Role of Magnetite (Fe₃O₄)-Titania (TiO₂) hybrid particle on mechanical, thermal and microwave attenuation behaviour of flexible natural rubber composite in X and Ku band frequencies

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Abstract
This current work aims to prepare a flexible elastomeric microwave shielding composite material with good mechanical and thermal properties by dispersing Fe₃O₄ and titania hybrid nano-particles. The primary aim of this research work is to prepare a high strength and high thermal stable flexible natural rubber composite with higher wave attenuation coefficient against 'X (8–12 GHz)' and 'Ku (12–18 GHz)' band microwave frequencies. The Fe₃O₄–Titania hybrids were prepared using low energy ball milling and the hybridization effect was confirmed using transmission electron spectroscopy. The prepared Fe₃O₄–Titania hybrids were surface-treated using 3-Aminopropyltriethoxysilane via wet solution method to avoid agglomeration. The natural rubber flexible composite was made by two-roll milling with recommended process parameters. The mechanical and thermal results showed improved tensile strength, modulus and mass decomposition. The highest tensile strength of 60 MPa was observed for composite contains 1.0 vol% of Fe₃O₄–TiO₂ particles. Similarly, the highest thermal stability of 385 °C is observed for composite contain Fe₃O₄–TiO₂ particles. The X and Ku band microwave attenuation behaviour revealed the highest attenuation of 28.1 dB for 1.0 vol% of Fe₃O₄–TiO₂ particle dispersed rubber composite in Ku band frequency. These mechanically toughened thermally stable and high microwave attenuation flexible composites could be used as EMI shielding material at antennae and other telecommunication devices where electromagnetic wave interference creates crucial issues.

1. Introduction
Flexible composites with high mechanical strength and microwave shielding abilities are required in modern technologies to minimize the electromagnetic interference between electronics devices and telecommunication devices [1]. The electromagnetic shielding may primarily required to improve the performance of electronic gadgets when they are indented to work together [2]. There are several researchers have investigated the polymer-based microwave shielding materials with conductive and magnetic particles [3, 4], But in polymeric shielding material the flexibility and deploy-ability could be a bigger issue due to the complexity in structure of electronic devices [5]. If the shielding material is flexible and easy to shape change they could be effectively deploy in high complex electronic devices [6]. There are several research were done on thermo and thermostet plastic based shielding material whereas the natural rubber based shielding material research is less. Based on this research gap this current study aims to develop a natural rubber-based flexible microwave shielding composite with ultra-fine high conductive TiO₂ and magnetic Fe₃O₄ particles. The conductive cum magnetic combination of particles could be more useful when requiring high performance on shielding effect [4]. prakash et al [5] conducted a study on copper-iron oxide particle hybrid for effective shielding performance on the polymeric material. The authors confirmed that the addition of the combined form of copper-iron oxide
particle on the epoxy matrix gives better shielding than composite contains only copper as filler. Similarly, Tahir et al[6] concluded the role of magnetic particle along with semi-conductive MWCNTs in kenaf-epoxy composite. The results outcome revealed that combined form of MWCNTs and magnetite particles offered better shielding effect than other composites. Based on the previous literatures in this current research to prepare high performance shielding material the TiO2 particle could select as conductive particle since it has a large dielectric constant of 85, the Fe3O4 could be selected as a magnetic counterpart since it contains high retentivity, susceptibility and coercivity[7, 8]. To embed these two particles the natural rubber could select as matrix material since it is abundant, ease of availability, simple manufacturing technique and degradable nature too. This natural rubber is highly resistant to chemical and water hence it is suggested in a lot of engineering applications[9]. These mechanically and thermally strengthened microwave shielding materials may serve better in electronics applications as an EMI shield[10].

