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RESEARCH ARTICLE

DIELECTRIC MATERIALS INCORPORATED MICRO IRON ROD REINFORCED POLYURETHANE COMPOSITES FOR MICROWAVE ABSORBING APPLICATIONS

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ABSTRACT

Now a day, a vast variety of ceramic and polymeric materials is used for the fabrication and designing of the microwave absorbing materials and RADAR absorbing materials (RAMs). The microwave has got two different parts. One is electric part and the other one is the magnetic component. Both of these two components are perpendicular to each other. To absorb the microwave radiation, it is essential to cancel both of these components and the material which will be used as a genuine candidate to absorb the microwave should absorb or nullify the two components. Generally these types of materials can be prepared by incorporating magnetically conducting filler and electrically conducting filler or dielectric materials into a continuous polymer matrix. Here we have prepared the microwave absorbing material with 33.3% filler loading in TPU matrix. The sample thickness was maintained at 2 mm. X ray diffraction studies and Scanning electron microscopy was used to determine the morphology of the as prepared composites. Scattering parameters were measured in X-band region by using a Vector Network Analyzer.

INTRODUCTION

The traditions of using RADAR absorbing materials (RAMs) or Microwave absorbing materials has been started since 1930s (Das, *et al*., 2011). Now a day it is used mainly for commercial and military applications. The dielectric properties of various dielectric materials like Barium Titanate, Strontium Titanate, Titanium dioxide, and Zirconium dioxide has been extensively studied. The magnetic properties of micro dimensional iron rod have now become an interesting and emerging field of material research. It is a promising candidate for microwave absorbing application due to its properties like high conductivity, small diameter, high aspect ratio and super huge mechanical strength. That is why the investigations of electro-dynamic properties of composites based on different polymer matrices are growing at a very fast rate. Different polymer matrix can be used for this research like epoxy (Li, *et al*., 2006; Matzui, *et al*., 2007), polystyrene (PS), polyaniline (PANI) (Wang, *et al*., 2005; Watts, *et al*., 2003; Ma, *et al*., 2005), polymethylmethacrylate (PMMA) (Wang, *et al*., 2005; Fan, *et al*., 2006), polypropylene (Liu, *et al*., 2007; Al-Saleh, *et al*., 2009), etc. Here we have studied the microwave absorbing properties or RADAR absorbing properties of various dielectric materials ($BaTiO_3$, $SrTiO_3$, ZrO_2, and TiO_2) incorporated and micro dimensional Iron rod loaded polyurethane composites. We thought that micro iron Rod will absorb the magnetic part and the dielectric material will absorb the electric components of the microwave. There is a problem with the iron rod because iron rod is very difficult to disperse and homogenize with the polymer matrix. So technically we have used the polypropylene coated iron rod. In this case the iron rod is first coextruded with polypropylene. The extruded material was directly used as filler where polypropylene compatibilizes between the iron rod and the polyurethane matrix. According to the previous work rod like materials is a better candidate for microwave absorption. Hence, we studied here the microwave absorption ability of five composites with different nature of dielectric material loading. We prepared those composites and their morphological study was investigated by Scanning electron microscopy (SEM) and X-ray diffraction analysis and its microwave characteristics was studied by Agilent vector network analyzer (ENA E5071C). The matrix used for the preparation of different RAMs is thermoplastic polyurethane (TPU). Total filler percentage was maintained around 33.3% and the thicknesses of the RAMs were 2mm.

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MATERIALS AND METHODS

Materials: Iron rod of diameter 5-8 nm was purchased from Ticona (Germany). The TPU utilized for developing RAMs belong to commercial medical grade aliphatic. The dielectric materials purchased from Sigma Aldrich and were used without further purifications.

