Available online at www.sciencedirect.com
SciVerse ScienceDirect
Ceramics International 39 (2013) 4945–4951
CERAMICS INTERNATIONAL
www.elsevier.com/locate/ceramint

Electrical and thermal properties of low permittivity $Sr_2Al_2SiO_7$ ceramic filled HDPE composites

K.M. Manu, S. Ananthakumar, M.T. Sebastian*

Materials Science and Technology Division, National Institute for Interdisciplinary Science and Technology (NIIST), CSIR, Thiruvananthapuram 695019, India

Received 2 November 2012; received in revised form 26 November 2012; accepted 27 November 2012
Available online 5 December 2012

Abstract

The feasibility of low permittivity $Sr_2Al_2SiO_7$ (SAS) ceramic filled high density polyethylene (HDPE) composites for substrate and packaging applications has been investigated in this paper. The composites were prepared by the melt mixing and hot pressing techniques. Scanning electron microscopic images of SAS filled HDPE showed the increased connectivity with filler loading. The composites showed excellent relative density ($>98\%$) with low bulk density ($<2.40\ g\ cm^{-3}$) and very low moisture absorption (<0.10 wt%). The relative permittivity (ε_r) and the dielectric loss ($\tan\delta$) at 1 MHz and at 5 GHz were found to be low and found to increase with filler volume fraction (V_f). The experimentally observed relative permittivity at 5 GHz was correlated with the values proposed by different theoretical models. Among them, effective medium theory (EMT) gave better fit with experimental values except at the highest filler loading (0.50 V_f). Improvement in the thermal properties was also observed with filler content. The coefficient of linear thermal expansion (CTE) was found to decrease with filler content. Thermal conductivity (TC) of the composite was greatly enhanced as a function of filler volume fraction. The composite with 0.50 filler volume fraction showed balanced thermal and dielectric properties with $\varepsilon_r=4.2$, $\tan\delta=3.9\times10^{-3}$, $TC=2.2\ W\ m^{-1}\ K^{-1}$ and CTE = 101 ppm/°C.

Keywords: C. Thermal properties; D. Silicate; E. Substrate; Microwave dielectric properties

1. Introduction

Recently, wireless communication using microwaves has become the dominant way of information exchange. The electronic consumer market is continuously seeking new low cost light weight microwave modules which can transfer large amount of information with a high signal speed. Electrical and thermal behavior of the dielectric substrate have great influence on the overall performance of the microwave module [1]. Low relative permittivity (ε_r) materials with low dielectric loss ($\tan\delta$) can be efficiently used as substrates or packages which can deliver high signal speed with good selectivity at microwave frequencies [2–4]. Moreover, a microwave substrate should have a low coefficient of linear thermal expansion (CTE) and a high thermal conductivity (TC) for practical applications [3,4]. Low bulk density is also required for producing light weight microwave devices. Several silicate based ceramics were reported as good dielectric materials with low ε_r and low $\tan\delta$ or high quality factor ($Q_u\times f$) [4–9]. Brittle nature of these ceramics prevents their immediate practical application in portable communication devices. Moreover, they need high processing temperatures. Polymers on the other hand, offer good flexibility, low processing temperature, light weight in nature and excellent electric insulations. Certain polymers show low ε_r and very low $\tan\delta$ [10]. However, poor thermal properties especially low TC and high CTE restrict their immediate use as substrates [11]. Recently, Button et al. [12] proposed a method of reinforcing the ceramic filler into the polymer matrix thereby achieving balanced thermal, mechanical and dielectric properties which cannot be achieved by the individual components alone. The main advantage of this method is that the properties can be designed as per the consumer interest by the proper selection of polymer and

---

*Corresponding author. Tel.: +91 471 2515294.
E-mail address: mailadils@yahoo.com (M.T. Sebastian).

0272-8842/$ - see front matter © 2012 Elsevier Ltd and Techna Group S.r.l. All rights reserved.
http://dx.doi.org/10.1016/j.ceramint.2012.11.090

ceramic and also by varying the filler volume fraction. Recent literature reports utility of several low loss, low permittivity polymer–ceramic composites for electronic packaging applications [11,13–18].

