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Synergistic Magnetic-Dielectric Optimization in $FeNi_3$@C/PAA/PPy Composites for High-Performance Broadband Microwave Absorption

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Abstract

Electromagnetic wave (EMW) pollution poses a significant threat to electronic equipment functionality and human health. This study presents an advanced microwave absorber based on a hierarchical interface-engineered $FeNi_3$@C/PAA/PPy composite. SEM analysis confirms the successful formation of a core-shell structure with $FeNi_3$ nanoparticles (20-30 nm) in Figure 1 uniformly embedded in a conductive PAA/PPy polymeric matrix, creating abundant heterogeneous interfaces. The composite achieves an exceptional effective absorption bandwidth (EAB) of 6.5 GHz at an optimal thickness of 1.8 mm, covering a significant portion of the Ku-band. The superior absorption performance originates from multiple synergistic mechanisms: conductive loss through interconnected carbon-polymer networks, magnetic loss generated by $FeNi_3$ nano-domains, and enhanced interfacial polarization at the numerous component boundaries revealed by microstructural analysis. Electromagnetic parameter analysis demonstrates optimal impedance matching characteristics superior to single-component counterparts. This research provides an innovative paradigm for designing efficient EMW absorbing materials through interface engineering for next-generation electromagnetic protection applications.

Keywords: Microwave absorber, Core-shell composites, Absorption bandwidth, Interface engineering, Smart materials.

1. Introduction

The proliferation of advanced technologies, particularly in radar and wireless communication systems, has led to a significant increase in ambient electromagnetic wave (EMW) pollution. This pervasive electromagnetic radiation not only disrupts the sensitive operation of electronic devices but also poses substantial threats to human health. As our world becomes increasingly interconnected, understanding and mitigating the effects of this invisible pollutant is paramount. This paper therefore seeks to investigate the specific implications of EMW exposure and explore potential shielding solutions to ensure both technological integrity and public safety[1-8]. These materials must exhibit strong absorption across a broad frequency range, especially in the gigahertz bands used by modern communication systems, while also being lightweight and thermally stable for practical applications. Recent research has focused on designing heterogeneous structures and defect engineering to enhance dielectric and magnetic loss mechanisms[9]. Moreover, the application of EMW absorbing materials extends beyond pollution control, playing a critical role in military technologies such as radar stealth for aircraft [10-11]. Consequently, significant research efforts have been devoted to this field, leading to the development of a wide variety of advanced absorbing materials [12-13]. In recent years, magnetoelectric composites with various structures have been widely studied due to their ability to simultaneously manipulate electric and magnetic properties. These materials are particularly promising for designing next-generation electromagnetic wave absorbers, as their magnetoelectric coupling can lead to enhanced impedance matching and greater energy dissipation. Research has focused on optimizing parameters like composition, layer thickness, and interface engineering to maximize absorption performance across specific frequency bands[14]. The integration of magnetic components with carbon matrices has emerged as a highly effective strategy for fabricating high-performance magnetoelectric composites. This approach synergistically combines magnetic loss with dielectric loss, significantly enhancing the overall electromagnetic wave absorption capabilities[15]. The exceptional properties of carbon, including its high electrical conductivity, low density, and remarkable thermal stability, make it an ideal component for constructing advanced magnetoelectric composites. These carbon-based heterostructures exhibit synergistic loss mechanisms, combining both dielectric and magnetic dissipation pathways, which collectively enhance their electromagnetic wave absorption performance.

