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Engineering of dielectric composites on electromagnetic and microwave absorbing properties for operation in the *X*-band

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In this study, carbon black (CB) powder-loaded polyurethane (PU) composites (CB–PU composites) were prepared by melt mixing method with different volume percentages (45, 50, 55, 58 and 61 vol.%) of CB in the PU matrix. The prepared CB–PU composites had been further studied for surface morphology using the field-emission scanning electron microscopy (FESEM) technique. Dielectric properties in terms of real permittivity (ε') and imaginary permittivity (ε'') of the fabricated composites were computed using an Agilent E8364B vector network analyzer in the frequency range of 8–12 GHz (*X*-band). Dielectric loss factor of the prepared CB–PU composites was computed in terms of the dielectric loss tangent ($\tan \delta_e = \varepsilon''/\varepsilon'$). Microwave absorbing properties were appraised in terms of the reflection loss (RL) which in turn was calculated for varying thicknesses of the prepared composites from the measured real and imaginary permittivity data. The minimum RL was observed as −20.10 dB for the absorber with a thickness of 2.2 mm and the bandwidth achieved was 1.92 GHz for RL ≤ −10 dB. Based on the above results these CB–PU composites have potential use as effective microwave absorbers in 8–12-GHz (*X*-band) frequency range.

Keywords: Carbon black; dielectric materials; polymeric composites; complex permittivity; absorbing properties.

1. Introduction

Nowadays, various research works have developed microwave absorbers and investigated their electromagnetic (EM) and radar-absorbing properties to protect the sensitive avionics and electrical equipment which are operating in the gigahertz frequency ranges.[1] Development of high-performance microwave absorbing materials (MAMs) is a great challenge for researchers across the globe so far. Electromagnetic wave transmitted from the EM sources with certain GHz frequency is utilized for different purposes like in medical applications, wireless information transfer, imaging, broadcasting, etc. Currently, various types of filler materials, dielectric, magnetic and conducting materials, have been applied in the design and development of microwave absorbing composites (MACs).[2,3] The outcomes of conducting absorbing materials have motivated researchers to develop MACs that are thin, light in weight, flexible and have moderate functional properties. Many parameters are related to the performance of filler materials and their composites, such as the ionic radius and structure of fillers, conductivity of fillers, state of dispersion, methods and most importantly, the dispersion ratio of materials. A variety of frequency-response dielectric and other relaxation parameters have been studied.[4,5] Dielectric composites offer extra reliable electrical polarization and better functional properties than the macroscopic polymeric composites.[6,7]

In this research work, carbon black (CB) powder is used as a conductive filler material to fabricate the polymeric composites. Polymeric composites are fabricated by dispersion of conductive filler materials into the polymeric matrix. These polymeric composites were prepared by incorporating the conductive filler (CB) into the polyurethane (PU) matrix applied using wet mixing method. The starting filler materials are commonly in the form of fibers, powder, flakes or layered shape. The aim of this study is to fabricate a series of CB–PU polymeric composites that can be utilized as dielectric microwave absorbers, applicable in high-GHz-frequency region.

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2. Experimental Process

2.1. *Fabrication of composites*

Carbon black (99%; Sigma-Aldrich, USA) powder was thoroughly mixed with two components of the polyurethane matrix, which is a two-component system that consists of polyol-8 (Ciba-Geigy, Switzerland) and hexamethylene diisocynate (E-Merk, Germany) in the ratio of 50:50. For the measurement of complex permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) of the desired rectangular-shaped composites, a series of CB–PU composites were prepared with the carbon black powder being loaded with different volume percentages (45, 50, 55, 58 and 61 vol.%) into the polyurethane matrix and thoroughly mixed in the mortar by a pestle. Wet mixing method has been applied for sample preparation subsequently; a semi-cured homogenized mixture of CB–PU was further poured into the desired rectangular shape of stainless steel mold. Poured steel mold was then hot-pressed using compression molding technique at the temperature of 40 °C and pressure of 10 MPa for a curing time of 8 h. Further, rectangular-shaped samples were finished into dimensions of 0.4 inch×0.9 inch to fit exactly into the *X*-band waveguide (WR90) for microwave measurements.[8,9] The schematic diagram is shown in Fig. 1.

