

Strategic Enhancement of Photocatalytic, Piezoelectric and Dielectric Properties of Solid-State Synthesized BaFe₁₂O₁₉ Nanoparticles Reinforced into PMMA Polymer Matrices Nanocomposites for Energy Storage Applications

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Strategic Enhancement of Photocatalytic, Piezoelectric and Dielectric Properties of Solid-State Synthesized BaFe₁₂O₁₉ Nanoparticles Reinforced into PMMA Polymer Matrices Nanocomposites for Energy Storage Applications

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ABSTRACT

This research focuses on the synthesis and characterization of nanocomposites consisting of barium hexaferrite (BaFe₁₂O₁₉) nanoparticles reinforced into poly(methyl methacrylate) (PMMA) polymer matrices. The nanocomposites were prepared using a solution-casting technique, while the barium hexaferrite nanoparticles were synthesized via a conventional solid-state reaction method, followed by calcination at 800°C for 1 hour. Structural, morphological, photocatalytic, piezoelectric, and dielectric properties were evaluated using several characterization techniques, including FTIR, XRD, SEM, UV-Vis spectroscopy, d₃₃ meter, and LCR analyzer. FTIR analysis identified absorption bands corresponding to Ba-O and Fe-O, as well as organic functional groups associated with the PMMA polymer matrices. XRD analysis confirmed the hexagonal structure of BaFe₁₂O₁₉ nanoparticles, with an average crystallite size of 15.23 nm. SEM analysis determined the particle size to be approximately 28 nm. UV-Vis spectroscopy evaluated the nanoparticles band-gap 1.76 eV and assessed photocatalytic activity. Under UV irradiation, all nanocomposites progressively degraded methylene blue (MB), where the 40 wt% BFO/PMMA nanocomposite achieving comparatively higher degradation efficiency of 67.22% than 61.78% for 50 wt% BFO/PMMA nanocomposite after 60 min. The maximum piezoelectric coefficient was determined 40 pC/N for 50 wt% BFO/PMMA nanocomposite with d₃₃ meter analysis. The dielectric, impedance and modulus characteristics were observed over the low-high frequency range 40-110 MHz. As barium hexaferrite nanoparticles reinforcement increased into PMMA polymer matrices, the 50 wt% composition exhibited relatively high dielectric constant 7.1 and ac conductivity $2.25 \times 10^{-3} \text{ S cm}^{-1}$, along with low dielectric loss 0.08 at high frequency range (1M-110M) Hz. Impedance and electrical-modulus analysis further revealed broad non-Debye relaxation behavior

associated with heterogeneous BFO/PMMA interfaces and localized charge-transport processes. The dispersion observed in the plots of Z'' and M'' with increasing frequency indicates the occurrence of dielectric relaxation. Overall, relatively higher $\text{BaFe}_{12}\text{O}_{19}$ reinforcement enhanced multifunctional properties of the nanocomposite, demonstrating their potential for energy storage, sensing, actuations, dielectric and photocatalytic pollutant-remediation applications.

Key words: Barium ferrite, PMMA, Nanocomposite, Dielectric, Piezoelectric, Photocatalytic

1. INTRODUCTION

The rapid development of miniaturized electronics, distributed sensing technologies, and advanced environmental remediation systems has intensified the demand for materials capable of delivering multiple functions within compact, processable architectures. Conventional dielectric, electromechanical, and photocatalytic technologies commonly rely on distinct classes of functional materials, increasing fabrication complexity and limiting their inclusion in multifunctional platforms [1]. Polymer–ceramic nanocomposites provide a beneficial route to overcome this drawback by combining the low density, electrical insulation, mechanical compliance, and processability of polymers with the high electric polarizability and functional responses of electroactive ceramics [2], [3]. Such integrated systems are particularly attractive because dielectric polarization, piezoelectric mechanical-to-electrical conversion, and photocatalytic photoinduced activity can be combined within a single processable material platform [1], [4].

Among multifunctional ferrite ceramics, barium hexaferrite ($\text{BaFe}_{12}\text{O}_{19}$, BFO) is attractive because of its useful dielectric response, ferrimagnetic character, and reported polarization-related behavior [5], [6], [7]. $\text{BaFe}_{12}\text{O}_{19}$ can also exhibit semiconductor-mediated photocatalytic activity under suitable irradiation, where photoexcitation generates electron–hole pairs, which participate in surface redox reactions and facilitate the formation of reactive oxygen species [4], [8], [9]. This photoactivity has extended $\text{BaFe}_{12}\text{O}_{19}$ -based materials toward degrading persistent organic contaminants, including methylene blue [8], [9]. Despite this adaptability, direct use of BFO has important functional restrictions. Bulk $\text{BaFe}_{12}\text{O}_{19}$ ceramics are rigid, brittle, and typically require high-temperature processing [7]. However, nanoscale $\text{BaFe}_{12}\text{O}_{19}$ particles frequently agglomerate due to their high surface energy and magnetic dipole interactions, which complicates their fabrication into continuous, mechanically robust structures [7], [10]. These limitations motivate combining $\text{BaFe}_{12}\text{O}_{19}$ with a suitable polymeric matrix.

Reinforcing $\text{BaFe}_{12}\text{O}_{19}$ within poly(methyl methacrylate) (PMMA) provides a rational strategy for retaining the functional characteristics of the ferrite while overcoming these processing and mechanical restrictions. PMMA offers low density, mechanical stability, optical transparency, and compatibility with solution-based fabrication. Although PMMA possesses relatively low dielectric permittivity and modest intrinsic electromechanical functionality, it provides a suitable and

processable matrix for functional enhancement through ferrite reinforcement [11]. BFO and PMMA therefore form a complementary architecture - the ferrite phase contributes dielectric polarization, mechanically responsive electrical behavior, and photoactive sites, whereas PMMA provides structural continuity, processability, mechanical support, and immobilization of the nanoparticles [4], [6], [7], [8], [9], [10], [11]. Previous studies have demonstrated that incorporating BFO into polymer matrices can tailor dielectric performance while preserving important polymer attributes [10], [12], [13], [14], [15]. Earlier work emphasized that homogeneous BFO distribution is vital for reliable functional behavior in BFO/PMMA nanocomposites [10]. Another study demonstrated that increasing the ferrite fraction can enhance dielectric permittivity while maintaining the processability of polymer-based composites [14], [15].

The multifunctional response of BFO/PMMA nanocomposites depends on the intrinsic properties of the constituent phases, as well as factors including ferrite-particle dispersion and the characteristics of the ferrite-polymer interface. A higher $\text{BaFe}_{12}\text{O}_{19}$ content increases the amount of polarizable material and the density of heterogeneous interfaces, which can enhance interfacial or Maxwell-Wagner-Sillars polarization. In contrast, reduced interparticle separation and agglomeration can intensify local electric-field distortion, interfacial charge accumulation, dielectric loss, and nonuniform charge transport [16], [17], [18]. These microstructural factors also influence mechanically induced electrical performance because the effective charge response depends on the active ferrite fraction, particle connectivity, interfacial adhesion, and mechanical stress transfer through the polymer matrix [4], [13], [19]. Furthermore, $\text{BaFe}_{12}\text{O}_{19}$ introduces a photoactive pathway under UV irradiation, in which photogenerated electrons and holes initiate surface reactions that generate reactive species capable of degrading organic dye molecules [4], [8], [9]. Within the composite, PMMA primarily provides structural support and immobilization, while dispersed $\text{BaFe}_{12}\text{O}_{19}$ serves as photoactive ferrite sites. Therefore, particle accessibility, dispersion, and polymer-ferrite interactions significantly influence the photocatalytic response [8], [9], [20]. Ferrite reinforcement and interfacial structure thus represent common design parameters that link the dielectric, mechanically induced electrical, and photocatalytic responses of BFO/PMMA nanocomposites. However, their combined dependence on $\text{BaFe}_{12}\text{O}_{19}$ concentration and ferrite-polymer interfaces remains underexplored [4], [10], [13], [14], [15], [20].

This study focuses on developing BFO/PMMA nanocomposites containing $\text{BaFe}_{12}\text{O}_{19}$ reinforcement of 30 wt%, 40 wt%, and 50 wt% and systematically evaluating their structural, morphological, dielectric, piezoelectric, and photocatalytic properties through FTIR, XRD, SEM, LCR meter, d_{33} meter and UV-Vis analyzer, respectively. $\text{BaFe}_{12}\text{O}_{19}$ nanoparticles were synthesized via a solid-state route and incorporated into PMMA by solution casting. Together, these synthesis and analysis clarified how $\text{BaFe}_{12}\text{O}_{19}$ reinforcement and ferrite-polymer interfaces govern multifunctional performance, thereby addressing the gap between predominantly single-function BFO-polymer studies and multifunctional composite design. The resulting structure-property relationships provide a basis for considering BFO/PMMA nanocomposites as a processable

multifunctional platform for dielectric and capacitive components, piezoelectric-type sensing and mechanical-energy transduction, and photoactive pollutant-remediation systems.

2. MATERIALS & METHODOLOGY

2.1. Synthesis of Nanoparticles:

BaFe₁₂O₁₉ precursor was synthesized via high-temperature solid-state reaction using barium oxide [BaO] ($\geq 99.5\%$ purity, Merck, Germany) and iron (III) oxide [Fe₂O₃] ($\geq 99.5\%$ purity, Merck, Germany) in a 1:12 molar ratio. The reagents were ground together using a mortar and pestle, and the resulting powder was dried at 80°C for 10 minutes to reduce moisture and contaminants. This grinding and drying cycle was repeated, with an additional 15 minutes of grinding each time, for a total duration of 3 hours. The mixture was then heated from room temperature to 800°C at a rate of 10°C/min, held at 800°C for 1 hour in a furnace, and subsequently cooled to room temperature at a rate of 10°C/min. The calcined powder was further ground in a mortar and pestle for 3 hours and stored in a vacuum box for subsequent characterization.

2.2. Synthesis of BFO/PMMA Nanocomposite:

The BFO/PMMA solution were created with great care using a precise solvent casting technique. The first phase was submitting polymers pellets to a comprehensive drying procedure using a vacuum oven, wherein a temperature of 90°C was maintained for a period of 24 hours. The implementation of this crucial pre-processing step facilitated the elimination of any remaining moisture, ensuring the integrity of the

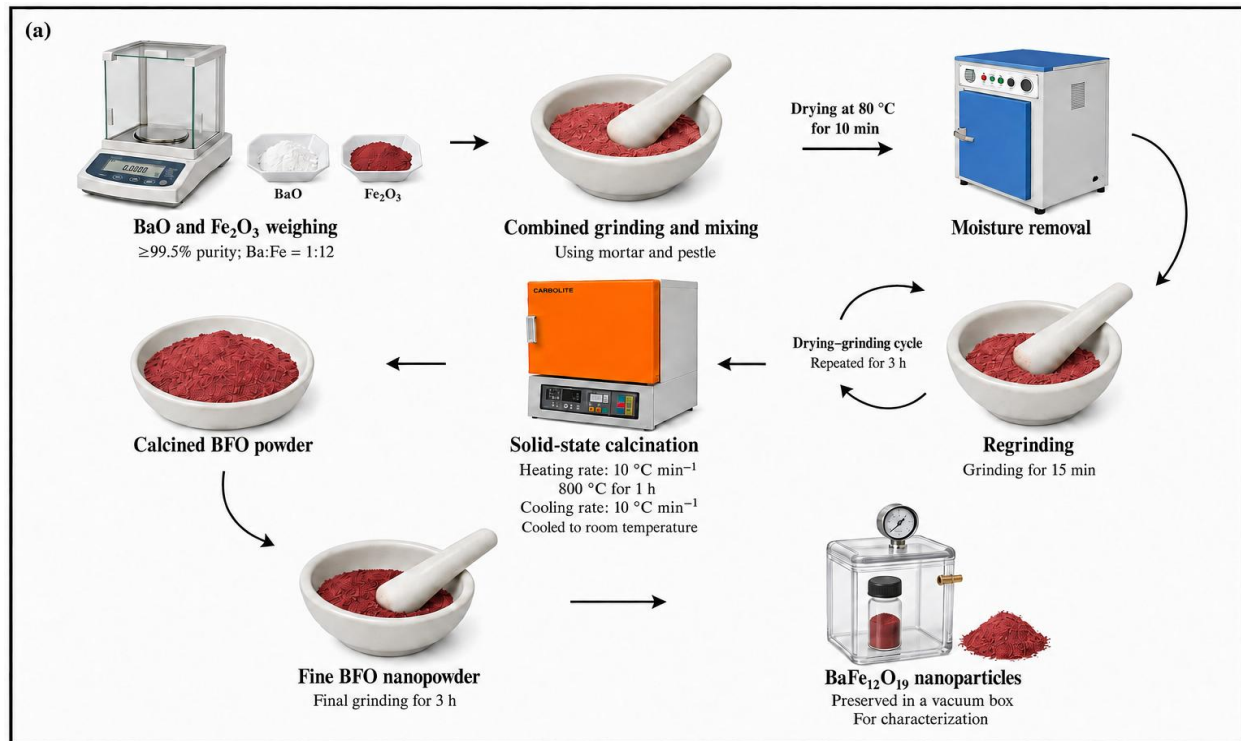


Figure 2.1. Schematic illustration of the solid state synthesis of BaFe₁₂O₁₉ nanoparticles

polymer blend. After the completion of the drying procedure, a composition consisting of PMMA was measured. The addition of a combination consisting of dimethyl sulfoxide (DMSO) and acetone, with a