2. Experimental part

2.1. Materials

The natural rubber matrix used in this current study was having a density of 1.15 g cm

−3 and purchased from Evonik Industries AG, Germany along with curing agent. The TiO2 nanoparticles of diameter <100 nm and density 3.78 g cm

−3 and silane surface modifier 3-Aminopropyltriethoxysilane was purchased from Sigma Aldrich, USA. Similarly, ferric chloride of 162 g mol

−1, NaOH pellets of 40 g mol

−1 and ethanol of 46 g mol

−1 were purchased form MERCK India, Ltd All the chemicals were used in the as-received condition without any post processes.

2.2. Co-precipitation process

The Fe3O4 magnetic particles were prepared via co-precipitation method. In this NaOH of 1 N concentration has been prepared by dissolving NaOH pellets into water-based on molecular weight. The required amount of ferric chloride salt was then added with NaOH solution and stirred continuously for 10 min. Oleic acid of 3 wt% was added with ferric chloride to control the particle growth in precipitation. After 10 min the NaOH solution was decanted and the precipitate was separated using Whatman filter paper. The precipitates were dried at a heating oven to form Fe3O4 molecular structure from Fe(OH)2. Again the oleic acid-coated nanoparticles were briefly dipped in NaOH solution to remove coated oleic acid by heating the particle dipped NaOH solution. Finally, the particle was separated by using Whatman filter paper and dried in an oven at 100 °C to remove moisture and make the particles dry[11].

Figure 1 shows the TEM images of co-precipitated Fe3O4 particles along with its XRD analysis. The TEM examination was done using a high-resolution transmission electron microscopy JEOL JEM S 3000 model with the recommended evaluation procedure. The particles were dispersed in ethanol and sonicated for 10 min. A drop of particle mixed ethanol was kept on the carbon-coated holey copper grid and gets dried off by heating...
oven and ready for analysis. The particles show spherical morphology and in the range of around 100 nm. Similarly, the XRD results confirmed the 2θ peaks at 36.2, 44.2, 51.5 and 67.8, which indicate the presence of Fe₃O₄ simple cubic crystal structure in the prepared particles [12].

2.3. Ball milling process

The TiO₂ and Fe₃O₄ particles of 25 g were mixed and ball milled in planetary ball mill for 4 h. The ball material used was WC with a ball diameter of 10 mm. The mill was set with a rotating rpm of 250 throughout the process. The powder weight to ball weight ratio was maintained at 1:15. The ball mill jar was made up of stainless steel and coated with rubber material to avoid abrasion of the parent material. A small quantity of powder sample at the end of milling was collected for further characterization. Figure 2 shows the transmission electron microscope image of cold-welded TiO₂ and Fe₃O₄ particles. The images revealed that both TiO₂ and Fe₃O₄ particles were fused and formed a hybrid nature. Figures 2(a) and (b) shows two different crystal structures, which indicates the presence of both TiO₂ and Fe₃O₄ [13].

2.4. Surface-treatment of particle hybrid

The raw TiO₂ particle and ball-milled TiO₂ and Fe₃O₄ were further surface-treated via a wet chemical method. In this 95 weight percentage of ethanol was mixed with 5 weight percentage of water for diluting the ethanol. A required amount of acidic acid was added with an ethanol-water solution to adjust the pH of silane solution between 4.8 to 5.4. The silane substance of 2 weight percentage is generally added with an ethanol-water solution and mild stirring applied via magnetic stirrer. The TiO₂ and TiO₂–Fe₃O₄ hybrid particles were immersed in the solution and soaked for 10 min. Finally, the particles were separated from the solution and heated by using an oven to form siliconized particle networks [14].

