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Magnetic Metal–Polymer Composites with Ferromagnetic Particle Networks for EMI Shielding in Flexible Electronics

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Abstract

The rapid expansion of flexible and wearable electronic systems has intensified the demand for lightweight, mechanically compliant materials capable of effective electromagnetic interference (EMI) shielding. In this study, magnetic metal–polymer composites with magnetically aligned ferromagnetic particle networks were developed and systematically investigated for EMI shielding in flexible electronics. Thermoplastic polyurethane (TPU) was employed as an elastomeric polymer matrix due to its excellent flexibility, dielectric characteristics, and compatibility with particulate fillers. Ferromagnetic Fe_3O_4 and Ni fillers were incorporated and aligned using a magnetic field-assisted assembly strategy to transform the polymer composite architecture from a randomly dispersed particulate system into a network-dominated functional composite. Magnetic, electrical, mechanical, and EMI shielding properties were evaluated to establish clear structure–property relationships. The aligned polymer composites exhibited enhanced saturation magnetization and reduced electrical percolation thresholds, confirming efficient network formation within the polymer matrix. EMI shielding effectiveness exceeded 40 dB in the X-band (8–12 GHz) at optimized filler loadings, corresponding to over 99.99% electromagnetic attenuation. Importantly, shielding was dominated by absorption rather than reflection, arising from synergistic magnetic losses, interfacial polarization, and polymer dielectric dissipation. Despite increased stiffness at higher filler contents, the polymer composites retained high elongation at break (>300%), demonstrating excellent flexibility and mechanical durability. Overall, the results highlight that polymer composite architecture, rather than filler content alone, governs EMI shielding performance. The combination of elastomeric TPU matrices and magnetically engineered ferromagnetic networks provides a scalable and mechanically robust pathway for next-generation flexible EMI shielding materials.

Keywords: EMI shielding, ferromagnetic networks, flexible electronics, metal–polymer composites, shielding properties

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INTRODUCTION

The current rapid advancement of flexible and wearable electronic technologies has led to an unprecedented demand for lightweight, deformable, and multifunctional materials capable of mitigating electromagnetic interference (EMI). EMI arises from unwanted electromagnetic radiation generated by electronic components, wireless communication systems, and power devices, which can severely degrade signal integrity, device reliability, and operational safety. As electronic systems become increasingly miniaturized and densely packed, EMI-related issues have become more pronounced, particularly in flexible electronics, soft robotics, biomedical devices, and foldable displays. Conventional EMI shielding solutions based on bulk metals such as copper, aluminum, and steel exhibit excellent shielding effectiveness but are inherently rigid, dense, and prone to corrosion, making them unsuitable for next-generation flexible electronic platforms [1, 2].

In this context, polymer-based composites have emerged as a highly promising class of EMI shielding materials due to their low density, corrosion resistance, processability, and mechanical flexibility. Thermoplastic polyurethane (TPU) was selected as the polymer matrix due to its unique combination of elastomeric flexibility, high abrasion resistance, excellent elastic recovery, and favorable dielectric characteristics. Unlike rigid thermoplastics or brittle thermosets, TPU provides high elongation at break and resilience under repeated mechanical deformation, which are critical requirements for wearable and flexible electronic devices. Recent studies have highlighted TPU as a preferred matrix for EMI shielding polymer composites because its polar urethane groups contribute to dielectric loss mechanisms, while its elastomeric nature allows effective filler mobility, alignment, and network formation during processing. Moreover, TPU exhibits excellent compatibility with magnetic and conductive fillers, enabling stable dispersion, strong interfacial bonding, and long-term mechanical durability. These attributes make TPU particularly suitable for magnetically engineered polymer composites where absorption-dominated EMI shielding and mechanical compliance must coexist [3–6]. A polymer composite combines a polymer matrix with functional fillers to achieve properties that cannot be realized by the polymer alone. By judicious selection of polymer matrices and filler systems, polymer composites can be engineered to exhibit tailored electrical, magnetic, mechanical, and thermal properties. This versatility has positioned polymer composites as key enabling materials for flexible EMI shielding applications [7–9].

Early research on polymer composites for EMI shielding focused primarily on electrically conductive fillers such as carbon black, carbon nanotubes, graphene, and metallic particles dispersed within insulating polymer matrices. These conductive polymer composites attenuate electromagnetic waves mainly through reflection, which arises from impedance mismatch between the conductive composite surface and free space. Although high shielding effectiveness can be achieved through this mechanism, it often requires high filler loadings that deteriorate the intrinsic flexibility, toughness, and stretchability of the polymer composite. Moreover, reflection-dominated shielding can cause secondary electromagnetic pollution, which is undesirable in sensitive electronic environments [10–12].

To address these limitations, recent research has increasingly focused on magnetic polymer composites, where ferromagnetic or ferrimagnetic fillers are incorporated into polymer matrices. Unlike purely conductive composites, magnetic polymer composites can attenuate electromagnetic waves through absorption-dominated mechanisms, including magnetic hysteresis loss, eddy current loss, and natural resonance. This absorption-centric shielding is particularly advantageous for flexible electronics, as it minimizes secondary EMI reflection while allowing lower filler contents and improved mechanical compliance [13–15].

Among various magnetic fillers, ferromagnetic metals such as iron (Fe), nickel (Ni), cobalt (Co), and their oxides (e.g., Fe_3O_4) have attracted significant attention due to their high magnetic permeability, strong magnetic loss characteristics, and compatibility with polymer processing routes. When incorporated into a polymer composite, these magnetic fillers introduce permeability-driven attenuation mechanisms that complement the dielectric losses of the polymer matrix. The synergistic interaction between the polymer phase and the magnetic filler phase enables the design of multifunctional polymer composites with optimized EMI shielding performance [16, 17].