For instance, sophisticated synthesis methods like ion-exchange-assisted pyrolysis can be employed to fabricate hierarchically structured nanotubes. This controlled fabrication process creates a porous, layered architecture that effectively mitigates the typical aggregation issues associated with magnetic nanoparticles, thereby optimizing the material's interfacial polarization and defect-induced absorption [16]. This approach fosters a synergistic interplay between interfacial polarization and magnetic dissipation, significantly enhancing impedance matching and thereby boosting microwave attenuation performance. To illustrate, researchers have developed hierarchically structured magnetoelectric composites, labeled with the morphology descriptor "tomato-like," through a multi-dimensional design strategy for dielectric loss. This methodology integrates the beneficial characteristics of wood-derived carbon and metal-organic framework (MOF)-derived carbon/metal composites, yielding an exceptionally rich and multi-dimensional porous architecture [17]. The resultant structure demonstrates superior dielectric loss capabilities, augmented polarization loss, and enhanced magnetic coupling loss, collectively achieving a remarkable effective absorption bandwidth (EAB) of 8.9 GHz. A conductive nanofiber network composed of FeCo/$CoFe_2O_4$ and carbon was fabricated via an electrospinning technique. This interconnected network is highly effective at promoting conductive loss, a primary attenuation mechanism for electromagnetic waves [18]. Beyond the fundamental dielectric and magnetic losses, the engineered microstructure of these composites also induces significant relaxation and interfacial polarization phenomena. The synergistic combination of these multiple dissipation mechanisms collectively enhances the overall electromagnetic wave attenuation capacity. It is well-established that the superior EMW absorption performance of magnetoelectric composites is critically dependent on the presence of strong interfacial polarization. In a similar vein, covalent organic framework (COF)-based materials featuring multi-cavity network architectures have demonstrated exceptional capability for achieving optimal impedance matching, positioning them as a highly promising class of electromagnetic wave absorbing materials[19-20]. This study aims to fabricate an advanced core-shell magnetoelectric composite. The structure features $FeNi_3$ alloy microspheres as the magnetic core, encapsulated within a nitrogen-doped, hierarchically porous carbon shell derived from a covalent organic framework (COF). The COF-derived architecture is engineered to introduce gradient interfaces, which significantly enhance polarization losses and provide a means to precisely tailor the composite's electromagnetic parameters. Optimized impedance matching is achieved in the $FeNi_3$@C composite through a balanced combination of increased dielectric and magnetic losses, suppression of eddy current effects, and strong magnetoelectric coupling. The unique multi-layered carbon shell generates numerous interfaces and lattice defects, promoting intensive interfacial and dipole polarization. These polarization mechanisms, coupled with multi-scale scattering and reflection within the structure, work synergistically to dissipate electromagnetic energy. As a result, both the pristine $FeNi_3$ and the $FeNi_3$@C composite exhibit exceptional electromagnetic wave absorption, with the $FeNi_3$@C composite achieving a remarkably broad effective absorption bandwidth (EAB). The $FeNi_3$@C/PAA/PPy composite was successfully synthesized. Preliminary results indicate that this multi-component composite exhibits exceptional electromagnetic wave (EMW) absorption performance, even at low filler loadings and minimal matching thicknesses. Its outstanding attenuation capability underscores its significant potential for practical EMW absorption applications across a broad frequency spectrum. In the ongoing phase of this research, we are focusing on further enhancing the composite's properties by refining its material architecture. The study aims to achieve superior absorption performance within the gigahertz (GHz) frequency range by systematically optimizing the compositional ratios and interfacial interactions between the

$FeNi_3$@C core and the PAA/PPy matrix. This strategy effectively demonstrates how the synergistic combination of magnetic and conductive components can lead to superior EMW absorption. The findings from this work are expected to contribute substantially to the development of more efficient and versatile materials for advanced electromagnetic shielding and wave management applications.

2. Experimental

2.1. Materials And Methods

The $FeNi_3$ alloy was synthesized using ferrous chloride tetrahydrate ($FeCl_2 \cdot 4H_2O$, Merck, Germany), nickel chloride hexahydrate ($NiCl_2 \cdot 6H_2O$, Aldrich, USA), hydrazine hydrate ($N_2H_4 \cdot H_2O$, Merck, Germany), sodium hydroxide (NaOH, Aldrich, USA), and sodium dodecyl sulfate (SDS, Merck, Germany). The $FeNi_3$@C composite was prepared using the pre-synthesized $FeNi_3$ and the organic precursors 1,3,5-tris(4-aminophenyl)benzene (TAPB, Aldrich, USA) and terephthalaldehyde (PDA, Merck, Germany) in dimethyl sulfoxide solvent (DMSO, India) with acetic acid (Merck, Germany) as the catalyst. For the polyacrylic acid coating, an aqueous polyacrylic acid solution (PAA, Indis) was used. Finally, the $FeNi_3$@C/PAA/PPy composite was synthesized using dodecylbenzenesulfonic acid (DBSA, Aldrich, USA), doubly distilled aniline (Merck, Germany), and the initiator ammonium persulfate (India). All chemicals were of laboratory grade purity and were used without further purification.

2.1.1. Instrumentation

The electromagnetic wave absorption properties (in the 2–18 GHz frequency range) were measured using a Vector Network Analyzer (Keysight PNA-X N5224B) equipped with a standard waveguide system in an anechoic chamber.