Preparation of the composites: At first we have taken the raw materials in required amount (Table 1). To disperse the iron rod we have used polypropylene coated iron rod. Then we first took the thermoplastic polyurethane along with polypropylene coated iron rod in the Brabender plasticorder. The whole material was mixed for 2-3 minutes at 220˚C. After the homogenization of the material, we introduced different dielectric materials in different cases and mixed again for 2 mins. Then the above mixture was removed from the Brabender and directly placed it in compression moulding machine. The pressure used was 5 MPa pressure and the temperature was 170°C for compression moulding process. Aluminum sheet was used as supporting material of the developed RAMs. Samples were prepared with 33.3% filler loading. Thickness of the RAM was maintained to 2mm. Both the samples were shaped into rectangular forms of size 0.4 inch x 0.9 inch to fit into X-band waveguide for microwave measurements.

Characterizations

All the micro structural details and the position of iron rods have been investigated by the Scanning electron microscopy (SEM) study carried out on Carl Zeiss-SUPRA 40. X-ray diffraction (XRD) analysis was carried out by Rigaku ULTIMA-III X-ray diffractometer, with Cu Kα radiation (λ =1.5418 A). The microwave absorption study was done by using Agilent vector network analyzer (ENA E5071C).

RESULTS AND DISCUSSION

Morphological Study

Figure 1 depicts the SEM images for the fractured cross-sections of all the samples. The first figure is for $BaTiO_3$ incorporated Fe-rod reinforced sample. The second one is for $SrTiO_3$ incorporated Fe-rod reinforced sample. The third one is for TiO_2 incorporated Fe-rod reinforced sample and the fourth one is for ZrO_2 incorporated Fe-rod reinforced sample respectively.

Table 1. Compositions of the asprepared microwave absorbing composites

Name	Sample-1 (S1)	Sample-2 (S2)	Sample-3 (S3)	Sample-4 (S4)	Sample-5 (S5)
TPU	20	20	20	20	20
FE-ROD	10	5	5	5	5
BARIUM TITANATE	0	5	0	0	0
Sr-TITANATE	0	0	5	0	0
ZrO_2	0	0	0	5	0
TiO_2	0	0	0	0	5