2.5. Composite preparation

The particulate flexible natural rubber composite has been prepared via two roll mill process. In the first step, the TiO₂ particles of 0.5 and 1.0 vol% is mixed with natural rubber and milling takes place. Similarly in second step TiO₂ and Fe₃O₄ particle hybrids of 0.5 and 1.0 vol% mixed with natural rubber and rolled continuously for near 40 passes. The stand-off-distance between the two rollers is maintained as 3 mm and the rollers are rotated at the rpm of 100. The resulted hybrid particle and natural rubber mix were further mixed with MBTS (Dibenzothiazole disulphide) of 10 g, TMT (Tetramethyl thiuram disulphide) of 5 g, Sulphur of 20 g and Zinc oxide of 25 g for curing and finally kept in a steel mould for high-temperature curing. The temperature was raised to 80 °C and soaked for 2 h in the mould itself. Thus particulate natural rubber composite was prepared [15, 16].

2.6. Specimen preparation

The two rolled milled hybrid TiO₂ and Fe₃O₄ particle dispersed natural rubber composite was taken out from the mould and checked for visual defects. ASTM test specimens were cut using a shearing machine with careful attention. The sectioned samples from cured rubber composite were again evaluated with standards and ready for testing and evaluation [17].

Figure 2. (a) and (b) shows cold-welded TiO₂ and Fe₃O₄ hybrid particles.
3. Characterizations

3.1. Mechanical properties
The tensile strength and tear toughness of pure natural rubber, TiO₂ particle dispersed natural rubber composite and TiO₂–Fe₃O₄ hybrid particle dispersed natural rubber composite was tested using a universal testing machine INSTRON 5900, UK by following ASTM D 638 and D 624 respectively. The microhardness of natural rubber composite was tested using a Shore-durometer (A scale) followed by ASTM D 2240.

3.2. Thermal properties
The mass decomposition analysis of natural rubber and its hybrid composites were done using a thermogravimetric analyzer NETZSCH STA Jupiter 409, Germany. The samples were scanned at the heating rate of 10 °C min⁻¹.

3.3. Magnetic analysis
The Hysteresis analysis on electro-magnetic TiO₂–Fe₃O₄ hybrid particle dispersed natural rubber composite was tested using a vibrato sample magnetometer, Lakeshore, 7407 (USA). The applied magnetic field was increased up to 15000 G and the mode of acquisition was point by point method with 101 positions.

3.4. Microwave shielding analysis
The microwave shielding analysis at the proposed microwave frequencies (X band 8–12 GHz and Ku band 12–18 GHz) were investigated via transmission line technique using vector network analyzer (Agilent/HP E8362B Network Analyzer, 10 MHz to 20 GHz) in room temperature. The sample holder was in rectangular shape with a length of 22 mm, a width of 11 mm and a thickness of 3 mm. The reflection parameter of S₁₁ and transmission parameter of S₂₁ was calculated from vector network analyzer (VNA) reading. Similarly, the relative permittivity (εᵣ) and the relative permeability (μᵣ) were calculated using Nicolson-Ross-Weir (NRW) by using the measured S₁₁ and S₂₁ values [18]. Based on the output parameters S₁₁ and S₂₁ from the vector network analyzer the shielding effect by absorption and reflection was calculated using the formulae (1) and (2) [19].

$$SE_{A} = 10 \log_{10} \left( \frac{1 - S_{11}}{S_{21}} \right)$$

$$SE_{R} = 10 \log_{10} \left( \frac{1}{(1 - S_{11})} \right)$$

thus the total shielding effect was calculated based on the equation (3).

$$SE_{total} = SE_{A} + SE_{R}$$

Where,

- $SE_{total}$ — Total shielding effect
- $SE_{A}$ — Shielding effect by absorption
- $SE_{R}$ — Shielding effect by reflection