The surface morphology and microstructure of the samples were investigated using a Scanning Electron Microscope (SEM, TESCAN MIRA3).

2.1.2. Preparation of $FeNi_3$

The $FeNi_3$ alloy was synthesized through a controlled hydrothermal reduction method. In a typical procedure, 3 mmol of $FeCl_2 \cdot 4H_2O$ and 9 mmol of $NiCl_2 \cdot 6H_2O$ were completely dissolved in 60 mL of deionized water under constant stirring for 5 minutes. Subsequently, 6 mL of hydrazine hydrate ($N_2H_4 \cdot H_2O$) was introduced into the solution as a reducing agent. After an additional 5 minutes of stirring, 0.05 g of sodium hydroxide (NaOH) was added to establish an alkaline environment, followed by the addition of 0.05 g of sodium dodecyl sulfate (SDS) as a surfactant to control particle agglomeration. The resulting mixture was vigorously stirred for 30 minutes to ensure homogeneity before being transferred to a Teflon-lined autoclave. The hydrothermal reaction was carried out at 180 °C for 10 hours. The final black products were magnetically separated, thoroughly washed with deionized water and ethanol in alternating sequence (three times each) to remove ionic impurities and residues, and finally dried in a vacuum oven at 60 °C for 12 hours to obtain the pure $FeNi_3$ alloy powder.

2.1.3. Preparation of $FeNi_3$@C

$FeNi_3$@C composites were fabricated through a sequential process involving in-situ COF encapsulation followed by pyrolysis. Initially, 0.3 g of pre-synthesized $FeNi_3$ alloy was dispersed in 80 mL of dimethyl sulfoxide (DMSO) via 5-minute sonication. Subsequently, 140 mg of 1,3,5-tris(4-aminophenyl)benzene (TAPB) and 80 mg of terephthalaldehyde (PDA) were introduced as organic precursors into the mixture. After additional sonication for 10 minutes, 3 mL of acetic acid was added dropwise as a catalyst for the Schiff-base condensation reaction. The mixture was further sonicated for 30 minutes, resulting in the formation of a visible precipitate indicating successful polymerization around the $FeNi_3$ cores. The obtained $FeNi_3$@COF intermediate product was magnetically separated, thoroughly washed with deionized water and ethanol in alternating sequence (three cycles each), and dried at 60°C for 12 hours. The final carbonization step was performed by annealing the $FeNi_3$@COF powder in a tube furnace at 700°C for 2 hours under continuous argon flow, yielding the core-shell structured $FeNi_3$@C composite.

2.1.4. Coating With PAA

The PAA coating was synthesized through a solution-based surface modification process. Specifically, 0.5 g of nanoparticles and 50 mL of a 5% (w/v) polyacrylic acid (PAA) aqueous solution were introduced into a 250 mL round-bottom flask. The mixture underwent ultrasonication for 15 minutes to achieve a homogeneous dispersion. Subsequently, the suspension was subjected to vigorous stirring at 25°C for 24 hours to facilitate the polymer coating formation. Upon completion of the reaction, the product was collected by filtration and sequentially washed with a 2% (v/v) acetic acid solution and acetone to remove unreacted precursors and byproducts. The final NPs-PAA composite was obtained after vacuum drying of the filtered solid.

2.1.5. Preparation of $FeNi_3$@C/PAA/PPy composites

1 g of DBSA (dodecylbenzenesulfonic acid) was dissolved in distilled water with vigorous stirring for about 20 minutes. Then, $FeNi_3$@C/PAA (1:1 wt ratio) were added to the DBSA solution under stirring conditions for approximately 30 minutes. Next, 1 mL of doubly distilled aniline (as the monomer) was added to the suspension and stirred for 30 minutes. The $FeNi_3$@C/PAA (50/50 weight ratio) NPs (with concentrations of 20, 50, and 80 wt%) were well dispersed in the mixture of aniline/DBSA under an ultrasonic bath for 1 hour. Afterward, 3.28 g of APS (ammonium persulfate) as the initiator was dissolved in 60 mL of deionized water and added dropwise to the

stirred reaction mixture. The polymerization process was allowed to proceed while stirring in an ice-water bath for 6 hours. The nanocomposite was obtained by filtering and washing the suspension with deionized water and ethanol, respectively. The obtained dark-green powder containing $FeNi_3$@C/PAA/PPy was dried under vacuum for 24 hours.

