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Effect of Carbonyl Iron Particle Concentration on The Printability and Stiffness of Magnetorheological Elastomer Filaments

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Abstract

Magnetorheological elastomer (MRE) are increasingly valued for their field-responsive mechanical properties, making them ideal for adaptive components in automotive, aerospace, and vibration control systems. However, conventional mould-based fabrication methods limit design flexibility, produce excess material waste, and are unsuitable for complex or customized geometries caused challenges that have hindered broader industrial adoption. Although 3D printing offers a compelling alternative, research on printable MRE filaments remains limited, particularly regarding how carbonyl iron particle (CIP) content influences printability and mechanical performance. This study investigates the fabrication and characterization of MRE filaments using fused filament fabrication (FFF), incorporating CIP concentrations of 20 wt.%, 40 wt.%, 60 wt.%, and 80 wt.%. Filaments were extruded and printed into standardized geometries, then tested using Dynamic Mechanical Analysis (DMA) in tensile mode to assess viscoelastic behaviour. The results show that increasing CIP content leads to a progressive enhancement in mechanical stiffness, with Young's modulus rising from 14 MPa for pure Thermoplastic Polyurethane (TPU) to approximately 32 MPa at 80 wt.% CIP. Stress-strain analysis revealed steeper curves and higher load resistance at elevated particle loadings, while surface quality and print consistency were maintained up to 60 wt.% before minor defects emerged due to particle agglomeration. These findings confirm that FFF can produce structurally consistent, functionally tunable MRE and that CIP content is a critical parameter for tailoring mechanical properties. This work lays the groundwork for the development of 4D-printed MRE systems, offering new possibilities for smart, load-responsive devices in sectors such as automotive damping, adaptive infrastructure like railway tracks with tunable dampers, isolators and suspension systems in the automotive and aerospace industries.

Keyword: 4D printing; fused filament fabrication; magnetorheological elastomer; printability; carbonyl iron particles; mechanical stiffness

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INTRODUCTION

Magnetorheological elastomer (MRE) are a distinct class of smart materials capable of changing their mechanical behaviour such as stiffness and damping, in response to a magnetic field. This ability comes from the rearrangement of magnetic particles inside the elastomer when exposed to magnetic stimuli, allowing for rapid and reversible changes in mechanical performance [1–3]. These tunable properties make them highly suitable for applications that require real-time adaptability, especially in systems designed to control vibration. As a result, they are increasingly used in technologies like adaptive dampers, isolators, and suspension systems in the automotive and aerospace industries, where tailored mechanical response improves both functionality and user experience [4]. MRE also represent a broader trend in material science, moving from passive components to materials that interact intelligently with their environment.

Traditionally, these materials are made by mixing magnetic fillers, usually carbonyl iron particles (CIP), into elastic carriers like thermoplastic polyurethane (TPU) or synthetic rubber [5]. Selecting an appropriate filler is essential in polymer composite design [6–8], as it governs the interaction with the matrix, dispersion stability, and the resulting functional performance [9,10]. Among the various options, CIP are predominantly utilized in MREs due to their exceptional magnetic characteristics, including high saturation magnetization, low residual magnetism, softness, and high magnetic permeability. These properties make CIP highly effective in enhancing magnetic responsiveness and optimizing the overall performance of MRE [11]. Although this process is well understood, it depends heavily on conventional moulding techniques, which present several limitations. These include high material waste, slow production speeds, and the need for expensive, rigid moulds [12]. In addition, mould-based methods restrict geometric flexibility, making it difficult to produce custom or complex parts. Such constraints make conventional methods less compatible with modern demands for efficient, flexible, and sustainable manufacturing, particularly for applications that require fast prototyping or customized solutions. In contrast, additive manufacturing (AM) is commonly known as 3D printing has emerged as a promising alternative. It enables digital designs to be turned into physical objects through a controlled, layer-by-layer process, offering substantial freedom in shape and structure while minimizing waste and tooling requirements [6-7]. Among the various AM techniques, fused filament fabrication (FFF), or fused deposition modelling, is widely used due to its simplicity and cost-effectiveness, especially for processing thermoplastic-based composites [15,16]. Its compatibility with functional fillers like magnetic particles makes it a strong candidate for producing responsive elastomer-based systems [17]. By overcoming the limitations of moulding, this technique supports a shift toward more efficient, on-demand production of smart material components.