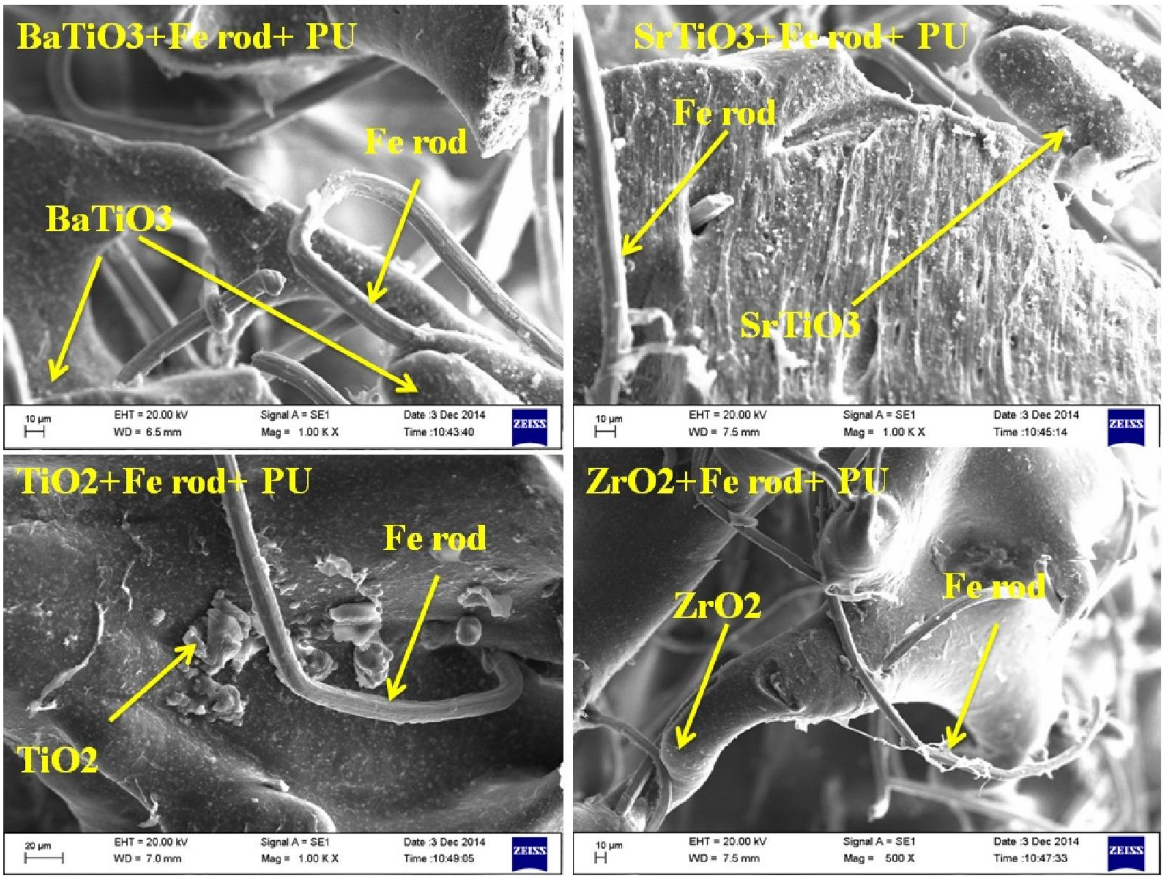

Fig. 1. SEM images of all the as prepared composites

The morphology shows a very clear distinguishable phase of Iron rod, which is situated in continuous PU matrix. Small fibre like polypropylene is also found surrounds the iron micro rods. The diameter of the iron rod is near 6-8 micrometer range in all the samples. The dielectric materials are properly dispersed in the continuous PU matrix. The dielectric materials in PU matrix form a type of matrix droplet morphology.

X-ray diffraction analysis

Figure 2 depicts the X-ray diffraction pattern of all the as prepared samples. All the samples show characteristic peaks of dielectric materials along with the presence of Iron rod. The peak of iron rod appears near 2Ɵ= 45-55˚. The peak should be much sharper but due to the presence of amorphous polymer matrix, the intensity of the peak is little less. The peak appears at 2Ɵ=21˚, clearly resembles the presence of $BaTiO_3$. This is the characteristic peak of $BaTiO_3$. The peak appears at 2Ɵ= 24˚, clearly resembles the presence of $SrTiO_3$. This is also the characteristic peak of $SrTiO_3$. The peak appears at 2Ɵ=28.7˚, clearly resembles the presence of TiO_2 and the peak appears at 2Ɵ=27˚, clearly resembles the presence of ZrO_2.

