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Dielectric and magnetic properties of YIG/PMMA nanocomposites

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Abstract.

Yittrium iron garnet, $Y_3Fe_5O_{12}$ (YIG), is a material used widely in electronic devices for the microwave region as well as the magnetic bubble domain-type memories. Yittrium iron garnet ($Y_3Fe_5O_{12}$) was produced by mechanochemical synthesis from Y_2O_3 and Fe_2O_3 with particle size of around 150 nm. PMMA/YIG composite films were prepared by solution casting method at diffferent concentration (i.e. 10%, 20% and 40%) of YIG filler. Dielectric permitivity of composite materials were studied over a wide a range of frequency and temperature as a function of filler concentration. The electrical properties of composites were explained by in terms of molecular mobility and interfacial polarization.

1. Intoduction

Composite systems consisting of an insulating matrix and randomly dispersed fine inorganic particles have generated significant research interest, mostly, due to their electrical and electromagnetic performance [1-3]. The primary applications of these polymer composites refer to electromagnetic interference (EMI) shielding, radio frequency interference (RFI) shielding and electrostatic dissipation of charges (ESD). Composites consisting of inorganic particles embedded in a polymer matrix have attracted more and more attentions due to integrative advantages of the two constituents. Introducing high dielectric constant (ε) ceramic particles into polymer matrix will result in high ε composite with the processibility and flexibility similar to polymers [4–8]. Such kind of composites could contribute to the promising candidates for dielectric materials in flexible capacitors with high performance and as a microwave absorber.

The dielectric properties of polymer/ceramic composites are influenced not only by the dielectric constant of the polymer and the ceramic, but also by the dispersion and loading of the ceramic in the polymer matrix. Using higher dielectric constant particles and polymer matrix, and increasing the loading of particle fillers can increase the effective dielectric constant of a composite. As well, the morphological and structural optimization of particles will enhance their contribution to dielectric constant improvement [9]. Though lifting the filler content is a choice, however, too high loading of fillers would lead to poor quality of the composite. Polymers and polymer matrix composites are basically electrical insulators, due to their low concentration of free charge carriers. Thus their electrical response is, mainly, associated with relaxation phenomena occurring under the influence of ac field. The observed relaxation processes are related to dipolar orientation effects or space charge migration [10, 11]. Molecular mobility and interfacial polarization are regarded as the origin of

dielectric effects. At sufficient high temperatures, in the vicinity of the glass transition temperature, large segments of the polymer chains are able to move trying to follow the alternation of the field, while at lower temperatures polar side groups are contributing to the electrical performance of the system. Interfacial polarization is the result of the heterogeneity of the system, such as mobile charges accumulated at the fillers–polymer matrix interface, form large dipoles. Recently, adding conductive fillers (with infinitively large ε) into polymer was reported on preparation of high ε composites [12–17], but the insulating properties of the composites will be reduced in this case. So, the inorganic fillers with super high ε should be the key consideration in obtaining higher ε composites with integrative properties needed in practical applications.

The ferromagnetic garnet, yttrium iron garnet (YIG), is a well-known material, and it has been widely used in electronic devices, such as circulators and phase shifters for microwave and magneto-optical devices [18, 19]. Additionally, YIG has controllable saturation magnetization, low dielectric loss tangent (tan δ) in microwave regions, and small line-width (ΔH) in ferromagnetic resonance [20]. Oscillations and waves of magnetization or spin waves in yttrium–iron garnet (YIG) ferrite films and mono-crystals have very interesting and useful properties such as a high Q-factor, a wide variety of dispersion laws dependent on the orientation of bias magnetic fields, and a possibility to tune their frequencies in the range of several GHz by changing the magnitude of the bias magnetic field. Another important property of ferrite films is low (of the order of several microwatts) power threshold of nonlinear spin wave processes. The nonlinear properties of ferrite films can be used for the development of novel microwave signal processing devices based on the parametric interaction of spin wave packets propagating in the films with localized electromagnetic fields of the microwave pumping. Excellent features of the ferrite/polymer composites, such as sharply reduced dielectric loss compared to in the bulk ferrites, while uninfluenced microwave absorption properties owing to the domination of natural ferromagnetic resonance absorption in the loss mechanism of the ferrite absorber materials [21, 22], make them quite attractive for applications not only as inductive and capacitive materials but also as microwave absorber materials.

In this work, Yittrium iron garnet, $Y_3Fe_5O_{12}$ (YIG) is synthesized using mechanochemical synthesis method and the polymethyl methacryalate polymer-based composites consisting of YIG nanoparticle fillers with varying concentrations were reported, aiming to investigate how nano fillers affects the electrical properties of such composites which could be explained by in terms of molecular mobility and interfacial polarization.