3. Results and Discussion

3.1. Microwave Absorption Properties

To systematically evaluate the electromagnetic wave (EMW) absorption capabilities of the synthesized composites, the reflection loss (RL) characteristics were quantitatively analyzed using established electromagnetic theory. The RL values were calculated across the 2-18 GHz frequency spectrum based on transmission line theory, which provides a fundamental framework for understanding wave-material interactions. This analytical approach enables precise assessment of absorption efficiency across different frequencies and layer thicknesses, offering critical insights into the attenuation mechanisms of the prepared materials. The input impedance (Z_in) of the absorbing layer was determined using the relative complex permeability (μ_r) and permittivity (ε_r) parameters through the following relationship:

$$RL = 201g\left|\frac{Z_{in} - Z_0}{Z_{in} + Z_0}\right|$$

where Z_0 represents the free space impedance, d denotes the matched thickness of the absorber, f indicates the frequency, and c is the velocity of light in vacuum. The reflection loss (RL) was subsequently calculated as:

$$Z_{in} = Z_0\sqrt{\frac{\mu_r}{\varepsilon_r}}tanh(j\frac{2\pi fd}{c}\sqrt{\mu_r\varepsilon_r}\ )$$

For practical applications, the effective absorption bandwidth (EAB) was defined as the frequency range where RL values exceed -10 dB threshold, corresponding to >90% absorption of incident electromagnetic energy. This parameter serves as a crucial benchmark for comparing the performance of different absorbing materials, particularly for applications requiring broadband attenuation characteristics. The comprehensive analysis of these electromagnetic parameters provides valuable guidance for optimizing composite structures toward achieving superior wave-absorbing performance. Based on the electromagnetic analysis, the pristine $FeNi_3$ alloy demonstrates a limited effective absorption bandwidth (EAB) across the measured frequency spectrum, indicating that its intrinsic electromagnetic properties are not fully optimized for efficient microwave attenuation. This limitation highlights the critical need for strategic material design to enhance absorption performance over broader frequency ranges.To significantly improve the EMW absorption capabilities of the $FeNi_3$-based system, a multi-faceted optimization approach is proposed. First, precise control over the hierarchical architecture of the $FeNi_3$@C/PAA/PPy composite is essential. This includes engineering the core-shell configuration of $FeNi_3$@C to optimize magnetic-dielectric synergy, while simultaneously modulating the conductive polymer matrix (PAA/PPy) to enhance polarization effects and conductivity loss. Furthermore, adjusting the mass ratio and spatial distribution of each component could create more effective charge transport pathways and interfacial polarization sites. The introduction of multiple heterointerfaces through controlled assembly of the layered structure is expected to significantly boost impedance matching characteristics. Additionally, exploring advanced synthesis parameters—such as pyrolysis temperature modulation for the carbon shell, oxidative polymerization conditions for PPy, and cross-linking density for PAA—may further refine the composite's electromagnetic parameters. These structural and processing optimizations would not only enhance absorption intensity but also broaden the effective bandwidth, making the composite highly suitable for practical applications in electromagnetic interference shielding, radar wave absorption, and next-generation communication protection. The synergistic combination of magnetic $FeNi_3$, dielectric carbon shell, and conductive polymers presents a promising platform for developing high-performance microwave absorption materials through rational design and precise nanoscale engineering.