Still, incorporating MRE into this printing method presents notable challenges. Research by Al Rashid and Koç [18] points to ongoing issues with surface quality and bonding between layers, where both crucial to ensuring mechanical reliability. These difficulties largely stem from the uneven distribution of magnetic particles in the filament, which disrupts flow and leads to poor extrusion and weak structural connections [19]. These challenges are not solely attributable to suboptimal printer settings or equipment limitations but are inherently linked to the behaviour of highly filled composite systems. Uniform dispersion of CIPs is difficult to achieve due to strong inter-particle magnetic attractions that encourage agglomeration, particularly at higher loadings. Insufficient wetting by the polymer matrix or inadequate shear during mixing can further promote clustering, which disrupts flow uniformity, increases melt viscosity, and leads to surface defects in printed components. Therefore, achieving improved print quality necessitates optimization of both material preparation methods and printing parameters. To address these gaps, this study explores how different concentrations of CIP affect the print performance of MRE filaments using FFF. The work focuses on key print quality indicators for extrusion consistency, layer bonding, and surface finish, and aims to identify the optimal combination of material formulation and printing conditions. Although earlier efforts have shown that MRE can be printed, few have investigated in detail how filler content affects the entire printing outcome. The novelty of this study lies in its systematic evaluation of material behaviour across a range of particle loadings, helping to build a foundation for reliable processing. The insights gained are expected to support future development of 4D-printed components capable of responding dynamically to magnetic fields, marking progress in the integration of functional materials with advanced manufacturing.

METHODS AND MATERIALS

This section outlines the materials, formulation processes, fabrication techniques, and evaluation methods employed in the development and characterization of MRE filaments. A systematic approach was adopted to investigate the influence of CIP concentration on printability and mechanical behaviour. The methodology encompasses material preparation, extrusion processes, printability assessments, and tensile testing using dynamic mechanical analysis (DMA) to ensure a comprehensive evaluation of the filament performance.

Materials and Specifications

The materials used in this study include CIP as the magnetic filler, TPU as the polymer matrix, and tetrahydrofuran (THF) as the solvent. The TPU filament, characterized by a Shore hardness of 85A and a standard filament diameter of 1.75 mm, was sourced from Fabbxible (Malaysia). The TPU elastomeric matrix serves as the continuous phase that binds and encapsulates the CIP, providing structural cohesion and elasticity to the composite. Its inherent flexibility ensures that the filament remains pliable enough for feeding through the FFF system, while its thermoplastic nature supports repeated melting and solidification cycles. The CIP, with a particle size distribution ranging from 3.9 µm to 5.2 µm and a density of 7.874 g/cm^3, was procured from BASF SE (Germany). THF was employed to dissolve TPU due to its high volatility and compatibility with polyurethane systems, enabling effective dispersion of magnetic fillers in the composite matrix [20].

To explore the influence of magnetic content on filament quality, the MRE compositions were prepared with varying CIPs concentrations: 20, 40, 60 and 80 wt.%. These proportions were selected to capture a wide range of particle loadings, from moderately filled to near-saturation levels. THF was added to facilitate TPU dissolution in a solvent-to-polymer weight ratio of 8:1, allowing sufficient viscosity control and uniform mixing. The mass fraction of CIPs in each mixture was calculated using Equation (1):

$$\text{CIP (wt\%)}=((\text{Weight of CIP})/(\text{Wight of CIP}+\text{Weight of TPU}))\times 100 \quad (1)$$

The complete material breakdown by weight for each formulation is presented in Table 1, which includes calculated THF quantities required for consistent processing.