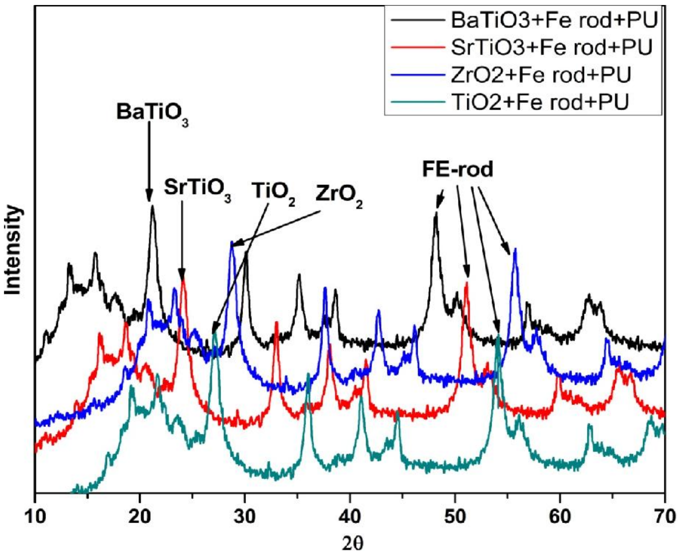

Fig. 2. X ray diffraction pattern of all the as prepared composites

Microwave Absorptivity Spectra

To evaluate the microwave absorbing efficiency of the as-prepared samples, the return loss (RL) value of the nanocomposites was calculated using the relative complex permittivity and permeability values at given frequency according to the transmission line theory (Michielssen *et al*., 1993 and Min Zhou *et al*., 2011). Complex relative permittivity $(\varepsilon_r = \varepsilon' - j\varepsilon'')$ and permeability $(\mu_r = \mu' - j\mu'')$ of the developed composites were measured by employing vector network analyzer (model PNA E8364B). The characteristic input impedance of the absorbers was calculated with the help of complex permittivity and complex permeability values by using the following equation (Tyagi *et al*., 2004, 2011)-

$$z_{in} = z_0\sqrt{(\mu_r/\varepsilon_r)}\tanh[(-j2\pi/c)(\sqrt{\varepsilon_r\mu_r})fd] \quad \text{...... (i)}$$

Where z_{in} is the characteristic input impedance, c is the velocity of light, f is the frequency of microwave and d is the thickness of absorber. The surface reflectance, i.e. the Return loss was calculated by the following equation (Tyagi *et al*., 2004, 2011)-

$$R_L = -20\log_{10}[|\frac{(z_{in}-z_0)}{(z_{in}+z_0)}|] \quad \text{...... (ii)}$$

So, the surface reflectance of an absorber depends on six characteristic parameters, namely ε', ε'', μ', μ'', f and d. The absorbing properties of the absorbers with different thickness in the X-band region are shown in Figure 3.

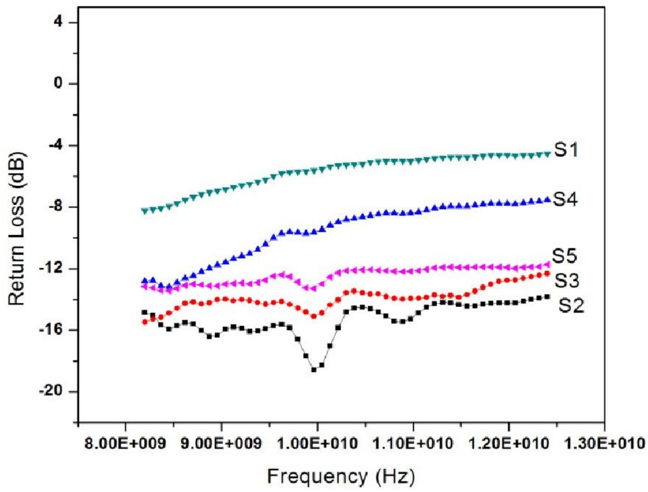

Figure 3. Microwave absorbing properties of S1, S2, S3, S4 and S5

Sample-1 showed a minimum return loss (MRL) of 8.24 dB at 8.2 GHz. Sample-2 achieved the MRL of 18.60 dB at 9.95 GHz. Minimum return loss showed by the Sample-3 was 15.47 dB at 8.2 GHz. Sample-4 and 5 showed the minimum return loss of 13.20 dB at 8.42 GHz and 13.30 dB at 9.96 GHz respectively. From the above results, it can be seen that the composites based on both magnetic iron rod and dielectric material achieved superior microwave absorbing performance than the composite based on only iron rod. The return loss values of all the Samples except Sample-1 were less than 10 dB i.e. the composite materials based on both magnetic and dielectric materials absorbed more than 99% microwave energy. Sample-2 (iron rod and BaTiO3 based composite) showed the maximum absorbing capability compare to other magnetic-dielectric based composites because in this case the dielectric property of BaTiO3 is better than other dielectric materials (Abbas *et al*., 2005). The mechanism of absorption was clearly represented with the help of real and imaginary parts of permittivity and permeability values.