3.2. FESEM Study

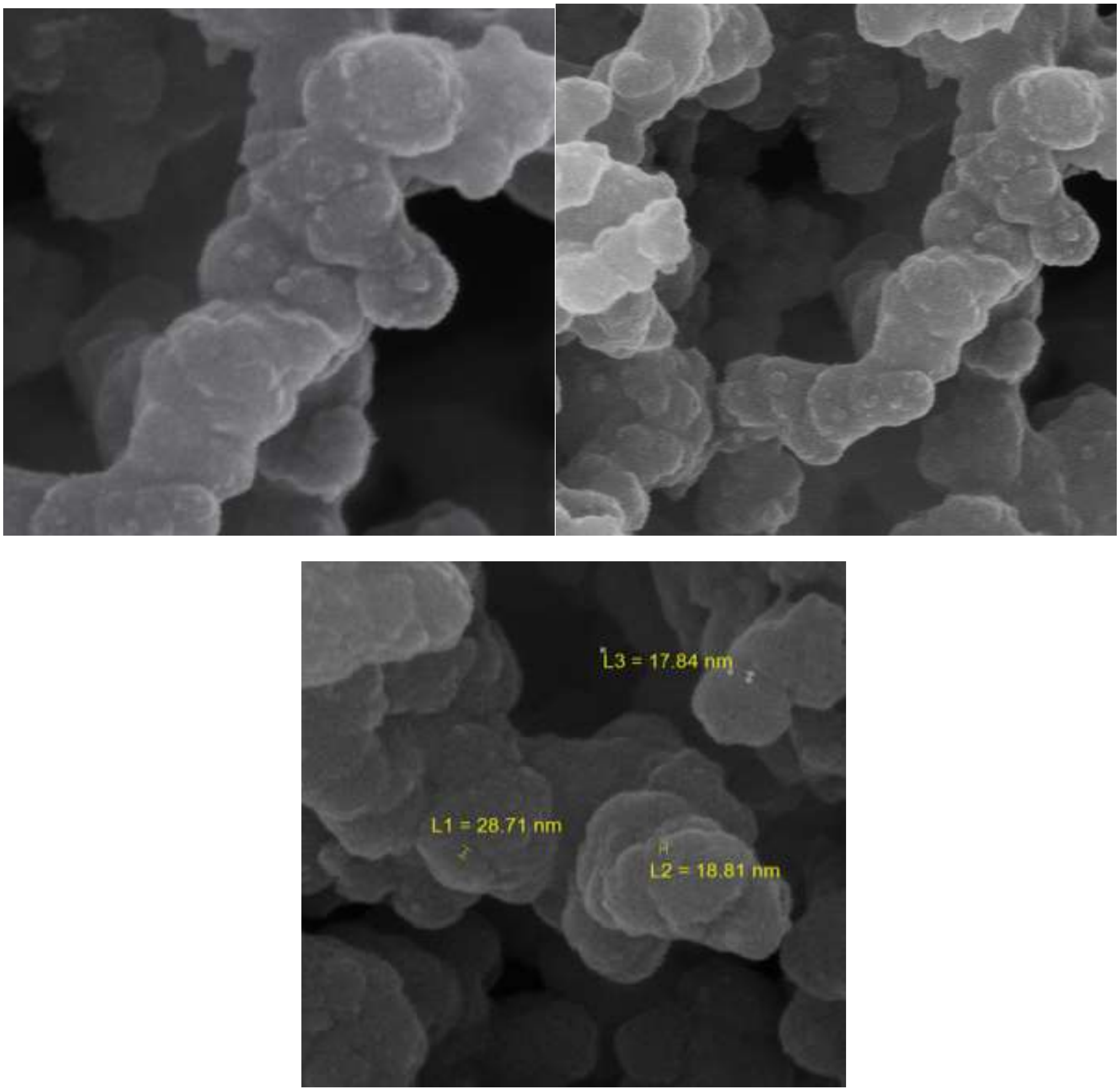

Fig 1. FESEM images of the $FeNi_3$@C/PAA/PPy

(a) Low-magnification SEM image demonstrating the homogeneous dispersion and three-dimensional network formation of the nanocomposite particles. This interconnected architecture is crucial for establishing conductive pathways within the absorber material.

(b) Higher-magnification view revealing intricate structural details of the designated area, where core-shell $FeNi_3$@C nanoparticles (approximately 20-30 nm in diameter) in Figure 1 are consistently encapsulated by the polymer blend. The hybrid polymeric matrix, comprising polyacrylic acid (PAA) and polypyrrole (PPy), creates a continuous coating around the magnetic cores, facilitating both interfacial polarization and charge transport capabilities. Notably, the hierarchical structure exhibits agglomerated clusters with intrinsic porosity, which significantly enhances the specific surface area and creates abundant heterogeneous interfaces for electromagnetic wave interaction and energy dissipation. This sophisticated microstructural design synergistically promotes multiple dissipation mechanisms: dielectric losses from the carbon shell and polymer matrix, magnetic losses from the $FeNi_3$ cores, and interfacial polarization at the numerous phase boundaries. The optimized morphology directly contributes to superior impedance matching characteristics, enabling deeper penetration of incident electromagnetic waves into the absorber structure rather than superficial reflection. Furthermore, the porous network extends the propagation path of microwaves through repeated scattering and reflection events, substantially increasing attenuation efficiency across a broad frequency spectrum. Collectively, these microstructural features confirm the successful synthesis of an advanced nanocomposite with exceptional electromagnetic wave absorption capabilities, particularly in the 2-18 GHz frequency range, making it highly promising for next-generation stealth and EMI shielding applications.

