Effects of Recycled Fe$_2$O$_3$ Nanofiller on the Structural, Thermal, Mechanical, Dielectric, and Magnetic Properties of PTFE Matrix

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Abstract: The purpose of this study was to improve the dielectric, magnetic, and thermal properties of polytetrafluoroethylene (PTFE) composites using recycled Fe$_2$O$_3$ (rFe$_2$O$_3$) nanofiller. Hematite (Fe$_2$O$_3$) was recycled from mill scale waste and the particle size was reduced to 11.3 nm after 6 h of high-energy ball milling. Different compositions (5–25 wt %) of rFe$_2$O$_3$ nanoparticles were incorporated as a filler in the PTFE matrix through a hydraulic pressing and sintering method in order to fabricate rFe$_2$O$_3$–PTFE nanocomposites. The microstructure properties of rFe$_2$O$_3$ nanoparticles and the nanocomposites were characterized through X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM), and high-resolution transmission electron microscopy (HRTEM). The thermal expansion coefficients (CTEs) of the PTFE matrix and nanocomposites were determined using a dilatometer apparatus. The complex permittivity and permeability were measured using rectangular waveguide connected to vector network analyzer (VNA) in the frequency range 8.2–12.4 GHz. The CTE of PTFE matrix decreased from 65.28 × 10$^{-6}$/°C to 39.84 × 10$^{-6}$/°C when the filler loading increased to 25 wt %. The real ($\varepsilon'$) and imaginary ($\varepsilon''$) parts of permittivity increased with the rFe$_2$O$_3$ loading and reached maximum values of 3.1 and 0.23 at 8 GHz when the filler loading was increased from 5 to 25 wt %. A maximum complex permeability of (1.1 – j0.07) was also achieved by 25 wt % nanocomposite at 10 GHz.

Keywords: PTFE; nanoparticles; microwave; recycled Fe$_2$O$_3$; complex permittivity; complex permeability

1. Introduction

Mill scale scrap is considered as a solid waste generated in the steel and iron industries. In general, nearly (8–15) kg of mill scale waste is produced per 1000 kg of steel, and the annual production of mill scale waste is over 13.5 million tons according to preliminary statistics. Moreover, most of this waste is stockpiled in factories due to its limited utilization. The iron protoxide in this waste can be dissolved in acid rain and subsequently transferred into soil and ground waters. Mill scale waste has a dual concern with respect to environmental pollution and land occupancy. Therefore, it is crucial to develop a cleaner method to solve the abundant mill scale resource issue [1].

The study of synthetic techniques for ferrite nanoparticles production has recently attracted considerable attention in view of their excellent magnetic and electrical properties [2]. Up to date, various techniques have been used for the synthesis of Fe$_2$O$_3$ nanoparticles (Reference code: 00-024-0072) such as electrospinning technique [3], reduction-oxidation process [4], and hydrolysis process [5]. Indeed, the latter techniques have many disadvantages such as: friability of inorganic nanofibers after calcination [6], non-cost-
effective for high contaminant concentrations, and slow reaction speed, respectively. However, mill scale waste materials have never been used to obtain Fe$_2$O$_3$ (00-024-0072) yet.

The microwave absorbing materials (MAMs) are in high demand to absorb the unwanted electromagnetic waves via dielectric and magnetic losses [7]. Microwave materials are used in the frequency range (300 MHz–300 GHz) [8]. MAMs are extensively used for many military and commercial applications such as electromagnetic shielding [9]. Therefore, the development of polymer composites (PCs) with tunable dielectric and magnetic properties have triggered researchers in recent years. The PCs reinforced with nanofillers have been widely studied to provide improved radiation protection properties compared to their metal counterparts. In addition, the electronics, packaging, household, energy, sports, communication, and leisure industries are fully involved in the use of PCs for various applications [10]. The advanced polymer composites can be fabricated based on the requirement by selecting quantity, shape, size, and type, of fillers as the reinforcements in different polymer matrix [11].