The properties of polymer–ceramic composites depend on the individual components and also the size and shape of the filler particles. Low loss polymer with good flexibility, ease of processing at relatively low temperatures can be used as the matrix. The microwave dielectric properties of several low loss polymers have been reported in the literature [10]. Fluropolymers like polytetra fluroethylene (PTFE) shows excellent microwave dielectric properties [10]. Several PTFE based 0–3 connected low loss polymer–ceramic composites suitable for substrate applications have been reported in the literature with excellent dielectric properties [11,13,16,19,20]. However, PTFE is very difficult to process due to its high melt viscosity [11]. High density polyethylene on the other hand can be easily processed at a relatively low temperature (~180 °C). Moreover, it shows excellent dielectric properties at microwave frequencies ($\varepsilon_r = 2.3$ and $\tan\delta \sim 10^{-4}$) [10]. A few recent reports show excellent utilization of ceramic filled HDPE composites as microwave substrates [15,17,21–23]. However, the increase in demand for low cost and light weight substrates with balanced electrical and thermal properties stimulated us to develop new HDPE based polymer–ceramic laminates. Low relative permittivity ceramic with a low dielectric loss is an ideal choice as the ceramic filler for the development of polymer–ceramic substrate. In our previous paper, we have reported the microwave dielectric properties of $Sr_2Al_2SiO_7$ ceramic with $\varepsilon_r = 7.5$ (porosity corrected) and quality factor ($Q_u \times f$) = 33000 GHz [7]. Hence in the present investigation, we have selected $Sr_2Al_2SiO_7$ ceramic as the filler. The paper reports the effect of SAS filler content on the thermal and dielectric properties of HDPE composites for the first time.

2. Experimental

The $Sr_2Al_2SiO_7$ ceramic filler was prepared by the conventional solid state ceramic route. High purity powders of $SrCO_3$ (99.9%, Sigma Aldrich, Inc., Milwaukee, WI, USA), Al_2O_3 (99.7%, Sigma Aldrich) and SiO_2 (99.6%, Sigma Aldrich) were weighed in stochiometric proportions and ball milled for 24 h. The slurry was dried and the powder was calcined at 1350 °C for 4 h. Calcined powder was again ball milled in water medium for 24 h for reducing the particle size and sieved using a 25 μm sieve. The melt mixing technique using a kneading machine was adopted to mix the different volume fractions (V_f) of SAS ceramic with high density polyethylene (HDPE) (Kerala Plastics, Kerala, India) at 180 °C for 30 min. The composite thus obtained were hot pressed at 180 °C for 30 min by applying a pressure of 2 MPa for the microwave dielectric (length = 50 mm, breadth = 50 mm and thickness = 0.8 mm), radio frequency (diameter = 11 mm and thickness = 1.5 mm), thermal conductivity (diameter = 12.85 mm and thickness = 2.5 mm) and linear thermal expansion (diameter = 8 mm and thickness = 10 mm) measurements.

The phase purity of the ceramic filler was characterized by the powder X-ray diffraction (XRD) techniques (PANalytical X'Pert PRO Diffractometer having Ni filtered CuKα radiation, Netherlands). The microstructure of the composites was studied by a scanning electron microscope (SEM) (JEOL-SEM 560lv, Tokyo, Japan). The bulk densities of the specimen were measured by the Archemedies method. The moisture absorption measurement was done for the samples used for the microwave measurements (length = 50 mm, breadth = 50 mm and thickness = 0.8 mm). The weights of the samples were noted initially and the samples were dipped in distilled water for 24 h. The samples were then taken out and the surface water was wiped out and again weighed. The percentage of water absorption can be calculated using the equation

$$\%\text{water absorbed} = \frac{W_f - W_i}{W_i} \times 100 \tag{1}$$

where W_f and W_i are weight of the samples before and after immersing in distilled water. The dielectric properties at 1 MHz were measured by an LCR meter (Hioki 3532-50 LCR HiTESTER, Nagano, Japan) using the parallel plate capacitor method by silver pasting the samples on both sides. The microwave dielectric properties were measured by the $TE_{01\delta}$ resonant mode of the split post dielectric resonator (SPDR) using a vector network analyzer (E 5071C, Agilent, USA). The quality factor and the resonant frequencies of the SPDR before and after inserting the planar samples were measured. The iterative solutions to the following equations proposed by Krupka [24,25] were used for the calculation of relative permittivity and the dielectric loss of the composites.

$$\varepsilon_r = 1 + \frac{f_0 - f_s}{h f_0 K_\varepsilon(\varepsilon_r, h)} \tag{2}$$

$$\tan\delta = \left(Q_u^{-1} - Q_{DR}^{-1} - Q_C^{-1}\right)/\rho_{es} \tag{3}$$

where h is the thickness of the sample under test, f_0 is the resonant frequency of the fixture without sample, f_s is the resonant frequency of fixture with the sample, K_ε (ε_r, h) is a function of ε_r and h that has to be evaluated using Rayleigh–Ritz technique, Q_u is the unloaded quality factor of resonant fixture containing the dielectric sample, Q_c^{-1} and Q_{DR}^{-1} are losses of metallic and dielectric parts of the resonator respectively and ρ_e is the electric energy filling factor of the sample. These calculations were done with the aid of a high precision software (QWED, Poland). The temperature variation of relative permittivity at 1 MHz was also measured in the temperature range 25–70 °C using the LCR meter. The coefficient of linear thermal expansion were measured using a Dilatometer (DIL 402 PC, Netzsch, Selb, Germany) in the temperature range 30–80 °C using the relation

$$\text{CTE} = \frac{1}{L} \times \frac{\Delta L}{\Delta T} \tag{4}$$

where L is the length of sample at 30 °C, ΔL difference in length of samples for the difference in temperature ΔT.