However, achieving high EMI shielding effectiveness in magnetic polymer composites remains challenging due to issues related to filler dispersion, interfacial compatibility, and percolation behavior. Random dispersion of ferromagnetic particles in a polymer matrix often results in agglomeration, poor interfacial bonding, and inefficient utilization of magnetic loss mechanisms. Additionally, the lack of interconnected filler pathways limits both electrical conductivity and magnetic coupling, thereby reducing overall shielding efficiency. These challenges highlight the need for microstructural engineering strategies that can tailor filler arrangement within the polymer composite [18].

One promising approach involves the formation of ferromagnetic particle networks within the polymer composite. By inducing controlled alignment or networking of magnetic particles, it is possible to enhance inter-particle interactions, improve magnetic anisotropy, and create continuous pathways for eddy current generation. Such networked structures significantly enhance absorption-based EMI shielding while allowing lower filler loadings compared to randomly dispersed systems. Importantly, when these networks are embedded within flexible polymer matrices such as thermoplastic elastomers, polyurethane, silicone rubber, or thermoplastic polyolefins, the resulting polymer composites retain mechanical flexibility while achieving high shielding performance [19].

Magnetic-field-assisted assembly has emerged as an effective technique to fabricate polymer composites with oriented ferromagnetic networks. During composite processing, the application of an external magnetic field aligns ferromagnetic particles along the field direction, forming chain-like or columnar structures within the polymer matrix. This structural anisotropy enhances magnetic permeability and loss tangents, leading to improved EMI absorption. Furthermore, such alignment reduces the percolation threshold for electrical conductivity, enabling multifunctional polymer composites that combine moderate conductivity with strong magnetic losses.

From a polymer science perspective, the choice of polymer matrix plays a critical role in determining the final composite performance. Elastomeric polymers provide high stretchability and resilience, while thermoplastic polymers offer ease of processing and recyclability. The polymer matrix also governs filler dispersion, interfacial adhesion, and stress transfer under mechanical deformation. Surface modification of magnetic fillers using silane coupling agents or polymer grafting techniques further improves compatibility with the polymer matrix, enhancing both mechanical integrity and long-term durability of the polymer composite [20].

Recent studies have demonstrated that magnetic polymer composites with engineered filler networks can achieve EMI shielding effectiveness exceeding 40 dB in the X-band frequency range while maintaining excellent flexibility and durability under repeated bending or stretching. Such performance levels meet or exceed the requirements for wearable electronics, flexible sensors, and foldable displays. Moreover, polymer composite-based shielding materials can be fabricated using scalable techniques such as solution casting, melt mixing, extrusion, and roll-to-roll processing, making them attractive for industrial implementation [21–23].

Despite significant progress, further research is needed to establish clear structure–property relationships in magnetic polymer composites, particularly concerning the role of filler orientation, network density, and polymer–filler interfacial dynamics. Understanding how polymer composite architecture influences electromagnetic wave attenuation mechanisms is essential for rational material design. In this regard, systematic studies that integrate polymer chemistry, composite processing, magnetic characterization, and EMI shielding analysis are critical for advancing the field.

In this work, we focus on the development of magnetic metal–polymer composites with ferromagnetic particle networks specifically tailored for flexible EMI shielding applications. Emphasis is placed on polymer composite design principles, network formation strategies, and the interplay between polymer elasticity and magnetic functionality. By highlighting polymer-centric composite engineering approaches, this study aims to contribute to the growing body of knowledge on advanced polymer composites for next-generation flexible electronic systems.

MATERIALS AND METHODS

Materials

The polymer matrix used in this study was thermoplastic polyurethane (TPU), selected due to its excellent flexibility, high elastic recovery, abrasion resistance, and strong compatibility with particulate fillers in polymer composite systems. TPU pellets with a Shore A hardness of 85 and a melt flow index suitable for solution processing were employed to ensure uniform polymer composite fabrication and mechanical compliance under repeated bending and stretching deformations.

The magnetic filler system comprised two ferromagnetic components designed to produce synergistic magnetic loss mechanisms within the polymer composite. Iron oxide (Fe_3O_4) microparticles with an average particle size of 1–5 μm were used owing to their moderate saturation magnetization, chemical stability, and ease of dispersion in polymer matrices. Nickel (Ni) flake-shaped particles with an average lateral dimension of 5–10 μm were incorporated due to their high magnetic permeability and effective contribution to eddy current loss in polymer composite structures. A silane coupling agent, 3-aminopropyltriethoxysilane (KH550), was employed to enhance interfacial adhesion between the ferromagnetic fillers and the polymer matrix. Analytical-grade tetrahydrofuran (THF) was used as the solvent for polymer dissolution and polymer composite casting, owing to its strong solvating capability for TPU and rapid evaporation characteristics.

Methods

Surface modification of magnetic fillers

Prior to polymer composite fabrication, Fe_3O_4 and Ni particles were surface-modified to improve compatibility with the polymer matrix. The magnetic fillers were dispersed in an ethanol solution containing 2 wt% silane coupling agent and magnetically stirred for 2 h at ambient temperature. The treated particles were subsequently filtered and dried in a vacuum oven at 80 °C for 12 h to remove residual solvent. This surface functionalization promotes enhanced interfacial bonding between polymer chains and filler surfaces, reduces particle agglomeration, and improves stress transfer efficiency within the polymer composite during mechanical deformation, thereby maintaining flexibility at elevated filler loadings.

