Combinatorial surface coating and greatly-improved soft magnetic performance of Fe/Fe₃O₄/resin composites

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HIGHLIGHTS

- Fe/Fe₃O₄/resin composites were fabricated by combinatorial coating methods.
- Fe₃O₄ nanolayers can reserve high saturation magnetization for the composites.
- Uniform resin coating promoted the reinforced structures of the composites.
- High density and high TRS values were achieved in the compacted composites.
- Good soft magnetic properties were obtained in the Fe/Fe₃O₄/resin composites.

GRAPHICAL ABSTRACT

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ABSTRACT

Structure-reinforced Fe/Fe₃O₄/resin soft magnetic composites (SMCs) have been fabricated through coating micron-sized Fe powders with high-purity Fe₃O₄ nanolayers and silicon resin films by combinatorial controlled oxidation and physical coating method. High magnetization M_s of 201 A m² kg⁻¹ can be reserved in the Fe/Fe₃O₄/resin SMCs due to high-purity ferrimagnetic Fe₃O₄ shells and low-content resin layers. The smooth Fe/Fe₃O₄ interfaces and uniform silicone resin coating layers in the Fe/Fe₃O₄/resin SMCs also lead to their low coercivity of 11.4 Oe and high compressed density of 7.50 g/cm³. For traditional metal-based SMCs fabricated by various common coating methods, their reduced core loss is usually achieved but accompanying with low magnetic induction and poor mechanical properties. In this work, high transverse rupture strength value of 92 MPa and greatly enhanced soft magnetic performance with both decreased core loss in accompany with high magnetic inductions are achieved for the fully densified Fe/Fe₃O₄/resin SMCs due to their well-structured composite interfaces, which promise their great latent applications for high-power and low-loss magnetic components.

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Fig. 1. The schematic illustration of preparation for Fe/Fe₃O₄/resin composites.

1. Introduction

High-performance metal-based soft magnetic composites (SMCs) have consistently received much research interests due to the improved overall properties containing decreased core loss, desirable frequency-dependent magnetic performance and good processibility for the composites, which can be used for various high-power or low-loss magnetic components with complex shapes [1–4]. In the past few decades, many metal-based SMCs with core-shell structure have been prepared by coating Fe or Fe-based alloy particles (FeSi, FeCo, FeSiAl, FeNi and amorphous alloys) as the cores with high-resistivity or electrical insulating layers (epoxy resin, SiO₂, MgO, NiZnFe₂O₄, Al₂O₃, TiO₂, ZrO₂ and SiO₂–Na₂O–CaO silicate) as the shells by several chemical or physical methods [5–14]. Commonly, these metal-based SMCs can exhibit improved soft magnetic performance such as reduced core loss, improved frequency magnetic performance and three-dimensional isotropic ferromagnetic behavior due to the suitable coating layers. Especially, the SiO₂-coated iron submicrometer laminates show decreased core loss accompanying with greatly-improved high-frequency magnetic performance under a maximum applied frequency reaching 50 MHz [15]. However, decreased core loss is often achieved in these conventional core-shell structured SMCs by seriously decreasing their magnetic inductions because of low density of nonmagnetic coating layers [16–18]. Moreover, the most current researches on SMCs have always focused on the improvement of their soft magnetic properties, but very few studies have involved their mechanical properties. As is known, the bulk SMCs are generally obtained by directly compressing the initially prepared metal-based composite particles at room temperature but not by sintering at high temperature due to their poor heat resistance [19–21]. Thus, low interfacial bonding strength would occur between the cores and the coating layers, and deformation inconsistency also exists between the two parts in metal-based composite particles, both of which can lead to their poor compactness so as to result in low density and low transverse rupture strength (TRS) in bulk SMCs [22,23]. Most of recent research for metal-based SMCs has been still focused on the enhancement of their magnetic properties by decreasing their core loss while maintaining their high saturation magnetization, which can be successfully realized by optimizing various metal-based core-shell interface structures fabricated through various coating methods [24–28]. However, the mechanical properties for full-density metal-based SMCs also need to be improved. Thus, it is necessary to explore suitable preparation method to fabricate high-performance SMCs with the ideal composite structures, which can result in their both good magnetic and mechanical properties to accord the requirement for practical applications.

In this work, the Fe/Fe₃O₄ core-shell structures have been firstly fabricated through coating micron-sized Fe powders with high-purity Fe₃O₄ nanolayers by controlled oxidation. The reduced core loss along with high magnetic inductions can be obtained in the Fe/Fe₃O₄ SMCs due to the ferrimagnetic Fe₃O₄ nanolayers with relatively high electric resistivity. Then, suitable amount of silicone resin is coated on the initially oxidized Fe-based composites to form new-type Fe/Fe₃O₄/resin SMCs, which can exhibit high compaction density, greatly-improved soft magnetic performance and high TRS value due to the combinatorial

Fe₃O₄ and resin coatings, which promote their practical applications for many metal-based magnetic devices including particularly the complex-shaped motors or generators. The interfacial structures accompanying with their influences on magnetic and mechanical properties of the Fe/Fe₃O₄/resin SMCs have been also investigated systematically.