2.2. *Characterization*

Carbon black powder and prepared CB–PU composites were characterized and measured for their structural and EM properties. X-ray diffraction (XRD) patterns for CB powder and the CB–PU composite were recorded on an X'Pert Pro diffractometer ranging from 10° to 55°. Field-emission scanning electron microscopy (FESEM) was employed to examine the surface morphology of CB–PU composites. Dielectric and microwave absorption properties were evaluated using standard rectangular waveguide. The measurement setup consists of a rectangular *X*-band waveguide (10.15×22.85 mm^2) attached to an Agilent E8364B vector network analyzer having the software module 85071E. It is observed that the results are expressed in terms of the complex scattering parameters (S_{11}, S_{22}, S_{21} and S_{12}). Subsequently, complex permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) of the prepared composites was evaluated from the measured scattering parameters, resulting from the transmission/reflection line technique in the (*X*-band) frequency range of 8–12 GHz.

3. Results and Discussion

3.1. *Structural properties*

Figure 2 shows the XRD patterns of CB powder and fabricated CB–PU composite with maximum vol.% loading of CB powder recorded by the X-ray diffractometer (PANalytical, X'Pert Pro) with a Cu-Ka radiation source (wavelength k = 1.540598 Å) to identify the most intensive peaks at standard circumstance. The room-temperature peaks obtained for each interval of 0.02° in the scanning range from 10° to 50° for CB powder which were confirmed by the Miller indices (*hkl*), are in fair agreement with the previously reported value for carbon black and its composite. From the data of CB depicted in the corresponding curve, the most intensive peaks at the 2θ values of 25.4° and 43° can be indexed as the (002) and (100) crystal planes of CB powder, respectively.[10,11] The Debye–Scherrer equation [$D = 0.94\lambda/\beta\cos\theta$] has been used to evaluate the crystallite size of the high intense peak (002) and it is found to be 18 nm for carbon black powder.

Figure 3 reveals the FESEM micrographs for the lowest (45 vol.%) and highest (61 vol.%) contents of CB powder in polymeric composites. All the composites were coated with gold (Au) thin film using sputtering prior to scanning for surface morphology analysis. Figure 3(a) indicates a smoother surface for lower CB dispersion compared to higher CB dispersion [Fig. 3(b)] in the PU matrix indicating the percolation

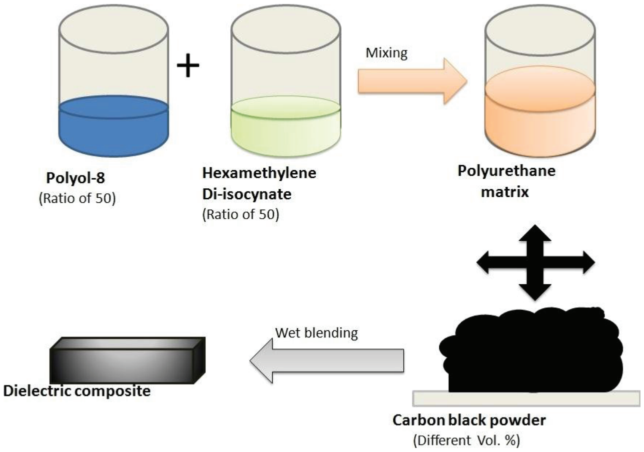


Fig. 1. Schematic diagram for the preparation of CB–PU composite applying wet mixing and compression molding techniques.

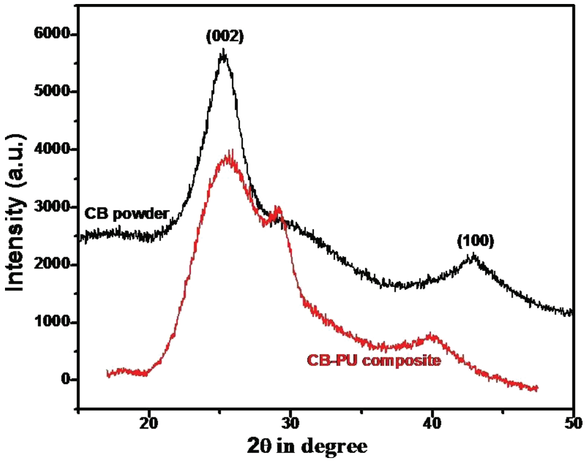


Fig. 2. X-ray diffraction patterns of carbon black powder and prepared CB–PU composite.