2. Experimental procedure

YIG ceramic was produced from Y_2O_3 and Fe_2O_3 by mechanochemical method. The synthesis was carried out in a Fritsch Pulverisette 5 Planetary high energy ball milling system. The following milling conditions were used: stainless steel vessel with a volume of 500 cm^3, stainless steel balls with diameter of 10 mm, ball-to-powder weight ratio was 40:1, air atmosphere, rotation speed of discs with vials was 320 min^{-1} and milling time was 8 h. Polymethyl methacrylate (PMMA) was used as the polymer matrix**.** Polymer-based nanocomposites were prepared by directly mixing YIG nanoparticles and PMMA together, by solution casting method. Tetrahydrofuran (THF) was used as a solvent to dissolve the polymer. The mixture of polymers and nanofillers were stirred with different volume fraction of YIG filler (i.e. 10%, 20% and 40%) at room temperature using magnetic stirrer at high speed of around 700 rpm until all polymer dissolved. After obtaining a homogeneous mixture, the mixture was poured into a clean glass trough. The solvent was evaporated at room temperature ($25^0C \pm 1^0C$) to get thin films of polymer nanocomposites which were then dried in vacuum oven at 30^0C. Powder X-ray diffraction analysis (Rigaku, Cu Ka, 2°/min) was used to determine the crystalline phases of milled powder. The particleshape and morphologies of YIG were investigated by SEM (JEOL-5910LV) after coating with gold. The particle sizes of milled powder were measured using Malvern particle size analyzer (Nano ZS Zeta Sizer). High frequency magnetic properties of the YIG particles were studied using Bruker MMX X-band ESR spectrometer. A small sample was located at the center of x-band cavity and field derivative FMR spectrum was recorded as a function of external

field. The direct microwave absorption spectrum of the YIG particles was investigated using a Network analyzer operating as a function of microwave frequency between 1-50 GHz. A small amount of sample was placed in a cavity in the microwave transmission line. The cavity was located in between and producing an electromagnetic field in the range of 0-2.2 Tesla. Microwave signal with different frequencies was applied and the external field was scanned from 0 up to 2.2 Tesla. After coating silver layers on the two surfaces of the composite films as electrode, the dielectric and electrical properties were measured using HP4980A LCR meter.

3. Results and discussion

3.1 The structure and morphology of YIG nanoparticles

Fig.1 shows the particle size distribution of powders after 8 hrs of high energy milling indicating a nearly homogeneous distribution. The mean particle size of the powders was around 150 nm. Fig. 2 shows XRD patterns of the YIG powders milled for different times indicated that after 8h of high energy milling nearly all of the phases converted to $Y_3Fe_5O_{12}$ (JCPDS: 43-0507) through formation of $FeYO_3$ (JCPDS: 39-1489). After 2 h of high energy milling, iron and yittrium oxides started to react to form $FeYO_3$ and the first peaks of $Y_3Fe_5O_{12}$ were observed in samples milled for 4 hrs but there were still unreacted iron and yittrium oxides in addition to $FeYO_3$.

Fig. 3 indicates the SEM micrograph of mechanochemically activated powders after 8 hrs of milling which shows nanoscale particles. The particles had nearly uniform size distribution but contained also some agglomeration. After producing a pellet from mechanochemically treated powder, it was then sintered at 1425°C for 10 hrs which gave a YIG ceramic with a theoretical density of around 95%. This indicated that mechanochemical method gave a high density YIG ceramic after sintering without any calcination step.

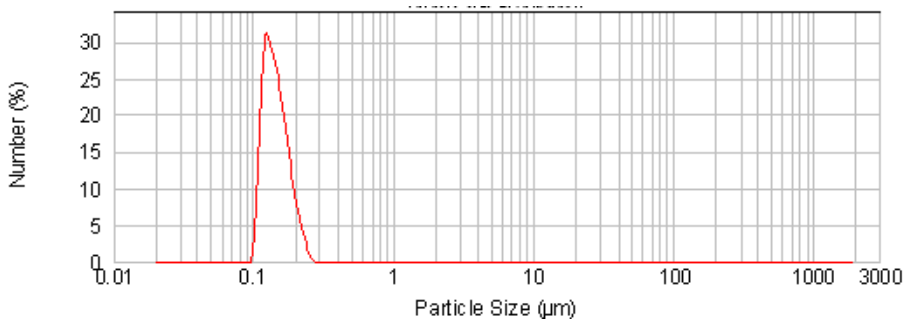

Figure 1. Particle size distribution of powders produced by mechanochemical synthesis after 8 hrs of high energy milling.