2. Experimental

2.1. Fabrication of Fe/Fe₃O₄/resin SMCs

The commercial high-purity Fe powders with average particle size of approximately 170 μm (bought from Titt Metal materials Co., Ltd, China) used as the raw materials. The commercial silicon resin solution containing 50 wt% solid silicone resin dissolved in toluene solvent was supplied by Shin-Etsu Chemical Co., Ltd. Japan.

The Fe/Fe₃O₄ core-shell composites were firstly prepared by properly oxidizing the micron-sized Fe powders under an appropriate controllable oxidation condition, and then the Fe/Fe₃O₄/resin SMCs were obtained by coating the properly oxidized Fe particles with suitable amount of silicon resin through a physical coating method. Fig. 1 displays the schematic preparation process for Fe/Fe₃O₄/resin SMCs. The main reaction is illustrated as followed:

Firstly, high-purity Fe₃O₄ shells with 60–80 nm thickness are in situ formed on the surfaces of unoxidized Fe parts to generate Fe/Fe₃O₄ core-shell structured composites under the controlled oxidation based on formula (1). The detailed controlled oxidation process for metal particles can refer to our previous work in Ref. [9]. In this work, only surface ferrimagnetic Fe₃O₄ phase excluding the other Fe oxides such as FeO, Fe₂O₃ and their mixtures is expected to form the micron-nano Fe/Fe₃O₄ composites with core-shell structure, which can result in their increased electrical resistivity along with good intrinsic magnetism and thus to improve the soft magnetic properties of the composites. In a typical controlled oxidation procedure, a certain amount of high-purity micron-sized Fe particles were loaded into a tubular furnace under a controlled atmosphere, and then were quickly heated at a temperature rate with 8 °C/min from 25 °C to 450 °C at a vacuum of 1.0×10^{-3} Pa. When the temperature approached 450 °C, the gas mixture containing high purity oxygen and argon gas with their volume ratios fixed at 2:8 was quickly pumped into the furnace, which was then sealed until the pressure for mixed gas approached 1 atm. Afterwards, the micron-sized Fe powders were heated continuously beyond the mixed oxygen-argon atmosphere at 450 °C for 50 min. The Fe/Fe₃O₄ composites were obtained when the continuously heated Fe particles were naturally cooled from 450 °C to room temperature. Secondly, the above properly oxidized Fe particles were put into a silicon resin solution diluted with certain amount of acetone and then quickly stirred until all the solvents in the solution completely evaporate to obtain the sample with wet surface. The Fe/Fe₃O₄/resin SMCs as the end product can be obtained by naturally drying the wet sample in the air for 24 h. The solid silicon resin content is 0.8% of the total mass of the products.

Fig. 2. Typical XRD patterns for original micron-sized Fe powders (a), the properly oxidized samples fabricated at 450 °C for 50 min (b) and the composites coated with silicon resin (c).

2.2. Material structure characterization

A D/max 2500 PC X-ray diffractometer (XRD, Cu K α with $\lambda = 1.5406$ Å) was used to examine the phase structures for the samples. The morphologies and microstructures for the samples were investigated using a JEOL JSM-7500F field emission scanning electron microscope (FESEM) along with a JEOL JEM-2100 transmission-electron microscope (TEM). A focused ion beam (FIB) technique was used for preparing the TEM samples of Fe/Fe₃O₄ composites for cross-section observation. The surface elemental states for the samples were detected by an X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi).

2.3. TRS and magnetic measurements

The TRS values for the fully compacted samples were measured by a three-point bending test using a compression testing machine. A LakeShore-7410 vibrating sample magnetometer (VSM) was used for measuring intrinsic magnetic properties for the samples under a maximum applied magnetic field reaching 10 kOe at room temperature. To measure the alternating-current (ac) and direct-current (dc) soft magnetic performance for the composites, the densified toroidal products with a 20-mm outer diameter, a 10-mm inner diameter and a 3.6-mm sample thickness were prepared by directly compressing the Fe/Fe₃O₄/resin composites at a pressure of 1200 MPa with no any lubricants. An Archimedes method was used to measure the density for the fully compacted products. The total core loss for toroidal samples tested at frequencies of 50–1000 Hz under an applied maximum induction with 1 T and their magnetic inductions tested at magnetic fields of 1200–10000 A/m were measured using an ac and dc test equipment for measuring magnetic hysteresis loops of soft magnets (provided by the National Institute of Metrology, China). A precision impedance analyzer (Agilent 4294A) was used to measure frequency-dependent magnetic characteristics of the products at a maximum applied frequency reaching 200 kHz.