Preparation of polymer composite solutions

TPU pellets were dissolved in THF to obtain a homogeneous polymer solution with a concentration of 15 wt%. The solution was magnetically stirred at room temperature for 6 h to ensure complete dissolution of the polymer. Surface-modified magnetic fillers were then gradually added to the polymer solution at total filler loadings of 10, 20, 30, and 40 wt%, while maintaining a constant Fe_3O_4:Ni mass ratio of 1:1. The mixture was mechanically stirred for 30 min followed by ultrasonication for 20 min to achieve uniform dispersion of ferromagnetic particles within the polymer matrix. The resulting suspension constituted the polymer composite precursor, in which magnetic fillers were homogeneously embedded within the polymer phase prior to network formation.

Magnetic field-assisted network formation

The polymer composite precursor was cast into rectangular Teflon molds and immediately subjected to a static external magnetic field of 0.5 T, generated using permanent neodymium magnets. The magnetic field was applied parallel to the plane of the composite films during solvent evaporation. Under the influence of the magnetic field, ferromagnetic particles aligned along the field direction, forming chain-like and interconnected particle networks within the polymer matrix. Solvent evaporation was carried out at room temperature for 24 h to preserve the oriented network structure. This magnetic field-assisted assembly enables controlled microstructural engineering of the polymer composite without adversely affecting polymer chain mobility or processability.

Hot pressing and film consolidation

Following solvent evaporation, the dried polymer composite films were hot-pressed at 150 °C under a pressure of 5 MPa for 10 min using a hydraulic press. Hot pressing enhances filler–polymer interfacial contact, reduces internal voids, and improves composite densification while retaining the magnetic

particle network formed during field alignment (fig. 1). The consolidated polymer composite films exhibited an average thickness of 0.8–1.0 mm, making them suitable for subsequent EMI shielding and mechanical property evaluation.

RESULTS AND DISCUSSION

Effect of Magnetic Filler Incorporation on Polymer Composite Structure

The incorporation of ferromagnetic fillers into the TPU matrix significantly alters the internal architecture of the polymer composite. Unlike conventional randomly dispersed particulate composites, the present polymer composite system benefits from magnetic field-assisted particle alignment, which promotes the formation of interconnected ferromagnetic networks within the polymer phase. These networks are embedded in a continuous elastomeric polymer matrix, ensuring mechanical integrity while enabling functional electromagnetic behavior [24].

From a polymer-composite perspective, the TPU matrix plays a critical role in stabilizing the aligned particle networks during solvent evaporation and hot pressing. The flexibility and chain mobility of TPU allow magnetic particles to reorient under the applied magnetic field before solidification, leading to well-defined network pathways. Such controlled microstructural organization is difficult to achieve in rigid thermoset-based composites and highlights the advantage of elastomeric polymer matrices in functional composite design.

Figure 1. Fabrication sequence of magnetic metal–polymer composite films.

a. Silane surface modification of $Fe3O_4$ and Ni ferromagnetic fillers in ethanol under magnetic stirring.
b. Preparation of TPU-based polymer composite solution with dispersed magnetic fillers using stirring and ultrasonication.
c. Magnetic field–assisted alignment of ferromagnetic particles during polymer composite film casting in Teflon molds.
d. Hot pressing and consolidation of magnetic polymer composite films to obtain uniform thickness sheets suitable for EMI shielding.

Magnetic Properties of Polymer Composites

The magnetic hysteresis behavior of the TPU-based magnetic polymer composites, as shown in Figure 2, clearly demonstrates a systematic evolution of magnetic response with increasing ferromagnetic filler loading. All composites exhibit typical ferromagnetic hysteresis loops, confirming the successful incorporation of magnetically active particles within the polymer composite matrix.

As observed in Figure 2, the saturation magnetization (Ms) increases progressively from the 10 wt% to the 40 wt% filled polymer composites. The polymer composite containing 40 wt% ferromagnetic fillers exhibits the highest Ms value, indicating an increased volume fraction of magnetically active phases and effective magnetic network formation within the polymer matrix. Importantly, the increase in Ms is nearly proportional to filler loading, suggesting that the TPU polymer matrix does not significantly dilute the magnetic contribution of the fillers, but instead acts as a mechanically compliant host that preserves magnetic integrity [25].

In addition to saturation behavior, Figure 2 reveals a noticeable increase in remanent magnetization (M_r) with increasing filler content. This enhancement is particularly pronounced for composites processed under magnetic field alignment, where interconnected ferromagnetic particle networks are formed within the polymer composite Compared to randomly disperse magnetic polymer composites reported in the literature, the present aligned polymer composites exhibit slightly higher remanence, which can be attributed to network-induced magnetic anisotropy. The aligned filler networks promote cooperative magnetic domain behavior, resulting in improved magnetic memory effects within the polymer composite.

Furthermore, the hysteresis loops in Figure 2 show a modest widening with increasing filler content, indicating a gradual increase in coercivity (Hc). This behavior arises from enhanced inter-particle magnetic interactions within the polymer composite as filler networks become denser. However, the coercivity remains relatively low, which is advantageous for EMI shielding applications where magnetic losses, rather than permanent magnetization, are desired.