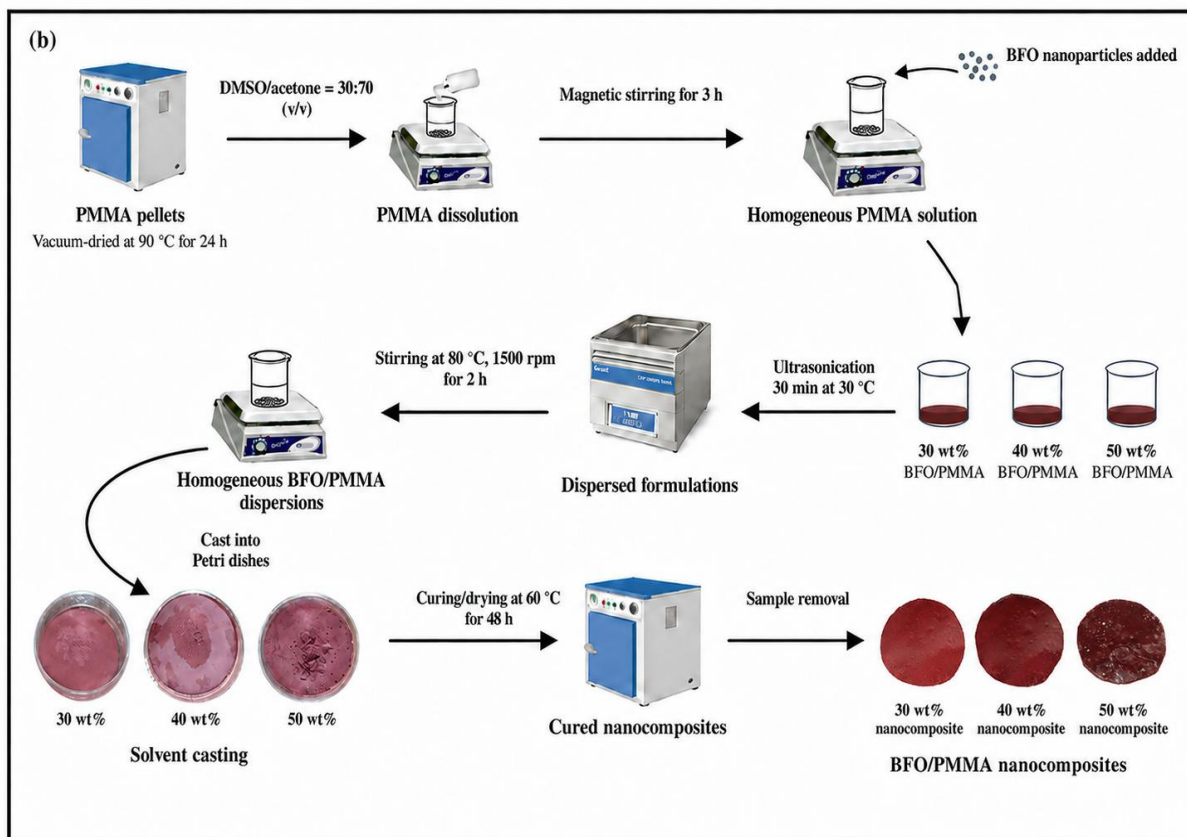


Figure 2.2. Schematic illustration of BFO/PMMA nanocomposite fabrication

volumetric ratio of 30:70, was performed in order to achieve a uniform blend. This mixture was added to the 5g weighted PMMA. Following this, the composite underwent vigorous stirring on a heated plate for a duration of 3 hours, facilitating the dissolution of the polymers in the solvent combination. In order to include the synthesized BFO nanoparticles into polymer matrices, same ratios of 30%, 40%, 50% were introduced into the respective beakers. The remaining weight% consisted of the solvents dimethyl sulfoxide (DMSO) and acetone, which were essential for preserving the intended viscosity. The uniformity of the mixture and dispersion of particles were facilitated with a 30- minute sonication procedure using an ultrasound sonicate machine, operating at of 30°C. Then, BFO nanoparticles were added at 30%,40%, 50% ratio and the mixes underwent continuous agitation at a temperature of 80°C for a prolonged period of 2 hours following 3 times the same process, utilizing a stirring speed of 1500 rpm. The extended duration of mixing facilitated the homogeneous dispersion of BFO particles within the PMMA matrices. After attaining the appropriate degree of uniformity and coherence, the amalgamation was cautiously transferred into a petri-dish and afterwards introduced into an oven. The petri- dish was subjected to a curing period of 48 hours, during which it was exposed to a constant temperature of 60°C. Thus, the successful synthesis of PMMA with reinforced BFO nanoparticles was achieved by the implementation of a complete method.

3. RESULT & DISCUSSION

3.1. Structural Analysis

3.1.1 Fourier Transform Infrared Spectroscopy (FTIR) Analysis

The FTIR spectrum of $\text{BaFe}_{12}\text{O}_{19}$ nanoparticles showed well defined peaks located at 513 cm^{-1} for Fe-O stretching vibration within the FeO_6 octahedral sites, confirming the formation of ferrite phase and the absorption band at 1126 cm^{-1} was assigned C-O stretching vibration, plotted in Figure 3.1, which was associated with residual carbonate from synthesis process [21], [22]. Figure 3.2 presented the FTIR spectra of three 30,40,50 wt% BFO/PMMA nanocomposites where materials structure assignments corresponding to each peak in all spectra were listed in Table 3.1 and Table 3.2. All prominent absorption peaks appear at the following wavenumber positions: 3775 cm^{-1} , 3367 cm^{-1} , 2922 cm^{-1} , 2352 cm^{-1} , 1728 cm^{-1} , 1611 cm^{-1} , 1446 cm^{-1} , 1246 cm^{-1} , 1145 cm^{-1} , 968 cm^{-1} , 850 cm^{-1} , 752 cm^{-1} , 521 cm^{-1} . The persistence of principal PMMA bands following BFO incorporation indicates that the polymer backbone remains intact [23]. The lack of new intense absorption bands suggested that composite formation primarily involves physical incorporation and interfacial interaction, rather than the formation of new covalent bonds [23], [24], [25], [26], [27]. The strong absorption peak at 1728 cm^{-1} was assigned to the ester carbonyl $\text{C}=\text{O}$ stretching vibration of PMMA, confirming the presence of the PMMA phase in all composites [24], [28]. The 2922 cm^{-1} band corresponded to C-H stretching of $-\text{CH}_2$ and $-\text{CH}_3$ groups, indicating the PMMA alkyl backbone. In the fingerprint region, absorptions at 1246 and 1145 cm^{-1} were attributed to C-O-C stretching of the PMMA ester group, while bands at 968 cm^{-1} , 850 cm^{-1} , and 752 cm^{-1} were associated with $\text{O}-\text{CH}_3/\text{C}-\text{H}$ rocking and C-C skeletal vibrations of the PMMA

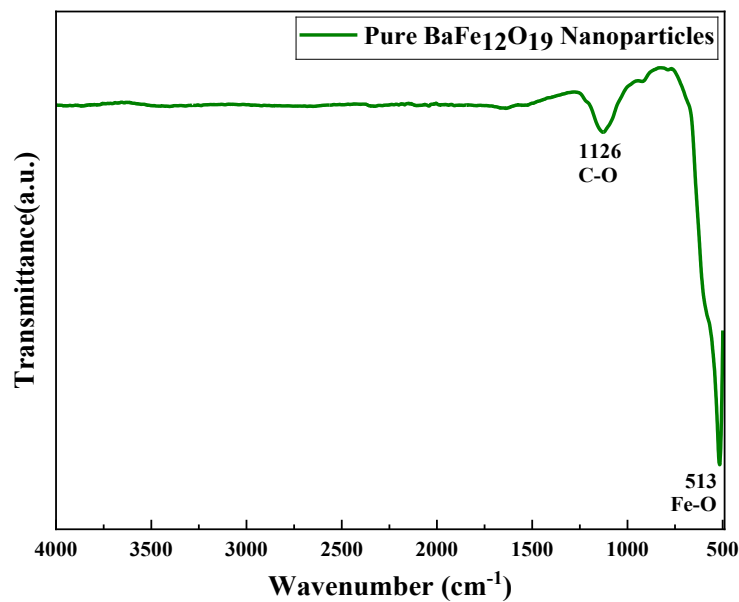


Figure 3.1. FTIR graph of pure $\text{BaFe}_{12}\text{O}_{19}$ nanoparticles after calcination

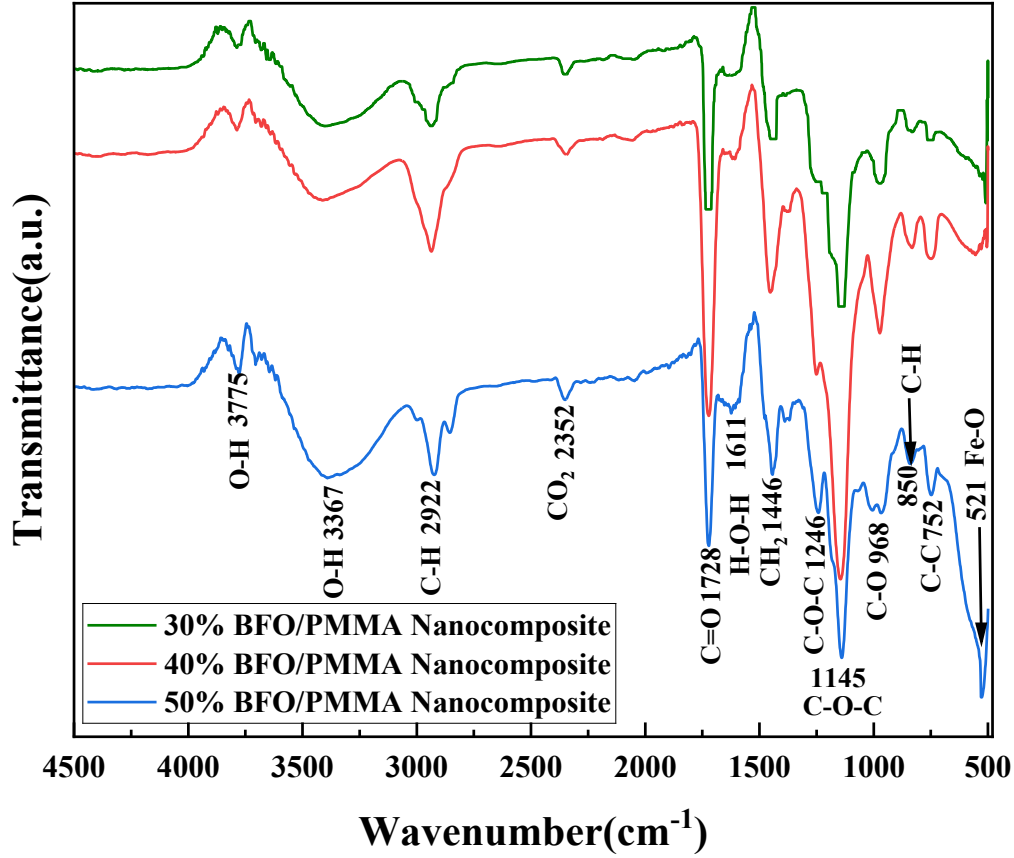


Figure 3.2. FTIR spectra for 30 wt%, 40 wt% and 50 wt% BFO/PMMA nanocomposites

chain [24], [25]. Here, The Fe–O stretching was attributed at 521 cm^{-1} in the barium ferrite lattice [26], [29]. This band confirmed BFO nanoparticles reinforcement into the PMMA matrix, and its increased intensity at higher BFO content is consistent with a greater proportion of ferrite nanoparticles in the composite [23]. The identified broad bands at 3367 cm^{-1} and 3775 cm^{-1} , together with the band near 1611 cm^{-1} , were attributed to O–H stretching and H–O–H bending arising from adsorbed moisture or surface hydroxyl groups, respectively [30]. The weak band at 2352 cm^{-1} was resulted from atmospheric adsorbed CO_2 adsorption [24], [31]. With increasing BFO content, the principal PMMA bands remain largely unchanged in intensity, indicating interfacial interactions between PMMA ester groups and BFO nanoparticle surfaces which contributed to interfacial polarization in dielectric polymer–ferrite nanocomposites [14], [27].