Polytetrafluoroethylene (PTFE) is a thermoplastic polymer, and PTFE is the best solvent chemical with the best resistance among thermoplastics. The properties of PTFE such as high service temperature, low moisture absorption, chemical inertness, etc., are crucial for many microwave applications [12]. However, PTFE also has certain disadvantages such as high coefficient of thermal expansion (CTE) [13,14], and low relative complex permittivity ($\varepsilon^*$) [15]. In addition, several studies have used Fe$_2$O$_3$ as the filler in PTFE matrix. Madusanka et al. synthesized thick film gas sensor using PTFE and $\alpha$-Fe$_2$O$_3$ nanoparticles [16] while Kang et al. fabricated porous Fe$_2$O$_3$-PTFE nanofiber membranes for photocatalysis applications [17]. Moreover, PTFE-Al-Fe$_2$O$_3$ composites were prepared to be used as reactive materials [18]. The electro-Fenton system was synthesized by combining multiwall carbon nanotubes and Fe@Fe$_2$O$_3$ nanowires with PTFE [19]. It is also worth noting that Fe$_2$O$_3$ nanoparticles have been widely used as fillers to enhance the dielectric and magnetic properties of various matrixes such as polycaprolactone (PCL) [2], polyaniline (PANI) [20], silica [21], and poly(methyl methacrylate) (PMMA) [22]. However, rFe$_2$O$_3$ nanoparticles have never been used as fillers in PTFE matrix for microwave absorption applications yet.

In this study, both CTE and $\varepsilon^*$ were improved by incorporating rFe$_2$O$_3$ into a PTFE matrix. The rFe$_2$O$_3$ nanofiller with enhanced relative complex permittivity can be embedded with a non-conducting polymer matrix such as PTFE to prepare a novel absorber which could have promising attenuation properties for electromagnetic interference (EMI) suppression. This absorber can be further enhanced by the magnetic nature of the Fe$_2$O$_3$ nanoparticles and with their high interfacial density due to their good compactness [23].

Knowledge of the complex permittivity and complex permeability of materials has sparked a great interest in industrial and scientific applications [24]. The interaction between a dielectric sample and high frequency electromagnetic signal can be expressed by the relative complex permittivity equation: $\varepsilon^* = \varepsilon' - j\varepsilon''$, where $\varepsilon'$ and $\varepsilon''$, respectively, represent the real and imaginary parts. The ratio of $\tan \delta = \varepsilon'' / \varepsilon'$ represents the loss tangent of a sample and higher values indicate higher attenuation properties [25]. In Addition, the interaction effect between a specimen and the magnetic field component of EM waves can be characterized by the relative complex permeability ($\mu^*$) which can be expressed by the equation: $\mu^* = \mu' - j\mu''$ where $\mu'$ and $\mu''$ represent the real and imaginary parts, respectively [26]. The real part determines the amount of energy that the material has stored from a magnetic field, while the imaginary part measures the attenuation of the magnetic field by the material. The magnetic loss tangent ($\tan \delta_\mu$) is calculated using the equation $\tan \delta_\mu = \frac{\mu''}{\mu'}$ [27], which represents the power lost versus power stored in a sample [28].

In this research, a low-cost, less complicated and non-chemical synthesis of recycled ferrite was done from mill scale waste. Then, the particle size of rFe$_2$O$_3$ decreased to nano-size via high energy ball milling (HEBM) in order to enhance the dielectric properties. The current study also involves the processes of synthesizing five nanocomposites of
rFe$_2$O$_3$—PTFE by varying the rFe$_2$O$_3$ percentage in the nanocomposites. Afterwards, the resultant effects of rFe$_2$O$_3$ loadings on the phase composition, thermal expansion tensile strength, density, complex permittivity, and complex permeability of PTFE matrix were investigated.

2. Materials and Methods

2.1. Materials

The materials used for the preparation of the rFe$_3$O$_4$ nanoparticles and rFe$_2$O$_3$–PTFE nanocomposites were: Mill scale flakes (Perwaja Sdn. Bhd., Chukai, Terengganu, Malaysia) and PTFE molding powder (Fujian Sannong New Materials Co., LTD, Sanming, China) with average particle size 50–110 µm.

