Effect of silicon carbide nanoparticles on dielectric (2.45 GHz) and thermal properties of epoxy nanocomposites for microwave curing

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Abstract

Efforts to use microwaves in material processing are gradually increasing. However, the phenomenon associated with the processing is less understood. The conversion of electromagnetic energy into heat depends largely on the dielectric properties of the material being treated. Therefore, the fundamental knowledge of these properties is essential for processing of materials using microwaves. In this study, first the dielectric evolution of silicon carbide (SiC) infused epoxy nanocomposites prepared at room temperature with 0-0.3 wt% content of SiC was measured. Secondly, the dielectric properties of the prepared nanocomposites after heating for 10 min in microwaves at a power of 500 W were investigated in order to see the effect of microwave curing. The dielectric properties of all the samples were measured at the microwave frequency of 2.45 GHz using the advanced cavity perturbation method attached to a vector Network Analyzer (VNA). The results indicate that the dielectric properties of the resultant nanocomposites increase with the increase in SiC content as compared to the neat epoxy sample. However, the dielectric properties were found to be decrease after microwave curing signaling the maximum possible extent of curing. This indicates that reinforcement of SiC nanoparticles in epoxy makes them ideal candidates for efficient microwave curing of nanocomposites. Lastly, the determination of thermal properties also confirms the maximum possible extent of curing of epoxy using SiC as nanofillers.

Keywords: Dielectric properties, silicon carbide, microwave curing, glass transition temperature.

Introduction

In today's world of science and technology, the polymer nanocomposites are the centre of attraction due to its economically low cost and light weight concerned [**1**]. The extensive use of polymer nanocomposites in engineering structures, aerospace, automobile industries and nuclear reactors has initiated strong need of efficient, time saving and environment-friendly manufacturing processes which are capable of overcoming the inherent challenges of processing these materials. In practice, many major manufacturing processes intended for processing of composite materials involve interaction with heat; consequently the thermal energy based conventional material processing techniques become highly dependent on effective heat transfer between source and target material which quite often takes long time requiring a huge amount of energy to obtain good quality cured product. Thus, processing limitations of the conventional thermal techniques call for development of novel techniques to process composite materials effectively and efficiently, at the same time, the new process also needs to be fast. The microwave material processing technique is one such solution which offers significant time saving and hence can be cost-effective and more importantly clean and environment friendly [**2**]. Originally, microwaves are used for communication/ telecommunication purposes but recently they have gained very fast popularity for processing of polymers based materials and advanced materials such as polymer matrix composites (PMC) [**3**]. Many research groups have investigated the accelerated curing of nanocomposites in order to gain high strength [**4**-**7**]. However, the success has been very limited due to lack of understanding the behavior of dielectric permittivity of polymer nanocomposites because most of the polymers exhibit very low dielectric losses in the GHz region and it is difficult to heat them by microwaves [**8**]. Basically, microwaves interact with the materials through dielectric permittivity resulting in rapid heating. To address this issue, the use of fillers as microwave absorber in the polymer matrix has been proposed to enhance the response of microwaves to polymeric materials [**9**]. The ability of the materials to be heated in the presence of microwave field is defined by its dielectric permittivity $\varepsilon^*=\varepsilon'-j\varepsilon''$. Each term from the above equation represents specific feature of the dielectric material undergoing microwave radiation. ε' represents the ability of the material to become polarized under the electric field. ε" represents the ability of the material to convert

electromagnetic energy into heat [**10**]. At present, researchers are focusing more on the nanofillers to improve the dielectric properties of composites. Silicon carbide (SiC) is one of the ceramic fillers which are extensively used because some of its inherent characteristics such as high impact radiation and oxidation resistance, high chemical stability, catalyst support, strengthening and increasing super plasticity of materials. These intrinsic properties facilitate SiC nanoparticles applicable in the field of semiconductor materials, optic devices, hard disk support etc. [**11**]. Numerous researchers investigated the compatibility of SiC nanoparticles into composites and have shown remarkable improvement in response to thermal and mechanical properties. But, to the best of author's knowledge a very little work is focused on using SiC nanoparticles to improve dielectric properties in MW region. In order to comply with the ISM frequency band, 2.45 GHz is chosen as the operating frequency. Therefore, the aim of this article is "to study the effect of SiC nanoparticles on dielectric properties at 2.45 GHz and microwave curing of epoxy nanocomposites in terms of thermal properties".