Filament Fabrication Procedures

The filament production process began by dissolving TPU pellets in THF under controlled stirring at 300 rpm and 50°C within a fume chamber. After full dissolution, CIPs were gradually introduced into the polymer solution according to the selected weight fractions. The mixture was stirred continuously until it achieved a uniform gel-like consistency. Following this, the mixture was cast into petri dishes and allowed to dry at ambient conditions for 16 hours to ensure complete solvent evaporation. The dried composites were cut into granules (0.5 - 1.0 cm) and thermally treated at 55°C for 2 hours to improve inter-particle bonding. The material was then extruded using a Noztek Pro single-screw extruder at 180°C, producing filaments with a target diameter of 1.75 ± 0.10 mm. The extrusion speed was regulated at 2.5 m/min. After extrusion, filaments were visually and physically inspected for uniformity, surface smoothness, and flexibility. Minor adjustments to nozzle temperature, feed rate, and bed adhesion methods were performed to enhance print stability, consistent with prior composite extrusion studies [5-11].

Table 1. Material composition for MRE filament fabrication.

Mixture composition (%wt)	TPU filament (g)	CIP content (g)	Tetrahydrofuran (g)
20%	16	4	128
40%	12	8	96
60%	8	12	64
80%	4	16	32

The printability of the resulting MRE filaments was assessed using three main criteria: extrusion stability, surface quality, and dimensional accuracy. Smooth filament flow through the printer nozzle was monitored to detect clogging or irregular extrusion, especially at higher filler concentrations. Surface inspection was conducted to evaluate roughness or particle-induced texture variations that may impact structural and functional quality. Dimensional accuracy was assessed by comparing printed part dimensions against the original CAD models to account for shrinkage, warping, or swelling due to varying particle content. To ensure experimental consistency, the following printing parameters were fixed throughout the study: nozzle temperature (240°C), bed temperature (60°C), print speed (30 mm/s), layer height (0.2 mm), and nozzle diameter (0.6 mm). These values were selected based on optimal settings for TPU-based materials and are consistent with AM studies involving filled composites [22].

Mechanical Testing via Dynamic Mechanical Analysis

To quantify the mechanical behaviour and validate the structural consistency of the printed MRE samples, tensile testing was performed using a Dynamic Mechanical Analyzer (DMA; Mettler Toledo, Switzerland) in tension mode. Stiffness was determined from Young's modulus, calculated from the initial linear region of the stress–strain curves, enabling precise evaluation of the elastic response and bonding quality across different CIP concentrations. Three different shapes; circular, rectangular, and dogbone, were printed, with rectangular samples chosen for the tensile test due to their consistent cross-sectional area and clamping ease. Each sample was measured using a precision vernier calliper and tested at 25°C under a constant load rate of 1 N/min. The setup aimed to simulate quasi-static tensile loading while capturing stiffness and bonding quality metrics across varying CIP concentrations. A total of 15 tests were conducted for each formulation to ensure statistical reliability. Young's modulus was derived from the stress–strain data to assess how particle content influences the elasticity and bonding performance of the printed composites [21].

RESULTS AND DISCUSSION

This section presents the outcomes of the printing trials, tensile testing, and mechanical characterization of the fabricated MRE filaments. The influence of CIP concentration on the print quality, stress-strain behaviour, and material stiffness was analysed to assess printability and structural performance. The findings are discussed in the context of their implications for future applications in load-bearing and smart structural components.