Table 3.1. FTIR band positions and assignments of pure $\text{BaFe}_{12}\text{O}_{19}$ nanoparticles

Wavenumber (cm^{-1})	Interpretation
513	Fe-O stretching vibration (FeO_6 octahedral sites)
1126	C-O stretching

Table 3.2. FTIR band positions and assignments BFO/PMMA nanocomposites

Wavenumber (cm ⁻¹)	Interpretation
521	Fe–O stretching
752	C–C vibration
850	C–H rocking
968	C–O stretching
1145	C–O–C asymmetric stretching
1246	C–O–C stretching
1446	CH ₃ asymmetric bending
1611	H–O–H bending
1728	C=O stretching
2352	CO ₂ asymmetric stretching
2922	C–H stretching
3367	O–H stretching
3775	O–H stretching

3.1.2. XRD Analysis

3.1.2.1. Major Peak Analysis

Figure 3.3 displays the X-ray diffraction pattern of BaFe₁₂O₁₉ nanoparticles calcined at 800 °C for 1 hour. Diffraction data were collected over a 2 θ range of 10–80° with a step width of 0.02°, utilizing Cu K α radiation (wavelength 0.1540 nm) at an operating voltage of 40 kV and a current of 30 mA. Distinct diffraction peaks were observed at 2 θ values of 24.0°, 33.2°, 34.1°, 37.1°, 40.4°, 46.7°, 49.8°, 54.1°, 57.4°, 62.3°, and 64.1°, corresponding to the (006), (107), (114), (203), (206), (0014), (220), (217), (1110), (304), and (311) crystallographic planes, respectively, as indexed in the supplied diffraction pattern. The observed reflections are in good agreement with the standard diffraction pattern of hexagonal BaFe₁₂O₁₉ reported in JCPDS card no. 00-039-1433, confirming formation of the BFO phase [8], [32].

No prominent additional diffraction peaks attributable to a major secondary crystalline phase were observed within the resolution of the measurement, indicating the successful formation of BaFe₁₂O₁₉ under the applied synthesis and calcination conditions. The sharp and well-defined diffraction peaks further demonstrate the highly crystalline nature of the synthesized powder. Among the observed reflections, the strong peak located at 2 θ = 33.15°, indexed as the (107) plane in the supplied pattern, was selected as the representative major reflection of the synthesized BFO nanoparticles. The crystallinity index was estimated to be approximately 92.82%, which is consistent with the pronounced diffraction peaks observed in the XRD pattern. Overall, the results support the formation of highly crystalline BaFe₁₂O₁₉ nanoparticles without a detectable major impurity contribution.

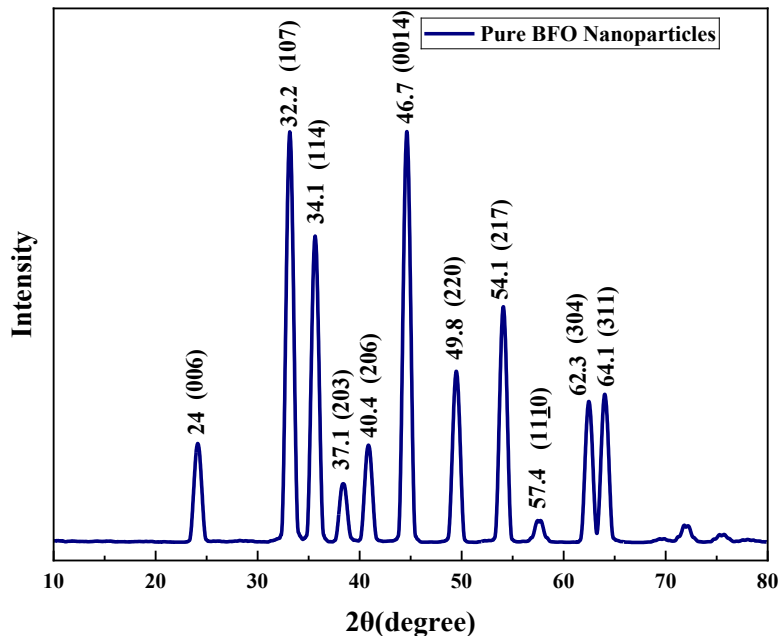


Figure 3.3. X-ray diffraction (XRD) pattern of pure BFO nanoparticles.

3.1.2.2. Williamson–Hall Analysis: Crystallite Size and Microstrain

The average crystallite size and lattice microstrain of the synthesized $\text{BaFe}_{12}\text{O}_{19}$ nanoparticles were determined using Williamson–Hall analysis according to the Uniform Deformation Model. This

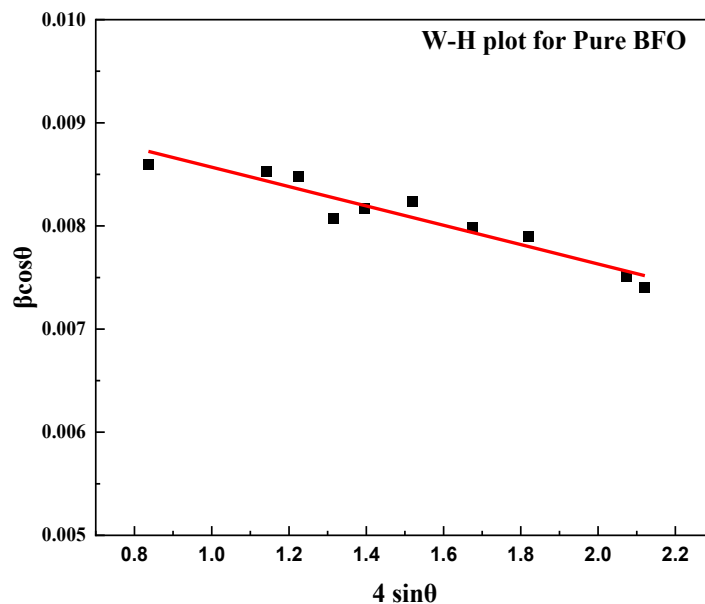


Figure 3.4. Williamson–Hall plot for pure BFO nanoparticles.

method considers diffraction-peak broadening arising from the combined contributions of finite crystallite size and strain developed within the crystal lattice. The model assumes that the strain is

uniformly distributed along the different crystallographic directions. Figure 3.4 presents the Williamson–Hall plot, which was constructed using the experimental full width at half maximum values of the major diffraction peaks. Linear fitting of this plot resulted in an average crystallite size of approximately 15.23 nm and an average microstrain magnitude of 0.94×10^{-3} . The negative slope of the fitted line indicates that the microstrain is compressive in nature, while its relatively low magnitude suggests a limited degree of average lattice distortion within the calcined $\text{BaFe}_{12}\text{O}_{19}$ crystallites [33]. The crystallite size determined from XRD represents a coherently diffracting domain and should not automatically be interpreted as the full particle size measured by electron microscopy. Thus, larger particles observed by SEM, if present, may comprise several smaller coherent crystallites or closely associated nanocrystals, a distinction commonly encountered in agglomerated ferrite powders.

Table 3.3: Structural and optical parameters of the synthesized $\text{BaFe}_{12}\text{O}_{19}$ nanoparticles obtained from XRD and UV-Vis analyses.

Sample name	2 θ of the dominant (107) peak	Crystallite size (nm)	Average microstrain ($\times 10^{-3}$)	Crystallinity index (%)	Band-gap energy (eV)
$\text{BaFe}_{12}\text{O}_{19}$	33.15	15.23	0.94	92.82	1.76

3.2. Morphological Analysis

3.2.1. SEM Analysis

SEM characterization was conducted to determine the morphology of the synthesized BFO nanoparticles (NPs), as this property significantly affects their dispersion and interfacial behavior in polymer nanocomposites. Figure 3.5 shows that the nanoparticles exhibit predominantly irregular to quasi-spherical morphologies with a relatively non-uniform distribution. Noticeable agglomeration and the formation of particle clusters are evident in several regions. The average

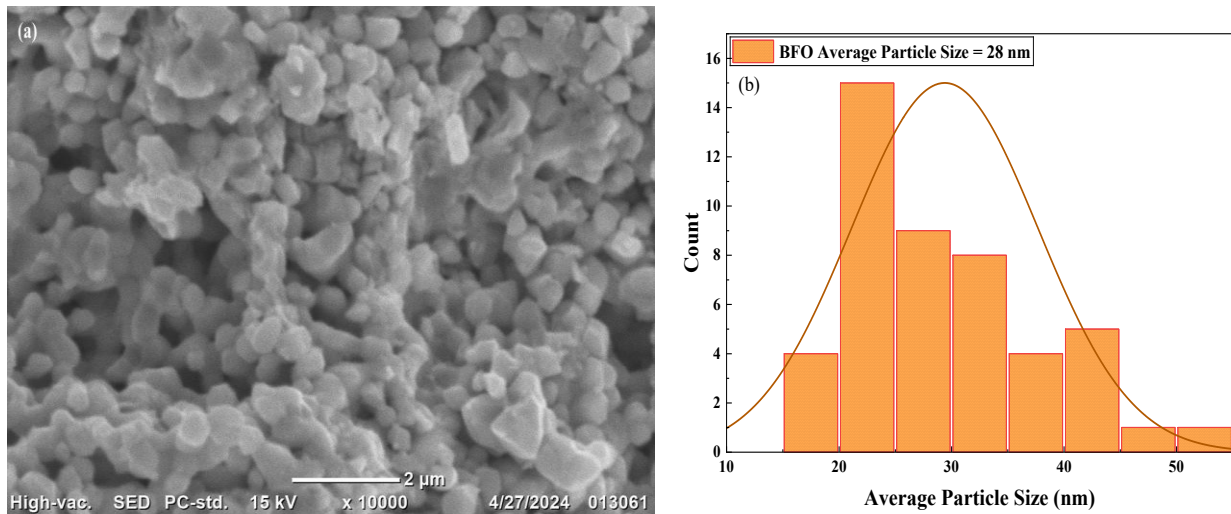


Figure 3.5. Morphology and particle size distribution of $\text{BaFe}_{12}\text{O}_{19}$ nanoparticles (a) SEM micrograph and (b) Particles size distributions of $\text{BaFe}_{12}\text{O}_{19}$ nanoparticles

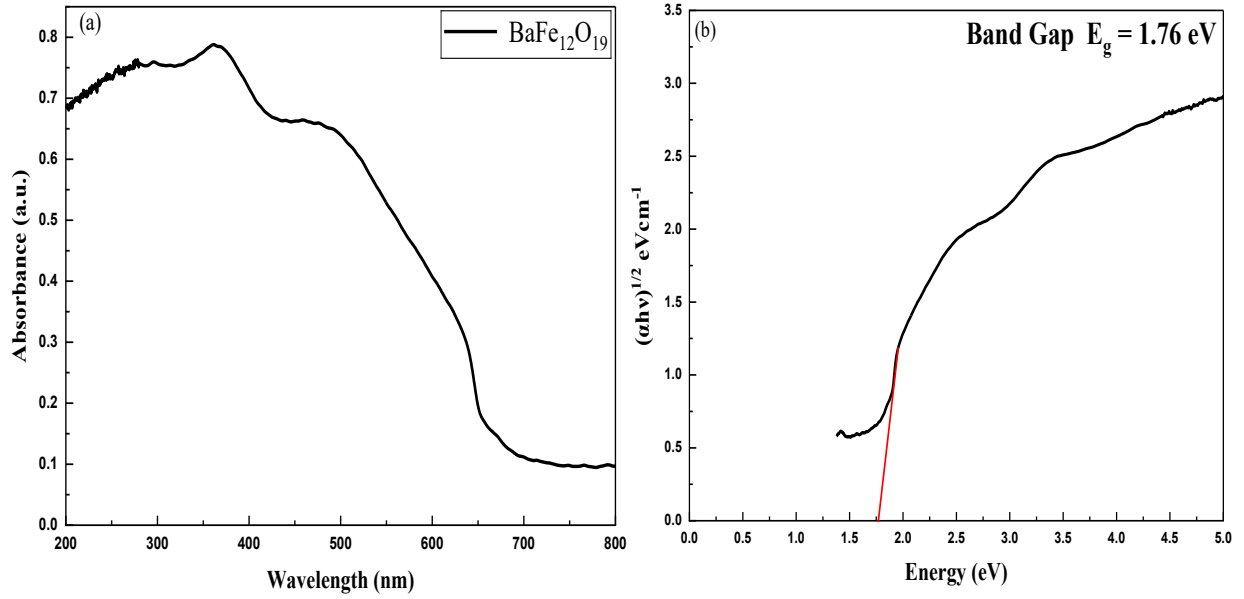


Figure 3.6. (a) UV–Vis absorbance spectrum of the synthesized BaFe₁₂O₁₉ nanoparticles (b) Tauc plot showing the indirect band-gap energy of the synthesized BaFe₁₂O₁₉ nanoparticles

particle size and particle-size distribution were measured using ImageJ and OriginLab Pro software, respectively, also shown in Figure 3.5. SEM analysis indicates an average particle size of approximately 28 nm.

