Sustainability in magnetic metal-carbon nanocomposites: a comprehensive review of manufacturing, characterization, and applications

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Abstract. Magnetic metal-carbon nanocomposites (MMCN) are emerging as sustainable materials, consisting of magnetic metals or alloys and carbon-based materials like CNT, graphene (Gr), carbon fiber (CF), and activated carbon. These materials possess unique magnetic properties that depend on various factors, such as preparation conditions, metal content, and phase composition. Incorporating carbon-based materials into magnetic metals has been observed to enhance their magnetic properties, including magnetic strength and moment. Researchers employ a range of tests to characterize these materials, such as FTIR, XRD, FESEM, TEM, BET, and VSM. Carbon-based materials such as CNT, graphene, etc., have been used as filler materials to reinforce the metal matrix because of their sustainability, tendency to integrate, and low cost. Further, they enhance the tribological performance and mechanical strength, provide corrosion resistance and improve electrical and thermal properties. Additionally, the addition of filler magnetic material in single or hybrid form into the carbon matrix increases the scope of application of MMCN. These composites are widely used in the application of biomedical, semiconductors, tribology, fuel cells, etc. In the present study, a comprehensive review has been carried out to provide a view of the fabrication aspect of the MMNC and to understand the role of the reinforcement method used to fabricate the composites. Finally, it covers different uses of the MMCN, which can lead to an eco-friendly environment.

Keywords: Nanocomposites; carbon allotropes; metal alloys; reinforcement; magnetic properties.

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1 Introduction

Magnetic material has gained its scope in various disciplines and has attracted many researchers in this field due to its potential in multiple applications [1]. The study of magnetic material was started in ancient Greek times, and people found out that lodestone has the capability to attract iron [2,3]. Later in the 19^{th} and 20^{th} centuries, many discoveries and findings were made, leading to new magnetic materials development [4,5]. These magnetic materials have found scope and usage in the fields of electronics, biomedical and energy generation instruments [6-8]. The MMCNs are among the magnetic materials which are composed of a combination of metals and reinforced carbon-based materials. This material provides excellent mechanical properties and is corrosion resistance [9, 10]. The combination of materials has emerged as facilities used in many of the latest technologies and applications [11]. The Carbon-based reinforcement includes graphene, CNT, carbon fibre and active carbon [12]. Apart from reinforcement content, MMCNS properties can also be influenced by the pretreatment methods and the material's magnetic characteristics [13, 14]. Many articles have pointed out that the addition of magnetic metals has more influence when compared with the addition of carbon-based materials [15-17]. Due to this, these composites are now used in the removal of heavy meals, dye adsorptions, and radiation-absorbing material [18–21].

In the last few decades, researchers have extended their scope to investigate the potential use of MMCNs and their synthesis methods [22]. Akbarbandari et al. [23] studied the synthesis of magnetic metal-organic framework (MOF) nanocomposites on magnetic activated carbon (MAC) to decrease methylene blue (MB) in water. The nanocomposites showed a core-shell structure and demonstrated significant magnetic potency. Multiple cost-effective methodologies were outlined to produce magnetic metal-encapsulated carbon nanobeads and nanotubes. Yakushko et al. [24] have synthesized the NiCo/C-based nanocomposites using IR heating and precursors of $NiCl_2$, $CoCl_2$, and Polyacrylonitrile (PAN). These nanocomposites displayed significant magnetic moments when rare-earth-metal elements were in contact with 3d transition metals, such as Fe and Cr. Tugirumubano et al. [14] have calculated the properties of Fe-Al-MWCNTs and Fe-Co-Al-MWCNT magnetic hybrid MMCs. It was found that adding MWCNTs, up to 2 vol%, did not affect the saturation magnetization of the nanocomposites. Incorporating Co nanoparticles in Fe-Al-MWCNT nanocomposites significantly improved their soft magnetic properties. This was achieved by reducing the coercivity and retentivity by up to 42% and 47%, respectively. However, it also decreased the electrical resistivity and mechanical transverse rupture strength.