3. Results and discussion

Fig. 2 presents the typical XRD patterns for the original micron-sized Fe powders, the properly oxidized products and the composites coated with silicon resin. From Fig. 2a, the original Fe powders display obvious XRD diffraction peaks located at three diffraction angles of 44.6°, 65.2° and 82.3°, which is in good agreement with the (110), (200) and (211)

Fig. 3. Fe 2p XPS narrow-scan spectrums for original micron-sized Fe powders (a) and properly oxidized products fabricated at 450 °C for 50 min (b).

crystal planes of the single bcc-Fe phase. From Fig. 2b and c, the XRD diffraction peaks of the silicon resin-coated oxidized particles are almost the same as those of the initially-oxidized particles due to the low resin silicon content (0.8 wt%) in this resin-coated sample, which shows that no further oxidation occurs during the resin coating process. For these two products, apart from the bcc-Fe characteristic peaks, all the remaining peaks are well indexed to a fcc-Fe₃O₄ phase, the lattice constant for which (8.3961 Å) nearly approaches that (8.3963 Å) for bulk Fe₃O₄ [29]. This indicates that pure Fe₃O₄ layers are in situ formed on the surfaces of unoxidized Fe parts by oxidizing high-purity Fe powders under a suitable oxidation environment. As discussed below, both high saturation magnetization and decreased core loss are achieved in these two Fe-based SMCs containing high-purity Fe₃O₄ layers due to their ferrimagnetism and relatively high resistivity.

As is known, the Fe₃O₄ phase exhibits almost the same XRD patterns but different XPS spectrums with those of γ -Fe₂O₃ phase [30,31]. The Fe chemical states for the initially-oxidized sample and original high-purity Fe powders have been examined with their XPS spectrums illustrated in Fig. 3. From Fig. 3a, two obvious Fe 2p XPS peaks (Fe 2p_{3/2} located at ~706.6 eV accompanying with Fe 2p_{1/2} located at ~719.8 eV) for the original Fe particles shows a pure Fe state [32]. An obvious broad bump located at ~711.3 eV in Fig. 3a is probably related to Fe²⁺ ions [33,34], which is attributed to the very weak surface Fe oxidation for the original materials. Furthermore, only two obvious Fe 2p XPS peaks located at ~723.8 eV for Fe 2p_{1/2} and at ~710.3 eV for Fe 2p_{3/2} observed in Fig. 3b for the oxidized product indicate the presence of high-purity Fe₃O₄ but no formation of γ -Fe₂O₃ phases on the surfaces of this initially-oxidized sample [35]. The XPS results for the oxidized products accord well with the above XRD investigation and further proved the in-situ formation of single-phase Fe₃O₄ layers on the surfaces of high-purity Fe powders processed under the controllable oxidation conditions.

Fig. 4a–c presents the representative SEM images for the original Fe particles, the Fe/Fe₃O₄ composites and the Fe/Fe₃O₄/resin composites. The Fe/Fe₃O₄ composites show very similar morphologies with those of the original Fe particles. However, different appearance color can be observed in these two samples with light gray for original Fe powders changed to dark blue for the oxidized powders, which is believed to result from the Fe₃O₄ coating layers. Moreover, the resin coating layers are completely and uniformly coated on the surface of Fe/Fe₃O₄ particles, which also result in their apparent color change from dark blue to dark gray for Fe/Fe₃O₄/resin composites. To further investigate microstructures of the Fe/Fe₃O₄ core-shell particles, An FIB technique was applied to prepare their cross-section TEM samples. For protecting the

Fig. 4. Representative SEM images for original Fe powders (a), the Fe/Fe₃O₄ composites (b) and the Fe/Fe₃O₄/resin composites (c), the cross-section TEM (d) and HRTEM (e) images for one typical Fe/Fe₃O₄ core-shell particle, and the back-scattered electron images with compositional (f) or topographic (g) contrast of the cross section of the fully compacted Fe/Fe₃O₄/resin composites.