3.3. UV-Vis Analysis

3.3.1. Band-gap Analysis

UV–Vis spectroscopy is employed to evaluate the optical properties of materials by measuring their absorption of ultraviolet and visible light. The optical properties of synthesized barium hexaferrite (BaFe₁₂O₁₉; BFO) nanoparticles were examined using a UV–visible spectrometer [UV-2600, SHIMADZU]. The absorbance spectrum, presented in Figure 3.6 (a), demonstrates strong absorption in the ultraviolet region and across a broad range of the visible region. The band-gap energy was analyzed from the absorbance data using the Tauc plot method [34]:

$$(\alpha h\nu)^n = A(h\nu - E_g)$$

Here, α denotes the absorption coefficient, h is Planck's constant, ν represents the photon frequency, A is the proportionality constant, and E_g is the optical band-gap energy. The γ value can be 1/2 for indirect allowed transitions and $\gamma = 2$ for direct allowed transitions. Figure 3.6 (b) shows the indirect band-gap energy of the synthesized BaFe₁₂O₁₉ nanoparticles, determined to be 1.76 eV for the indirect allowed transition. This value agrees closely with the band-gap energies of approximately 1.77 eV and 1.78 eV previously reported for BaFe₁₂O₁₉ [8], [32]. Oxygen vacancies can promote the partial reduction of Fe³⁺ to Fe²⁺ to maintain charge neutrality and thereby modify the local electronic environment near the absorption edge [21]. Furthermore, lattice strain and structural disorder in the synthesized nanoparticles, as indicated by the Williamson–Hall

and XRD analyses, may produce localized band-edge states and contribute to the small variation between the present band-gap energy and the previously reported values [35].

3.3.2. Photocatalytic Performance Evolution

The photocatalytic activity of BFO/PMMA nanocomposites was assessed by monitoring the photodegradation of methylene blue (MB) under light irradiation for 60 minutes, as illustrated in Figure 3.7. The absorption spectrum of the MB solution displayed two characteristic peaks at approximately 615 nm and 665 nm. Photodegradation was tracked at 665 nm, which corresponds to the main visible absorption band of MB. The absorbance at 665 nm progressively decreased with increasing irradiation time, indicating ongoing decolorization of the dye solution.

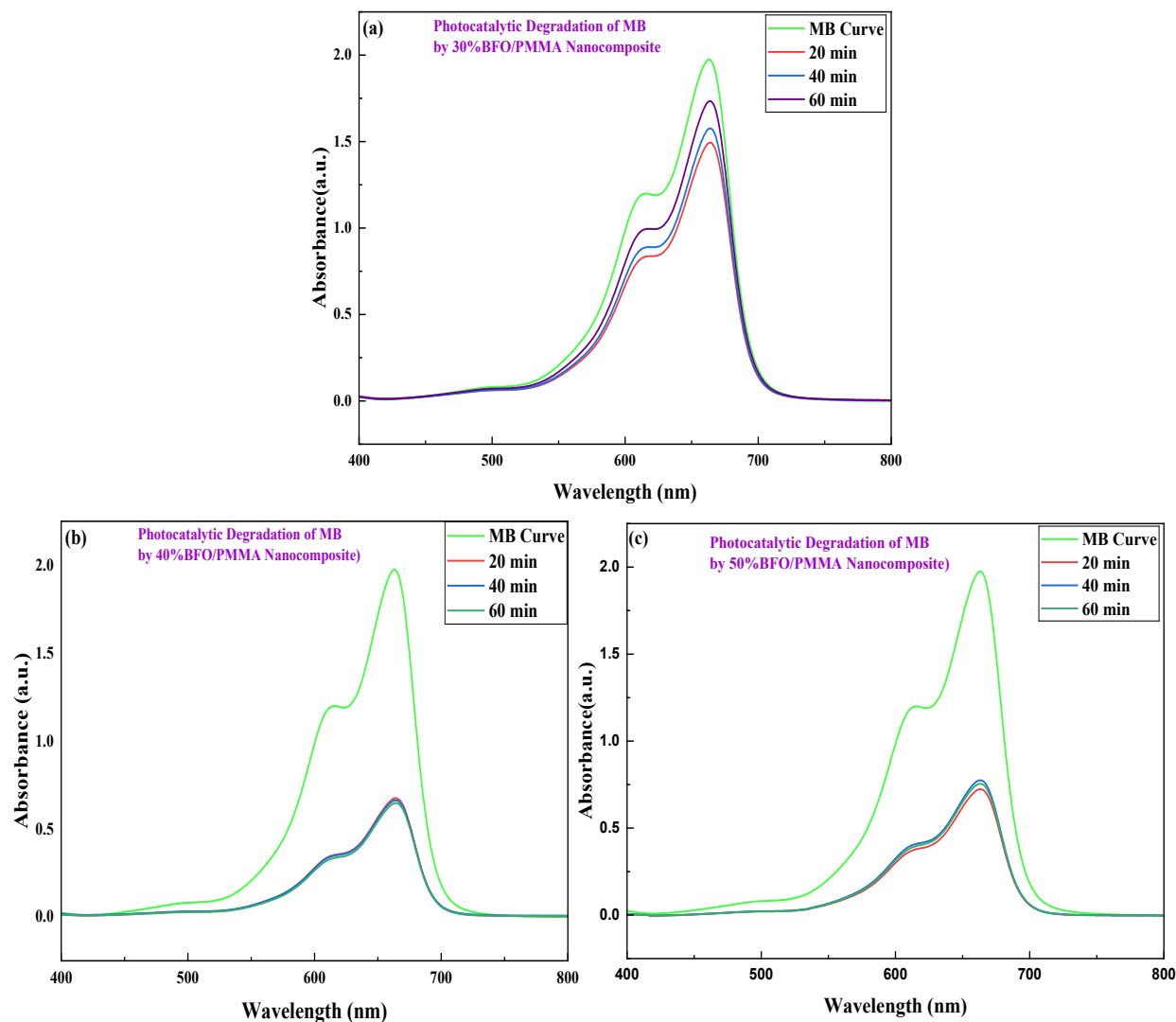


Figure 3.7. Time-dependent absorbance spectra of MB in the presence of (a) 30 wt% BFO/PMMA, (b) 40 wt% BFO/PMMA, and (c) 50 wt% BFO/PMMA nanocomposites.

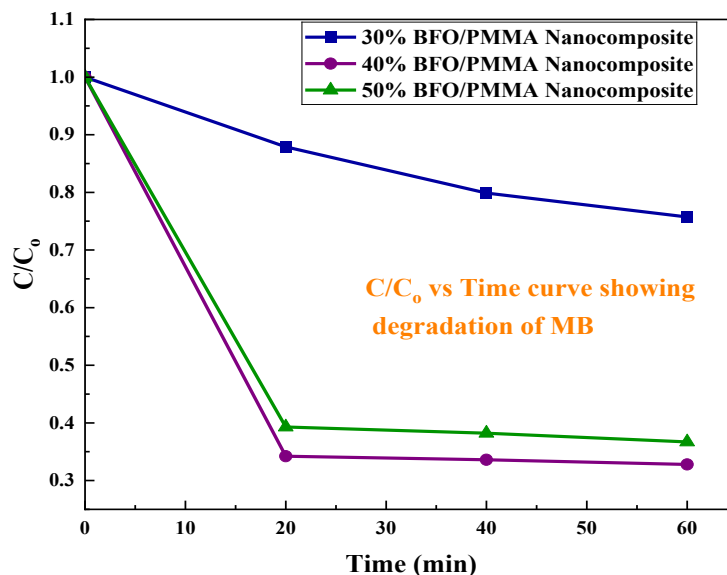


Figure 3.9. Plot of (C/C_0) versus irradiation time showing the photodegradation of methylene blue over the 30 wt%, 40 wt%, and 50 wt% BFO/PMMA nanocomposites under light irradiation.

Photocatalytic activity varied systematically with BFO reinforcement, as reflected by the decline in the characteristic MB absorbance. The 30 wt% BFO/PMMA nanocomposite showed a modest reduction, whereas the 40 wt% composition produced the most pronounced attenuation of the characteristic MB band. At 50 wt%, the composite retained substantial photocatalytic activity, but the extent of absorbance reduction remained slightly lower than that observed for the 40 wt% sample. This improvement associated with the increased photoactive BFO surface area and the greater availability of accessible photocatalytic sites [36], [37], [38]. This observation is consistent

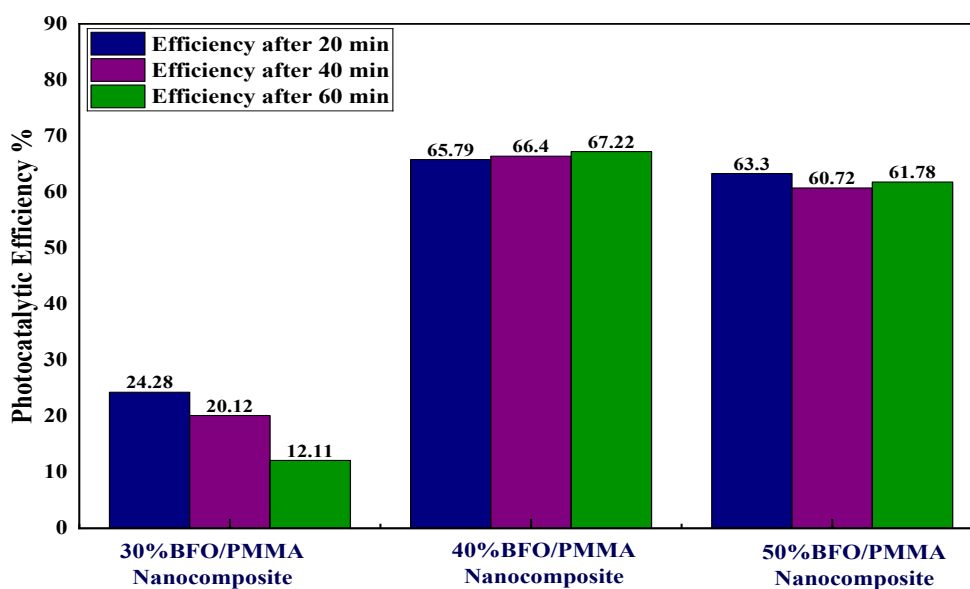


Figure 3.8. Photocatalytic degradation efficiencies of the 30, 40, and 50 wt% BFO/PMMA nanocomposites.

with previous findings that the metal-oxide particles reinforcement into PMMA polymer matrices enhances dye removal of photocatalytic system, attributing enhanced surface properties, increased pollutant adsorption, and greater accessibility of active species through π - π stacking and ionic interaction [36], [38], [39]. Under light irradiation, BFO acted as the photoactive semiconductor and generates electron-hole pairs. The synthesized BFO nanoparticles exhibited a band-gap energy of 1.76 eV, which enabled visible-light absorption and the subsequent formation of reactive oxygen species, including hydroxyl radicals ($\cdot\text{OH}$) and superoxide radicals ($\text{O}_2^{\cdot-}$), through surface redox reactions [38], [40], [41]. These reactive species attacked the conjugated chromophoric structure of MB and promote dye decolorization. In the BFO/PMMA nanocomposites, PMMA primarily serves as a supporting and immobilizing matrix, whereas BFO provided the active semiconductor sites required for photon absorption and interfacial redox reactions [36], [39]. Therefore, the improved degradation performance of the 40 wt% composite was attributed to an effective photoactive BFO content and the availability of photocatalytically active sites under light irradiation.