Magnetic metal-carbon nanocomposites have unique magnetic properties and potential applications in various fields. There are many aspects on which the properties of these nanocomposites depend. Such aspects are conditions during fabrication, metal content, and phase composition. Past research has significantly explored the fabrication and characterization of magnetic metal-carbon nanocomposites and the applications that they might have across a range of fields, like heavy metal ion removal, dye adsorption, etc. The objective of this article is to give a short introduction to what magnetic metal-carbon nanocomposites are, discussing their fabrication method, followed by their characterization. The study also briefly discusses the application of these nanocomposites.

2 Magnetic properties of metal-matrix nanocomposites

The process opted for synthesis, the reinforcement material and amount, and the phase composition of the metal-matrix are the key factors that determine the magnetic characteristics of metal-matrix nanocomposites [25–27]. Magnetic metal alloys and composites can be used differently across various industries and fields. Their distinctive magnetic properties benefit them in numerous applications, like aerospace equipment, bearings, sensors, actuators, refrigeration systems, magnetic storage devices, and high-performance magnets [28, 29]. The critical parameters to measure a material's magnetic characteristics are permeability, coercivity, and saturation magnetization. Here, coercivity refers to the magnetic field strength required to reduce the magnetization of a material to zero after it has been magnetized. It measures a magnetic material's resistance to demagnetization and is usually measured in Oersted (Oe). It is calculated using a magnetometer or vibrating sample magnetometer (VSM) by subjecting the material to an increasing magnetic field until it demagnetized [30]. The coercive strength of a material is defined as the magnetic field required to demagnetize the material, which is used to calculate its coercivity. Furthermore, the upper limit of the magnetic moment represents the saturation magnetization when the material is placed in a magnetic field. This magnetic moment is measured with the help of VSM and SQUID, which measure the maximum magnetic moment per unit volume until the material reaches its saturation [31]. Permeability is a dimensionless quality that checks the material's capability to conduct the magnetic flux. It is calculated by applying an external magnetic field, and by using VSM or permeameter, the magnetic flux density is calculated [11].

3 Principle

The hysteresis loop is a magnetization curve that establishes a relationship between magnetic field intensity and magnetic moment, which depicts the relationship between the applied magnetic field (H) and magnetic moment (M). Figure 1(a) shows a typical hysteresis loop. Various parameters can be calculated through this curve, including coercive strength, remanence, saturation magnetization, etc. Analysing this curve allows researchers to understand the magnetic material in detail [32].

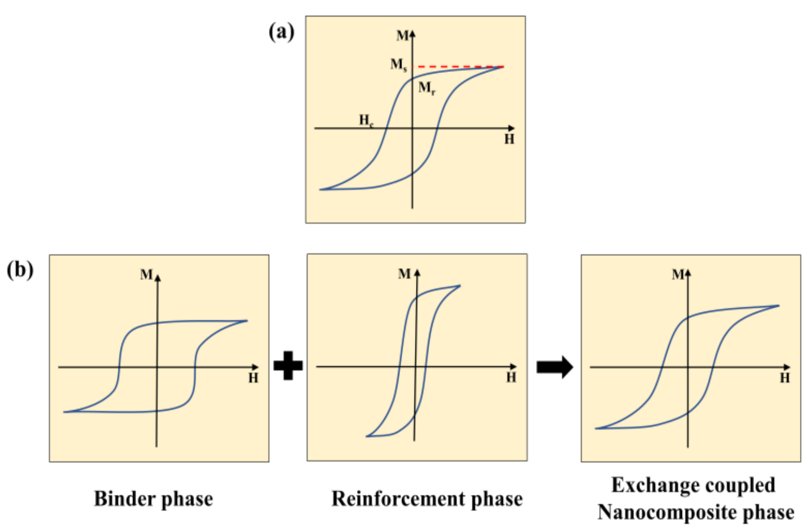

Fig. 1. (a) A typical hysteresis loop. (b) Superimposition of hysteresis loop.