Print Quality Assessment of MRE Filaments

The printability of the MRE filaments was initially evaluated by fabricating simple geometries; circular, rectangular, and dogbone shapes, intended for subsequent mechanical testing. All filament compositions were successfully extruded and printed using the FFF process, indicating a baseline compatibility between the TPU matrix and the integrated CIP. For filaments containing low CIP concentrations (20 and 40 wt.%), extrusion was stable, and the resulting prints exhibited smooth surface finishes, uniform layer adhesion, and consistent geometry. However, as the CIP content increased to 60 wt.% and above, minor surface defects and irregularities began to appear. These included visible layer roughness and occasional filament inconsistencies, which became more pronounced at 80 wt.%. These effects are likely the result of particle agglomeration, which has been widely documented in magnetically active composite materials during extrusion-based printing processes. As Wei et al. [20] discuss, high magnetic filler content increases the risk of interparticle magnetic interactions, which can lead to poor particle dispersion and localized clustering during melt processing. Such agglomerates not only disrupt the flow of the molten filament through the nozzle but also cause uneven filament deposition and weak interlayer fusion. Moreover, Díaz-García et al. [21] highlight that polymer-based magnetic composites with high filler loadings often experience non-Newtonian rheological behaviour, characterized by increased viscosity and poor flow homogeneity. This adversely impacts layer bonding and surface finish during extrusion, especially when the matrix does not provide sufficient wetting or flow alignment of the embedded particles. The compounded effects of viscosity elevation, particle clustering, and interrupted flow dynamics can explain the surface roughness observed in the higher-CIP samples of this study.

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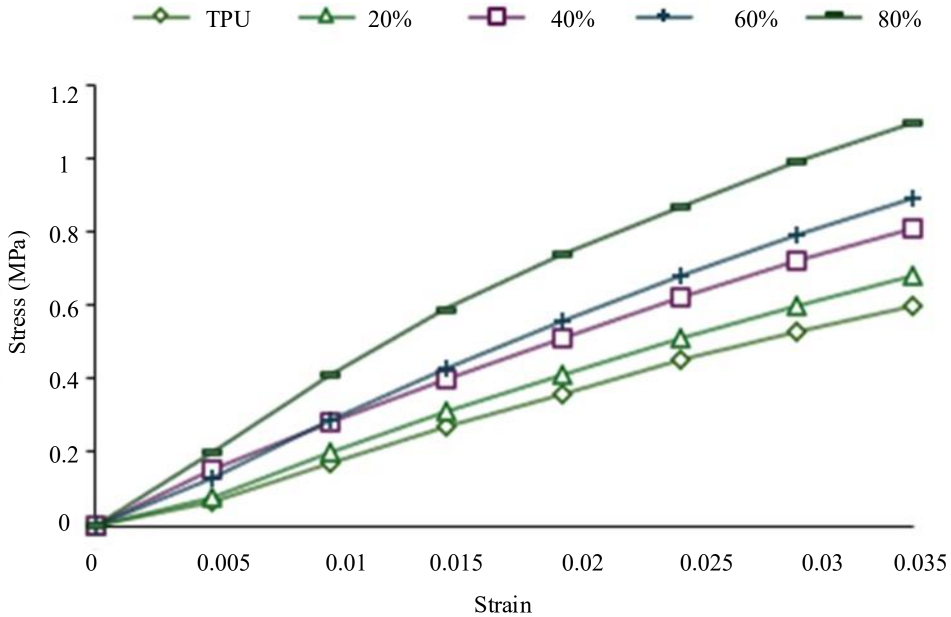

Figure 1. Stress–strain curves for MRE filaments with varying CIP concentrations.

Tensile Behaviour of Printed MRE Filaments

Tensile testing was performed to examine the mechanical response of the printed MRE filaments under uniaxial loading. The stress–strain curves for TPU and MRE samples with CIP contents of 20, 40, 60 and 80 wt.% are shown in Figure 1.