3.3.2.1. Effect of BFO nanoparticle reinforcement

A clear composition-dependent trend emerged over the 60 min irradiation period. At 20, 40, and 60 min, degradation efficiencies were 12.11%, 20.12%, and 24.28% for the 30 wt% composite, 65.79%, 66.40%, and 67.22% for the 40 wt% composite, and 60.72%, 61.78%, and 63.30% for the 50 wt% composite, respectively. As summarized in Figures 3.8 and 3.9, increasing the BFO content from 30 to 40 wt% produced a pronounced improvement in degradation efficiency, whereas a further increase to 50 wt% resulted in a modest decline. Thus, among the investigated compositions, the 40 wt% BFO/PMMA nanocomposite exhibited the highest measured photocatalytic performance. Increasing the BFO content from 30 to 40 wt% increased the effective photoactive ceramic fraction and the number of BFO/PMMA interfaces. These interfaces supported BFO dispersion and provide additional sites for photon absorption, electron-hole generation, and surface redox reactions, consistent with reported photocatalytic behavior of BFO-containing polymer composites [36], [39].

For determining photocatalytic degradation efficiency, the efficiency equation was calculated as follows:

$$\text{Efficiency (\%)} = \frac{C_o - C_t}{C_o} \times 100$$

Where C_o is the initial concentration and C_t is the dye concentration at irradiation time 't'.

The degradation efficiency provided a quantitative measure of photocatalytic performance over the irradiation period and enabled direct comparison between the investigated nanocomposites [40]. Furthermore, quantifying the degradation efficiency of photocatalytic activity is essential for a precise assessment of the photocatalyst's performance over time. [42]. If degradation efficiency is not considered, the reported photocatalytic activity efficiency may be misleading and fail to reflect the actual performance of the nanocomposites in practical applications [43].

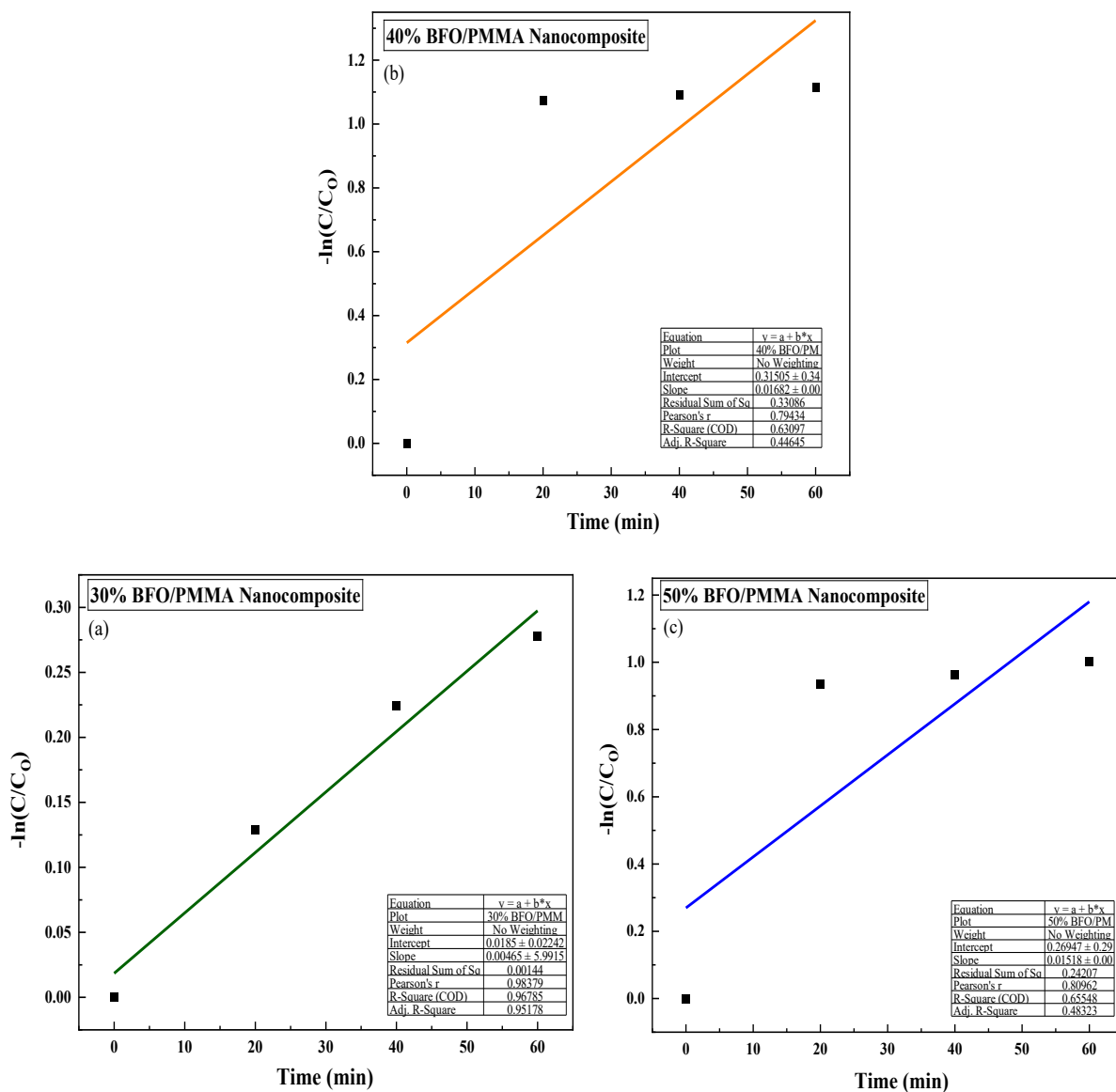


Figure 3.10: Pseudo-first-order kinetic plots of $\ln(C/C_0)$ versus irradiation time for MB degradation in the presence of (a) 30 wt% (b) 40 wt% and (c) 50 wt% BFO/PMMA nanocomposites.

3.3.2.2. Degradation kinetics

The degradation kinetics of MB in the presence of the BFO/PMMA nanocomposites were analyzed using the pseudo-first-order model described by equation:

$$\ln\left(\frac{C_t}{C_0}\right) = -kt$$

Here, C_0 is the initial concentration of dye, C_t represents the concentration of dye at time 't', and 'k' is the first order rate constant, which was calculated from the slope of the straight line obtained

by plotting $-\ln(C/C_0)$ vs time, in Figure 3.10(a,b,c) [37], [38]. The fitted pseudo-first-order rate constants (k) and the corresponding coefficients of determination (R^2) were summarized in Table 3.4. Among the investigated compositions, the 40 wt% BFO/PMMA nanocomposite exhibited the highest apparent rate constant. The apparent rate constant followed a clear composition-dependent trend, increasing from $(0.00465 \pm 5.991) \text{ min}^{-1}$ at 30 wt% BFO/PMMA nanocomposite to 0.01682 min^{-1} at 40 wt% BFO/PMMA nanocomposite, followed by a slight decrease to 0.01518 min^{-1} at 50 wt% BFO/PMMA nanocomposite. This variation in ' k ' was consistent with the corresponding degradation efficiencies, where the substantial improvement from 30 wt% to 40 wt% BFO nanoparticles reflected the enhanced photocatalytic performance of the BFO/PMMA nanocomposite, while the modest reduction at 50 wt% corresponded to its slightly lower degradation efficiency relative to the 40 wt% composition. Nevertheless, the R^2 values of 0.96785, 0.63097, and 0.65548 indicated strong linear agreement for the 30 wt% nanocomposite and only moderate linear agreement for the 40 wt% and 50 wt% nanocomposites. Accordingly, the calculated rate constants were best interpreted as comparative kinetic indicators under the present test conditions rather than as definitive evidence of strict pseudo-first-order behavior.

Table 3.4. Pseudo-first-order kinetic parameters and degradation efficiencies of the BFO/PMMA nanocomposites.

Sample	C/C_0 after 20 min	C/C_0 after 60 min	$K (\text{min}^{-1})$	R^2	Efficiency (%) After 20 min	Efficiency (%) After 60 min
30% BFO/PMMA Nanocomposite	12.11353	24.27775	0.00465 ± 5.991	0.96785	12.11353	24.27775
40% BFO/PMMA Nanocomposite	65.78814	67.21595	0.01682 ± 0.00	0.63097	65.78814	67.21595
50% BFO/PMMA Nanocomposite	0.3928	0.36695	0.01518 ± 0.00	0.65548	60.71972	63.30461

3.3.2.3. Mechanism

In general, the photocatalytic process involves three principal steps: light absorption, generation of electron-hole pairs, and interfacial redox reactions mediated by charge transfer [38], [41]. Under light irradiation, BFO serves as the photoactive component in the BFO/PMMA nanocomposite. In this role, electrons are promoted from the valence band (VB) to the conduction band (CB), resulting in the generation of electron-hole pairs [38]. PMMA functions as a supporting matrix, and its carbonyl and ester groups can interact with the BFO surface, assisting nanoparticle dispersion and improving the accessibility of photocatalytically active sites [36], [39]. The photogenerated charge carriers then migrate to the BFO surface, where CB electrons reduce

adsorbed oxygen to superoxide radicals ($O_2^{\cdot-}$), while VB holes react with H_2O or OH^- to generate hydroxyl radicals ($\cdot OH$). Protonation of $O_2^{\cdot-}$ produces hydroperoxyl radicals ($HO_2^{\cdot}$), which can subsequently form H_2O_2 and additional $\cdot OH$. The combined action of photogenerated holes, $O_2^{\cdot-}$, and $\cdot OH$ attacks the conjugated chromophoric structure of MB, resulting in decolorization and the generation of intermediate degradation products. Based on the reported BFO photocatalytic pathways [38], [40], [41], the proposed reactions are as follows:

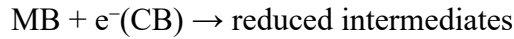
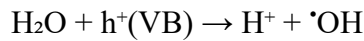
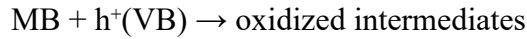
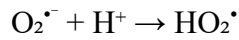
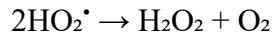
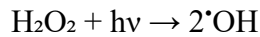
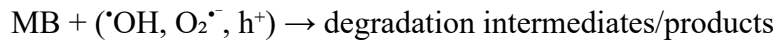
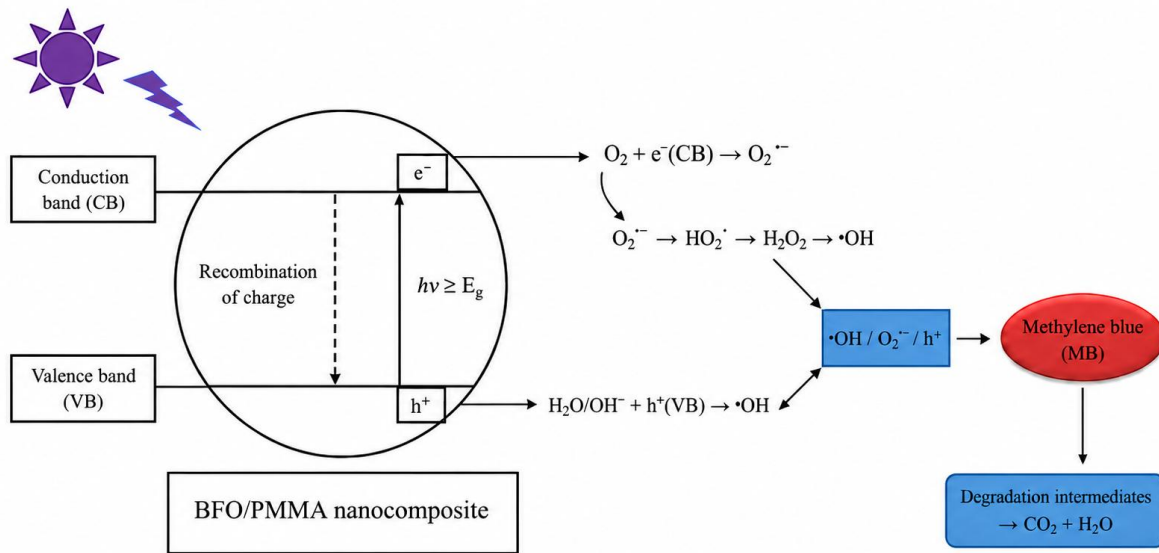


Figure 3.3.2.3.1. Illustration of proposed photocatalytic degradation mechanism of MB by BFO/PMMA nanocomposite

3.4. Piezoelectric Analysis:

The piezoelectric coefficient (d_{33}) of BFO/PMMA nanocomposites were measured with YE2730A d_{33} Meter (Sinocera). This meter is able to measure the d_{33} values over a wide range, at better resolution with a high degree of reliability. As previously discussed, the incorporation of ceramic nanoparticles into polymer matrices could enhance the piezoelectric response by promoting electroactive-phase formation and interfacial [44], [45], [46]. Figure 3.11 illustrated that BFO/PMMA nanocomposites enhanced d_{33} values 36 pC/N, 37 pC/N, 40 pC/N for gradually enhancing 30wt%, 40wt%, 50wt% BFO nanoparticles reinforcement into polymer matrices, respectively. Notably, BFO/PMMA nanocomposite evaluated higher piezoelectric response as reinforcement increased 30wt% to 50wt%, in amount.