Ceramic pellet (diameter = 8 mm and thickness = 10 mm) sintered at 1525 °C for 4 h was used for the measurement of CTE of the filler. Thermal conductivity of the samples were measured by the laser flash technique [26] using a thermal conductivity analyzer (Flash Line™ 2000, Anter Corporation, USA) by the relation

$$TC = \lambda \times C_p \times \rho \tag{5}$$

where λ is the thermal diffusivity, C_p is the specific heat capacity at room temperature and ρ is the density of the sample. The thermal conductivity of virgin HDPE was measured by coating a thin layer of silver on both sides of the sample in order to avoid its semi transparent nature and the ceramic pellet sintered at 1525 °C for 4 h having diameter 12.6 mm was used for the measurement of thermal conductivity of SAS filler.

3. Results and discussion

The ceramic filler must be phase pure for the exact prediction and control of dielectric as well as thermal properties of polymer–ceramic composites. Fig. 1 shows powder X-ray diffraction pattern of $Sr_2Al_2SiO_7$ filler calcined at 1350 °C for 4 h. The observed patterns were indexed and found to be matching with the standard ICDD file no: 38–1333. The absence of any extra peaks indicates the phase purity of SAS ceramic at this temperature.

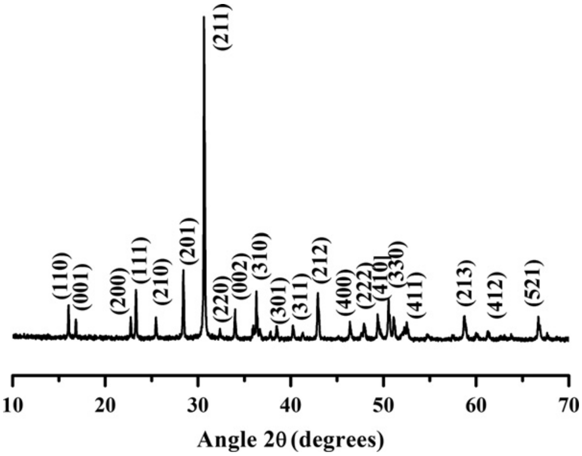

Fig. 1. Powder X-ray diffraction pattern of SAS filler.

The surface morphology of 0.10 V_f and 0.40 V_f SAS ceramic filled HDPE composites are shown in Fig. 2(a) and (b) respectively. Connectivity between the ceramic particles was found to be increasing with the increase in filler content. Agglomeration of ceramic particles can also be observed at higher filler loading which in turn increases the porosity and can be correlated with the reduction in the relative density with increase in filler content (Table 1).

Table 1 gives the bulk density, relative density, moisture absorption and the dielectric properties at 1 MHz for different filler volume fractions of HDPE–SAS composites. The bulk densities of all the compositions were relatively low compared to commercially available ceramic based substrate materials like Al_2O_3 (bulk density = 3.9 g cm^{-3}) which make them attractive for the development of light weight microwave modules. A gradual decrease in the relative density with increase in filler content might be attributed to development of porosity as a result of aggregation of ceramic filler (Fig. 2(b)).

Absorption of moisture seriously affects the microwave dielectric properties of polymer–ceramic composites [27]. The composite when exposed to humid environment can absorb a few weight percentages of water. The polar water molecule usually shows high ε_r and high tan δ. Moreover, the water molecule acts as an interlayer between ceramic and the polymer leading to interfacial polarization thereby causing an enhancement in the dielectric loss and relative permittivity [27,28]. The dipolar relaxation caused by the moisture absorption will degrade the dielectric properties at microwave frequencies [4,28]. Large moisture absorption may also degrade the mechanical properties. Hence low moisture absorption is preferred for practical applications. It was reported by Laverghetta [28] that the upper limit of moisture absorption for electronic packaging applications is 0.10 wt%. In the present investigation, the moisture absorption of all the compositions were less than 0.10 wt% (Table 1). It can be seen from the Table 1 that the moisture absorption increased with increase in filler content and reached a maximum value of 0.09 wt% for

