEE Trans. Microwave Theor. Tech **36**(5), 882 (1988)
5. A. Moulart, C. Marrett, J. Volton, Polym. Eng. Sci **44**(3), 588 (2004)
6. F. Xiang, H. Wang, X. Yao, J. Eur. Ceram. Soc **26**(10–11), 1999 (2006)
7. S.C. Tjong, Mater. Sci. Eng. B **53**, 73 (2006)
8. C-CM. Ma, Y-J. Chen, H-C. Kuan, J. Appl. Polym. Sci **100**, 508 (2006)
9. A.K. Tagantsev, V.O. Sherman, K.F. Astafiev, J. Venkatesh, N. Setter, Journal of Electroceramics **11**, 5 (2003)
10. L.F. Chen, C.K. Ong, C.P. Neo, V.V. Varadan, V.K. Varadan, *Microwave Electronics: Measurement and Materials Characterization* (Wiley, New York, 2004)
11. C.K. Huang, S.W. Chen, W.C.J. Wei, J. Appl. Polym. Sci **102**, 6009 (2006)
12. H. Tang, Q. Qi, Y. Wu, G. Liang, L. Zhang, J. Ma, Macromol. Mater. Eng **291**, 629 (2006)
13. S. Koulouridis, G. Kiziltas, Y. Zhou, D.J. Hansford, J.L. Volakis, IEEE Trans. Microwave Theor. Tech **54**, 4202 (2006)