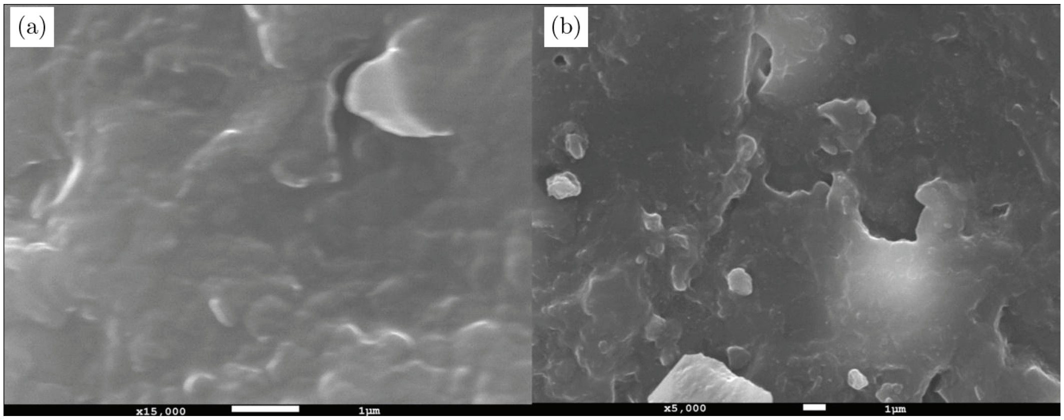
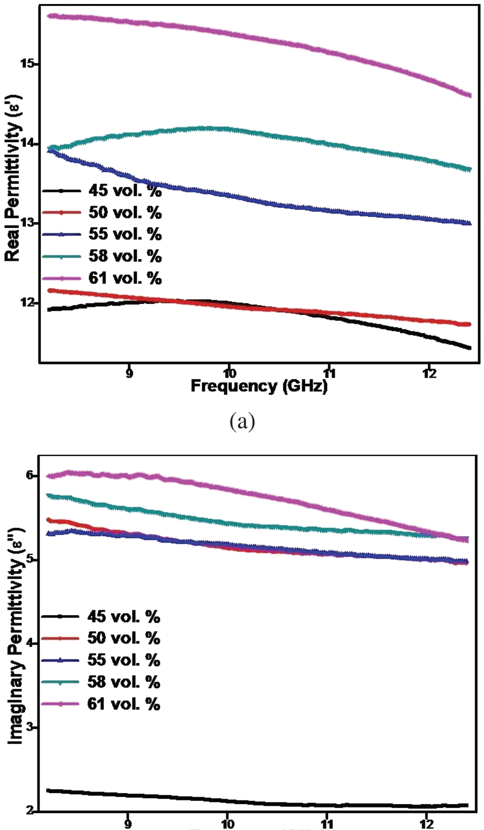


Fig. 3. FESEM images of polyurethane composites with (a) 45 vol.% and (b) 61 vol.% of CB powder.



(a)

(b)

Fig. 4. Frequency responses of (a) ε' and (b) ε'' of complex relative permittivity of the polyurethane composites with 45, 50, 55, 58 and 61 vol.% of CB powder.

effect. The surface morphologies of the prepared composites have shown uniform dispersion of CB powder in the polymeric matrix which reveals sufficient interaction between the filler and polyurethane matrix resulting in a conducting polymeric composite.

3.2. *Electromagnetic properties and absorption phenomenon*

EM properties of prepared CB–PU composites are usually described by their real permittivity (ε'), imaginary permittivity (ε'') and dielectric loss tangent ($\tan \delta_e = \varepsilon''/\varepsilon'$). These properties can be tuned to optimize the absorption phenomena of transmitted wave. The rectangular-shaped composites are placed inside a two-port waveguide attached with an Agilent vector network analyzer having a software module 85071E. This calibrated setup was employed to measure the loss scattering parameters S_{11} or S_{22} in 8–12-GHz-frequency region. Further, complex permittivity was calculated from the measured scattering parameters. Subsequently, loss tangent and absorbing properties were evaluated using the complex permittivity values.[12]

3.2.1. *Complex permittivity* ($\varepsilon_r = \varepsilon' - j\varepsilon''$)

The real part of complex permittivity of the PU matrix filled with CB was measured in the *X*-band frequency as shown in Fig. 4(a). The *X*-band calibration was performed in free space environment. It is observed that the percentage of CB in PU increased from 45 vol.% to 61 vol.%, the value of ε' increased from a value of 11.8 to 15.7 at 8-GHz frequency. This nonlinear spectrum of the real part of complex permittivity (ε_r) versus the frequency showed a resonance pattern for all the prepared CB–PU composites. The maximum value of real permittivity (ε'), i.e., 15.7, was obtained for 61 vol.% of CB–PU composite at a frequency of 8.2 GHz. It is also seen that the real permittivity (ε') declined from 15.7 to 14.5 for 61 vol.%, from 14.21 to 13.6 for 58 vol.%, from 13.9 to 13 for 55 vol.%, from 12.2 to 11.6 for 50 vol.% and from 11.9 to 11.4 for 45 vol.% in the whole *X*-band frequency range. The increment in the value of the dielectric constant (ε') is due to the conductivity factors (conduction and polarization) associated with carbon black, which appear only after an applied microwave frequency.