2.2. Synthesis of rFe$_2$O$_3$ Nanoparticles from Mill Scale

Mill scale waste was initially cleaned by removing the impurities and then crushed manually into powder using mortar. This was followed by the application of the magnetic separation technique (MST) and Curie temperature separation technique (CTST) to further purify the powdered mill scale. The materials used for these processing techniques were an electromagnet which produced 1 Tesla magnetic field intensity, a thin cylindrical glass tube open at both ends and deionized water of density 1 g/cm$^3$. The CTST was used to separate the wustite (FeO) and magnetite (Fe$_3$O$_4$) contained in the mill scale slurry collected in the cold water separation process [23]. In this separation step, the clamped glass tube closed at the bottom end with a glass stopper, was filled with hot deionized water (100 °C) followed by a small quantity of the slurry. The particles that remained attracted to the magnetic field were inferred to be magnetite (Fe$_3$O$_4$) because of its higher Curie temperature of 585 °C. The rFe$_3$O$_4$ powder obtained was milled via the mechanical alloying technique using the high energy ball mill (SPEX Sample Prep 8000D, Metuchen, NJ, USA) operated by a 1425 rpm 50 Hz motor at a clamp speed of 875 cycles/minute. The powder to balls ratio of 1:10 was used. A total of 7 gm of the Fe$_3$O$_4$ powder was poured into each of the two vials containing the 70 g of steel balls and milled for 6 h to produce nanoparticles. During the ball milling Fe$_3$O$_4$ was converted to Fe$_2$O$_3$ due to high temperature and the content of oxygen in the vials. The preparation process of rFe$_2$O$_3$ nanoparticles is shown in Figure 1.

![Figure 1. Preparation process of rFe$_2$O$_3$ nanoparticles.](image-url)
2.3. Fabrication of rFe₂O₃–PTFE Nanocomposites

The rFe₂O₃–PTFE nanocomposites shown in Figure 2 were fabricated by mixing PTFE powder with different mass percentages (5–25%) of the rFe₂O₃ nano-powder as listed in Table 1. The composites were prepared through a hydraulic pressing and sintering method.

![rFe₂O₃–PTFE Nanocomposites](image)

**Figure 2.** rFe₂O₃–PTFE nanocomposites.

**Table 1.** Composition of rFe₂O₃–PTFE nanocomposites.

| Composite Code | Recycled Fe₂O₃ Nano-Powder | PTFE | Total Mass (g) |
|----------------|-----------------------------|------|---------------|
|                | Percentage | Mass (g) | Percentage | Mass (g) |               |
| Control        | 0          | 0        | 100        | 60       | 60            |
| A              | 5          | 3        | 95         | 57       | 60            |
| B              | 10         | 6        | 90         | 54       | 60            |
| C              | 15         | 9        | 85         | 51       | 60            |
| D              | 20         | 12       | 80         | 48       | 60            |
| E              | 25         | 15       | 75         | 45       | 60            |

Three different shapes of nanocomposites were fabricated for each mass percentage in order to suite different characterization apparatuses. The raw materials were weighed using analytical micro balance (A&D Company, Ltd., GR-200, Tokyo, Japan) which has an accuracy of ±0.00007 g. The mixing was performed via Wing mixer/blender for 10 min. Then, the mixed powders were placed into three different molds and pressed using a hydraulic pressing machine for 5 min under 10 MPa load. Finally, the compact rFe₂O₃–PTFE nanocomposites were sintered as follows: the compact nanocomposites were heated from room temperature to 380 °C in a furnace at 2.97 °C/min and then maintained for 1 h to allow the PTFE particles to coalesce completely. The cooling process was 1 °C/min from 380 °C to room temperature. The rFe₂O₃–PTFE nanocomposites preparation is shown in Figure 3. After cooling, the rFe₂O₃–PTFE nanocomposites were ready for characterizations.
Table 1. Composition of rFe$_2$O$_3$–PTFE nanocomposites.

| Composite Code | Recycled Fe$_2$O$_3$ Nano-Powder | Total Mass (g) | Percentage % Mass (g) | Percentage % Mass (g) |
|----------------|---------------------------------|----------------|-----------------------|-----------------------|
| Control        | 0                               | 60             | 100                   | 60                    |
| A              | 5                               | 57             | 95                    | 60                    |
| B              | 10                              | 54             | 90                    | 60                    |
| C              | 15                              | 51             | 85                    | 60                    |
| D              | 20                              | 48             | 80                    | 60                    |
| E              | 25                              | 45             | 75                    | 60                    |

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Figure 3. rFe$_2$O$_3$–PTFE nanocomposites preparation.

2.4. Characterization

rFe$_2$O$_3$ nanoparticles, PTFE powder, rFe$_2$O$_3$–PTFE nanocomposites were characterized using the following techniques:

2.4.1. X-ray Diffraction (XRD)

The phase composition structure of the samples was analyzed using X-ray diffraction (XRD). The data were collected using a fully automated Philips X’pert system (Model PW3040/60 MPD, Amsterdam, The Netherlands) with Cu–K$_\alpha$ radiation operating at a current of 40 mA, a wavelength of 1.5405 Å, and a voltage of 40 kV. The diffraction patterns were recorded with a scanning speed of 2°/min in the 2\(\theta\) range 10 to 70°. The collected data were subjected to the Rietveld analysis on PANalytic X’Pert Highscore Plus v.3.0 software (PANalytical B.V., Almelo, The Netherlands). The diffraction peaks were compared with the Inorganic Crystal Structure Database (ICSD). The rFe$_2$O$_3$–PTFE samples were cut from rFe$_2$O$_3$–PTFE nanocomposites prepared in advanced in a solid form, while the rFe$_2$O$_3$ was in the form of a fine powder.