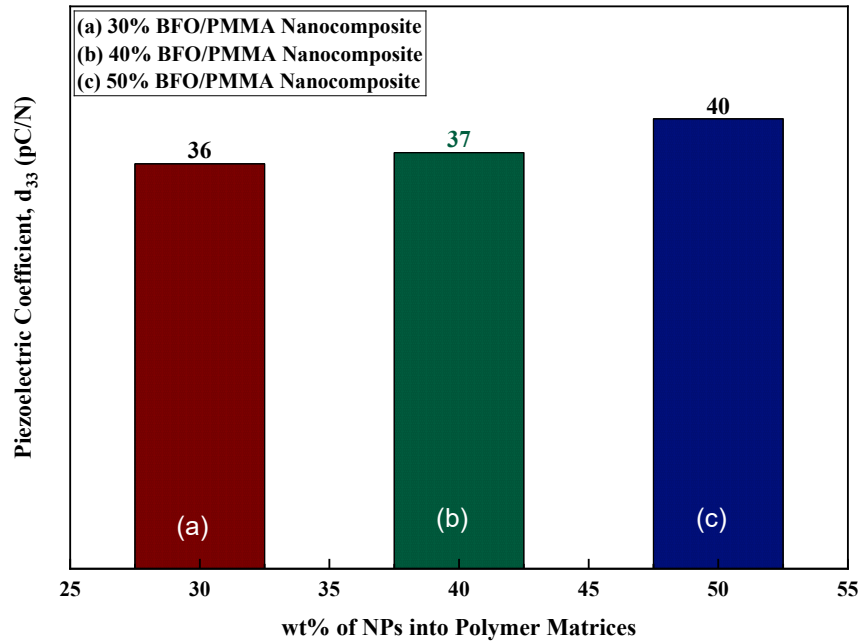


Figure 3.11. Comparison of piezoelectric coefficient (d_{33}) of BFO/PMMA nanocomposites for (a) 30wt%, and (b) 40wt% (c) 50wt% BFO NPs reinforced into Polymer matrices.

3.5. Electrical Properties Analysis

3.5.1. Dielectric Studies

Complex impedance spectroscopy is employed to investigate the electrical properties of electro-materials. This technique allows for effective analysis of the contributions of grains and grain boundaries to dielectric properties, including dielectric constant, dielectric loss, and conductivity. The frequency dependence of the dielectric constant (ϵ'), dielectric loss ($\tan\delta$), and conductivity (σ_{ac}) of the nanocomposites with varying BFO contents are presented in Figures 3.12, 3.13, and 3.14. As shown

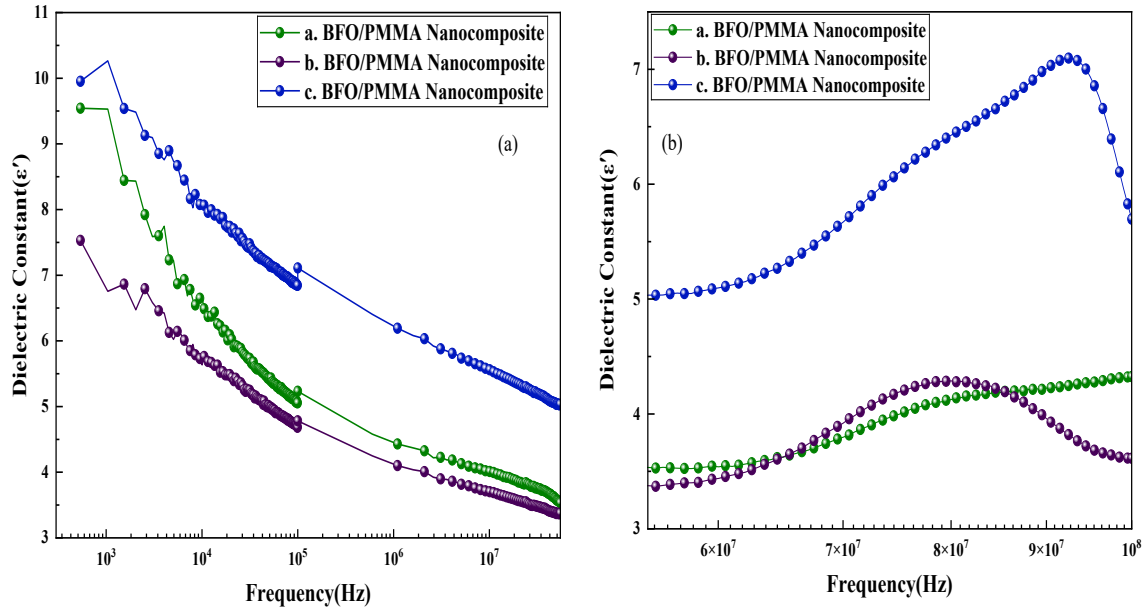


Figure 3.12. Dielectric constant of BFO/PMMA nanocomposites at (a) 40-55MHz, (b) 55MHz-100MHz at room temperature

in Figure 3.12(a), the dielectric constant values of all nanocomposites were dependent on frequency over the considered range 40Hz-100MHz. The dielectric constants of nanocomposites were varied with the BFO nanoparticles reinforcement [47], [48]. At 40 Hz, the dielectric constants of the 30, 40, 50 wt% BFO/PMMA nanocomposites were maximum approximately 9.54, 7.53 and 10.26, respectively. This improvement is attributed to higher dielectric constant of BFO nanoparticles relative to PMMA

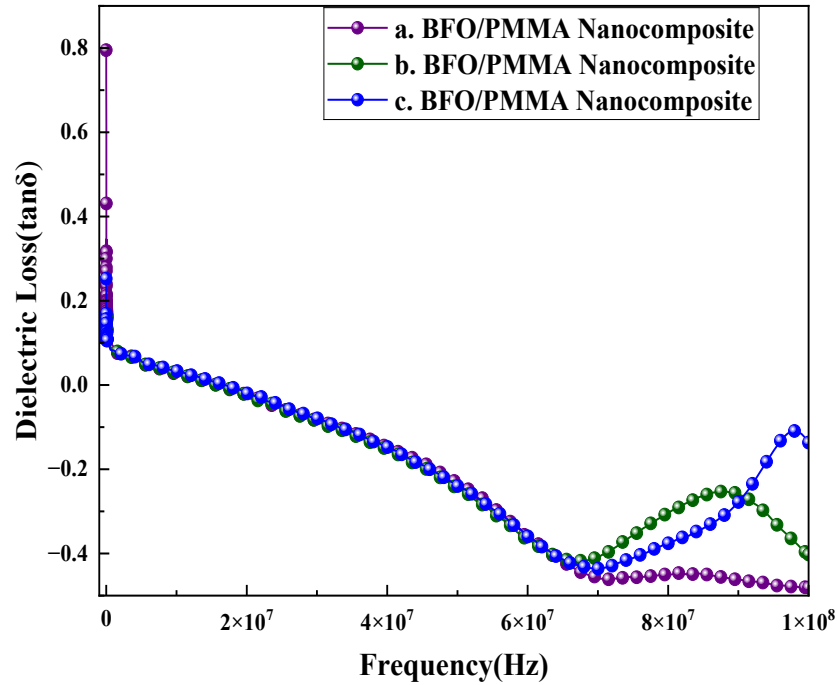


Figure 3.13. Dielectric loss of BFO/PMMA nanocomposites at 40-100MHz frequency region at room temperature

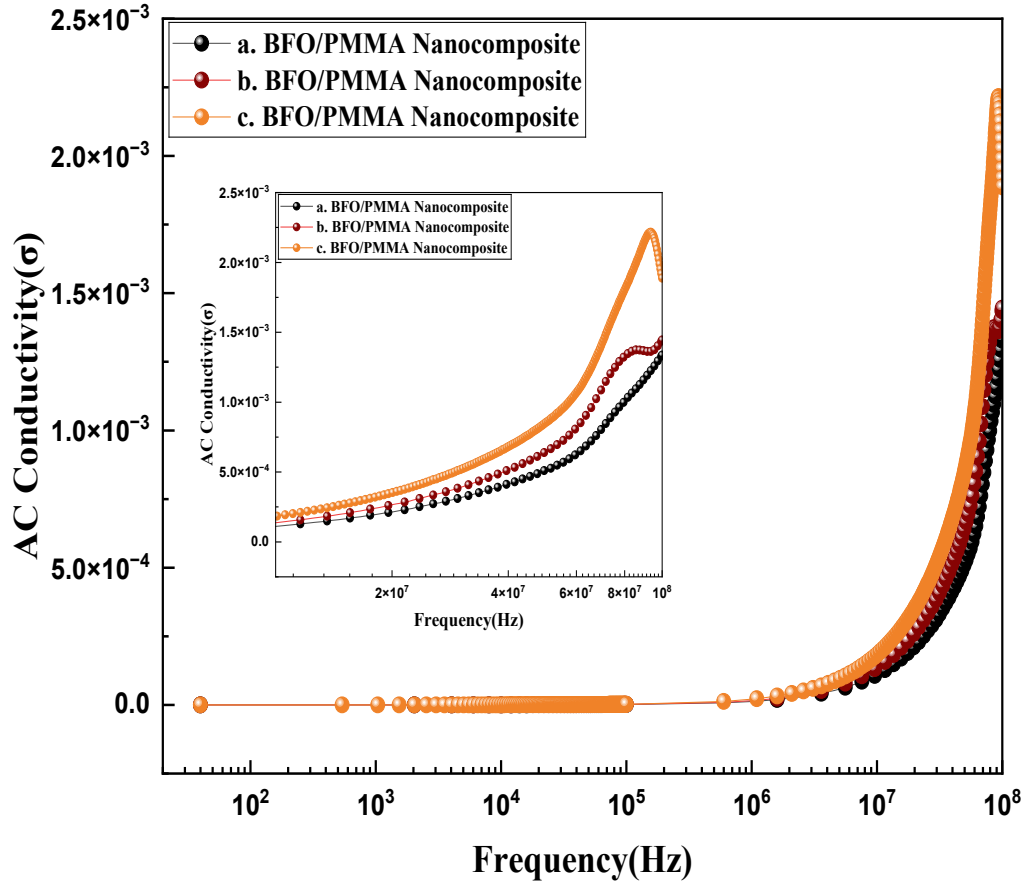


Figure 3.14. Frequency dependence of Ac conductivity at room temperature for for (a) 30wt%, and (b) 40wt% (c) 50wt% BFO/PMMA Nanocomposites

matrix, and the Maxwell-Wagner-Sillars (MWS) interfacial polarization effect [49], [50], [51]. MWS interfacial polarization was prominent at low frequency region due to its relatively long relaxation time [50]. At 10 kHz, the corresponding values of 30, 40, and 50 wt% BFO/PMMA nanocomposites were approximately 6.3, 5.7 and 8.0, whereas gradually decreased 4.0, 3.8 and 5.6 for 10 MHz, respectively. As measurement frequency increased, dielectric constant decreased from 40 Hz to the intermediate frequency region. In contrast, a slight anomalous increase was observed for BFO/PMMA nanocomposites at high frequency range 90-93 MHz, where dielectric constant of 50 wt% BFO/PMMA nanocomposite was approximately 7.10, compared with approximately 4.3 and 4.2 for 30 wt% and 40 wt% BFO reinforced nanocomposites, respectively. As shown in the inset of Figure 3.13, the low frequency dielectric loss (loss tangent) of nanocomposites showed values of approximately 0.79, 0.20 and 0.25 for 30, 40, 50 wt% BFO/PMMA nanocomposites, respectively. A comparable reduction in dielectric loss with increasing frequency is evident, as shown in Table 3.5 [52]. The findings indicate that incorporating BFO nanoparticles into PMMA matrices leads to significant modifications in the dielectric performance of the resulting nanocomposites [48]. At room temperature, the ac conductivity of all three nanocomposites exhibited frequency dependence, with conductivity increasing in the high frequency range. As summarized in Table 3.5, the conductivity of 30, 40, 50 wt% BFO/PMMA nanocomposites in the low frequency region was ($\approx 7.26 \times 10^{-4}$ S/m), ($\approx 5.69 \times 10^{-4}$ S/m) and ($\approx 9.59 \times 10^{-4}$