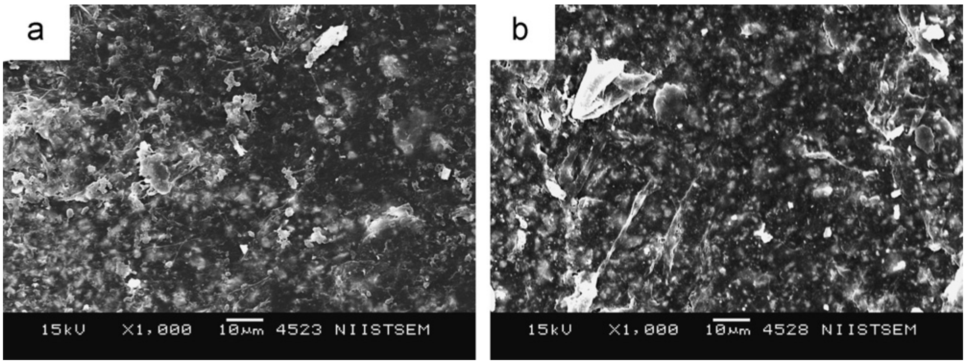

Fig. 2. Scanning electron microscopic images SAS ceramic filled HDPE composites for (a) 0.10 V_f and (b) 0.40 V_f of the filler.

Table 1
Bulk density, relative density, moisture absorption and the dielectric properties (at 1 MHz) of SAS ceramic filled HDPE composites.

Volume fraction of the filler (V_f)	Bulk density (g cm^{-3})	Relative density (%)	At 1 MHz		Moisture absorption (wt%)
			ε_r	tan δ	
0.1	1.2251	99.2	2.9	7.6×10^{-4}	0.03
0.2	1.5048	99.0	3.3	1.1×10^{-3}	0.03
0.3	1.6269	99.0	3.8	1.2×10^{-3}	0.03
0.4	2.0586	98.5	4.2	1.2×10^{-3}	0.05
0.5	2.3323	98.2	4.4	1.9×10^{-3}	0.09

0.50 filler volume fraction which is attributed to the increase in porosity.

Table 1 also gives the dielectric properties of HDPE–SAS composites at 1 MHz. The values of ε_r and tan δ were found to increase with increase in filler content. The relative permittivity of HDPE+0.10 V_f SAS is 2.9 and that of HDPE+0.50 V_f SAS is 4.4. It has been found that the relative permittivity of SAS ceramic at 1 MHz is 7.6 which is relatively higher than that of virgin HDPE (2.5). Moreover, as seen from the SEM images (Fig. 2), the particles come close to each other with the increase in filler content and the dipole–dipole interaction increases thereby enhancing the relative permittivity of the composites [23,29]. Various extrinsic and intrinsic factors contribute to the dielectric loss of the composite system. Intrinsic dielectric loss is due to the interaction of phonon with the electric field [30]. The extrinsic loss is associated with heterogeneity and connectivity of the system, porosity and agglomeration of ceramic particles [11]. The dielectric losses of individual components also affect the dielectric loss of the composite structure. The dielectric loss of SAS ceramic at 1 MHz sintered at 1525 °C for 4 h is 2.7×10^{-3} and that of virgin HDPE is 6.2×10^{-4}. In the present investigation, the value of tan δ was 7.6×10^{-4} for 0.10 V_f SAS filled HDPE and it increased to a value of 1.9×10^{-3} for 0.50 V_f of filler.

Fig. 3 shows variation of relative permittivity and the dielectric loss of SAS ceramic filled HDPE composites at 5 GHz. The dielectric properties varied in a similar manner as observed at 1 MHz with filler loading. The maximum value of relative permittivity was observed for HDPE+0.50 V_f SAS composite and it was found to be 4.2. The values of tan δ were found to be slightly higher than that observed at 1 MHz which can be attributed to the increased intrinsic loss with the increase in frequency [4]. All the composites showed good dielectric properties with low values of ε_r and tan δ. The observed properties of HDPE+0.50 V_f SAS composite ($\varepsilon_r=4.2$ and tan $\delta=3.9\times10^{-3}$) indicate its suitability for substrate and packaging applications.

Exact prediction of dielectric properties is desirable to develop composite structures as per the requirements of circuit designer. Various theoretical models for predicting the relative permittivity of composites have been proposed in the literature [11,13,31–33]. Fig. 4 shows a comparison of experimentally observed ε_r values at 5 GHz as a function of filler loading with the values obtained using the following theoretical models.

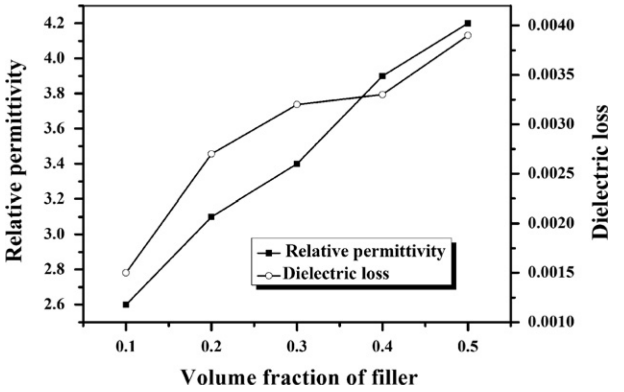

Fig. 3. Microwave dielectric properties of HDPE–SAS composites as a function of filler volume fraction at 5 GHz.