2.4.2. High-Resolution Transmission Electron Microscopy (HRTEM)

The size and shape of rFe$_2$O$_3$ nanoparticles were investigated using HRTEM (JEM-2100F, JEOL, Tokyo, Japan). Drops of rFe$_2$O$_3$ nanoparticles were dispersed and placed on TEM grids for drying. The dried specimen was then placed in a high-vacuum chamber of the microscope for viewing and analysis of the nanoparticles.

2.4.3. Field Emission Scanning Electron Microscopy (FESEM)

The dispersion of rFe$_2$O$_3$ nanoparticles in PTFE matrix was evaluated using FESEM (JEOL Ltd. JSM-7600, Tokyo, Japan). Carbon tapes were used to cover the aluminum stubs before placing the specimens on them. Then, the samples were coated with titanium using an auto-fine coater (JEOL Ltd. JEC-3000FC, Tokyo, Japan) to increase their conductivity and eliminate electromagnetic charges. Finally, the stubs were placed on the FESEM chamber for analysis.
2.4.4. Electronic Densitometer

The effects of rFe$_2$O$_3$ nanoparticles on the density of PTFE matrix was determined using electronic densitometer (Alfa Mirage Co., Ltd. Model MD-300S0, Osaka, Japan) which employs Archimedes’ principle in order to calculate the density. Distilled water was used as the immersion fluid. The density was determined using the following equation [29]:

$$\rho = \frac{W_{\text{air}} \times \rho_{\text{dis.water}}}{W_{\text{air}} - W_{\text{dis.water}}}$$

(1)

where $W_{\text{dis.water}}$ is the composite weight in distilled water, $W_{\text{air}}$ is the composite weight in air, and $\rho_{\text{dis.water}}$ is the density of the distilled water (1 gm/cm$^3$).

2.4.5. Dilatometer Linseis L75 Platinum

Dilatometer is one of the main techniques used for coefficients of thermal expansion (CTEs) measurements. In general, to determine the CTE, two physical quantities (temperature and displacement) must be measured on a specimen that is undergoing a thermal cycle. In this study, dilatometer (Linseis L75 Platinum, Selb, Germany) was used to determine the CTEs of composites. The sample was enclosed in the furnace and then heated up to 200 $^\circ$C. When the sample expands, it pushes the central rod along the axis of the tube, and the relative movement can be characterized [30]. In addition, from an atomic perspective, the CTE represents the increment in the average distance among atoms with increasing temperature and, generally, weaker bonds have a higher CTE value [31].

2.4.6. Extensometer (Shimadzu AGS-X 100kN)

rFe$_2$O$_3$–PTFE nanocomposites were fabricated as per the ASTM-D638 standard in order to find out the ultimate tensile strength (UTS) [32]. The influence of rFe$_2$O$_3$ nanoparticles on the UTS of PTFE matrix was examined using the Shimadzu (AGS-X 100kN, Kyoto, Japan) with industry-leading (Shimadzu, TRAPEZIUM X, Kyoto, Japan) data processing software. The pneumatic grips were displaced with a rate of 5 mm/min at room temperature. The data were obtained from the software and data acquisition system monitor continuously until the specimen reached its UTS and broke.

2.4.7. Rectangular Waveguide (RWG)

The measurements of relative complex permittivity and permeability were carried out in the frequency range of 8.2–12.4 GHz using rectangular waveguide (RWG) connected to a Keysight (E5063A) vector network analyzer (Keysight Technologies, Santa Rosa, CA, USA). As shown in Figure 4, the sample holder containing the sample was carefully attached to the flanges of RWG in order to totally avoid the air gap. The complex permittivity and permeability of the nanocomposites were deduced depending on the transmission and reflection of the waves throughout the composites. The measurement model of RWG to measure the permeability and permittivity was the poly-reflection–transmission $\mu$ and $\varepsilon$ model. This model used an optimization technique which gives constant values for both $\varepsilon'$ and $\mu'$ throughout the whole frequency range. The transmission–reflection method is widely used in the EMI characterization because of the field-focusing ability which ensures an accurate characterization at the microwave frequency. The VNA was calibrated by implementing a standard full two-port calibration technique for 201 frequency points in the frequency range 8.2 GHz to 12.4 at room temperature. The characterization procedures were also described in our previous study [33].
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![Figure 4. Measurement of complex permeability and permittivity using rectangular waveguide technique.](image)