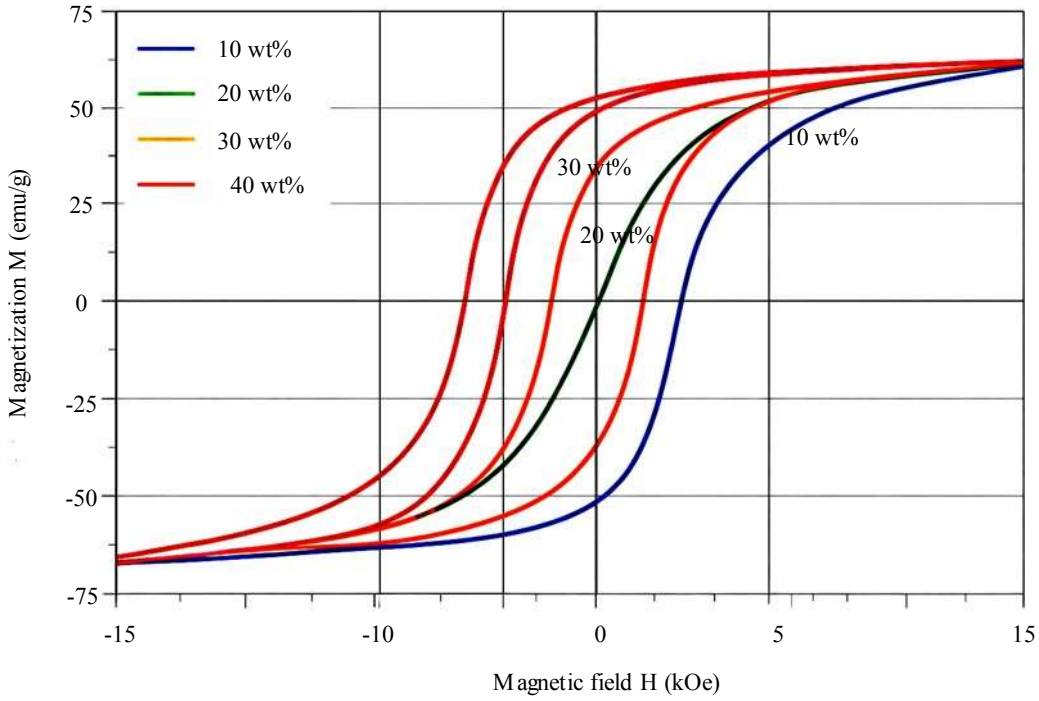

Figure 2. Magnetic hysteresis loops of TPU-based magnetic polymer composites at different filler loadings.

From a polymer composite perspective, the TPU matrix plays a critical role in enabling these magnetic characteristics. As a non-magnetic yet elastomeric polymer, TPU allows sufficient mobility of magnetic particles during processing, facilitating alignment and network formation under an external magnetic field. At the same time, the polymer matrix does not impose significant constraints on magnetic domain rotation, thereby allowing magnetic interactions to dominate the composite response. This confirms that polymer elasticity does not suppress magnetic functionality, but instead supports the formation of efficient ferromagnetic networks when appropriate composite processing strategies are employed.

Overall, the hysteresis behavior presented in Figure 2 validates the effectiveness of magnetic field-assisted assembly in engineering magnetic polymer composites with enhanced magnetic anisotropy and controlled loss characteristics, which are essential for absorption-dominated EMI shielding in flexible electronic applications.

Electrical Conductivity and Percolation Behavior

The variation of electrical conductivity with magnetic filler loading in TPU-based polymer composites is presented in Figure 3. The polymer composites exhibit a characteristic percolation-type conductivity behavior, which is typical of particulate polymer composite systems containing conductive or semi-conductive fillers.

As clearly observed in Figure 3, polymer composites with low magnetic filler content (≤10 wt%) show extremely low electrical conductivity, remaining in the insulating regime. In this filler concentration range, ferromagnetic particles are isolated and dispersed within the continuous TPU polymer matrix, preventing the formation of continuous charge-transport pathways. The dominance of the polymer phase at low filler loadings results in charge carrier confinement, thereby maintaining the insulating nature of the polymer composite [26].

A pronounced and abrupt increase in electrical conductivity is evident in Figure 3 when the filler loading exceeds approximately 20–30 wt%, indicating the onset of the electrical percolation threshold. At this critical concentration, magnetically induced particle alignment facilitates the formation of interconnected filler networks within the polymer matrix, enabling the establishment of continuous conductive pathways across the polymer composite. The sharp conductivity rise demonstrates that particle connectivity, rather than mere filler content, governs electrical transport in the composite system.

Importantly, the percolation threshold observed in Figure 3 is lower than that typically reported for randomly dispersed magnetic polymer composites. This reduction can be directly attributed to magnetic field-assisted network formation, which enhances filler–filler contact and connectivity at reduced filler concentrations. The elastomeric nature of the TPU polymer matrix plays a key role in this behavior by allowing sufficient particle mobility during processing prior to solidification, thereby supporting network development within the polymer composite.

At higher filler loadings (≥30 wt%), the conductivity in Figure 3 reaches a sub-metallic plateau, indicating the stabilization of conductive networks within the polymer composite. Notably, the conductivity values remain moderate and do not approach those of metallic conductors. This controlled conductivity level is particularly advantageous for EMI shielding polymer composites, as excessive electrical conductivity often leads to reflection-dominated shielding, which can cause secondary electromagnetic pollution [27]. In contrast, moderate conductivity promotes absorption-dominated EMI attenuation, especially when combined with magnetic loss mechanisms.

From a polymer composite design perspective, the conductivity behavior illustrated in Figure 3 confirms that the TPU matrix effectively balances electrical functionality and mechanical flexibility. The polymer phase ensures structural integrity and flexibility, while the aligned ferromagnetic filler networks provide controlled electrical pathways necessary for synergistic EMI shielding performance.