Exchange coupling is a powerful tool to adjust the magnetism of nanosystems for various applications, especially in designing and developing magnetic nanocomposites [33]. The distribution of reinforcement should be highly homogeneous in the nanocomposites, which comprise binder and reinforcement components. The K (or Hc) values can be tailored by controlling the magnetic elements. According to a theoretical study [34], Hc of the nanosystem can be expressed as,

$$H_c = \frac{K_b f_b + K_r f_r}{M_b f_b + M_r f_r}. \tag{1}$$

Where f is the volume fraction, subscripts b and r denote the binding and nanofiller phases. The hysteresis loop for nanocomposite is illustrated in Fig. 1b. Different applications of magnetic materials in various industries:

- Magnetic storage devices, such as hard disk drives and magnetic tape.
- Magnetic sensors and actuators used in various industries, including automotive, aerospace, and medical.
- Magnetic refrigeration, which has the potential to be more energy-efficient and environmentally friendly than traditional refrigeration methods.
- Magnetic bearings can reduce friction and improve energy efficiency in rotating machinery such as turbines and motors.
- Electromagnetic shielding, which is used to protect sensitive electronic equipment from electromagnetic interference.
- High-performance magnets are used in various applications such as electric motors, generators, and speakers.
- Aerospace applications, including satellite components and spacecraft propulsion systems.

Table 1. Various magnetic metal-carbon nanocomposites and their applications.

Metal type	**Form of carbon allotropes**	**Magnetic Properties in terms of magnetization**	**Application**	**Ref.**
Cu	Graphene/$CoFe_2O_4$	489 emu/cm^3	Dye absorption	[3]
Fe	Carbon nanorod	30 emu/ g	Chemical oxidative catalyst in water	[12]
Fe-Al	MWCNT	100 emu/cm^3	Magnetic cores for electromagnetic devices	[14]
Fe	MWCNT (oxidized)	40 emu/cm^3	MRI contrast agents	[15]
Ni	CNT	300 emu/cm^3	Hydrogen storage and EM absorption	[16]
Cu-Co-Ni	Graphene	-	Electromagnetic shielding	[17]
Co	Graphitic intercalated compound (GIC)	600 emu/cm^3	Data storage device	[36]
Fe	Graphene	224 emu/cm^3	Ion separation	[37]

Magnetic metal alloys and composites can be used in radar-absorbing materials [35], especially in military applications. The properties of the nanocomposites, such as magnetic, electromagnetic, and radar-absorbing properties, are directly dependent on their synthesis process and composition.

The various magnetic metal-carbon nanocomposites and their applications are shown in Table 1. These nanocomposites have a wide range of applications, including ion separation, dye absorption, MRI contrast agents, electromagnetic shielding, hydrogen storage, data storage, and magnetic cores for electromagnetic devices. The table shows the metal type, form of carbon allotropes, magnetic properties in magnetization, and their respective applications [36, 37]. This information can be helpful for researchers and engineers working on developing new materials for various applications

3.1 Fabrication of MMCs

Magnetic metal-carbon nanocomposites have been studied due to their unique magnetic properties and potential applications in various fields. Studies have investigated different synthesis and characterization of these nanocomposites [38]. The fabrication methods of MMCs depend on the type of nanofillers, distribution type, quantity, and ratio of reinforcement in the metal matrix.

3.2 Processing temperature-based MMC fabrication methods

MMC fabrication methods can be classified based on the processing temperature of the metal matrix during the reinforcement of nanofillers, as shown in Fig. 2. The three processing temperature-based methods are liquid-state, two-phase (solid-liquid) processes, and solid-state. Liquid-state methods include stir casting [39], infiltration [40], and squeeze casting [41]. Two-phase methods include Osprey deposition [42] and composting [43]. Solid-state methods include diffusion bonding [44] and powder metallurgy [45]. The powder metallurgy process involves ball milling [46] and modified electrochemical co-deposition [47, 48], a more facile and fast method for fabricating MMCs.