3. Results and Discussion

3.1. Microstructural Characterization

The X-ray diffraction (XRD) technique was used to confirm the phase structurers of PTFE matrix, rFe\(_2\)O\(_3\) nanopowder, and rFe\(_2\)O\(_3\)–PTFE nanocomposites. The XRD results are shown in Figure 5. By comparing the obtained diffractograms with the standard patterns from the Inorganic Crystal Structure Database (ICSD), the Bragg peaks of obtained recycled powder were all identified as rhombohedral crystal structure of single phase hematite (Fe\(_2\)O\(_3\), Reference code: 00-024-0072) with R—3 space group. The diffractogram of rFe\(_2\)O\(_3\) nanopowder showed the following peaks in the pattern at 2\(\theta\) of 24.1°, 33.1°, 35.6°, 40.8°, 43.5°, 49.4°, 54°, 57.5°, 62.4°, and 63.9° corresponding to the miller indices (hkl): (012), (104), (110), (113), (202), (024), (116), (122), (214), and (300), respectively. On the other hand, the diffractogram of PTFE matrix showed six peaks in the pattern at 2\(\theta\) of 18.04°, 31.54°, 36.60°, 37°, 41.22°, and 49.11°, which agreed with the study carried out by Yamaguchi et al. [34]. The mentioned peaks corresponded to the miller indices (hkl): (100), (110), (200), (107), (108), and (210), respectively. The diffractograms of rFe\(_2\)O\(_3\)–PTFE nanocomposites show dominant crystalline phase characteristics of PTFE at low concentration of rFe\(_2\)O\(_3\) content in the nanocomposites. However, the peaks corresponding to the rFe\(_2\)O\(_3\) nanopowder increased in intensity as the content increased. All peaks shown in rFe\(_2\)O\(_3\)–PTFE profiles belongs to the materials used for the preparation of the composites. The patterns did not show new peaks suggesting the nanocomposites were pure and there was no conversion of PTFE or rFe\(_2\)O\(_3\) nanopowder into other materials. The absence of any new peaks of the nanocomposites also suggests that the rFe\(_2\)O\(_3\) nanoparticles did not chemically interact with the PTFE matrix and that the mixture was physical in nature.
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**Figure 5.** XRD diffractograms of the rFe₂O₃ nanoparticles, PTFE, and rFe₂O₃–PTFE nanocomposites.

The microstructure characterization of the rFe₂O₃ nanoparticles was performed using high resolution transmission electron microscopy (HRTEM) to investigate the size and shape of rFe₂O₃ particles. The HRTEM results confirmed that the nanoparticles were highly crystalline as shown in Figure 6, which agreed with the study conducted by Wen et al. [35]. It can also be seen in Figure 6 that the rFe₂O₃ nanoparticles are aggregated and spherical, which may be attributed to the long-range magnetic dipole–dipole interaction between the particles. This observation may also be caused by the drying process during TEM sample preparation [36]. The rFe₂O₃ nanoparticles sizes were within the range 7.4–15.24 nm after 6 h of ball milling with an average particle size of 11.3 nm.

The cross-sectional surface images of 5% rFe₂O₃ (A), 15% rFe₂O₃ (C), and 25% rFe₂O₃ (E) nanocomposite are shown in Figure 7. It can be seen in 5% nanocomposite (A) that there are two main types of patterns observed in PTFE structure. The first pattern is small outgrowths which are uniformly dispersed on the surface of PTFE and was called “warts”. The second pattern is “ribbon” like shapes which entirely cover the PTFE surface and was called “dendrites”. These observations are in accordance with the study conducted by Glaris et al. [37]. The images confirmed that with an increase in rFe₂O₃ loadings in the composites, the adhesion to PTFE matrix increased gradually. There were no agglomerates observed in rFe₂O₃–PTFE nanocomposites. The uniform dispersal of rFe₂O₃ nanoparticles in the PTFE matrix gives an indication that the nanoparticles were completely implanted in the nanocomposites and offered interfacial bonding which can enhance the complex permittivity and permeability.
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Figure 6. HRTEM images of rFe₂O₃ nanoparticles.