Figure 4(b) depicts the dependence of imaginary part of complex permittivity (ε'') on the *X*-band frequency. As the frequency is enhanced from 8 GHz to 12 GHz, the corresponding value of ε' increases with the enhancement of CB content in CB–PU composites. Initially, for 45–61 vol.% of CB–PU composites, the increase in the value of ε'' is from 2.2 to 6 at 8-GHz frequency, whereas it decreases monotonically for all composites from their maximum values to 4.96 expected for 45 vol.%. Because of the association of conduction and polarization factors, complex permittivity (ε_r) is expressed in terms of DC and AC conductivities for the prepared CB–PU composites. Additionally, Joule-heating loss is also expected due to the conducting behavior of carbonaceous fillers (CB) in the PU matrix, consequently, the dielectric loss factor (ε'') is responsible for static conductivity (σ''_{dc}) and electrical conductivity (σ''_{ac}) related to the molecular polarization phenomena of the conducting materials as given by the following equation:

$$\begin{aligned}\varepsilon'' &= \varepsilon''_{\mathrm{polarization}} + \varepsilon''_{\mathrm{conduction}} \\ &= \varepsilon''_{\mathrm{ac}} + \frac{\sigma_{\mathrm{dc}}}{2\pi f \varepsilon_0}.\end{aligned} \tag{1}$$

In the above Eq. (1), $\varepsilon''_{\mathrm{ac}}$ is the AC loss contribution, ε_0 is the permittivity of free space and f is the frequency of electromagnetic wave. In the case of CB–PU composite, the contributions to ε' and ε'' also occur due to interfacial polarization and relaxation as the CB contents separated by dielectric matrix molecules give rise to heterogeneity behavior.

3.2.2. *Dielectric loss tangent*

Figure 5 demonstrates the frequency responses of dielectric loss tangent (tan δ_e) in terms of ε' and ε''. It is observed that the value of tan δ_e ($\varepsilon''/\varepsilon'$) varies in the frequency range from 8 GHz to 9.8 GHz and further becomes constant up to the 12-GHz frequency for all composites. Electromagnetic waves are attenuated at maximum distance inside the absorbers according to the impedance matching conditions of absorption mechanism given by the following equation:

$$\tan\delta_e = \frac{\varepsilon''}{\varepsilon'} = \frac{\sigma}{\omega\varepsilon'\varepsilon_0}, \tag{2}$$

where d is the thickness of absorbers and ε is the permittivity in vacuum. The value of tan δ_e reaches a maximum value of 0.45 from 8.2 GHz to 10 GHz and further stays constant with a value of 0.42 from 10 GHz to 12.4 GHz for 50 vol.% of CB content, which indicates that dielectric loss makes major contribution to the absorption mechanism. In the external electromagnetic field, dipole–dipole interactions between the CB fillers and dielectric elements of polyurethane allow to skip the charge mutually by dipoles in the CB–PU composites. Consequently, an increase in the value of dielectric loss factor was observed with optimizing CB contents in the composites. The dielectric loss tangent is tuned to optimize by reinforcement in all CB–PU composites.[13]

3.2.3. *Absorption phenomena*

Figure 6 shows the measured absorption spectra in terms of RL of fixed thickness for all the prepared composites of 2.2-mm thickness. RL values of dielectric composites have been measured inside a rectangular waveguide, which are a function of characteristic parameters; scattering components, thickness of absorbers and frequency of the transmitted wave. The maximum absorption or minimum loss ($\mathrm{RL_{min}}$) of electromagnetic interferences/radiations under normal impinges of the transmitted waves on the surface of an absorbing