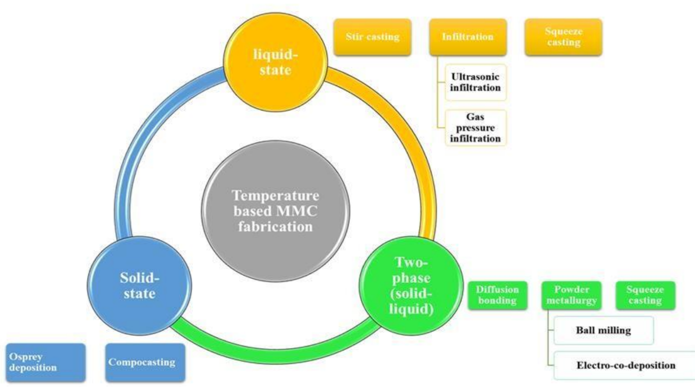

Fig. 2. Temperature-based MMC fabrication.

Powder metallurgy has been identified as the most suitable and cost-effective method for producing zinc-based composites. However, the mechanical processing involved in this technique, such as ball milling, can cause damage to the nanosheet carbon structure of graphene nanofillers. To address this issue, a modified electrochemical co-deposition method has been proposed. This method can uniformly distribute the reinforcement in the metal matrix for powder metallurgy without compromising the integrity of the graphene nanosheet structure [49]. This development holds promise for selecting a fabrication method for the next generation of graphene-based composite coatings and structural materials.

3.4 Sophisticated fabrication methods for magnetic MMC coatings

Coatings are widely used in magnetic MMC fabrication to modify and enhance surface properties such as corrosion resistance, wear resistance, toughness, microhardness, and biocompatibility. Advanced coating methods like chemical vapor deposition (CVD) [50], physical vapor deposition (PVD) [51], direct vapor deposition (DVD) [52], self-propagating high-temperature method (SHS) [53], laser-cladding [54], electro-less deposition [55], and electro-co-deposition [56] are commonly used. Spraying techniques like cold-spraying [57], thermo-plasma [58, 59], plasma-transferred wire arc [60, 61], and high-velocity oxy-fuel (HVOF) [62] are also used. However, cracks, distortion, delamination, substrate contaminations, and variations in physical properties can pose significant challenges during coating in magnetic MMC fabrication. The detailed descriptions of sophisticated fabrication methods for magnetic MMC coatings, including working, advantages-limitation, sophistication level, and possible application, are shown in Table 2. The electro-co-deposition technique is a low-cost, facile, moderately sophisticated method for co-deposing magnetic composite material on metallic surfaces.

4 Reinforcement patterns

Reinforcement patterns for magnetic MMCs can vary depending on the desired properties and performance of the final material [63]. However, some common reinforcement patterns for magnetic MMCs include [64, 65]:

1. Random reinforcement: This pattern randomly distributes magnetic particles throughout the metal matrix. As the reinforcement particles have a random distribution, these patterns are more accessible to achieve and eventually result in isotropic properties. The disadvantage of random allocation is that it may not provide the desired performance in specific applications.
2. Aligned reinforcement: In an aligned reinforcement pattern, the magnetic particles in the metal matrix are oriented in a particular direction. Due to a specific alignment of the magnetic properties, this pattern leads to anisotropic properties. Thus, this reinforcement pattern can be utilized in applications that demand directional properties, like magnetic shielding.
3. Layered reinforcement: A metal matrix contains multiple layers of magnetic particles in such a pattern. This pattern provides enhanced properties, like improved strength and stiffness, but these patterns can be challenging to synthesize.

The selection of reinforcement patterns is a crucial factor in determining the properties and intended use of the final material. The desired characteristics and application of the material play a significant role in deciding which reinforcement pattern to choose.

Table 2. Advanced fabrication techniques for magnetic metal-matrix composite coatings: Working, advantages limitations, the degree of complexity, and applications.