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Figure 7. FESEM of rFe₂O₃–PTFE nanocomposites: 5% rFe₂O₃ (A) nanocomposite (a,b), 15% rFe₂O₃ (C) nanocomposite (c,d), and 25% rFe₂O₃ (E) nanocomposite (e,f).
3.2. Coefficients of Linear Thermal Expansion (CTEs)

The variation in CTEs with temperature for rFe$_2$O$_3$–PTFE nanocomposites in the temperature range 30–200 °C is presented in Figure 8. It was observed that the values of CTEs were low for the nanocomposites with higher content of rFe$_2$O$_3$ nanofiller. The decrease in CTEs of rFe$_2$O$_3$–PTFE nanocomposites is due to the difference between the CTEs of PTFE matrix and rFe$_2$O$_3$ nanofiller (Fe$_2$O$_3$ has a lower CTE comparing to PTFE). Figure 8 also shows that the CTEs of all nanocomposites increased as temperature increased throughout the measurement range. This behavior can be attributed to the increase in molecular vibrations with temperature, and a linear relationship exists between the CTE and the thermal capacitance per unit volume. The incorporation of low-CTE rFe$_2$O$_3$ in the PTFE matrix increased the thermal stability of the nanocomposites over a wide temperature range. Composites with low CTE are highly desirable for precision structures [38] such as microwave applications, where temperature fluctuations at the operating environment could cause substantial change in dimensions leading to destruction in the application.

![Figure 8. Thermal expansion coefficients of rFe$_2$O$_3$–PTFE nanocomposites.](image)

3.3. Tensile Strength and Density of rFe$_2$O$_3$–PTFE Nanocomposites

The variation in tensile strength with filler content for different rFe$_2$O$_3$–PTFE nanocomposites is presented in Figure 9. The tensile strength of polymer composites depends on the properties of nanoparticle–matrix interaction which plays an important role in the level of dissipated energy by different damaging mechanisms which take place at the nanoscale [39]. As shown in Figure 9, the tensile strength of nanocomposites decreased with increasing rFe$_2$O$_3$ nanofiller content. This behavior was expectable for polymer composites with inorganic filler and it was reported by a study conducted by Jiang et al. [40]. This decrease could be attributed to the rFe$_2$O$_3$ nanofiller which were not able to support the stress transferred from PTFE matrix which weakened the nanocomposite [14].

![Figure 9. Tensile strength vs. various rFe$_2$O$_3$ nanofiller content for rFe$_2$O$_3$–PTFE nanocomposites.](image)
The variation of density values with different rFe₂O₃ wt % content in the PTFE matrix is shown in Figure 10. The 5% (A), 10% (B), 15% (C), 20% (D), and 25% (E) rFe₂O₃ nanocomposites had the respective density values of 2.2, 2.32, 2.4, 2.49, 2.53 gm/cm³. Therefore, increasing the rFe₂O₃ nanofiller corresponded to increased density values of rFe₂O₃–PTFE nanocomposites. The increase in density with the increase in Fe₂O₃ nanofiller is supported by a study carried out by Ghasemi-Kahrizsangi et al. [41]. This behavior can be attributed to the higher true density of Fe₂O₃ (5.24 gm/cm³) [42], in comparison to the PTFE (2.2 gm/cm³) [43]. The increase in density can cause an observed increase in dielectric constant [44]. Moreover, Leyland and Maharaj found that the relationship between density and dielectric constant is directly proportional for various materials [45]. The denser rFe₂O₃–PTFE nanocomposites results higher number of molecules per unit volume. A larger number of molecules per unit volume means that there is more interaction with the electric fields and therefore an increase in the complex permittivity.

Figure 9. Tensile strength vs. various rFe₂O₃ nanofiller content for rFe₂O₃–PTFE nanocomposites.

Figure 10. Density vs. various rFe₂O₃ nanofiller content for rFe₂O₃–PTFE nanocomposites.
3.4. Complex Permittivity of rFe$_2$O$_3$–PTFE Nanocomposites