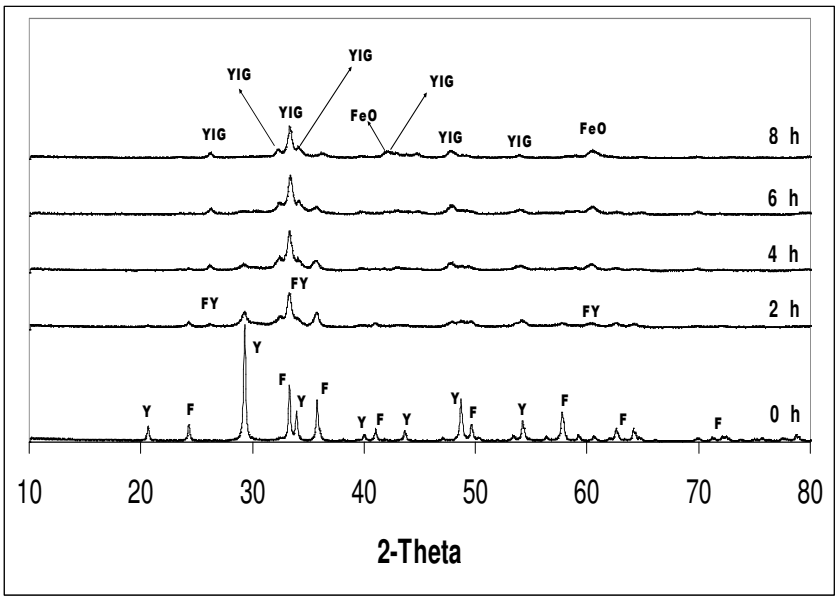

Figure 2. XRD patterns of the YIG powders milled for different times (Y: Y_2O_3, F: Fe_2O_3, FY: $FeYO_3$ (JCPDS Card No:39-1489), YIG: $Y_3Fe_5O_{12}$ (JCPDS Card No:21-1450).

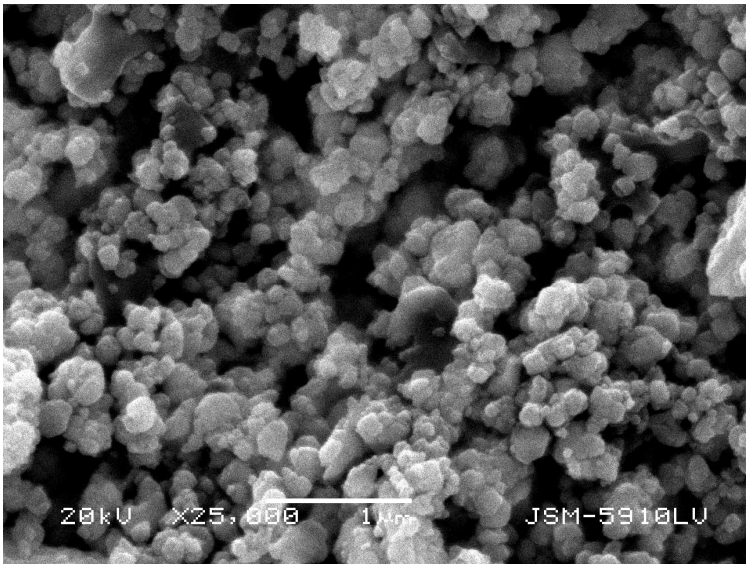

Figure 3. SEM of mechanochemically activated powder with 8 hrs of milling.

3.2 Magnetic properties of YIG particles

Ferro-magnetic resonance (FMR) spectrum of YIG particles is given in Fig. 4. As seen from the spectrum, FMR signal is very strong even at possible minimum microwave energy. This means that the individual particles are strongly magnetic as expected from YIG particles. The FMR absorption curve is considerably wide as well. This behavior might be attributed partially to crystalline anisotropy of the individual particles and partly to the magnetostatic long range interactions among the magnetic particles. Since magnetic axes are randomly oriented, resonance field of individual particles might be distributed in the field up to some extent determined by the magnetic anisotropy field. The asymmetric character of the resonance peaks could be originated from the conducting properties of the sample at higher frequencies.