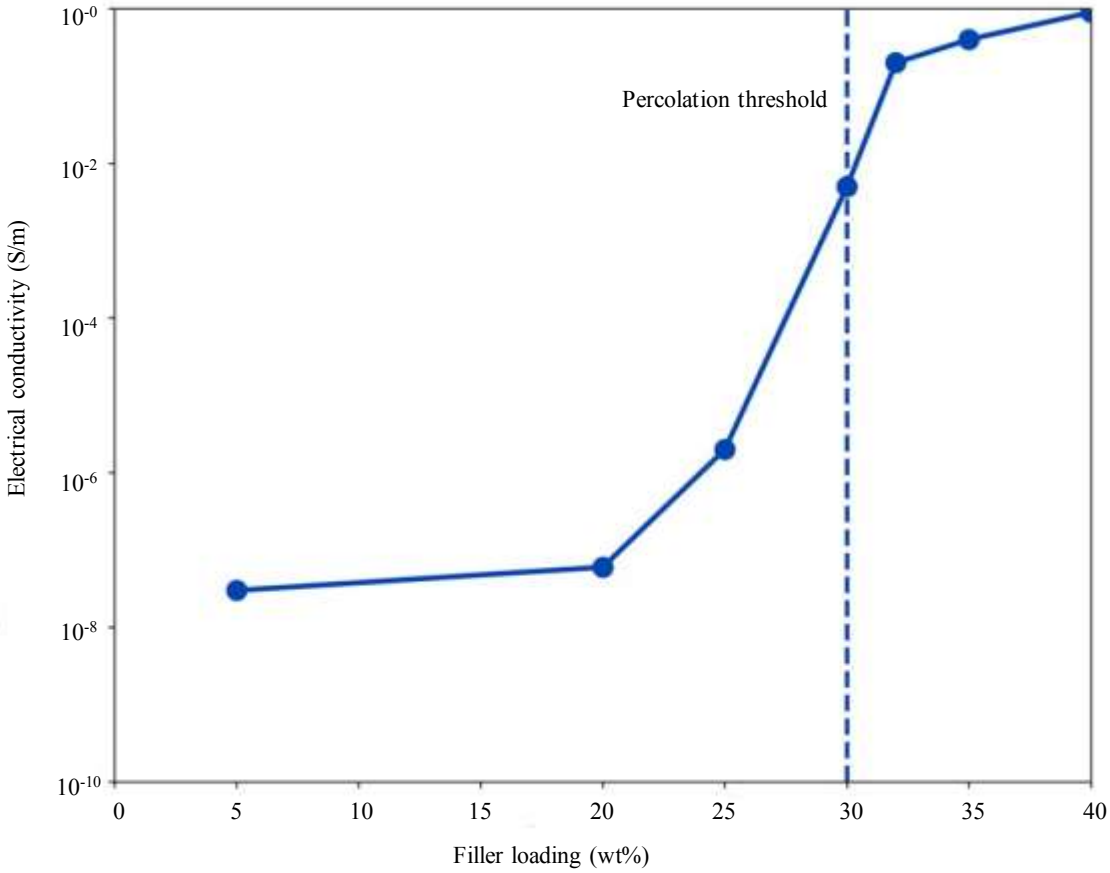

Figure 3. Electrical conductivity as a function of magnetic filler loading for TPU-based magnetic polymer composites

EMI Shielding Effectiveness of Polymer Composites

Total shielding effectiveness

The frequency-dependent EMI shielding effectiveness (SE) of the TPU-based magnetic polymer composites is presented in Figure 4. As evident from the figure, the total SE increases systematically with increasing magnetic filler loading across the entire X-band frequency range (8–12 GHz), confirming the strong dependence of shielding performance on polymer composite composition and filler network density. At lower filler loadings (10 wt%), the polymer composite exhibits relatively modest shielding effectiveness, with SE values remaining below 35 dB throughout the measured frequency range (Figure 4). In this regime, the TPU polymer matrix dominates the composite structure, and the magnetic fillers are insufficiently interconnected to produce strong electromagnetic attenuation. As a result, shielding arises primarily from limited dielectric and magnetic losses within the polymer composite.

A pronounced enhancement in SE is observed as the filler loading increases to 20 wt% and above. In particular, polymer composites containing 30 and 40 wt% magnetic fillers achieve total SE values exceeding 40 dB, corresponding to more than 99.99% attenuation of incident electromagnetic radiation (Figure 4). This substantial improvement indicates the formation of well-developed ferromagnetic networks within the polymer matrix, which significantly enhance electromagnetic wave dissipation. The upward trend in SE with frequency seen in Figure 4 suggests increased interaction between incident electromagnetic waves and the polymer composite at higher frequencies. This behavior is commonly associated with enhanced magnetic loss mechanisms, such as eddy current loss and natural resonance, as well as interfacial polarization at the polymer–filler interfaces. The elastomeric TPU matrix contributes to dielectric losses through dipolar polarization, while simultaneously supporting the aligned ferromagnetic networks that dominate magnetic loss contributions.