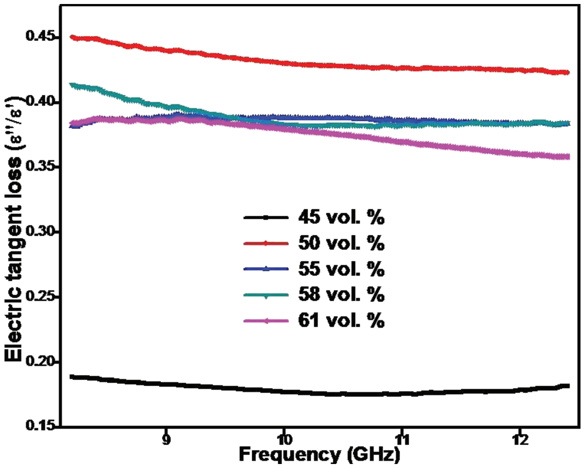


Fig. 5. The frequency responses of dielectric loss tangent for polyurethane composites with 45, 50, 55, 58 and 61 vol.% of CB powder of 2.2-mm thickness.

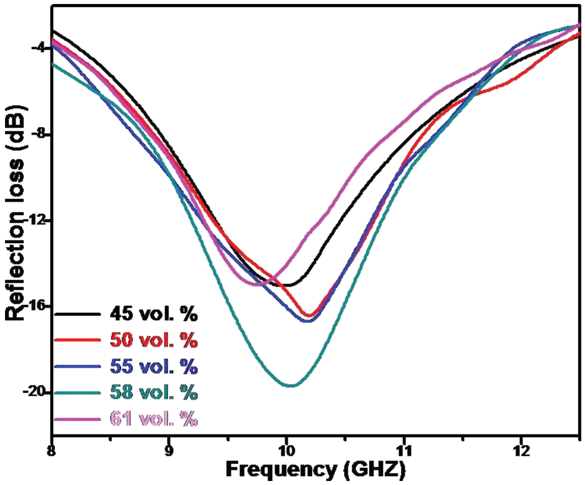


Fig. 6. The frequency responses of measured reflection losses for polyurethane composites with 45, 50, 55, 58 and 61 vol.% of CB powder of 2.2-mm thickness.

sample backed with a metallic plate can be defined through the proposed equation by Naito and Suetake[14]:

$$\mathrm{RL(dB)} = 20\log_{10}\left[\frac{\frac{Z_{\mathrm{in}}}{Z_o}-1}{\frac{Z_{\mathrm{in}}}{Z_o}+1}\right] = 20\log_{10}\left[\frac{\sqrt{\left(\frac{\mu_r}{\varepsilon_r}\right)}\tanh\left\{\frac{-j2\pi f\cdot d}{c}\right\}\sqrt{(\mu_r\varepsilon_r)}-1}{\sqrt{\left(\frac{\mu_r}{\varepsilon_r}\right)}\tanh\left\{\frac{-j2\pi f\cdot d}{c}\right\}\sqrt{(\mu_r\varepsilon_r)}+1}\right], \quad (3)$$

where the normalized impedance: $Z = \frac{Z_{\mathrm{in}}}{Z_o}$ is the ratio of absorber's impedance to the impedance of free space and ε_r and μ_r are the relative complex permittivity and permeability of the absorbing medium, respectively. In the present case, with the CB–PU composites being dielectric absorbers, the complex permeability ($\mu_r = \mu' - j\mu''$) is taken as unity; one for real permeability (μ') and zero for imaginary permeability (μ''). j is an imaginary quantity for the standard value ($j = \sqrt{-1}$). Frequency of the incident EM wave is given by f, while the velocity of EM wave in free space is given by c and d is the thickness of CB–PU composites.

In Fig. 6, a dip in the measured RL is equivalent to the minimal reflection at fixed thickness (d = 2.2 mm) of polyurethane composites with 45, 50, 55, 58 and 61 vol.% of CB powder dispersion. Dips of the minimal RL for all composites were tuned in the central frequency of X-band to optimize the thicknesses. This exhibits a minimum RL of −20.10 dB at a matching frequency (f_m = 10 GHz) with −10-dB bandwidth over the broad frequency range of 9.04–10.96 GHz for the optimized matching thickness (d_m = 2.2 mm). The shift of matching conditions for microwave absorption beyond the X-band for 1.5 mm > d_m > 2.5 mm is confirmed from the software module 85071E using the Nicolson–Ross algorithm.