Permittivity and Permeability Spectra

To investigate the possible mechanisms of the microwave absorption of the present composites, the real and imaginary parts relative complex permittivity and permeability values were studied as shown in Figure 4. The real parts of complex permittivity (ε′), permeability (μ′) stand for energy storage part and the imaginary parts of permittivity (ε″) and permeability (μ″) stand for energy loss part. The real parts of permittivity (ε′) value i.e. electric energy storage parts for Sample-1, 2, 3, 4 and 5 were approximately 13.63, 22.78, 17.67, 16.35 and 16.63 respectively with some resonance peak points (Figure 4a).

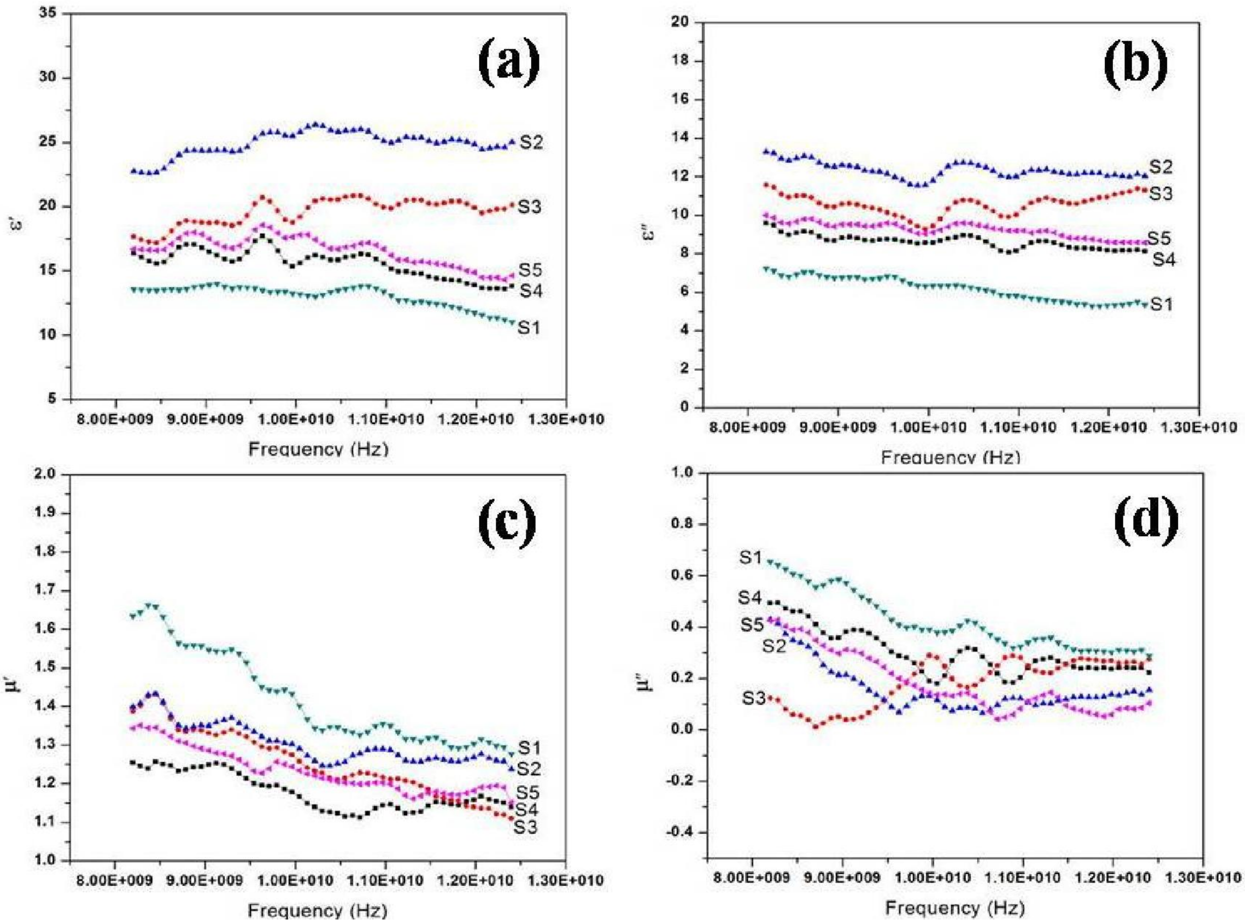

Figure 4. (a) Real (ε′) and (b) imaginary (ε″) parts of relative complex permittivity, (c) real (μ′) and (d) imaginary (μ″) parts of relative complex permeability of prepared samples

Dielectric constant depends on two factors, orientation and interfacial polarizations. In case of Sample-2, the orientation and interfacial polarization should be high. The imaginary parts of permittivity (ε″) i.e. electric energy loss parts for Sample-1, 2 were ~7.26, 13.28 and for Sample-3, it was ~11.54. The ε″ values for Sample-4 and 5 were approximately 9.56 and 10 respectively (Figure 4b). Similar to real part of permittivity, the imaginary parts of permittivity also have some resonance peaks. The multi-nonlinear resonance may cause the absorbing proficiency in a wider frequency region. The variation of both real and imaginary parts of permittivity with frequency may be happen due to space charge polarization (Wagner, 1913). The magnetic and dielectric particles embedded into the thermoplastic polyurethane polymeric matrix can act as a large number of charge domains and can contribute to increase the dielectric constant values due to interfacial polarization (Muhammad Abdul Jamal *et al*., 2009). The dielectric loss mechanisms occur due to the permanent and dominant polarization and their associated relaxation phenomena. The composite materials showed the microwave absorbing properties according to their dielectric loss values. $BaTiO_3$/Fe rod based composite (S2) have better dielectric loss value than other composites and it achieved better absorbing properties. The real parts of permeability (μ′) i.e. magnetic energy storage parts for S1 lie from 1.66 to 1.27 and the value was highest compare to other composite materials.