Experimental

Materials / chemicals details

Spherical shape silicon carbide (β-SiC, 99.8% pure) nanoparticles (average particle size less than 60 nm) was purchased from M/S Kulkarni Brothers Ltd. The TEM micrograph of as received SiC nanoparticles is shown in **Fig. 1**. Epoxy resin was purchased from M/S Resinova Chemie, India. It is a two part system. Part A is PG100, (Diglycidyl Ether of Bisphenol-A) and Part-B is PHY161 (Aeromatic Amine based curing agent).The density of PG100 is 1.1g/cc. The mixing ratio was Part A (10): Part B (1) by weight.

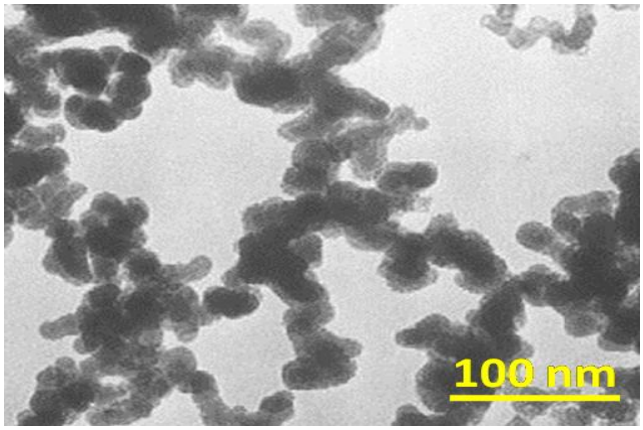

Fig.1. TEM micrograph of as received Silicon carbide (SiC) nanoparticles.

Material synthesis / reactions

SiC nanopowder was mixed with Part A of the epoxy resin system as per required weight ratio 0-0.3wt%.The mixture was then placed on a magnetic stirrer at 50ºC at a speed of 1500rpm for 2 hours. The mixture was then sonicated (Sonics Vibra Cell ultrasonic processor) for 20 min at 35% amplitude in order to obtain the homogeneous distribution of SiC nanoparticles into epoxy resin. After magnetic stirring and sonication Part B of the epoxy resin was added as per stoichiometric ratio (PartA: PartB=10:1) and then again sonicated for 5 min. To remove entrapped air and volatile materials from the mixture, a degasification process was introduced by placing the mixture in a vacuum oven for 35 min at room temperature [**12**]. The prepared mixture was then slowly poured in the Teflon mold and left for 8 hrs at room temperature to solidify. After initial 8 hrs room temperature curing, half of the test species were removed from the mould and dielectric properties was measured. The other half of the samples was post-cured in a microwave oven (Sharp Appliances Co. Ltd., Model R-212 operating at 2.45 GHz with a maximum power output of 1KW) for 10 minutes using a fixed power level of 500 W. The dielectric and thermal were then again measured in order to see the effect of microwave curing.

Characterizations / device fabrications / response measurements

The dielectric properties of SiC/epoxy nanocomposites were investigated using the microwave cavity perturbation method (CPM) attached to an Agilent vector network analyzer (VNA). The CPM is well known for the precise measurement of the complex permittivity of various types of materials. To observe the thermal stability of microwave cured nanocomposites, the Thermogravimetric analysis (TGA) was carried out under nitrogen gas atmosphere using a Perkin-Elmer TGA/DTA. The TGA measurements were carried out from 35 to 600ºC at a heating rate of 10º C/min. All the samples were cut into small pieces of 3 to 10 mg using a surgical blade. The glass transition temperature and cross linking density of microwave cured nanocomposites were obtained using Mettler Toledo Dynamical mechanical analysis (DMA) from 25 to 250ºC at a heating rate of 10ºC/min under nitrogen atmosphere. For DMA testing fresh samples of dimensions (15mm × 25mm × 1mm) were prepared.