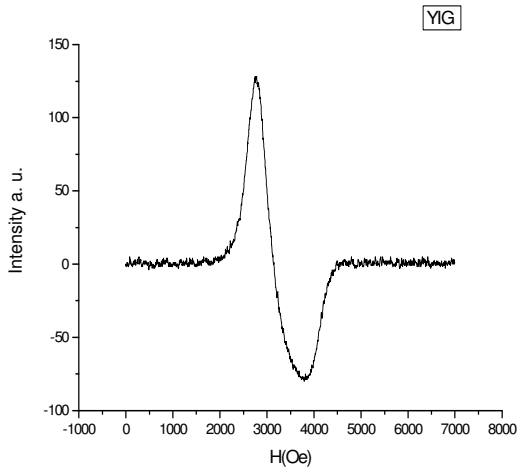

Figure 4. FMR spectrum of YIG nanoparticles

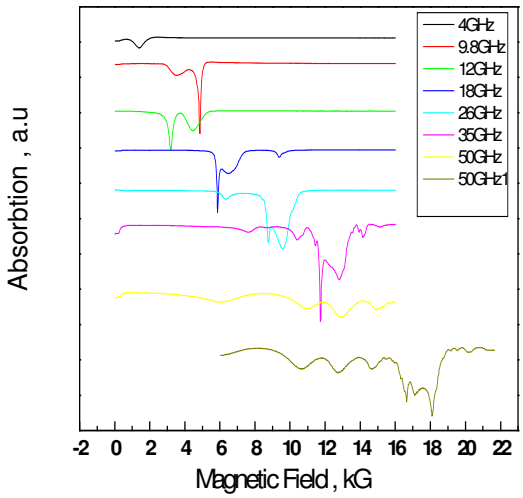

Figure 5. Microwave absorption spectrum of the sample at different frequencies and magnetic fields.

The FMR absorption spectrum with different applied magnetic fields is shown in Fig. 5. It was observed that the signal exhibits a resonance character at each frequency; however the location of the peak at the field axes monotonically shifts to higher field with increasing frequency. This behavior is consistent with FMR theory that the sample is ferromagnetic in nature even at very low amount of sample. Further additional peaks beside main FMR mode were observed and the number of these peaks increases with frequency. The shape and the number of these peaks depend on the external geometry of the sample in the cavity which is a basic property for spin wave resonance. It indicates that there is an existence of a long wave length spin wave i.e. magnetostatic wave modes throughout the sample.

3.2 The dielectric and conductivity properties of the PMMA/YIG nanocomposites

Fig.6 shows the dielectric constants and the electrical conductivities of the PMMA/YIG composites while varying the concentration of the YIG, frequency of applied fields and temperature. Both the dielectric constant and the electrical conductivity of the composites increased with the increase of YIG content. These phenomena could be interpreted from interfacial polarization inside the composites in applied alternating field [23]. The dielectric constant of the composites decreases with increasing the frequency of applied alternating filed. The reason is higher the frequency field, the shorter time the YIG particles have to polarize, and then the smaller the dielectric constant of the composite performs. Meanwhile, the conductivity of the composites increases with lifting the frequency. According to the

percolation theory [24], this property is the result of the direct proportion of YIG particles with each other. The variations in the dielectric constant, dielectric loss and the electrical conductivity of the PMMA/YIG composites with temperature are shown in Fig. 7. Similar to the most case for other polymer/ceramic composites, which indicated a behavior that dielectric constant increased with temperature [25-27], the PMMA/YIG composites display an increasing tendency in the dielectric constant with increasing temperature. This phenomenon is explained from the occurrence of two kinds of behaviors, which would yield converse effects on dielectric constant of the composite by changing the temperature [28]. First, increasing the temperature would improve the segmental mobility of the polymer, facilitate the polarization of polar fillers and consequently, increase the dielectric constant. Secondly, the obvious differential in thermal expansion coefficient between polymer and filler would change the aggregations of polar components, and thus increase the dielectric constant. PMMA is a kind of polymer with a low glass transition temperature, and both behaviors are the main effects in the present case. Fig. 7c shows the plot of dielectric loss versus temperature at a frequency of 10 kHz, 1 MHz and 2 MHz. It is observed that dielectric loss increases with the increase in temperature. The dielectric loss is due to the perturbation of phonon system by an electric field, the energy transferred to the phonon is dissipated in the form of heat. The growth in tan δ and thus increase in conductivity is brought about by an increase in conduction of residual current and the conduction of absorption current.