As expected, the neat TPU sample displayed the lowest tensile stress across the strain range, reflecting its flexible, unreinforced nature. Its gradual, linear response corresponds to elastic deformation behaviour commonly associated with thermoplastic elastomers. With the addition of CIPs, the mechanical stiffness and stress-bearing capacity of the filaments increased significantly. This trend is attributed to the introduction of rigid magnetic particles into the soft matrix, which act as load-bearing agents, reducing polymer chain mobility and enhancing stress distribution during deformation. Among the composite samples, the 80 wt.% CIP filaments exhibited the highest tensile stress, particularly at higher strain values, indicating a pronounced reinforcement effect. Similarly, the 60 wt.% sample showed a substantial increase in stiffness when compared to the 20% and 40% variants, suggesting that the particle network becomes more continuous and effective at higher loadings. At lower strains, the differences between samples were less prominent, implying that the initial elastic response is still dominated by the TPU matrix. However, as strain increased, the reinforcing effects of CIPs became more evident. The TPU sample showed the highest elongation while 20 wt.% CIP samples exhibited a steady and gradual stress increase with moderate elongation.

In contrast, the 60% and 80% CIP samples presented steeper stress–strain, indicating increased rigidity and reduced deformation ability. The 40 wt.% sample displayed intermediate behaviour, balancing stiffness with flexibility. This composition may be optimal for applications requiring a compromise between mechanical reinforcement and elastic compliance, such as in semi-flexible vibration damping systems. These results confirm that increasing CIP content systematically enhances the mechanical strength of MRE filaments. However, this enhancement comes at the cost of reduced flexibility and ductility, a trade-off commonly reported in magnetically filled elastomer systems. As Bastola and Hossain [23] highlight, higher particle loading restricts matrix mobility and may lead to stress concentration sites, thereby reducing elongation and deformability. Therefore, careful optimization of filler content is crucial, especially when designing materials for applications requiring a balance between mechanical rigidity and adaptability under load.

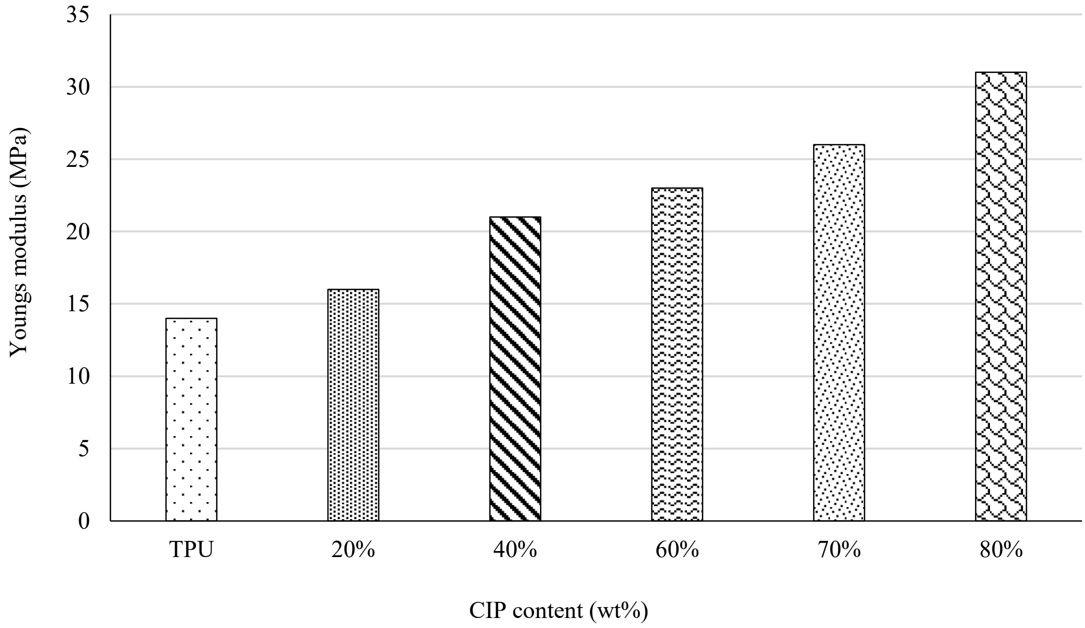

Figure 2. Young's modulus (MPa) of MRE filaments as a function of CIP concentration.

Modulus of Elasticity Analysis

To further understand the stiffness characteristics of the fabricated MRE filaments, Young's modulus was calculated based on the tensile test results. Figure.2 presents the relationship between CIP concentration and the corresponding modulus values, illustrating how increasing magnetic particle content affects the rigidity of the printed MRE.