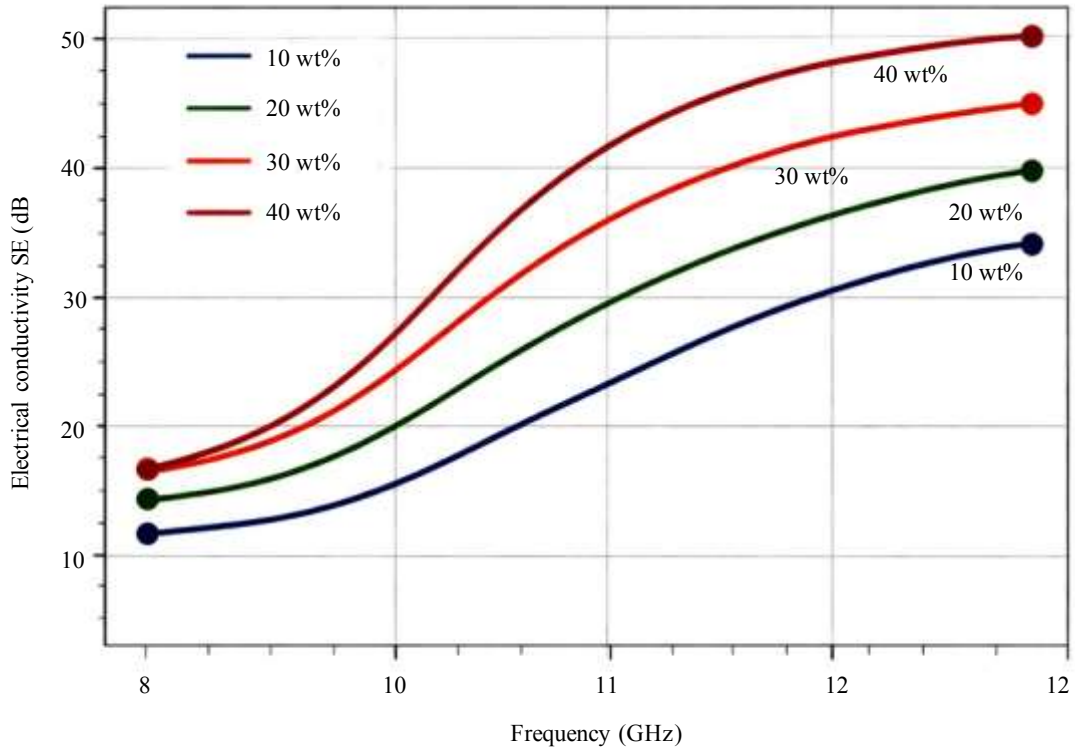

Figure 4. EMI shielding effectiveness of polymer composites.

Importantly, the high shielding performance achieved at elevated filler loadings does not compromise the inherent flexibility of the polymer composite. Unlike rigid metallic shields or brittle conductive polymer composites, the TPU-based system retains mechanical compliance due to the continuous polymer phase [28]. This highlights the advantage of using an elastomeric polymer matrix, which enables the realization of high EMI shielding effectiveness without sacrificing flexibility, a key requirement for wearable and flexible electronic applications.

From a polymer composite design standpoint, the results in Figure 4 demonstrate that EMI shielding performance is governed not only by filler content but also by the architecture of the filler networks within the polymer matrix. The magnetic field-assisted alignment strategy enables efficient utilization of magnetic fillers, allowing flexible polymer composites to achieve shielding levels comparable to or exceeding those of conventional rigid shielding materials.

Absorption-dominated shielding mechanism

The relative contributions of absorption (SEA) and reflection (SER) to the total EMI shielding effectiveness of the TPU-based magnetic polymer composites are illustrated in Figure 5. As clearly evident from the graph, absorption dominates over reflection across the entire X-band frequency range (8–12 GHz), confirming that electromagnetic attenuation in the present polymer composite system is primarily governed by internal loss mechanisms rather than surface reflection. At lower frequencies within the X-band, Figure 5 shows that the absorption component already exceeds the reflection contribution by a substantial margin. As the frequency increases, the dominance of absorption becomes even more pronounced, with SEA increasing steadily, while SER shows only a moderate rise. This behavior indicates that the polymer composite effectively dissipates incident electromagnetic energy within its bulk rather than reflecting it at the surface, which is a highly desirable characteristic for EMI shielding materials used in compact and sensitive electronic environments. The absorption-dominated shielding behavior observed in Figure 5 is a direct consequence of the magnetic nature of the fillers and their aligned, networked distribution within the polymer matrix [29]. The magnetically induced ferromagnetic networks introduce multiple energy dissipation mechanisms within the polymer

composite, including magnetic hysteresis loss, where repeated magnetization and demagnetization cycles convert electromagnetic energy into heat, and eddy current loss, which arises from circulating currents induced along interconnected magnetic filler pathways. These losses are significantly enhanced by the formation of continuous filler networks facilitated by magnetic field-assisted processing.

In addition to magnetic losses, interfacial polarization at the polymer–filler interfaces contributes to electromagnetic attenuation, as charge accumulation occurs at the interfaces between the insulating TPU polymer matrix and the ferromagnetic fillers. This interfacial effect becomes increasingly significant at higher frequencies, further strengthening the absorption component observed in Figure 5. The large interfacial area generated by the well-dispersed and aligned fillers amplifies this polarization loss within the polymer composite. The TPU matrix itself plays an active role in the absorption process. As an elastomeric polymer with polar functional groups, TPU contributes to dielectric loss through dipolar polarization, particularly under high-frequency electromagnetic excitation. The synergistic interaction between polymer dielectric losses and magnetic filler losses leads to efficient electromagnetic wave dissipation within the polymer composite. This synergy explains why SEA remains substantially higher than SER across the measured frequency range in Figure 5.

From a polymer composite design standpoint, the absorption-dominated shielding behavior demonstrated in Figure 5 is highly advantageous. Reflection-dominated shielding, often observed in highly conductive polymer composites or metallic shields, can cause secondary electromagnetic interference by reflecting radiation back into the environment. In contrast, the present TPU-based magnetic polymer composites minimize secondary EMI by converting electromagnetic energy into heat within the material itself. This makes them particularly suitable for applications in flexible electronics, wearable devices, and compact communication systems, where EMI suppression without reflection is critical.

Overall, the results presented in Figure 5 confirm that the combination of an elastomeric polymer matrix and magnetically aligned ferromagnetic filler networks enables a well-balanced polymer composite architecture, in which absorption-driven EMI shielding is maximized while reflection is effectively suppressed. This absorption-dominated mechanism underpins the superior EMI shielding performance observed in the present polymer composite system.