Results and discussion

Effect of SiC content on dielectric properties at 2.45 GHz

The complex permittivity of the prepared nanocomposites at ambient temperature is evaluated at S band of microwave frequency which resonates at 2.45 GHz. The fabricated cavity is then connected to the vector network analyzer (VNA) with the two coaxial to waveguide adapters which generally provides electric coupling between the coaxial cable coming out from VNA to the waveguide joint together with the rectangular cavity. The CPM is well known for the precise measurement of the complex permittivity of various types of materials. In the CPM, the complex dielectric properties of the sample are calculated by measuring the resonating frequency and the quality factor of the rectangular cavity (RC) in the

unperturbed and perturbed condition. In this paper, the S-band rectangular cavity of volume V_c (90 mm × 45 mm × 400 mm) with the sample test arrangement at the center of the cavity is employed. The TE_{103} (f_0 = 2.4, Q_0 = 1193) mode is chosen for the measurement, and the frequency data are accurately measured using the vector analyzer (VNA). The VNA is calibrated with the help of CALIBRATION KIT being provided by KEYSIGHT. The loaded and the unloaded parameters of the fabricated cavity are measured using the VNA by means of recording the transmission coefficient. The frequency corresponding to the maximum value of the insertion loss (dB) is taken as the resonant frequency, whereas the quality factor is calculated with the 3dB method. The measured transmission coefficients of each successive step are given in **Fig. 2 (a).**

Now, the measured S_{21} data are used to determine the dielectric properties of pure epoxy and composite samples. The plots of real and imaginary parts of complex permittivity vs. SiC content (wt %) is given in **Fig. 2 (b).** which shows the variation of real and imaginary part of complex permittivity with different SiC nanoparticles concentrations at the measured frequency. It can be observed from **Fig. 2 (b)** that both the dielectric constant (ε') and the loss factor (ε") are increasing with higher values of SiC content. The enhancement in dielectric constant and loss factor are explained on the basis of interfacial polarization generated in between epoxy matrix and SiC nanofillers. With the increase of SiC content, the number of accumulated charges will be increased and hence an additional contribution to the dielectric properties occur [13].

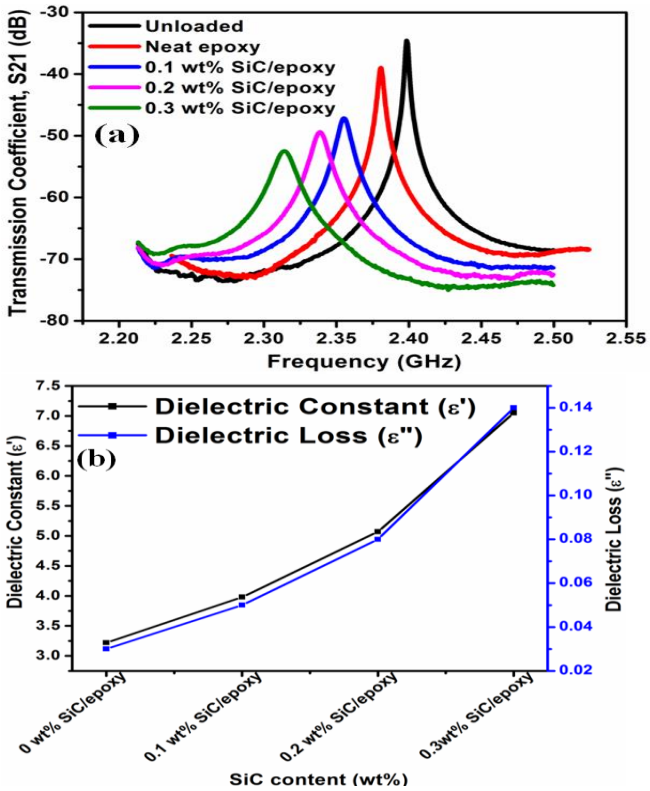

Fig. 2. Effect of SiC content on (a) measured transmission coefficient, S_{21}using VNA (b) dielectric constant (ε') and dielectric loss factor (ε").