4. Conclusion

Rectangular-shaped dielectric composites have been successfully prepared by using wet mixing method. The FESEM confirms the homogeneous dispersion of CB in the PU polymeric matrix. Complex permittivity values of the prepared CB–PU composites were significantly increased by increasing the volume percentage of CB, which is clear from the nature of real and imaginary permittivity spectra in the 8–12-GHz-frequency region. The optimal vol.% of carbon black powder in CB–PU composites achieves a microwave absorption of more than 99.3%. The dielectric tangent loss contributed to improved microwave absorption mechanism in the CB–PU composites. It is observed from the measured reflection loss curves (Fig. 6) for different vol.% of CB in the CB–PU composites that 58 vol.% shows a minimum reflection loss of −20.10 dB at 10 GHz, and a bandwidth of 1.92 GHz over the frequency range of 8–12 GHz. The prepared absorbers have shown better microwave absorbing performance as compared to other reported CB–PU-based absorbers in the literature.

References

[1] J. Rezania and H. Rahimi, Investigating the carbon materials' microwave absorption and its effects on the mechanical and physical properties of carbon fiber and carbon black/polypropylene composites, *J. Compos. Mater.* **51**, 2263 (2017).

[2] D. D. L. Chung, Electromagnetic interference shielding effectiveness of carbon materials, *Carbon* **39**, 279 (2001).

[3] I. R. Ibrahim, K. A. Matori, I. Ismail, Z. Awang, S. N. A. Rusly, R. Nazlan, F. M. Idris, M. M. M. Zulkimi, N. H. Abdullah, M. S. Mustaffa, F. N. Shafiee and M. Ertugrul, A study on microwave absorption properties of carbon black and $Ni_{0.6}Zn_{0.4}Fe_2O_4$ nanocomposites by tuning the matching-absorbing layer structures, *Sci. Rep.* **10**, 3135 (2020).

[4] A. P. Singh, M. Mishra and S. K. Dhawan, *Nanomagnetism*, ed. J. M. G. Estevez, Chapter 10 (One Central Press, 2017), pp. 246–276.

[5] A. C. Kumbharkhane, S. M. Puranik and S. C. Mehrotra, Dielectric relaxation of tert-butyl alcohol-water mixtures using a time-domain technique, *J. Chem. Soc. Faraday Trans.* **87**, 1569 (1991).

[6] K. K. Gupta, S. M. Abbas and A. C. Abhyankar, Carbon black/polyurethane nanocomposite-coated fabric for microwave attenuation in X & Ku-band (8–18 GHz) frequency range, *J. Ind. Text.* **46**, 510 (2015).

[7] V. Pratap, B. Singh, S. Kumar, A. K. Soni, M. Katiyar, K. Agarawal, Y. K. Sharma, A. M. Siddiqui, S. M. Abbas and N. E. Prasad, Electromagnetic and microwave absorbing properties of U-type barium hexaferrite/polyaniline-epoxy composites, *AIP Conf. Proc.* **2220**, 080003 (2020).

[8] G. G. Raju, *Dielectrics in Electric Fields*, Chapter 3 (CRC Press, 2019), pp. 83–136.

[9] S. M. Abbas, A. K. Dixit, R. Chatterjee and T. G. Goel, Complex permittivity, complex permeability and microwave absorption properties of ferrite-polymer composites, *J. Magn. Magn. Mater.* **309**, 20 (2007).

[10] T. Ungar, J. Gubicza, G. Ribarik, C. Pantea and T. W. Zerda, Microstructure of carbon blacks determined by X-ray diffraction profile analysis, *Carbon* **40**, 929 (2002).

[11] R. Jaiswal, K. Agarwal, V. Pratap, A. Soni, S. Kumar, K. Mukhopadhyay and N. E. Prasad, Microwave-assisted preparation of magnetic ternary core-shell nanofiller ($CoFe_2O_4$/rGO/SiO_2) and their epoxy nanocomposite for microwave absorption properties, *Mater. Sci. Eng. B* **262**, 114711 (2020).

[12] V. Pratap, A. K. Soni, A. M. Siddiqui, S. M. Abbas, R. Katiyar and N. E. Prasad, Dielectric and radar absorbing properties of exfoliated graphite dispersed epoxy composites, *J. Electron. Mater.* **49**, 3972 (2020).

[13] V. Pratap, A. K. Soni, S. Dayal, S. M. Abbas, A. M. Siddiqui and N. E. Prasad, Electromagnetic and absorption properties of U-type barium hexaferrite epoxy composites, *J. Magn. Magn. Mater.* **465**, 540 (2018).

[14] Y. Naito and K. Suetake, Application of ferrite to electromagnetic wave absorber and its characteristics, *IEEE Trans. Microw. Theory Tech.* **19**, 65 (1971).