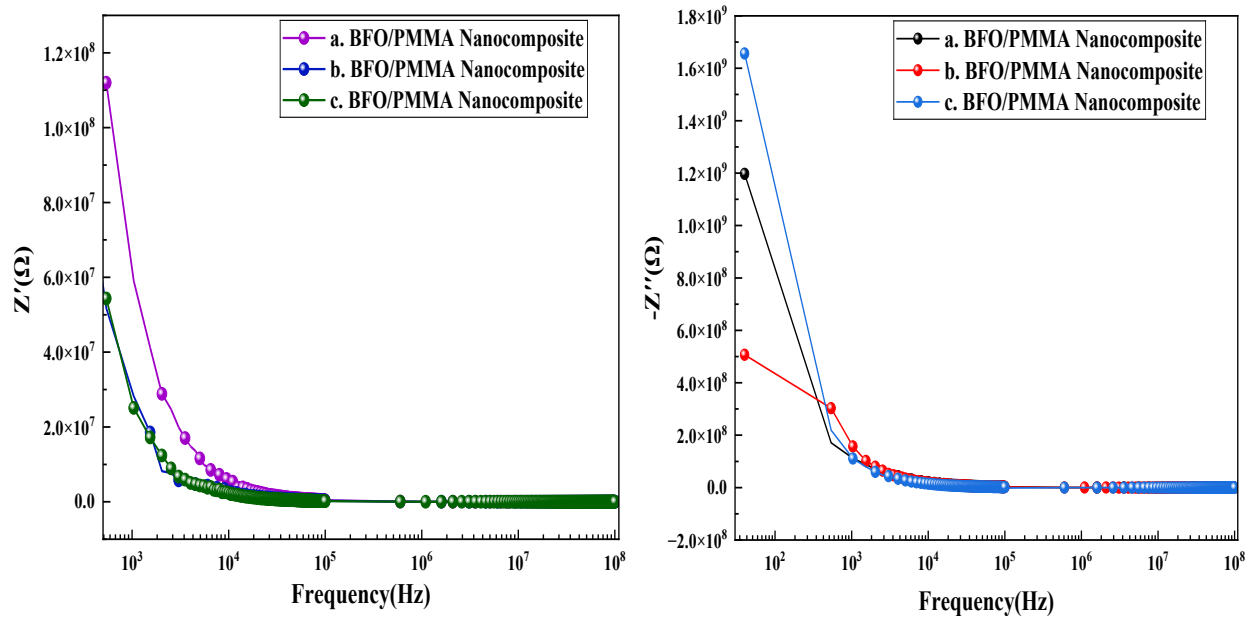


Figure 3.15. Variation of real part of impedance Z' , and imaginary part of impedance Z'' with frequency at a room temperature

S/m), respectively, whereas increased to $(\approx 1.45 \times 10^{-3} \text{ S/m})$, $(\approx 1.34 \times 10^{-3} \text{ S/m})$ and $(\approx 2.22 \times 10^{-3} \text{ S/m})$ in the 55-100MHz high frequency region. Here, conductivity increases with frequency for all BFO/PMMA nanocomposites. The ac conductivity remains low in the low-frequency region and exhibits pronounced dispersion at higher frequencies [53]. The variation in conductivity among the

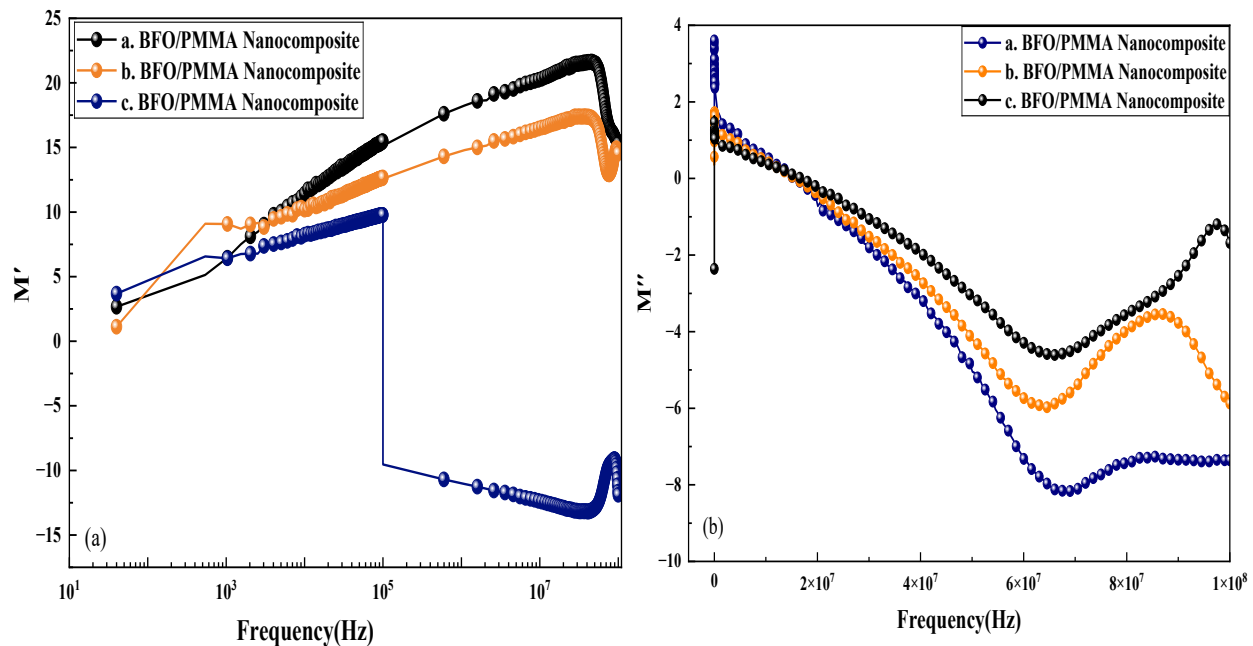


Figure 3.18. Variation of (a) real part modulus (M') and (b) imaginary part modulus (M'') with frequency of BFO/PMMA nanocomposites at room temperature.

BFO/PMMA nanocomposites was associated with interfacial polarization and localized charge transport across the BFO/PMMA interfaces [49], [51].

3.5.2. Impedance analysis

Complex impedance spectroscopy was employed to investigate the electrical properties of BFO/PMMA nanocomposites across the frequency range of 40 Hz to 100 MHz at room temperature. [54], [55]. Complex electric modulus analysis was conducted to examine the frequency dependence of the electrical response in BFO/PMMA nanocomposites [56]. In our analysis, we considered (1) series resistance (R_s), representing overall bulk resistance, is contributions of current collector, bulk electrode material, electrolyte and separator mediated resistance, (2) polarization resistance (R_p), which consisted of interfacial resistance (mainly SEI-mediated resistance, R_{SEI}), charge transfer resistance (R_{ct}), (3) Constant Phase Element (CPE_1), was placed

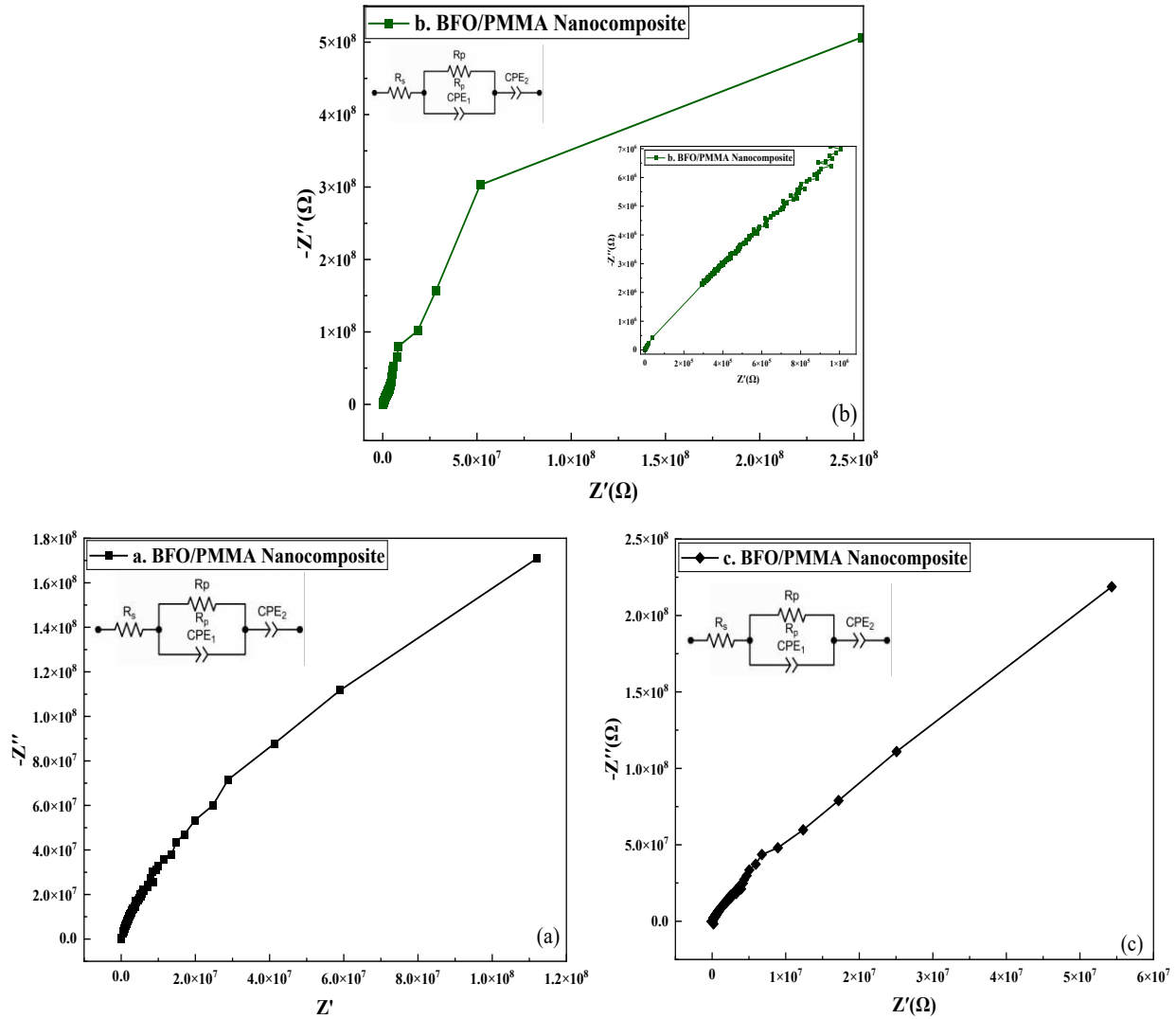


Figure 3.16. Complex impedance plots for the ($Z''=f(Z')$) for the (a) 30wt% BFO/PMMA, (b) 40 wt% BFO/PMMA, and (c) 50 wt% BFO/PMMA nanocomposites at 40Hz – 100MHz

with parallel with R_p , and (4) CPE_2 accounted for low frequency diffusive response [57], [58]. Figure 3.15(a)-(b) presents the variation in real impedance (Z') and imaginary impedance (Z'') within the high-frequency range (40 Hz to 100 MHz). The magnitude of Z' decreases and the values converge as frequency increases. The observed decrease in Z' for all three nanocomposite samples is attributed to increased alternating current (ac) conductivity at higher frequencies. The convergence of impedance responses at high frequencies is ascribed to the release of space charges, which leads to reduced barrier properties in the nanocomposite [54], [55]. It was observed that the initial value of Z'' might be indicated as high then it decreased as the frequency increased for all three nanocomposite samples [56]. The 50 wt% BFO/PMMA nanocomposite exhibited the higher imaginary Z'' value in the low frequency region, which decreased gradually over higher frequency region because of space-charge polarization and interfacial polarization [54], [56]. The plot of Z'' versus Z' (Nyquist plot) at low to high frequency range was shown in Figure 3.16, which expressed depressed and incomplete arc-like trajectories confirmed deviation from Debye behavior with a