4. Results and discussions

4.1. Mechanical properties
Table 1 shows the tensile strength and tear strength of natural rubber and its hybrid particle composites. It is observed that the pure natural rubber gives a very lower tensile strength of 28 MPa. This lower tensile strength is the cause of the absence of a strengthening mechanism in soft natural rubber material. There is no load absorbing reinforcements were dispersed thus lower tensile strength is observed. It is noted that the tear strength also measures as very lower as tensile strength. The measured tear strength is 27.4 Kg mm⁻¹ for pure natural rubber. Similarly, the shore durometer results show very lowest microhardness of 42. This poor tear strength

| Rubber vol%/TiO₂ vol%/TiO₂–Fe₃O₄ | Tensile strength (MPa) | Tear Strength (N/mm) | Micro hardness (Shore-A) |
|-----------------------------------|------------------------|-----------------------|-------------------------|
| 100/0/00                          | 30                     | 27                    | 39                      |
| 99.5/0.5/00                       | 48                     | 34                    | 43                      |
| 99.0/1.0/00                       | 55                     | 30                    | 48                      |
| 99.5/0/0.5                        | 44                     | 32                    | 46                      |
| 99.0/0/1.0                        | 60                     | 30                    | 50                      |
and hardness value are because of no micro-crack suppression mechanism within the material. Thus the propagation of microcracks is rapid enough and making immediate fracture [20]. It is further observed that the addition of 0.5 and 1.0 vol% of TiO₂ particle alone into natural rubber the tensile and tear strength improved along with improvement in hardness. The improved tensile strength, tear strength and hardness of 38%, 21% and 9.3% were observed for natural rubber composite, which consists of TiO₂ nanoparticle of 0.5 vol%.

Similarly, the improvement of 46%, 10% and 19% were observed for composite contains 1.0 vol% of TiO₂ nanoparticle. This improvement is because of the improved load sharing phenomenon of TiO₂ particle in the rubber matrix. The presence of ultrafine TiO₂ particles filled in void content of rubber and reduces the microcrack formation. This phenomenon improves the tensile and tear strength of the composite, contain 1.0 vol% of TiO₂ particles. The hardness also found to be increasing by dispersing 1.0 vol% of TiO₂ particles into rubber matrix. This improvement is the cause of arresting mobility of polymer chains when the load is applied. The presence of nanoparticle increases the penetration resistance thereby higher microhardness is observed [21].

It is observed that the addition of hybrid TiO₂–Fe₃O₄ particles into natural rubber increases its thermal stability. The improvement of 32% and 50% in tensile strength, 17% and 10% in tear strength and 16% and 22% in microhardness were observed for natural rubber composite, which contains TiO₂–Fe₃O₄ particles of 0.5 and 1.0 vol%. These significant improvements in these properties are because of the presence of TiO₂–Fe₃O₄ hybrids in natural rubber, which hinders the propagation of microcracks under loading condition. This hybrid particle uniformly settled in the voids of the rubber matrix and facilitates the applied load to transfer more uniformly. This phenomenon reduces the stress concentration factor on the matrix thus larger tensile strength, tear strength and microhardness was observed [22].

Figure 3 shows the fractograph of tensile and tear specimen after the test. Figure 3(a) and (b) shows the particle distribution on the matrix. The particles are not agglomerated much and dispersed uniformly. This uniform dispersion is the cause of more number of passes deployed in the two roll mill process. Similarly, figures 3(c) and (d) indicates a wavier fractured surface on the rubber matrix indicates matrix-particle interfacial fracture due to phase mixing.

### 4.2. Thermal Properties

Figure 4 shows the TGA thermogram of pure natural rubber and its composites. It is observed that the pure rubber gives rapid mass loss when the temperature increases. This rapid mass loss is the cause of the presence of isoprene monomer in a large volume, which gets evaporates at larger volume in the temperature range between 300 °C to 320 °C. The initial mass decomposition was seen at 300 °C with huge mass loss of 6.8%. It is observed that the addition of 0.5 and 1.0 vol% of TiO₂ particles into natural rubber increased its thermal stability. The
initial mass loss is delayed up to 340 °C and 350 °C, which is mere 12% and 14% of improvement on comparing with pure natural rubber. This improvement is because of the high heat-carrying capacity of TiO2 particle [23]. When temperatures increase the presence of TiO2 start absorbing the heat energy and hinder the rotation of natural rubber molecules. This hindrance in molecular lever leads the natural rubber to retain its current form for a long time. It is further noted that the mass loss at initial temperatures also reduces with the presence of hard titania [24]. The initial mass loss of 3.5% and 3.2 were noted for the composites, which is dispersed with 0.5 and 1.0 vol% of TiO2 particles in the rubber matrix. Similar, improvements were noted in rapid and final mass loss also. Thus the addition of hard TiO2 particles improved the thermal stability of natural rubber composite [25].