The μ′ values of S2, S3, S4, and S5 were reduced because of the addition of non-magnetic materials into the polymeric matrix and the values were decreased with increasing frequency which presents excellent frequency dispersion (Figure 4c). The value of imaginary parts of permeability (μ″) i.e. magnetic energy loss parts for S1 lie from 0.69 to 0.29 and the μ″ values were reduced for other composite materials due to decrease of bulk concentration of magnetic iron rods (Figure 4d)and magnetic energy loss occurred due to eddy-current loss and residual loss (Vander Zaag, 1999). In case of present absorbing materials, dielectric loss was mainly responsible for the microwave power absorption but the real and imaginary parts of permeability were also responsible for absorption. Return loss values are calculated from the expression:

$$R_L = -20\log_{10}[|\frac{(z_{in}-z_0)}{(z_{in}+z_0)}|]$$

When the characteristic input impedance (Z_{in}) is close to the free space impedance (Z_0 = 377 Ohms), a material exhibit better microwave absorbing performance. Characteristic input impedance depends on both the dielectric and magnetic contribution of an absorber. So an absorber achieved better absorbing properties when the dielectric contribution matches the magnetic contribution. For this reason, S1 showed very poor absorbing properties and its absorbing efficiency was tremendously improved after the addition of dielectric materials.

Conclusions

$BaTiO_3$, $SrTiO_3$, TiO_2 and ZrO_2 nanoparticles were synthesized successfully by co-precipitation and hydrothermal method. The composite materials were prepared by mixing the particles into the thermoplastic polyurethane polymeric matrix. The structure, morphology, and electromagnetic properties of the obtained products were characterized by XRD, SEM, TEM, and vector network analyzer. The results show that there have been significant changes in the microwave absorbing properties of the magnetic-dielectric based composites when compared with magnetic composite. Impedance matching effect is responsible for achieving the high performance microwave absorbing properties of the composite materials. Hence the present composite materials have great potential in the field of microwave absorption.

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