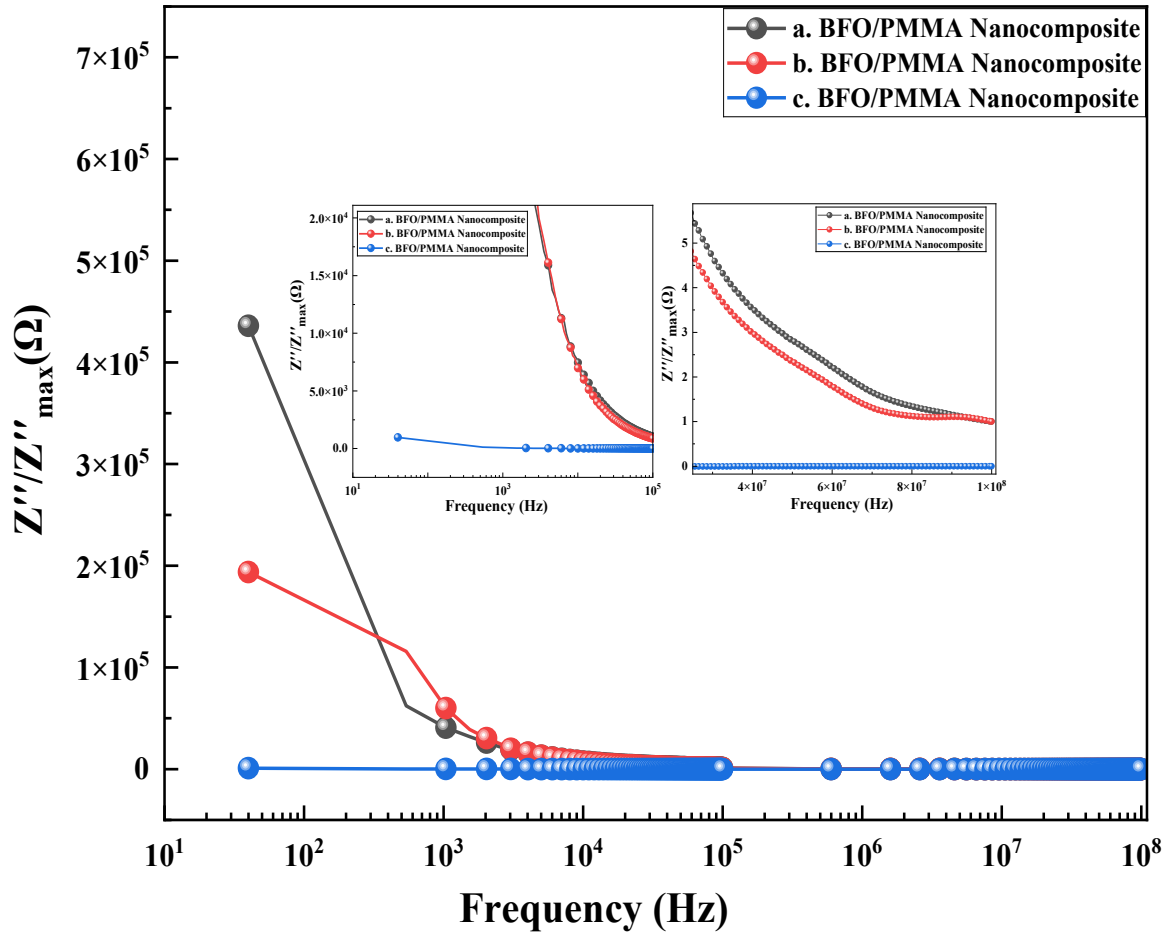


Figure 3.17. Imaginary part of impedance ($Z''/Z''_{\max}$) with frequency of BFO/PMMA nanocomposites at room temperature.

distribution of relaxation time. This behavior was associated with heterogeneous interfaces and multiple polarization process of nanocomposites. The plotted imaginary part of impedance $Z''/Z''_{\max}$ as a function of frequency for all BFO/PMMA nanocomposites exhibited a broad relaxation response, indicating deviation from ideal-Debye type relaxation, plotted in Figure 3.17. This behavior attributed to the heterogeneous BFO/PMMA interfaces and multiple polarization process [54], [56].

3.5.3. Modulus analysis

Figure 3.18(a) illustrated the frequency dependence of the real part of electric modulus M' for all BFO/PMMA nanocomposites. The 30 wt% BFO/PMMA nanocomposite exhibited a higher positive magnitude of M' compared to the 40 wt% BFO/PMMA nanocomposite across most of the frequency range, whereas the 50 wt% BFO/PMMA nanocomposite showed a distinct sign reversal in the high frequency region. In the very low frequency region, the magnitudes of M' were

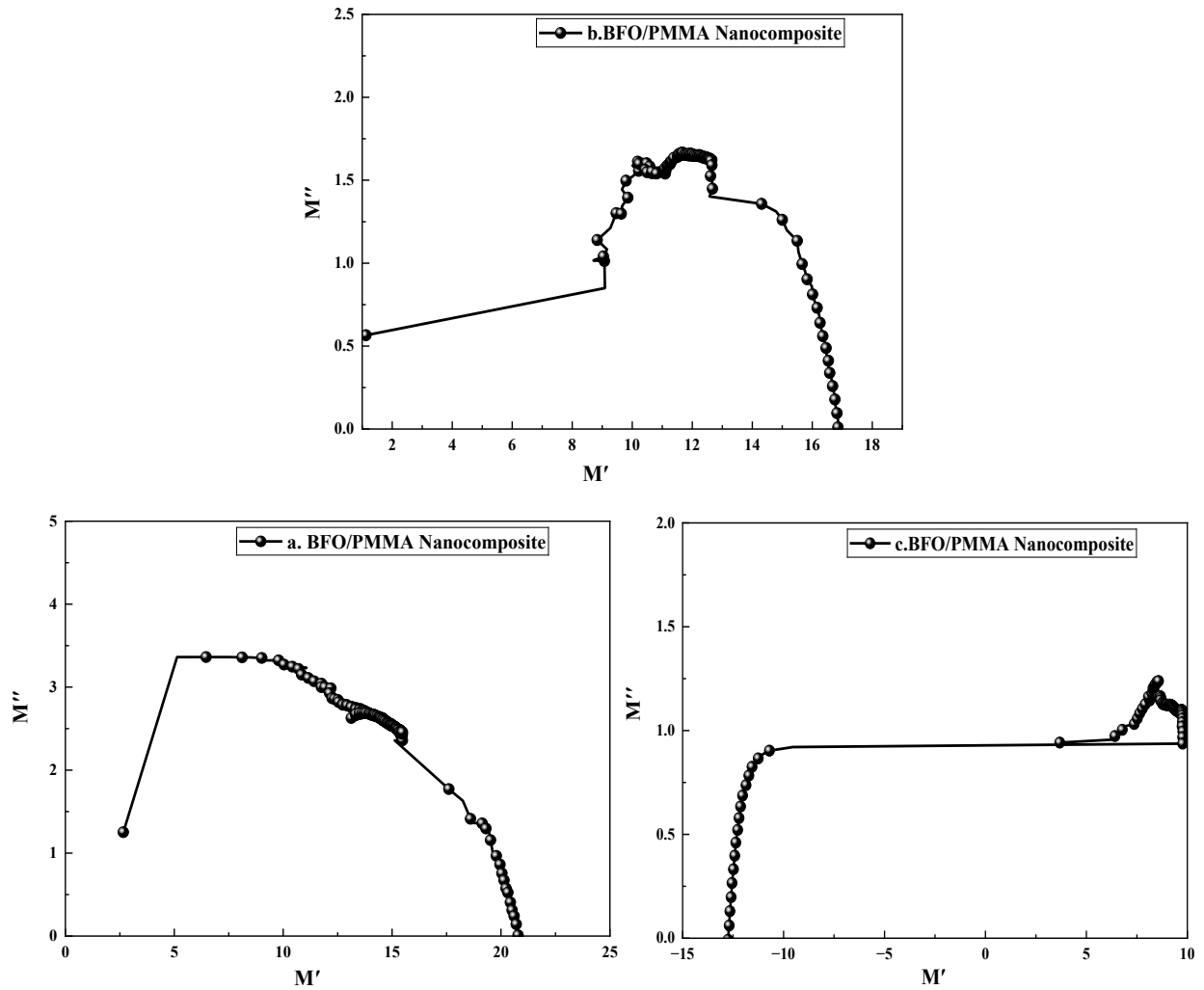


Figure 3.19. Nyquist plots of imaginary part modulus M'' vs real part modulus M' of (a) 30 wt% BFO/PMMA, (b) 40 wt% BFO/PMMA, and (c) 50 wt% BFO/PMMA nanocomposites

comparatively low, which evaluated a lower contribution of electrode effect. It exhibited continuous frequency dependent dispersion, suggested short-range mobility of charge carriers. With the increase of frequency, the M' attained higher values in the intermediate-frequency region and decreased gradually at high frequency region for 30 wt% and 40 wt% BFO/PMMA nanocomposites, while 50 wt% BFO/PMMA nanocomposite changed to negative M' values. Besides, a pronounced dispersion was observed at high frequency region, which indicated the contribution of localized charge-carrier relaxation process within nanocomposite samples [56].

Figure 3.18(b) represented the frequency dependence variation of imaginary part of electric modulus M'' for 30, 40 and 50 wt% BFO/PMMA nanocomposites. All nanocomposites exhibited a broad negative peak in M'' , indicating frequency dependent dielectric relaxation. The higher magnitude M'' response of BFO/PMMA nanocomposite samples, together with the composition dependent conductivity, suggested composition dependent relaxation and charge transport behavior [56]. Figure 3.19. showed M'' vs M' graph for all BFO/PMMA nanocomposites was deviation from Debye behavior and depressed semi-circles confirmed the presence of modest electrical relaxation phenomenon in the materials [54], [56].

Table 3.5. Summary of the dielectric constant, dielectric loss, and AC conductivity of BFO/PMMA nanocomposites

Sample	Frequency range	ϵ'	$\tan\delta$	$\sigma_{ac} \times 10^{-3}$ (S/cm)
a. BFO/PMMA Nanocomposite	40–1kHz	9.54	0.79	0.01
	1k–100 kHz	8.44	0.62	0.02
	100k–1 MHz	5.20	0.16	0.04
	1M–110 MHz	4.42	0.09	1.2
b. BFO/PMMA Nanocomposite	40–1kHz	7.53	0.18	0.013
	1k–100 kHz	6.86	0.21	0.03
	100k–1 MHz	4.75	0.12	0.05
	1M–110 MHz	4.28	0.08	1.5
c. BFO/PMMA Nanocomposite	40–1kHz	10.26	0.25	0.018
	1k–100 kHz	9.54	0.20	0.04
	100k–1 MHz	6.85	0.11	0.06
	1M–110 MHz	7.10	0.08	2.25

CONCLUSION

This study clarified how BFO nanoparticles concentration enhanced the functional behavior of solution-cast BFO/PMMA nanocomposites. Structural analysis as FTIR and XRD established the formation of crystalline hexagonal $\text{BaFe}_{12}\text{O}_{19}$ and its successful incorporation into PMMA, while SEM ensured nanoparticles morphology. Increasing the BFO nanoparticles reinforcement from 30 wt% to 50 wt% increased the proportion of the polarizable ceramic phase and the density of heterogeneous interfaces, resulting in higher dielectric and better piezoelectric response with notably high frequency-dependent ac-conductivity. The 40 wt% BFO/PMMA nanocomposite demonstrated higher photocatalytic activity, whereas 50 wt% BFO reinforcement might induce particle agglomeration and reduce accessible photoactive sites. The degradation of MB demonstrates its usefulness in waste management. Besides, 50 wt% composition exhibited enhanced piezoelectric performance which is potential for sensor and actuator applications. Such these nanocomposites with low reinforcement percentage have significant impact and offer new opportunities for designing scalable, high-energy-density polymer dielectric materials.

Declaration of generative AI and AI-assisted technologies in the manuscript preparation process

During the preparation of this work, the authors used generative AI tools (ChatGPT, OpenAI, and Gemini by Google) to assist with language refinement, clarity improvement, and organization of the manuscript text. These AI tools were not used to generate scientific results, data, analyses, figures, or interpretations. All AI-assisted content was thoroughly reviewed, edited, and validated by the authors to ensure accuracy, originality, and scientific integrity. The authors take full responsibility for the content of the published article.

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Declarations of Competing Interests:

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. Furthermore, the authors declare that they have no relevant financial or non-financial interests to disclose covering the last three years of conducting and preparing this research.

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