Effect of microwave curing on dielectric properties of resultant composites

In order to see the effect of MW curing on the SiC reinforced epoxy composites the remaining half of the samples were placed for 10 minutes in a microwave oven for curing. For MW curing of composites laboratory oven (Sharp Appliances Co., Ltd, Model: R-212) operating at 2.45 GHz was employed with a maximum output power of 1KW.

Fig 3 (a) shows measured S_{21} transmission curve for MW cured composites of neat epoxy, 0.1 wt% SiC/epoxy, 0.2 wt% SiC/epoxy, and 0.3wt% SiC/epoxy. The extracted dielectric properties of these cured samples are shown in fig 3 (b). It was found that for pure epoxy sample, there was no significant change in ε', and ε" after curing. However, for SiC infused composites there is a decrease in dielectric properties of these nanocomposites as compared to their values measured before the microwave treatment. The decrease in dielectric properties of these samples after microwave curing may be attributed to the reduction in the overall polarization of cured samples [14].

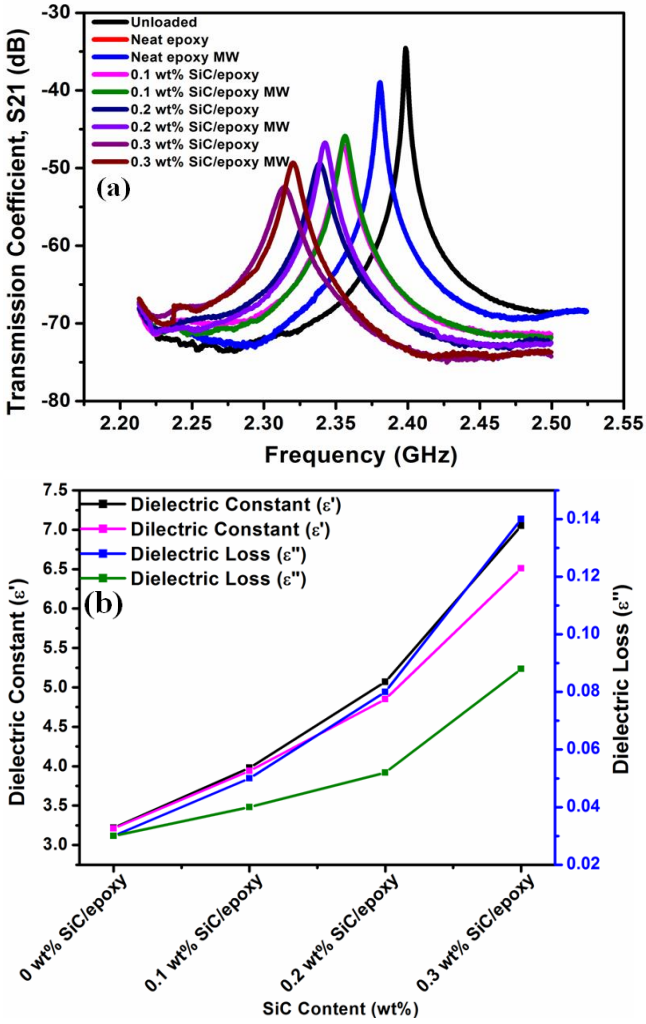

Fig. 3. Effect of microwave curing wrt various SiC content (a) measured transmission coefficient, S_{21} (dB) and (b) dielectric constant, ε' and loss factor, ε"

Thermal properties

Thermogravimetric analysis

In order to study the thermal stability of MW cured composites, its weight is monitored by raising the temperature at the rate of 10°C/min in the nitrogen atmosphere. The results are summarized in **Table 1**. These results show that for MW cured pure epoxy, the initial decomposition temperature (IDT) and maximum decomposition temperature of pure epoxy are ~ 310° C and 336°C. However, in case of SiC infused nanocomposites both onset and maximum decomposition temperature increase with the increase in content of SiC. This increase may be attributed to the good microwave absorption by the sample in the presence of SiC particles which improved the adhesion between SiC particles and epoxy resin.