It is observed that the further addition of Fe3O4–TiO2 particles of 0.5 and 1.0 vol% into natural rubber increased the thermal stability further. The improvement of 17% and 22% were observed for composite, which contains 0.5 and 1.0 vol% of Fe3O4–TiO2 respectively. This improvement is because of the presence of hybrid Fe3O4–TiO2 in the rubber matrix. The high heat capacity of Fe3O4 with simple cubic spinel structure has very high hardness and high heat-absorbing capacity [25]. Thus the further delay in initial mass decomposition is observed. It is again noted that adding more volume of hybrid Fe3O4–TiO2 particles in natural rubber matrix improved the initial thermal stability. This improvement is the cause of the more active surface area of hard ceramic particles in the matrix. Thus adding a hybrid form of Fe3O4–TiO2 particles in natural rubber improved the thermal stability of natural rubber.

4.3. Dielectric properties

Figure 5 shows the real part and imaginary part of relative permittivity of natural rubber and its composites. It is observed that the pure natural rubber gives very lower relative permittivity among all composites. This lower value is the cause of the absence of dipole molecules in the rubber matrix. The natural rubber usually contains saturated hydrocarbons and benzene rings thus a small amount of permittivity is observed via electronic polarization. But it is observed that adding 0.5 and 1.0 vol. % of TiO2 particles into natural rubber increased the permittivity. This improvement is because of dipole attraction between inclusive TiO2 nanoparticles in rubber matrix against of frequency. It is noted that the permittivity value decreases with the increase of frequency. This reduction is the reason for poor relaxation time given to the natural rubber composite to be polarized. The poor relaxation time led poor polarization effect thus decrement in relative permittivity is observed. The maximum relative permittivity of 4 (imaginary part) and 3.8 (real part) were observed for composites, which contains 1.0 vol% of TiO2 particle in 8 GHz microwave region. Further improvement of frequency marginally reduces the relative permittivity from 4 to 2.5 at 18 GHz frequency [26]. It is further observed that adding hybrid Fe3O4–TiO2 particles of 0.5 and 1.0 vol% in to natural rubber gives a significant improvement in relative permittivity. The relative permittivity (real part) of 0.5 and 1.0 vol% of hybrid Fe3O4–TiO2 particle dispersed
natural rubber composite is 2.8, 2.2, 1.8, 1.6, 1.4 and 3.2, 2.6, 2.2, 2.0, 1.8. This improved relative permittivity is the reason of presence of hybrid Fe$_3$O$_4$–TiO$_2$ particle in natural rubber. This incurred dielectric constant is improving the EMI shielding by blocking the incoming microwaves by faradays shielding. It is observed that in all composites the real part measures little lower value than imaginary part. Since the base matrix is the insulation based substance the amount of heat generation is more which is considered as a conduction loss. The poor relaxation time offered due to higher frequency is the cause of this heat loss. Moreover the excess vibration due to high degree of polarization changes also a cause of large dielectric loss in elevated frequency. It is observed that the hybrid form of Fe$_3$O$_4$–TiO$_2$ particle in natural rubber gives relatively lower permittivity in both real and imaginary part compare than natural rubber composite filled with 1.0 vol% of TiO$_2$ nano-particle. This difference in permittivity is the cause of availability of large dipole molecules with in natural rubber [27].