The results reveal a clear upward trend in Young's modulus with the increase in CIP loading. The unfilled TPU sample recorded the lowest modulus at approximately 14 MPa, indicative of its soft and elastic nature. The addition of 20 wt.% CIP resulted in a moderate increase, suggesting that even a relatively low volume of rigid particles can begin to restrict the mobility of polymer chains and provide localized reinforcement. As the CIP content rose to 40 and 60 wt.%, the modulus increased more substantially, reflecting a shift toward a more rigid material system. This behaviour is consistent with typical composite reinforcement mechanisms where it can be attributed to improved load transfer between the matrix and the dispersed particles, which act as mechanical anchors that inhibit chain relaxation under tensile stress. Wei et al. [20] highlighted that in polymer-based magnetic composites, such reinforcement becomes more effective as particles start to form continuous or semi-continuous networks, enabling collective load-bearing behaviour.

At 70 and 80 wt.% CIP content, the modulus peaked at approximately 27 and 32 MPa, respectively. These values represent more than a two-fold increase over pure TPU and demonstrate a significant enhancement in stiffness. This is consistent with findings from Peng et al. [19], who reported that highly filled MRE exhibit considerable increases in mechanical rigidity due to particle agglomeration and percolation effects. At these higher concentrations, the physical contact between particles intensifies, forming reinforcing microstructures that dominate the deformation behaviour. However, while increased particle content improves stiffness, it also introduces notable trade-offs. Firstly, excessive CIP loading leads to particle agglomeration, a phenomenon driven by inter-particle magnetic interactions and insufficient dispersion during mixing. This causes local variations in stiffness and disrupts homogeneous deformation, by slight inconsistencies in modulus across samples at 80 wt.%. Secondly, high filler concentrations elevate the composite's viscosity, which adversely affects flow uniformity during extrusion and increases the likelihood of nozzle clogging or surface roughness in printed parts [6,10].

From a materials design perspective, the trend in modulus values demonstrates that mechanical stiffness in MRE can be effectively tuned through CIP content, offering a powerful tool for customizing functional properties. Under an applied magnetic field, the CIPs within the elastomeric matrix tend to align along the field lines, forming chain-like microstructures that restrict polymer chain mobility. This reorientation enhances load transfer efficiency and reduces material compliance, thereby increasing the apparent stiffness of the MRE structure. Such tunable stiffness can be strategically leveraged for different applications. As for example, higher stiffness may benefit vibration isolation or structural health monitoring, while moderate stiffness may be preferred in soft robotic actuators to preserve elasticity. However, this tunability should be applied with caution, as excessive stiffness may compromise flexibility, surface finish, and print quality. Therefore, the selection of filler content and magnetic field conditions should be optimized to achieve the desired balance between printability, structural integrity, and functional responsiveness.

CONCLUSION

This study successfully demonstrated the fabrication of MRE filaments using AM, specifically through FFF, and evaluated their mechanical behaviour across varying concentrations of CIP. The findings revealed that particle concentration has a substantial impact on the mechanical properties and printability of the filaments. Higher CIP loadings consistently enhanced stiffness and tensile strength, confirming the reinforcing effect of magnetic fillers. However, these gains were accompanied by minor challenges in extrusion stability and surface uniformity, particularly at the highest filler concentrations. The integration of 3D printing into MRE fabrication presents a promising pathway for producing complex, customized components with reduced material waste and improved design flexibility with advantages that traditional moulding methods cannot offer. This approach opens new opportunities for the development of responsive, load-bearing, and smart material systems tailored for use in automotive, aerospace, and robotic applications. Future research should focus on refining the balance between stiffness and flexibility by optimizing print parameters and exploring hybrid or multi-material strategies. In addition, long-term performance studies under operational environments are essential to assess the durability, functional stability, and responsiveness of printed MRE in real-world applications.

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