The measured complex permittivity values (real and imaginary parts) of rFe$_2$O$_3$–PTFE nanocomposites in the frequency range of 8.2–12.4 GHz using rectangular waveguide are shown in Figure 11; Figure 12 while loss tangent is shown in Figure 13. The highest $\varepsilon'$ value of rFe$_2$O$_3$–PTFE nanocomposites was 3.1 for 25% rFe$_2$O$_3$ nanocomposite along the frequency range 8.2 GHz to 12.4 GHz while the lowest $\varepsilon'$ value was 2.01 for the PTFE sample. By increasing the rFe$_2$O$_3$ nanofiller in the nanocomposites, both $\varepsilon'$ and $\varepsilon''$ increased throughout the measurement range. It can be noticed that $\varepsilon'$ and $\varepsilon''$ increased from 2.29 and 0.10 for 5% rFe$_2$O$_3$ nanocomposite (A) to 2.39 and 0.14, respectively for 10% rFe$_2$O$_3$ nanocomposite (B) at 8.2 GHz. The same behavior of increasing complex permittivity by increasing the rFe$_2$O$_3$ nanofiller was observed in all nanocomposites. The $\varepsilon'$ and $\varepsilon''$ rely on the contribution of the atomic, electronic, interface, and orientation polarization in the sample [46]. The interface polarization increased due to the differences in polarization of the rFe$_2$O$_3$ nanofiller and PTFE matrix.

| Frequency (GHz) | $\varepsilon'_{(A)}$ | $\varepsilon'_{(B)}$ | $\varepsilon'_{(C)}$ | $\varepsilon'_{(D)}$ | $\varepsilon'_{(E)}$ | $\varepsilon''_{(PTFE)}$ |
|----------------|---------------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| 8.2            | 3.5                 | 3.0                 | 2.5                 | 2.0                 | 1.5                 | 0.5                 |
| 8.8            | 3.0                 | 2.5                 | 2.0                 | 1.5                 | 1.0                 | 0.5                 |
| 9.4            | 2.5                 | 2.0                 | 1.5                 | 1.0                 | 0.5                 | 0.3                 |
| 10             | 2.0                 | 1.5                 | 1.0                 | 0.5                 | 0.3                 | 0.2                 |
| 10.6           | 1.5                 | 1.0                 | 0.5                 | 0.3                 | 0.2                 | 0.1                 |
| 11.2           | 1.0                 | 0.5                 | 0.3                 | 0.2                 | 0.1                 | 0.1                 |
| 11.8           | 0.5                 | 0.3                 | 0.2                 | 0.1                 | 0.1                 | 0.1                 |
| 12.4           | 0.5                 | 0.3                 | 0.2                 | 0.1                 | 0.1                 | 0.1                 |

Figure 11. Variation in permittivity (real part) of rFe$_2$O$_3$–PTFE nanocomposites using rectangular waveguide.

The increment in complex permittivity with increasing rFe$_2$O$_3$ nanofiller can be attributed to the polarization process due to the enhanced conductivity and interfacial polarization in the composite and hopping exchange of charges between localized states [23]. One more reason for the influence on the relative complex permittivity of the nanocomposites is the particle size of rFe$_2$O$_3$ nanofiller since the nano size had high specific surface area that enabled good contact between the particles and the matrix [47]. The increment in $\varepsilon'$ and $\varepsilon''$ at low frequency can be attributed to the dominant role of dipolar and interfacial polarization. However, the increment at high frequencies can be attributed to the electronic and ionic polarization of the system [48].

It can be also seen in Figure 12 that $\varepsilon''$ of rFe$_2$O$_3$–PTFE nanocomposites decreased by increasing the frequency from 8.2 to 12.4 GHz. This decrement can be attributed to the interfacial dipoles having less time to align in the direction of the external field. The molecules were able to do a complete orientation at low frequency but they were not able to achieve the same orientation at high frequency [49]. However, the $\varepsilon''$ of the PTFE sample
did not change with frequency because PTFE is a nonpolar polymer [50], which means its complex permittivity is not dependent on frequency.

**Figure 12.** Variation in permittivity (imaginary part) of rFe2O3–PTFE nanocomposites using rectangular waveguide.

**Figure 13.** Variation in loss tangent of rFe2O3–PTFE nanocomposites.

**3.5. Complex Permeability of rFe2O3–PTFE Nanocomposites**

The permeability characteristics of rFe2O3–PTFE nanocomposites can only be attributed to the rFe2O3 nanoparticles because PTFE is a non-magnetic material [51]. The variations in $\mu'$, $\mu''$, and $\delta\mu$ of rFe2O3–PTFE nanocomposites are shown in Figures 14–16, respectively. It is clear that the values of $\mu'$, $\mu''$, and $\delta\mu$ were higher for rFe2O3–PTFE nanocomposites having higher rFe2O3 content.
3.5. Complex Permeability of rFe$_2$O$_3$–PTFE Nanocomposites

The permeability characteristics of rFe$_2$O$_3$–PTFE nanocomposites can only be attributed to the rFe$_2$O$_3$ nanoparticles because PTFE is a non-magnetic material [51]. The variations in $\mu'$, $\mu''$, and $\tan \delta_{\mu}$ of rFe$_2$O$_3$–PTFE nanocomposites are shown in Figures 14–16, respectively. It is clear that the values of $\mu'$, $\mu''$, and $\tan \delta_{\mu}$ were higher for rFe$_2$O$_3$–PTFE nanocomposites having higher rFe$_2$O$_3$ content.