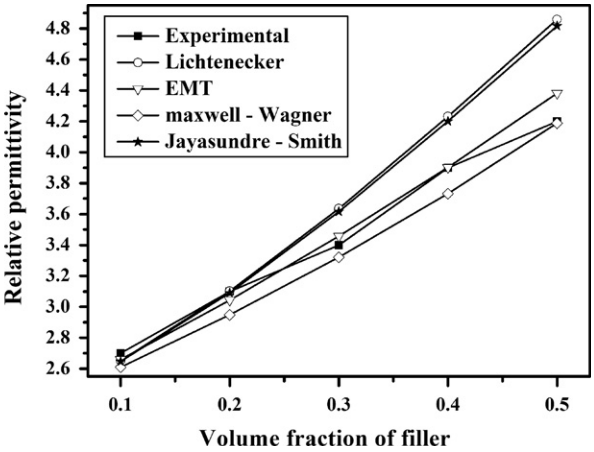

Fig. 4. Theoretical modelling of relative permittivity HDPE–SAS composites at 5 GHz.

Jayasundare–Smith equation [31]

$$\varepsilon_{eff}=\frac{\varepsilon_m\left(1-V_f\right)+\varepsilon_f V_f\left[\frac{3\varepsilon_m}{\varepsilon_i+2\varepsilon_m}\right]\left[1+\left(\frac{3V_f\left(\varepsilon_f-\varepsilon_m\right)}{\varepsilon_i+2\varepsilon_m}\right)\right]}{1-V_f+V_f\left[\frac{3\varepsilon_m}{\varepsilon_f+2\varepsilon_m}\right]\left[1+\left(\frac{3V_f\left(\varepsilon_f-\varepsilon_m\right)}{\varepsilon_f+2\varepsilon_m}\right)\right]} \tag{6}$$

Lichtenecker equation [33]

$$\ln\varepsilon_{eff}=\left(1-V_f\right)\ln\varepsilon_m+V_f\ln\varepsilon_f \tag{7}$$

Maxwell–Wagner equation [13]

$$\varepsilon_{eff} = \varepsilon_m \frac{2\varepsilon_m + \varepsilon_f + 2V_f\left(\varepsilon_f - \varepsilon_m\right)}{2\varepsilon_m + \varepsilon_f - V_f\left(\varepsilon_f - \varepsilon_m\right)} \tag{8}$$

Effective medium theory equation [32]

$$\varepsilon_{eff} = \varepsilon_m \left[1 + \frac{V_f\left(\varepsilon_f - \varepsilon_m\right)}{\varepsilon_m + n\left(1 - V_f\right)\left(\varepsilon_f - \varepsilon_m\right)}\right] \tag{9}$$

where, ε_{eff}, ε_f, and ε_m are the relative permittivity of the composites, filler and matrix respectively and V_f is the volume fraction of the filler. Different models follow different theoretical assumptions. These assumptions are related to the dielectric properties of individual components, size and shape of filler, morphology and the connectivity of polymer–ceramic composites. In the present investigation, all the models agree with experimental values at lower filler loading and the deviation was observed at higher filler loading. Imperfect dispersion of ceramic particles and the porosity observed in the composite system might be the reason for this deviation. Jayasundare–Smith model showed large deviation from the experimental values. This model considers the interaction between neighboring particles only which could be the reason for this poor fitting. Also in this model, particles are assumed to be exactly in spherical which may not be true in the present study. The SEM images support this conclusion (Fig. 2). The most common mixing rule in a bi-phase system is the logarithmic mixing rule which is used in the Lichtenecker equation. The values obtained in this model also make poor correlation with the experimental values. In the Lichtenecker equation, composite can be considered as a random mixture of dielectric spheres in the continuous polymer matrix. In the present investigation, the Maxwell–Wagner model also did not give the best fit. This model is suitable for a system in which individual components have similar properties. Significant difference between the relative permittivities of the SAS filler and HDPE matrix might be the reason for this poor fitting. The values obtained by the effective medium theory (EMT) proposed by Rao et al. gave better fit with experimentally observed compared to other models. EMT model assumes the composite system as a random unit cell consisting of each filler particle surrounded by the continuous matrix layer [32]. In EMT model the morphology factor '*n*' was determined empirically and was found to be 0.202. The small *n* value indicates the nearly spherical shape of the filler particle which is essential for the exact prediction of relative permittivity. In the present system, EMT model gives best fit up to 0.40 V_f above which agglomeration of ceramic particles occurs leading to poor fitting. At 1 MHz also, EMT model gave best fit up to 0.40 V_f of SAS ceramic with the same '*n*' value.