Table1. Summarizes initial decomposition temperature (IDT), Maximum decomposition temperature (MDT) MW cured nanocomposites.

Sample	*Microwave cured*	
	IDT (°C)	*MDT (°C)*
Neat epoxy	310	336
0.1 wt% SiC/epoxy	328	350
0.2 wt% SiC/epoxy	348	367
0.3wt% SiC/Epoxy	351	372

Glass transition temperature (T_g)

The glass transition temperature (T_g) is a key property of epoxy systems and is dependent on the extent of curing. In this test, the sample is subjected to an oscillatory stress, and the material response is measured. The temperature ranges from 25 to 250°C and T_g is obtained by the peak value of the *tan*δ curve. The results are summarized in **Fig. 4.** It is observed that the lowest value of T_g ~74°C is observed for pure epoxy. However, with the addition of SiC nano filler content, T_g is increased to ~ 17°C for 0.1wt% SiC/epoxy, ~28°C for 0.2 wt% SiC/epoxy, and ~ 38°C for 0.3 wt% SiC/epoxy, respectively as compared to pure epoxy. The obtained results are in very good agreement with the decrease in dielectric properties measured after microwave curing and can be understood as follows. The presence of SiC in epoxy resin enhances the microwave absorbing ability of pure epoxy which further increases the cross link density of polymer chains and hence increase in T_g [15].

Conclusion

The SiC/epoxy nanocomposites have been successfully prepared in the laboratory. Several characterization techniques have been employed for the study of microwave dielectric and thermal properties of a number of nanocomposites. For investigating the overall effect of microwave curing, the dielectric properties of SiC/epoxy nanocomposites having various content of SiC

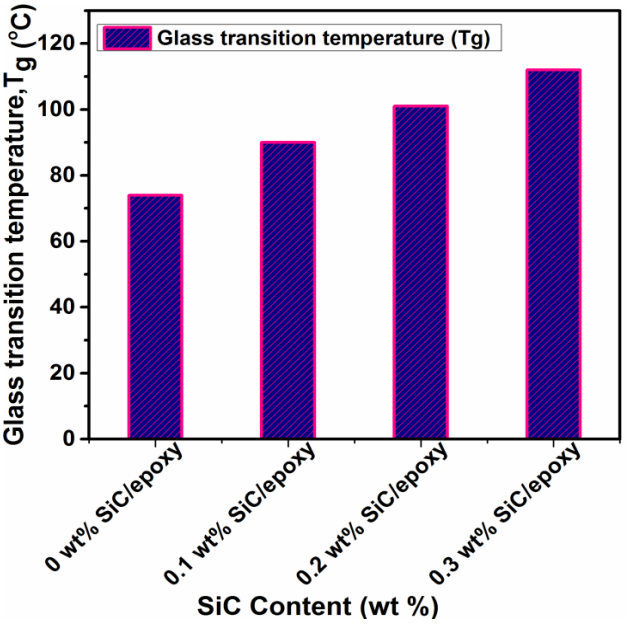

Fig. 4. Effect of SiC content on glass transition temperature (T_g) of microwave cured SiC/epoxy nano composites.

(0-0.3 wt %) have been measured before and after the microwave curing at 2.45 GHz. It has been found that the dielectric constant as well as loss factor of SiC/epoxy in the S band of microwave frequency is increasing with the increase of SiC nanofiller content. This rise in the complex permittivity of epoxy matrix nanocomposites with SiC nanofiller may be attributed to the interfacial polarization mechanism of the heterogeneous system. Hence, higher lossy materials can be obtained with SiC filler concentration. It has been found that the presence of SiC is beneficial for microwave curing and thus making the heating process more efficient. The dielectric constant and loss factor decreases after microwave curing which gives an idea about extent of curing. The thermal stability of the SiC/epoxy nanocomposites enhances with higher values of SiC content, as observed from thermogravimetric analysis. The glass transition temperature increases with increasing SiC content, which is in good agreement with the measured dielectric properties. The enhancement in glass transition temperature is attributed to increase cross linking density of polymer chains due to microwave curing.

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(a) Scientific article

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