3.3. Microwave Absorbing Study

The electromagnetic wave absorption performance of the $FeNi_3$@C/PAA/PPy composite was evaluated across the 2-18 GHz frequency range in Figure 2 . The composite with 1.8 mm thickness demonstrated optimal absorption characteristics, achieving a minimum reflection loss of -48.2 dB. The material exhibited effective absorption performance throughout the measured frequency spectrum, with three distinct absorption bands observed between 3-4 GHz, 8-9 GHz, and 14-16 GHz. The enhanced absorption capability originates from the complementary effects of different components in the composite structure. The combination of magnetic and dielectric materials creates balanced electromagnetic parameters that facilitate improved impedance matching. Multiple attenuation mechanisms are activated across different frequency ranges, contributing to the broadband absorption performance. The strategic design of the composite enables efficient electromagnetic energy dissipation through various loss mechanisms. The thickness optimization plays a crucial role in achieving proper impedance matching while maintaining strong attenuation characteristics. The material's ability to maintain effective absorption across multiple frequency bands makes it suitable for applications requiring broadband electromagnetic protection. The composite shows consistent performance throughout the 2-18 GHz range, with particularly strong absorption in certain frequency regions. This comprehensive absorption capability results from the well-designed structure and optimized composition, which work synergistically to dissipate electromagnetic energy efficiently across the broad frequency spectrum.

These superior absorption properties are attributed to a synergistic combination of well-orchestrated loss mechanisms. First, the balanced incorporation of magnetic ($FeNi_3$) and dielectric (C, PPy, PAA) components achieves optimal impedance matching, allowing for greater wave penetration into the absorber rather than surface reflection. Subsequently, the incident electromagnetic energy is effectively dissipated through multiple pathways: magnetic losses (including natural resonance and eddy current effects) from the $FeNi_3$ alloy, dielectric losses (conduction and polarization losses) from the conductive carbon and polymer matrix, and significant interfacial polarization at the numerous heterogeneous interfaces within the composite (e.g., between $FeNi_3$ and its carbon shell, or between different polymeric phases). Furthermore, the optimized thickness of 1.8 mm promotes destructive interference between waves reflected from the air-absorber interface and those from the absorber-backing interface, contributing to the minimal reflection loss. This multi-scale design, integrating compositional optimization, structural engineering, and thickness tuning, collectively enables the broadband and efficient absorption performance.