The variation of relative permittivity at 1 MHz as a function of temperature is shown in Fig. 5. The ε_r values were found to decrease with increase in temperature for all the compositions. On the other hand, SAS ceramic showed a positive temperature coefficient of relative permittivity (τ_ε) of 72 ppm/°C. Earlier reports of virgin HDPE [34] and HDPE based polymer–ceramic composites [22] showed a decrease in relative permittivity as a function of temperature similar to the observation made in the present investigation. Thermal expansions of individual components also play a dominant role in this behavior. It has been reported earlier that composites having large mismatch between the thermal expansion of filler (1.1 ppm/°C in the present investigation) and the matrix (230 ppm/°C in the present investigation) would disturb the aggregation of polar components thereby reducing the relative permittivity with temperature [20,35,36].

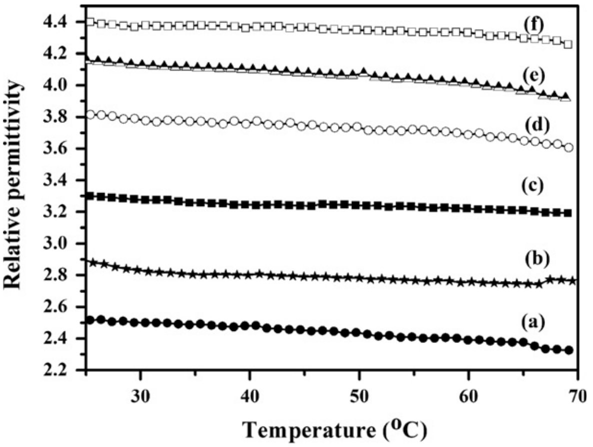

Fig. 5. Temperature variation of relative permittivity of HDPE–SAS composites for (a) 0.00 V_f (b) 0.10 V_f (c) 0.20 V_f (d) 0.30 V_f (e) 0.40 V_f and (f) 0.50 V_f of SAS filler.

The thermal properties of polymeric materials can be improved with the addition of ceramic filler. Fig. 6 shows variation of thermal conductivity and coefficient of linear thermal expansion as a function of filler loading. The thermal conductivity of HDPE+0.10 V_f of SAS is 0.82 W m^{-1} K^{-1} and reached a maximum value of 2.2 W m^{-1} K^{-1} for 0.50 V_f of filler. Usually ceramics show high TC values compared to that of polymers [37]. In the present study, TC values of HDPE matrix and SAS filler were found to be 0.6 W m^{-1} K^{-1} and 4.2 W m^{-1} K^{-1} respectively. The number of connecting paths between the filler particles increases with increase in filler content which in turn increases transfer of heat through the matrix causing an enhancement in the thermal conductivity value. The CTE value of HDPE with 0.10 V_f SAS is 234 ppm/°C and it decreased to a value of 101 ppm/°C for 0.50 V_f of filler content. The movement of polymer chains will be arrested with the increase in filler content and unable to expand with temperature thereby reducing the CTE. CTE value of SAS ceramic is very low (1.1 ppm/°C) compared to HDPE matrix (230 ppm/°C) which can also be correlated with the observed reduction in CTE for HDPE–SAS composites.

4. Conclusions

Low permittivity $Sr_2Al_2SiO_7$ ceramic filled HDPE composites were developed for substrate and packaging

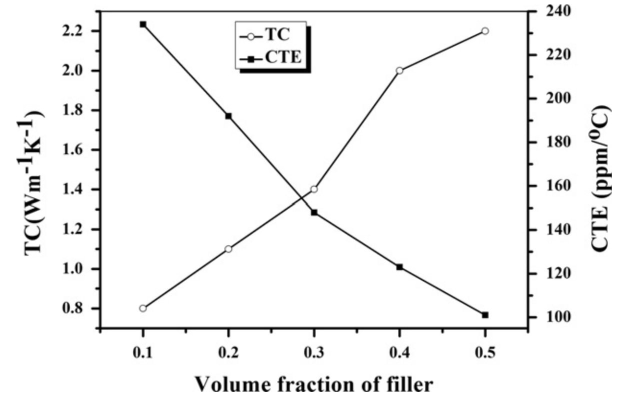

Fig. 6. Variation of thermal conductivity (TC) and the coefficient of linear thermal expansion (CTE) of HDPE–SAS composites as a function of SAS loading.