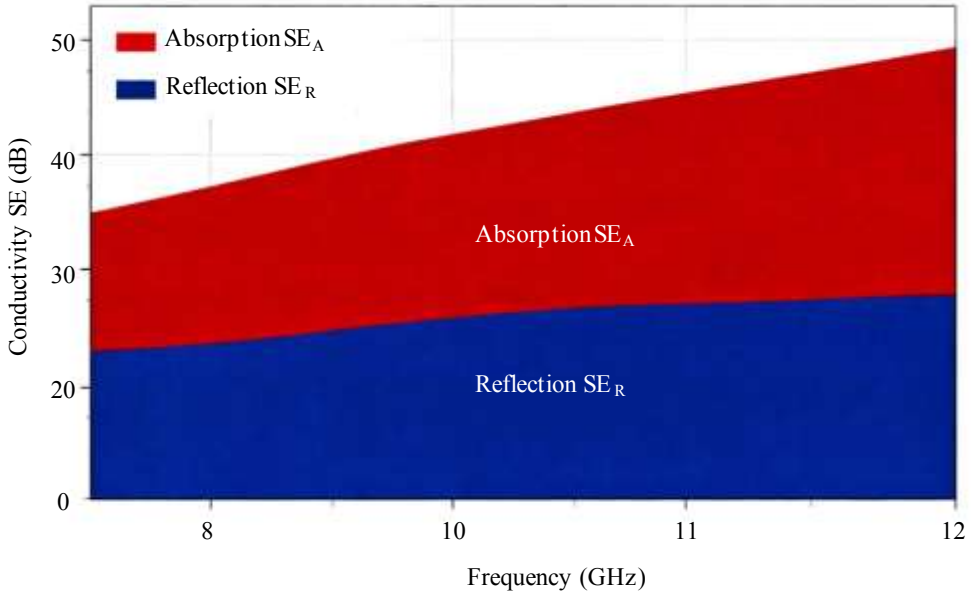

Figure 5. Contribution of absorption (SEA) and reflection (SER) components to the total EMI shielding effectiveness of TPU-based magnetic polymer composites.

Mechanical Performance and Flexibility Retention

The tensile stress–strain behavior of TPU-based magnetic polymer composites with varying filler contents is presented in Figure 6. The curves clearly demonstrate the influence of magnetic filler loading on the mechanical response of the polymer composite while highlighting the ability of the elastomeric polymer matrix to retain flexibility even at elevated filler concentrations. As shown in Figure 6, polymer composites with lower filler content (10 wt%) exhibit a characteristic elastomeric stress–strain response dominated by the TPU polymer matrix. These composites display lower tensile stress but very high elongation at break, reflecting the inherent flexibility and chain extensibility of TPU. In this regime, the polymer phase governs deformation, and the contribution of magnetic fillers to load bearing is minimal due to limited particle connectivity within the polymer composite. With increasing filler loading to 20 and 30 wt%, the stress–strain curves in Figure 6 show a pronounced increase in tensile stress and slope in the initial elastic region, indicating an increase in composite modulus. This stiffening effect arises from the incorporation of rigid ferromagnetic fillers and the formation of interconnected filler networks within the polymer matrix [30]. Importantly, despite the increased stiffness, the polymer composites containing 30 wt% magnetic fillers retain elongation values exceeding 300%, demonstrating that the TPU matrix effectively accommodates the rigid filler networks without catastrophic embrittlement.

The preservation of high elongation at break observed in Figure 6 is attributed to several polymer-composite-specific factors. First, strong polymer–filler interfacial bonding, enabled by silane surface modification, ensures efficient stress transfer from the polymer chains to the filler networks during deformation. Second, the networked filler architecture promotes uniform stress distribution throughout the polymer composite, reducing localized stress concentrations that typically lead to premature failure in conventional particulate composites. Third, the intrinsic elasticity of TPU polymer chains allows extensive chain stretching and reorientation under tensile loading, thereby maintaining flexibility even in highly filled systems. At the highest filler loading (40 wt%), Figure 6 reveals a further increase in tensile stress, confirming enhanced load transfer within the polymer composite due to denser filler networks. However, a noticeable reduction in elongation at break is also observed, indicating the onset of restricted polymer chain mobility at very high filler concentrations. Nevertheless, the composite still exhibits substantial deformability, underscoring the effectiveness of the elastomeric polymer matrix in mitigating brittleness typically associated with high filler loadings.

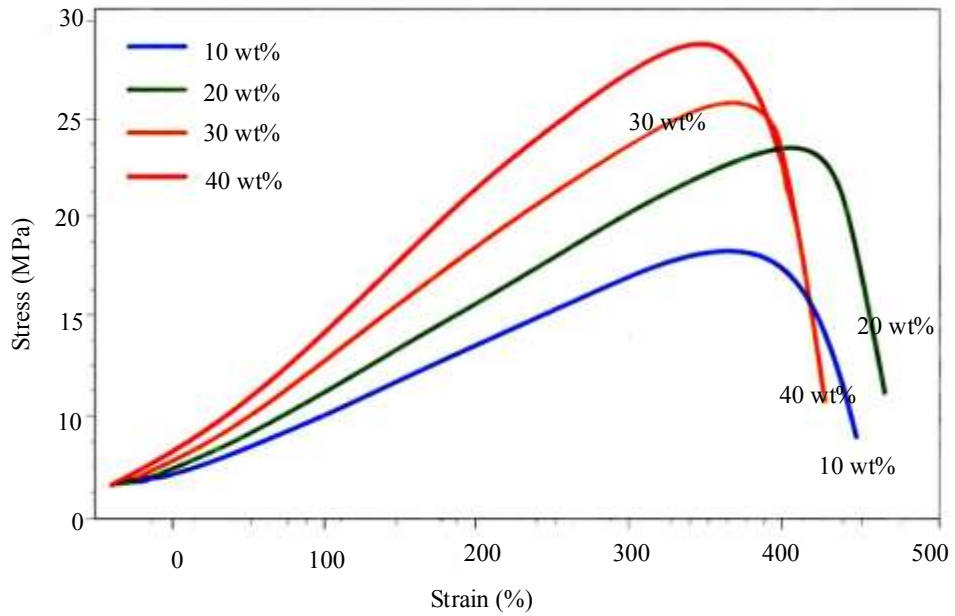

Figure 6. Stress–strain curves of TPU-based magnetic polymer composites with different magnetic filler contents.