4.4. Magnetic behaviour

Figure 6 shows the B–H curve of natural rubber composites made of 0.5 and 1.0 vol% of hybrid Fe$_3$O$_4$–TiO$_2$ particles. It is observed that the addition of ferromagnetic Fe$_3$O$_4$ particles into natural rubber along with TiO$_2$ as a hybrid form improved the degree of magnetization. The incurred magnetic moments of 625 emu and 875 emu were observed for 0.5 and 1.0 vol% of Fe$_3$O$_4$–TiO$_2$ particles respectively. This improvement is because of the presence of magnetic particles. On comparing with 0.5 vol% the 1.0 vol% of Fe$_3$O$_4$ particles gives large magnetic moments because of its active surface area and ability to become a magnet. It is noted that the 0.5 vol% of Fe$_3$O$_4$ particles dispersed natural rubber composite gives residual magnetism of 385 emu with higher coercivity of 610 G whereas the 1.0 vol% of Fe$_3$O$_4$ particle in natural rubber composite offers residual magnetism of 625 emu with coercivity of 2580 G. This large difference is the reason of adding large volume of strong Fe$_3$O$_4$ particles. It is observed that the 1.0 vol% magnetic Fe$_3$O$_4$ dispersed natural rubber composite gives a wider curve than 0.5
vol%. This high width is because of strong magnetization effect, which offers large coercive force on the composite via the inclusions of Fe₃O₄ particles [28].

Figure 7 shows the real and imaginary permeability values of hybrid Fe₃O₄–TiO₂ particle dispersed natural rubber composite. It is observed that presence of magnetite Fe₃O₄ particles in natural rubber significantly increase the magnetic permeability. A maximum magnetic permeability of 2.8 is observed as imaginary part in natural rubber composite, which contain 1.0vol% of TiO₂ and Fe₃O₄ particles whereas the real permeability measures 1.9. This larger magnetic permeation induce residual magnetism thus hinder the propagation of incoming microwave by affecting the magnetic component [29]. It is observed that the imaginary part giving higher magnetic permeability than real part. This phenomenon is occurring due to the magnetic loss in higher frequency ranges. It is further observed that the magnetic loss (hysteresis loss) in high frequency region is larger than low frequency region. At 8 GHz the magnetic loss is lower and at higher frequency region (18 GHz) it is found to be higher. This larger magnetic loss is the cause of very high frequency, which induces the rubber composite to become magnetize and de-magnetise at very rapidly [30].

4.5. Microwave shielding behaviour

Figure 8 show the shielding effect due to absorption, reflection. Transmittance and the total shielding effect of various composite materials investigated in this present study. It is observed that the pure natural rubber gives maximum wave attenuation of 2.7 dB. This small observation in shielding is the cause of no microwave shielding mechanisms in-built in pure rubber. It is further noted that adding conductive TiO₂ particles into natural rubber gives improved shielding effectiveness. The improved shielding effectiveness of 12.5 dB and 18.3 dB were noticed for composites made of 0.5 and 1.0 vol% of TiO₂ particles respectively. This improved shielding effect is the cause of the occurrence of more conductive losses due to eddy current and more heat loss. Moreover, the reflection mechanism also higher for TiO₂ dispersed natural rubber composite. The presence of TiO₂ particles increases the chances of multi reflection within shielding material thus improved shielding effectiveness is observed [31].