applications by melt mixing and hot pressing techniques. The surface morphology using scanning electron microscope was done which confirms the increased connectivity of the ceramic particles with increase in filler loading in the composite. The composites showed excellent densification and it was found to be decreasing with the increase in filler content. Moisture absorption of all the composites were found to be very low (<0.10 wt%). Different theoretical models were used to predict the relative permittivity at 5 GHz. Among them, only the EMT model gave best fit with the experimentally observed values. The thermal properties of HDPE matrix was improved with filler loading. All the composites showed low relative permittivity (<4.3) and low dielectric loss ($\sim 10^{-3}$) at 5 GHz. The composite HDPE + 0.50 V_f SAS showed balanced dielectric and thermal properties along with low moisture absorption and low bulk density. The observed physical, thermal and dielectric properties indicate its possibility for substrate and packaging applications in radio as well as microwave frequencies for the development of light weight electronic modules.

Acknowledgments

K.M. Manu is grateful to Council of Scientific and Industrial Research (CSIR), India for the award of senior research fellowship. The authors are grateful to Dr. P. Prabhakar Rao and Mr. M.R. Chandran of NIIST (CSIR) for extending XRD and SEM facilities.

References

[1] R. Tummala, M. Kosec, W.K. Jones, D. Belavic, Electronic Packaging for High Reliability; Low Cost Electronics, Kluwer Academic Publishers, The Netherlands: Dordrecht, 1999.

[2] K. Maexa, M.R. Baklanov, D. Shamiryan, F. Iacopi, S.H. Brongersma, Z.S. Yanovitskaya, Low dielectric constant materials for microelectronics, Journal of Applied Physics 93 (2003) 8793–8841.

[3] D.D.L. Chung, Materials for Electronic Packaging, Butterworth, Heinemann: Boston, 1995.

[4] M.T. Sebastian, Dielectric Materials for Wireless Communication, Elseiver Publishers, Oxford, UK, 2008.

[5] H. Ohsato, T. Tsunooka, T. Sugiyama, K. Kakimoto, H. Ogawa, Forsterite ceramics for millimeterwave dielectrics, Journal of Electroceramics 17 (2006) 445–450.

[6] T. Tsunooka, T. Sugiyama, H. Ohsato, K. Kakimoto, M. Andou, Y. Higashida, H. Sugiura, Development of forsterite with high Q and zero temperature coefficient TF for millimeterwave dielectric ceramics, Key Engineering Materials 269 (2004) 199–202.

[7] K.M. Manu, T. Joseph, M.T. Sebastian, Temperature compensated $Sr_2Al_2SiO_7$ ceramic for microwave applications, Materials Chemistry and Physics 133 (2012) 21–23.

[8] S. Thomas, M.T. Sebastian, Microwave Dielectric Properties of $SrRE_4Si_3O_{13}$ (RE = La, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Er, Tm, Yb, and Y) ceramics, Journal of the American Ceramic Society 92 (2009) 2975–2981.

[9] K.M. Manu, P.S. Anjana, M.T. Sebastian, Low permittivity $SrCuSi_4O_{10}$—LMZBS glass composite for LTCC applications, Materials Letters 65 (2011) 565–567.

[10] A.J. Bur, Dielectric properties of polymers at microwave frequencies: a review, Polymer 26 (1985) 963–977.

[11] M.T. Sebastian, H. Jantunen, Polymer–ceramic composites of 0–3 connectivity for circuits in electronics: a review, International Journal of Applied Ceramic Technology 7 (2010) 415–434.

[12] D.P. Button, B.A. Yost, R.H. French, W.Y. Hsu, J.D. Belt, M.A. Subrahmanian, H.-M. Zhang, R.E. Geidd, A.J. Whittacker, D.G. Onn, Ceramic-Fiber/Polymer Laminates: Thermally Conductive Composites With Low Dielectric Constants, vol. 26, American Ceramic Society, Westerville, OH, 1989, pp. 353–373.

[13] K.P. Murali, S. Rajesh, O. Prakash, A.R. Kulkarni, R. Ratheesh, Comparison of alumina and magnesia filled PTFE composites for microwave substrate applications, Materials Chemistry and Physics 113 (2009) 290–295.

[14] Y.M. Wang, D.C. Jia, Y. Zhou, Preparation and properties of PTFE/$Ba_2Ti_9O_{20}$ composites, Journal of Polymer Research 10 (2003) 247–258.

[15] P.S. Anjana, V. Deepu, S. Uma, P. Mohanan, J. Philip, M.T. Sebastian, Dielectric, thermal and mechanical properties of CeO_2—filled hdpe composites for microwave substrate applications, Journal of Polymer Science Part B: Polymer Physics 48 (2010) 998–1008.