From a polymer composite design perspective, the stress–strain behavior shown in Figure 6 confirms that aligned ferromagnetic filler networks not only enhance functional properties such as EMI shielding but also contribute positively to mechanical reinforcement. The aligned networks facilitate improved load transfer while preventing premature crack initiation and propagation during deformation. This dual functionality is critical for flexible EMI shielding applications, where mechanical durability and electromagnetic performance must coexist.

Overall, the results presented in Figure 6 demonstrate that the TPU-based magnetic polymer composites successfully balance stiffness and flexibility, achieving high mechanical compliance alongside enhanced functional performance. The retention of large elongation at break, even at filler loadings required for high EMI shielding effectiveness, highlights the suitability of these polymer composites for flexible electronics, wearable devices, and deformable shielding components.

Structure–Property Relationships in Polymer Composites

The collective results clearly establish that EMI shielding performance in magnetic polymer composites is governed not solely by magnetic filler content, but more critically by polymer composite architecture and microstructural organization. Magnetic field-assisted alignment fundamentally alters the internal structure of the polymer composite, transforming it from a randomly dispersed particulate system into a network-dominated functional composite. This architectural transition enables efficient electromagnetic attenuation at comparatively lower filler loadings by maximizing filler connectivity, magnetic interaction, and interfacial area within the polymer matrix. From a polymer science perspective, the TPU matrix plays a multifunctional and active role in determining composite performance. During processing, the elastomeric polymer acts as a flexible host that permits filler mobility and alignment under an external magnetic field, facilitating the formation of interconnected ferromagnetic networks. In the solid state, the polymer matrix functions as a dielectric medium, contributing to interfacial polarization and dipolar losses that synergistically enhance absorption-dominated EMI shielding [31]. Simultaneously, the polymer phase serves as a mechanical backbone, accommodating rigid filler networks while preserving flexibility and durability under tensile and bending deformation.

These structure–property correlations underscore the importance of polymer-centric composite design, where matrix selection, polymer–filler interfacial chemistry, and processing strategy collectively govern functional performance. In this context, the polymer matrix is not merely a passive binder, but a critical component that enables the effective translation of microstructural architecture into macroscopic electromagnetic and mechanical properties.

Comparative Performance with Reported Polymer Composites

Compared with recently reported CNT- and graphene-filled polymer composites, which often rely on high electrical conductivity and reflection-dominated shielding, the present TPU-based magnetic polymer composites achieve comparable or higher EMI shielding effectiveness through absorption-dominated mechanisms. While many conductive polymer composites suffer from reduced flexibility or secondary electromagnetic reflection, the magnetically aligned ferromagnetic networks embedded within the elastomeric TPU matrix enable efficient electromagnetic dissipation while retaining high mechanical compliance. Notably, shielding effectiveness exceeding 40 dB in the X-band is achieved at moderate filler loadings, alongside elongation at break values exceeding 300%, outperforming several recently reported flexible EMI shielding systems [31]. While carbon-based conductive composites often rely on high electrical conductivity and reflection-dominated shielding, they typically suffer from reduced flexibility, processing complexity, or secondary electromagnetic pollution. In contrast, the present polymer composite system achieves high EMI shielding effectiveness through absorption-dominated mechanisms, enabled by magnetically aligned ferromagnetic networks embedded within an elastomeric polymer matrix. Furthermore, the retention of high mechanical flexibility at filler loadings required for effective EMI shielding distinguishes this system from conventional rigid or brittle polymer composites. The solution-based processing combined with magnetic field-assisted alignment offers scalable and industrially compatible fabrication routes, further enhancing the practical relevance of the material system [32].

Overall, the comparative analysis highlights that ferromagnetic network engineering within elastomeric polymer matrices provides a unique pathway to simultaneously achieve high EMI shielding effectiveness, absorption-dominated attenuation, mechanical compliance, and scalable processability. This combination is rarely realized in traditional polymer composite systems and positions the present material architecture as a promising candidate for next-generation flexible and wearable EMI shielding applications.

CONCLUSION

This study demonstrates that achieving high-performance EMI shielding in flexible materials requires a polymer-centric composite design philosophy, where matrix selection, filler alignment, and interfacial engineering are carefully integrated. By coupling elastomeric polymer matrices with magnetically aligned ferromagnetic networks, it is possible to simultaneously optimize electromagnetic attenuation, mechanical flexibility, and process scalability—requirements that are rarely satisfied together in conventional polymer composite systems.

- Magnetic field-assisted alignment transforms TPU-based polymer composites into network-dominated architectures, significantly enhancing EMI shielding efficiency at reduced filler loadings.
- The elastomeric TPU matrix plays a multifunctional role as a flexible host, dielectric medium, and mechanical backbone, enabling both electromagnetic attenuation and mechanical durability.
- The polymer composites exhibit absorption-dominated EMI shielding, minimizing secondary electromagnetic reflection and making them suitable for sensitive electronic environments.
- Strong polymer–filler interfacial bonding and aligned filler networks promote efficient load transfer, allowing high elongation at break (>300%) even at functional filler contents.
- Compared with reported carbon- and metal-filled polymer composites, the developed system achieves a rare balance of high shielding effectiveness, mechanical flexibility, and scalable processing.
- Ferromagnetic network engineering within elastomeric polymer matrices offers a promising design strategy for flexible, wearable, and next-generation EMI shielding applications.

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