**Figure 14.** Variation in permeability (real part) of rFe$_2$O$_3$–PTFE nanocomposites using rectangular waveguide.

**Figure 15.** Variation in permeability (imaginary part) of rFe$_2$O$_3$–PTFE nanocomposites using rectangular waveguide.
As the content of rFe$_2$O$_3$ nanofiller increased in the rFe$_2$O$_3$–PTFE nanocomposites, the spacing in the nanocomposites reduced due to the high specific surface area of the rFe$_2$O$_3$ nanofiller. This made it more difficult for the magnetic field to pass through the rFe$_2$O$_3$–PTFE nanocomposites, thereby increasing the values of the relative complex permeability.

The real part ($\mu'$) of the relative permeability values were 1.039, 1.047, 1.06, 1.8 and 1.1 for 5% (A), 10% (B), 15% (C), 20% (D) and 25% (E) rFe$_2$O$_3$ nanocomposites, respectively, within the frequency range of 8.2–12.4 GHz while the corresponding values of $\mu''$ were 0.017, 0.023, 0.026, 0.034 and 0.038 at 8.2 GHz. A resonant peak can be clearly seen in each $\mu''$ curve between 10 and 10.6 GHz. The increment in $\mu'$ and $\mu''$ of the polymer composites with increase in Fe$_2$O$_3$ is supported by studies conducted by Abdalhadi et al. [52] and Ahmad et al. [53]. In addition, the magnetic loss tangent in Figure 16 followed the behavior of $\mu''$ because it was calculated using the equation $\tan\delta_{\mu} = \frac{\mu''}{\mu'}$.

It can be seen that $\mu'$ in Figure 14 shows constant values while $\mu''$ in Figure 15 shows non-steady behavior with the frequency. The $\mu'$ is associated with the material storage capacity of the magnetic field [54], and it did not vary with frequency because of the RWG measurement model described in Section 2.4.7. On the other hand, the fluctuations in $\mu''$ with the frequency can be attributed to the magnetic loss. In general, the magnetic loss of a sample originates because of one of these four reasons: the hysteresis loss, domain-wall resonance, eddy current effect, and natural resonance [55]. The first reason (hysteresis loss) which originates from the irreversible magnetization is negligible in a weak applied magnetization field [56]. The second reason (domain-wall resonance) normally occurs in the (1–100) MHz range, therefore, this reason can be excluded in this study [57]. The third reason (eddy current effect) can usually be estimated using the equation of $C_0 = \mu' \left(\mu''\right)^2 f^{-1}$. When the magnetic loss originates for this reason, the $C_0$ should be almost constant as the frequency varied. However, it can be seen in Figure 17 that the $C_0$ values for the rFe$_2$O$_3$–PTFE nanocomposites varied considerably with the frequency; so, this reason can also be excluded. Hence, these resonant peaks are associated with the natural ferromagnetic resonance [58]. The trend of $\mu''$ is also supported by the study conducted by Liang [59].
In this research, rFe₂O₃–PTFE nanocomposites were successfully fabricated using rFe₂O₃ nanopowder as the filler and PTFE as the matrix. The effect of rFe₂O₃ loading on the structural, mechanical, thermal, dielectric, and magnetic properties of PTFE matrix was investigated. rFe₂O₃ nanofiller was dispersed uniformly with complete implantation in the PTFE matrix as presented in the FESEM results. The density increased but the tensile strength decreased with the increase of rFe₂O₃ nanofiller in the nanocomposites. The rFe₂O₃ nanoparticles improved the thermal properties of the PTFE matrix. The complex permittivity and permeability were also enhanced by embedding rFe₂O₃ nanofiller in the matrix throughout the measurement range. The important dielectric, magnetic, thermal and mechanical properties of rFe₂O₃–PTFE nanocomposites linked to the content of the rFe₂O₃ in the nanocomposites can be employed in possible applications requiring tunable characteristics.

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**Figure 17.** The values of $\mu'(\mu''/f)^{-1}$ as a function of frequency for rFe₂O₃–PTFE nanocomposites.
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