MMC coating method	Working	Advantages	Applications	Ref.
Dipping, brushing, cold spray, and handroll painting.	A mixture of solutions, resins, composite materials, and additives are painted on the surface	Offers organic and inorganic coatings Provides a simple and fast method Can be used with large, heavy structures	Surface coating of magnetic materials	[57]

Electro-co-deposition	Metal ions are reduced on a cathode substrate in an electrolyte bath to form a coating or powder	Offers a low-cost method Provides controlled thickness Can be used with a variety of nanofillers	Production of magnetic thin films and coatings	[66]
Sol-gel method	Precursors form a suspension that can gel on the substrate surface	Can be used at room temperature Offers controlled composition Can be used with organic and non-inorganic coatings	Surface coating and functionalization of magnetic nanoparticles	[67]
Spray based coatings	Nanofillers of composite materials are sprayed onto heated surfaces	Produces micro-sized structures Offers control over phase structure and thickness	Production of magnetic coatings, thermal barrier coatings for magnetic materials	[68]
Vapour deposition: atomic, physical, and chemical	The constituents are evaporated or vaporized under high vacuum and condensed on the surface	Produces coatings with uniformly distributed, dense compositions Can be used with organic and non-inorganic coatings	Production of magnetic thin films and coatings	[69]

4.1 Reinforcement of magnetic metal matrix

The properties needed in the final material and its eventual application in the real world determine the suitable choice of reinforcement pattern. Some are chosen for the improvement they provide in the tribological properties like improved wear and tear resistance, some for their ability to enhance tensile strength, and some for the improved electrical or magnetic properties they introduce in the metal matrix. Magnetism also becomes essential in some applications, like making magnetic data storage materials or permanent magnets. Different carbon-based reinforcements widely used in magnetic metal-matrix applications are listed in Table 3 below. The information in the table below explains the type of reinforcement used for specific magnetic applications and how the reinforcement helps achieve the desired application. Understanding the properties of various carbon-based reinforcement materials and their potential applications is very important for manufacturers and researchers to choose the correct material to manufacture products or composites with specific properties and characteristic performance.

Advanced carbon allotrope reinforcement, such as Graphene and carbon nanotubes, possess significant potential in improving those material properties that are application-dependent, such as their magnetic properties. For instance, graphene is a perfect reinforcement material due to its remarkable thermal, electrical, and mechanical properties. It can be added to magnetic materials like cobalt or iron to improve their magnetic properties by enhancing the material's magnetic anisotropy and decreasing its coercivity. It enables us to achieve high magnetic energy density materials. Such materials have valuable applications in magnetic refrigeration and energy storage. Apart from graphene, carbon nanotubes (CNTs), on the other side, exhibit outstanding electrical conductivity and excellent tensile strength. They can be added to a polymer matrix to enhance the resulting composite's electrical conductivity and mechanical. CNT reinforcement can be helpful in situations where high electrical conductivity material is needed, like shielding against electromagnetic interference.

In general, advanced carbon allotrope reinforcement can also improve other application-reliant properties of materials, such as thermal conductivity, specific surface area, and gas adsorption capacity. By tailoring the properties of the reinforcement material to the particular application requirements, it is possible to develop materials with enhanced performance and functionality.

Table 3. Various Carbon-based reinforcement materials for magnetic metal-matrix composites: their properties and applications.

Reinforcement type	Application	Role in magnetism	Purpose of reinforcement	Properties	Ref.
Carbon fibers	Electromagnetic interference (EMI) shielding	Conducts electricity	Shields against EMI	High tensile strength, lightweight	[70]
Graphene	Magnetic sensors, data storage	Enhances magnetic properties	Increases magnetic properties of the matrix	High surface area, high electrical conductivity	[71]
CNTs	Permanent magnets, magnetic sensors	It provides a strong magnetic field	Increases magnetic properties of the matrix	High aspect ratio, high tensile strength	[15]