[16] T. Joseph, S. Uma, J. Philip, M.T. Sebastian, Electrical and thermal propeties of PTFE—Sr $_2ZnSi_2O_7$ composites, Journal of Materials Science: Materials in Electronics 22 (2011) 1000–1009.

[17] G. Subodh, V. Deepu, P. Mohanan, M.T. Sebastian, Dielectric response of $Sr_2Ce_2Ti_5O_{15}$ ceramics reinforced high density polyethylene, Journal of Physics D 42 (2009) 225501–225506.

[18] J. Chameswary, M.T. Sebastian, Butyl rubber—Ba $_{0.7}Sr_{0.3}TiO_3$ composites for flexible microwave electronic applications, Ceramics International, http://dx.doi.org/10.1016/j.ceramint.2012.09.047, in press.

[19] G. Subodh, C. Pavithran, P. Mohanan, M.T. Sebastian, PTFE/Sr_2 $Ce_2Ti_5O_{16}$ polymer ceramic composites for electronic packaging applications, Journal of the European Ceramic Society 27 (2007) 3039–3044.

[20] P.S. Anjana, M.T. Sebastian, M.N. Suma, P. Mohanan, Low dielectric loss PTFE/CeO_2 ceramic composites for microwave substrate applications, International Journal of Applied Ceramic Technology 5 (2008) 325–333.

[21] S. George, P.S. Anjana, J. Krupka, S. Uma, J. Philip, M.T. Sebastian, Dielectric, mechanical and thermal properties of low-permittivity polymer–ceramic composites for microelectronic applications, International Journal of Applied Ceramic Technology 7 (2010) 461–474.

[22] S. George, V.N. Deepu, P. Mohanan, M.T. Sebastian, Influence of $Ca(Li_{1/3}Nb_{2/3})(0.8)Ti-0.2O_3-$delta filler on the microwave dielectric properties of polyethylene and polystyrene for microelectronic applications, Polymer Engineering and Science 50 (2010) 570–576.

[23] T. Joseph, S. Uma, J. Philip, M.T. Sebastian, Dielectric, thermal and mechanical properties of $Sr_2ZnSi_2O_7$ based polymer/ceramic composites, Journal of Materials Science: Materials in Electronics 23 (2012) 1243–1254.

[24] J. Krupka, R.G. Geyer, J. Baker-Jarvis, J. Ceremuga, Conference on Precise Measurement Method for Complex Permittivity of Microwave Substrate, CPEM, 1988.

[25] J. Krupka, Frequency domain complex permittivity measurements at microwave frequencies, Measurement Science and Technology 17 (2006) R55–R70.

[26] W.J. Parker, R.J. Jenkins, C.P. Butler, G.L. Abbot, Flash method of determining thermal diffusivity heat capacity and thermal conducutivity, Journal of Applied Physics 9 (1961) 1679–1684.

[27] P. Gonon, T.P. Hong, O. Lesaint, S. Bourdelais, H. Debruyne, Influence of high levels of water absorption on the resistivity and dielectric permittivity of epoxy composites, Polymer Testing 24 (2005) 799–804.

[28] T.S. Laverghetta, Microwave Materials and Fabrication Techniques, Artech House, USA, 1984.

[29] D.H. Kuo, C.C. Chang, T.Y. Su, W.K. Wang, B.Y. Lin, Dielectric properties of three ceramic/epoxy composites, Materials Chemistry and Physics 85 (2004) 201–206.

[30] V.L. Gurevich, A.K. Tagantsev, Intrinsic dielectric loss in crystals, Advances in Physics 40 (1991) 719–767.

[31] N. Jayasundere, B.V. Smith, Dielectric-constant for binary piezoelectric 0–3 composites, Journal of Applied Physics 73 (1993) 2462–2466.

[32] Y. Rao, J.M. Qu, T. Marinis, C.P. Wong, A precise numerical prediction of effective dielectric constant for polymer–ceramic based on the effective-medium theory, IEEE Transactions on Components and Packaging Technologies 23 (2000) 680–683.

[33] K. Wakino, T. Okada, N. Yoshida, K. Tomono, A new equation for predicting the dielectric constant of a mixture, Journal of the American Ceramic Society 76 (1993) 2588–2594.

[34] B. Riddle, J.B. Jarvis, J. Krupka, Complex permittivity measurements of common plastics over variable temperatures, IEEE Transactions on Microwave Theory and Techniques 51 (2003) 727–733.

[35] M.A. Berger, R.L. McCullough, Characterization and analysis of the electrical properties of a metal-filled polymer, Composites Science and Technology 22 (1985) 81–106.

[36] G.M. Tsangaris, G.C. Psarras, The dielectric response of a polymeric three-component composite, Journal of Materials Science 34 (1999) 2151–2157.

[37] W.D. Kingery, Introduction to Ceramics, John Wiley and Sons, New York, 1960.