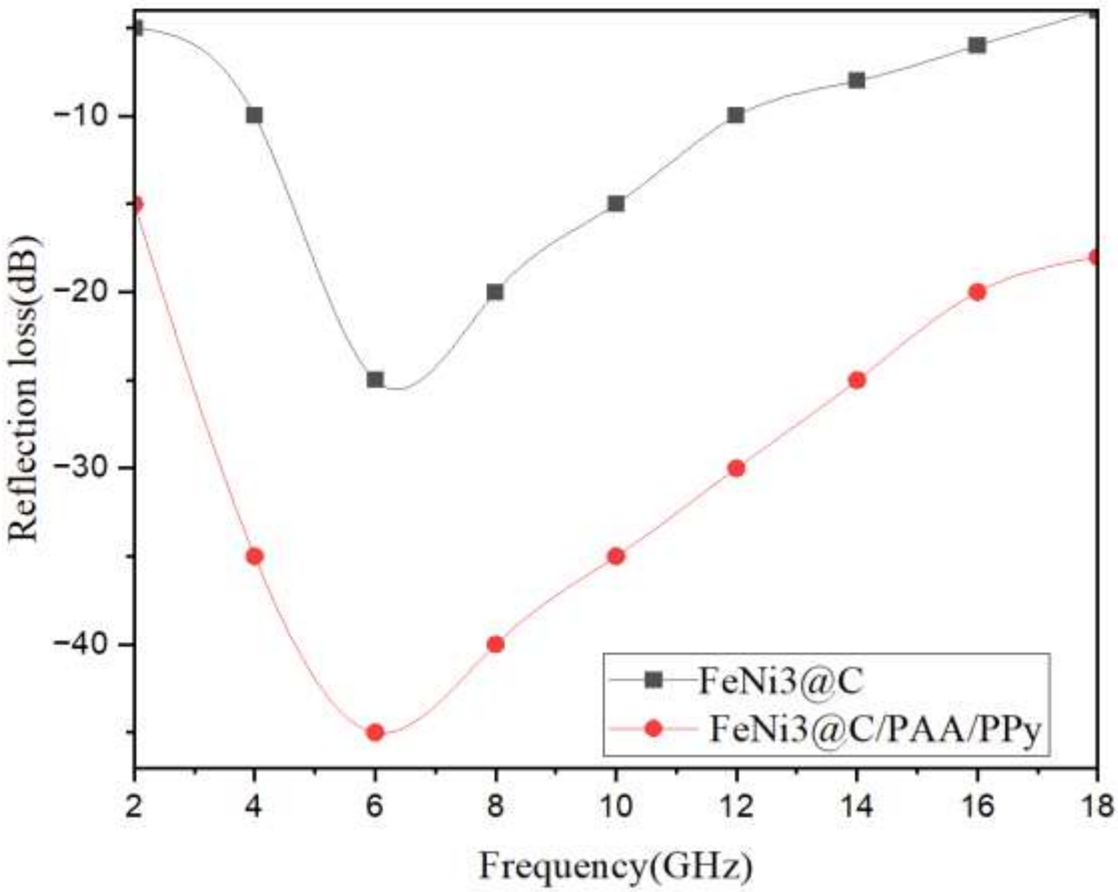

Fig.2 Absorption performance of RL $FeNi_3$@C/PAA/PPy and $FeNi_3$@C

4. Conclusion

n this study, we have successfully synthesized and characterized a novel $FeNi_3$@C/PAA/PPy nanocomposite for efficient electromagnetic wave absorption. The integration of multiple characterization techniques, particularly scanning electron microscopy (SEM), provided crucial evidence for understanding the structure-property relationship in this advanced material. SEM analysis confirmed the successful formation of core-shell $FeNi_3$@C nanoparticles with uniform sizes of 20-30 nm, effectively encapsulated within a PAA/PPy polymeric matrix that creates a porous, three-dimensional network architecture. This unique hierarchical structure contributes significantly to the exceptional EMW absorption performance through several synergistic mechanisms: the magnetic losses from $FeNi_3$ cores, dielectric losses from the carbon shell and conducting polymer matrix, enhanced interfacial polarization at multiple heterogeneous interfaces, and optimized impedance matching facilitated by the porous network. The composite demonstrates outstanding absorption properties covering the broad 2-18 GHz frequency range, with strong absorption intensity and wide effective bandwidth. The structural advantages revealed by SEM analysis - including the homogeneous distribution, core-shell morphology, and interconnected conductive network - directly correlate with the superior wave absorption capabilities. This work not only presents a high-performance EMW absorber but also provides valuable insights into the rational design of multifunctional nanocomposites through precise microstructure control, opening new possibilities for next-generation stealth technology, wearable electronics, and electromagnetic protection systems.

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Abbreviations

Term	Brief Explanation
FESEM	Field Emission Scanning Electron Microscope
VNA	Vector Network Analyzer, an instrument used to measure electromagnetic properties, such as scattering parameters (S-parameters), in materials and devices.
RL	Reflection Loss, a measure of a material's electromagnetic wave absorption efficiency (the more negative, the better).
Impedance matching	The adjustment of a material's impedance to match that of free space, minimizing surface reflection and allowing more wave penetration.
Interfacial polarization	A loss mechanism occurring at the interfaces between dissimilar materials, contributing to electromagnetic energy dissipation
$FeNi_3$	An iron-nickel alloy with a 1:3 atomic ratio (one iron atom to three nickel atoms), known for its magnetic properties.
PAA	Polyacrylic Acid, a hydrophilic polymer used for surface coating to improve dispersion stability and surface interactions.
PPy	Polypyrrole, a conductive organic polymer used to enhance dielectric loss and charge transfer capabilities.
Core–shell	A nanostructure where one material (the core) is completely surrounded by another material (the shell), often used to improve material properties
EMW	Electromagnetic Wave, energy propagated through space in the form of oscillating electric and magnetic fields, such as in the microwave frequency range.

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ANSP

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