5 Future scope and implications

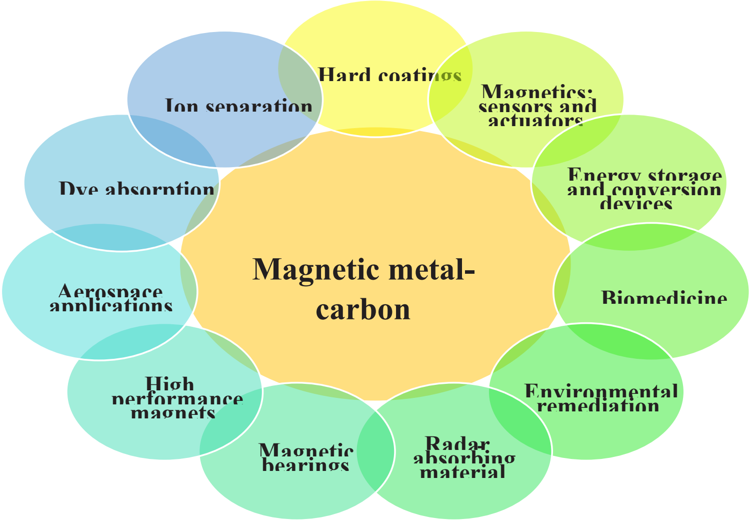

Fig. 3. Future scope and implications of magnetic metal-carbon nanocomposites.

Magnetic metal-carbon nanocomposites have a wide range of applications, including hard coatings, magnetics, energy storage, and conversion, and have shown great potential in fields such as biomedicine and environmental remediation [72, 73], as illustrated in Fig. 3. The magnetic properties of carbon-metal nanocomposites are influenced by various factors such as preparation conditions, metal content, and phase composition. With advancements in synthesizing magnetic nanoparticles of metals and alloys, the potential use of these materials in radar-absorbing applications has increased. Nanocomposites consisting of multi-walled carbon nanotubes and polyaniline, combined with magnetic metal oxide nanoparticles, can catalyze the oxidation of peroxidase substrates. Combining magnetic and photoluminescent properties, multifunctional nanocomposites offer significant benefits for applications in nanomedicine. A recent study has found that the addition of carbons to Fe_3O_4@ZnO magnetic metal-carbon nanocomposites resulted in a decrease in the degree of crystallinity and a broadening of the diffraction peaks. On the other hand, elastomer nanocomposites made of carbon nanofiber decorated with iron oxide have been shown to possess excellent thermal, mechanical, and magnetic properties at a low reinforcement volume fraction. Not much has been studied in MMCs, leaving much scope for further study and advancement. One such scope is the development of various new fabrication processes of metal matrix composites, which impact the quality and desired properties of the MMCs. Others use various magnetic metals and carbon reinforcements to fabricate MMCs to achieve different enhanced properties. Optimizing MMCs' composition and structure can also improve performance across multiple applications. Also, the commercialization and broad application of MMCs depend on developing scalable and affordable fabrication techniques. Green synthesis techniques, sustainable material usage, and sustainable development processes can help us prepare ecological and environmentally- safe MMCs. Thus, in a nutshell, magnetic metal-matrix nanocomposites have a shining bright future with applications across fields. With further study and research into significant gaps in this field, new composite materials with enhanced features can be created with various uses across domains.

6 Conclusion

This review discusses the magnetic metal-carbon nanocomposites' unique magnetic properties and potential applications in various fields. The conclusions drawn from the discussion are as follows:

1. The preparation conditions, metal content, and phase composition affect magnetic metal-carbon nanocomposite properties.
2. Adding advanced carbon allotropes such as graphene, CNT, and carbon fibres to metal matrix can affect their degree of crystallinity. Also, the decoration of carbon allotropes can improve the metal matrix's thermal, mechanical, and magnetic properties.
3. Multicomponent 3d transition metal-based binding matrix and reinforcement in MMCs can further improve magnetic properties and other application-reliant properties.
4. Magnetic metal-carbon nanocomposites have been characterized in various studies using advanced characterization techniques such as XRD, FTIR, FESEM, TEM, BET, and VSM for metallurgical and magnetic characteristics.
5. The Electro-co-deposition technique was found to be a low-cost, facile, and scalable method to reinforce advanced carbon allotropes into the metal matrix to prepare magnetic metal-carbon nanocomposites.

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