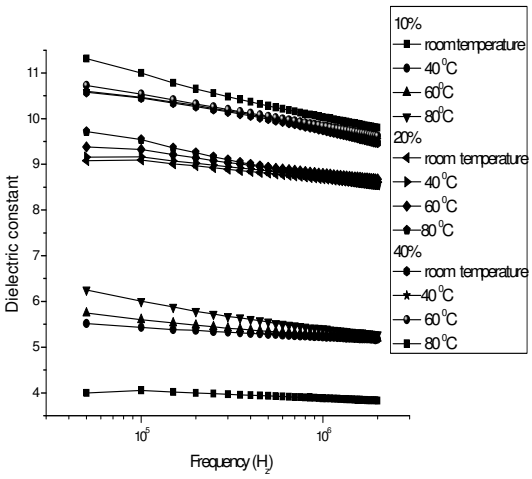

Figure 6(a). Frequency dependence of dielectric constant of the PMMA/YIG composite films on the different concentration of YIG at different temperature.

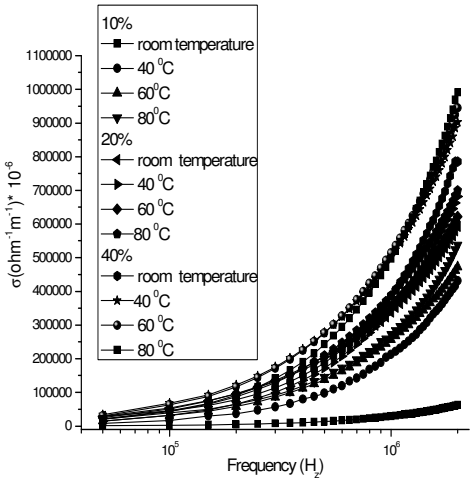

Figure 6(b). Frequency dependence of electrical conductivity of the PMMA/YIG composite films on the different concentration of YIG at different temperature.

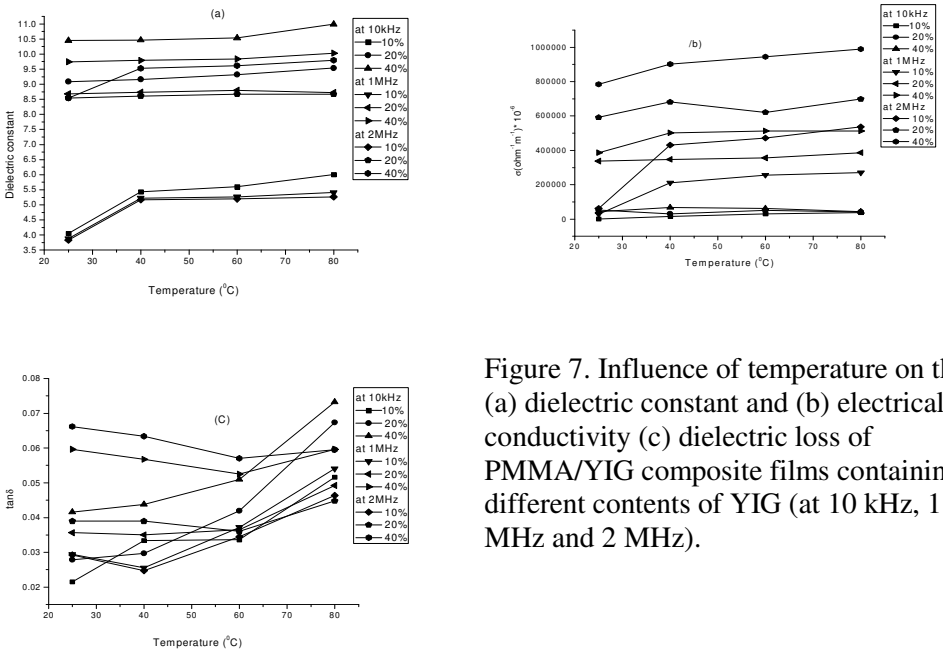

Figure 7. Influence of temperature on the (a) dielectric constant and (b) electrical conductivity (c) dielectric loss of PMMA/YIG composite films containing different contents of YIG (at 10 kHz, 1 MHz and 2 MHz).

4. Conclusion

In this paper, we prepared YIG material by mechanochemical synthesis from oxides of iron and yittrium. Electrical and dielectric properties of PMMA/YIG composites were studied over a wide range of frequencies and temperatures as a function of filler concentration. Both the dielectric constant and the electrical conductivity of the composites increased with the increase of YIG content. These phenomena could be interpreted from interfacial polarization inside the composites in applied alternating field. It was also observed that PMMA/YIG composites display an increasing tendency in dielectric constant and dielectric loss while increasing temperature. It is attributed to improving the segmental mobility of the polymer, facilitating the polarization of the polar fillers and increasing the dielectric constant.

5. Acknowledgements

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