Figure 8 shows the SEA and SER values of 0.5 and 1.0 vol% TiO₂–Fe₃O₄ particle dispersed natural rubber composite. The high permittivity and permeability of composite produced maximum shielding effectiveness of 24.1 and 28.1 dB at 18 GHz frequency. This higher dB is the reason for blocking both the electromagnetic fields of the incoming microwave by providing heat loss, eddy current, hysteresis loss and multi reflection between particles. When high-frequency incoming microwave is hitting the specimen the residual magnetism is created due to the presence of Fe₃O₄ particles. This residual magnetism further affects the magnetic component in the wave and produce large hysteresis loss. Thus larger attenuation is observed. On comparing with TiO₂ particle alone in natural rubber the TiO₂–Fe₃O₄ particle hybrid gives a significant effect on total shielding. Both the absorption and reflection properties were improved via enhancing conductance and magnetic effect by the inclusion of TiO₂–Fe₃O₄ hybrid particle. Thus the addition of magnetic Fe₃O₄ particles along with TiO₂ greatly improved the total shielding effect via blocking both electrical and magnetic components of an incoming microwave [32]. It is noticed that the transmittance getting increases with respect to frequency. A maximum
transmittance value of 29.6 dB is observed for composite, which contains hybrid form of TiO\textsubscript{2}–Fe\textsubscript{3}O\textsubscript{4} particles in natural rubber. All composites show relatively higher transmittance than shielding effect due to absorption and reflectance. This phenomenon is the cause of absence of multi reflection mechanism with in shielding material. Thus the transmittance is still equal with the total shielding effect of composite. Based on the graphs around 50% of shielding is achieved by the incorporation of hybrid TiO\textsubscript{2}–Fe\textsubscript{3}O\textsubscript{4} particle in rubber matrix. It is observed that the total shielding effect is larger for high frequency than lower frequencies. This phenomenon is because of very high frequency, which amplitude maximum number of time within shielding material and producing more loss due to loss of energy. It is understood from results that the absorption mechanism dominates more on total shielding compare than reflection mechanism. The absorption loss is maximum in high-frequency waves thus larger wave attenuation is observed in 18 GHz frequency.

5. Conclusions

A flexible natural rubber composite for high-performance shielding is fabricated with TiO\textsubscript{2} and TiO\textsubscript{2}–Fe\textsubscript{3}O\textsubscript{4} hybrid particles and their effectiveness is measured in mechanical, thermal and microwave shielding effect. The following conclusions were made from the present study.

1) The addition of hybrid TiO\textsubscript{2}–Fe\textsubscript{3}O\textsubscript{4} particle into natural rubber gives the highest tensile strength, tear strength and shore hardness.

2) The thermal stability of hybrid TiO\textsubscript{2}–Fe\textsubscript{3}O\textsubscript{4} dispersed natural rubber gives very high stability. The initial mass losses were minimum on comparing with other composites tested. Hence, the TiO\textsubscript{2}–Fe\textsubscript{3}O\textsubscript{4} hybrid particle dispersed natural rubber composites are highly stable against external temperature and suitable for shielding effectiveness under elevated temperature zones too.

3) The TiO\textsubscript{2}–Fe\textsubscript{3}O\textsubscript{4} hybrid particle dispersed natural rubber composite gives higher permittivity and loss at higher frequency (18 GHz).

4) The hysteresis loop shows broader and lengthier ‘S’ curve for 1.0 vol% TiO\textsubscript{2}–Fe\textsubscript{3}O\textsubscript{4} hybrid particle dispersed natural rubber composite which, shows higher magnetization, residual magnetism and high coercive force.
5) The scattering parameters like reflection \( (S_{11}) \) and transmittance \( (S_{21}) \) were significant for \( \text{TiO}_2-\text{Fe}_3\text{O}_4 \) hybrid particle dispersed natural rubber composite. Subsequently, higher shielding via absorption and reflection is observed for rubber composite with 1.0 vol\% of \( \text{TiO}_2-\text{Fe}_3\text{O}_4 \) hybrid particle at 18 GHz microwave region. Whereas the composite, which contains only \( \text{TiO}_2 \) particles gives nominal shielding effect.

6) Thus the addition of \( \text{Fe}_3\text{O}_4 \) particle along with \( \text{TiO}_2 \) yield better results in mechanical, thermal and microwave shielding effects. Hence, the magnetic particle addition is mandatory when preparing high-performance shielding materials for high-frequency microwave regions.

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