sample surface of Fe/Fe₃O₄ particles for TEM observation from the FIB disruption, the Au layers of appropriate thickness were sprayed upon the surfaces of the initially-oxidized powders using a special sputter coater. Fig. 4d shows a representative cross-section TEM image for one typical initially-oxidized particle, which shows a distinct Fe/Fe₃O₄ core-shell structure with 70-nm thick Fe₃O₄ shells. Moreover, as also showed in Fig. 4d, the observation of no obvious cracks or pores but a very dense and smooth state for Fe/Fe₃O₄ interface in initially-oxidized products indicates their good interface continuity, which is sure to lead to their high compaction density and low coercivity of the two composites as discussed below. The typical HRTEM image of the Fe/Fe₃O₄ composites illustrated in Fig. 4e shows the single HRTEM diffraction patterns of Fe₃O₄ phase with the measured interplanar spacing value of 0.48 nm matching closely with the distances for the (111) planes [36]. The TEM results also confirm the in situ formation of single-phase Fe₃O₄ layers on the surfaces of high-purity Fe particles during their controlled oxidation. To further investigate the interface structures of silicon resin coated composites, the back-scattered electron images with compositional (BSE-COMP) or topographic (BSE-TOPO) contrast of the cross section of the fully Fe/Fe₃O₄/resin samples are observed as shown in Fig. 4f and g. From the BSE-COMP image in Fig. 4f, a uniform interface with deformed composite particles isolated from each other by the coating layers is observed in the densified Fe/Fe₃O₄/resin sample. From the elemental mapping of a selected interface area inserted in Fig. 4f, the Si and C elements originating from silicon resin are concentrated between the

interface of deformed particles, which indicates that silicon-resin shells are uniformly coated on the surfaces of the oxidized particles. The suitable amount of silicon resin coating layers in the Fe/Fe₃O₄/resin composites may act as a lubricant during their compaction to improve their mechanical properties with high TRS values, and can also act as the grain boundaries with relatively high resistivity to greatly reduce their magnetic loss as discussed below. From the BSE-TOPO image in Fig. 4g, the fully compressed Fe/Fe₃O₄/resin composites also show good compactness with no obvious cracks between the interfaces of deformed particles, which would lead to their high compaction density so as to improve their relatively high magnetic inductions as investigated later.

As is known, high compaction density is an important material parameter for metal-based SMCs, which is correlated with their high magnetic induction. Generally, low density of common organic and inorganic coating layers (epoxy resin, SiO₂, MgO and Al₂O₃ et al.) in metal-based SMCs would inevitably decrease their compaction density [17]. However, good interface continuity exhibiting no apparent cracks or pores between material interfaces in the composites can maintain their relatively high compaction density [9]. In this work, a very high compacted density value (7.55 g/cm³) achieved for fully compacted Fe/Fe₃O₄ composites nearly approaches that (7.68 g/cm³) of high-purity Fe powder cores compressed at the same pressures of 1200 MPa, which is attributed to good interfacial continuity of the dense Fe/Fe₃O₄ core-shell structure as discussed in Fig. 4. The fully compacted Fe/Fe₃O₄/resin composites also achieve a relatively a high density value

Fig. 5. The density and TRS values of the three compacted samples from the original Fe particles, the Fe/Fe₃O₄ composites and the Fe/Fe₃O₄/resin composites.

Fig. 6. Hysteresis loops tested under room temperature for the original Fe powders (a), the initially-oxidized Fe/Fe₃O₄ particles (b) and the Fe/Fe₃O₄/resin composites (c).

of 7.50 g/cm³ because of low content resin coated on surfaces of the initially-oxidized composites. High TRS values is another important use requirement for metal-based SMCs. From Fig. 5, the fully compacted Fe powder core exhibits a high TRS value of 126 MPa due to good compacting ability of high-purity Fe particles. However, the compacted Fe/Fe₃O₄ composites show a very low TRS value of 28 MPa, which is believed to result from deformation inconsistency of the ceramic Fe₃O₄ coating layers and unoxidized Fe parts in the composites. It is interesting to point out that a high TRS value of 92 MPa can be obtained in the compressed Fe/Fe₃O₄/resin SMCs indicating their better compacting ability. A suitable amount of silicone resin coated on the surface of Fe/Fe₃O₄ particles may act as a lubricant during the compaction, which would be beneficial to deformation between the composite particles and promote the contact and occlusion of deformed particles so as to lead to the higher TRS value for the fully compacted Fe/Fe₃O₄/resin SMCs. It can be concluded that both high density and high TRS value can be achieved in the compacted Fe-based SMCs by coating the oxidized metal particles with an appropriate amount of silicone resin layers, which can promote their practical applications in magnetic devices. As discussed below, the resin coating can also increase the resistivity of

Fig. 7. DC magnetic induction (B) values for three toroidal samples compacted from the Fe/Fe₃O₄/resin SMCs, the Fe/Fe₃O₄ composites and the original Fe particles.

Fe/Fe₃O₄/resin SMCs and result in their low core loss.

Fig. 6 displays the hysteresis loops for the original Fe particles, the initially-oxidized Fe/Fe₃O₄ particles and the Fe/Fe₃O₄/resin composites measured under room temperature. All the three samples show typical single-phase magnetic hysteresis along with good intrinsic magnetism. The Fe/Fe₃O₄ core-shell particles show a high saturated magnetization value ($M_s = 206 \text{ A m}^2 \text{ kg}^{-1}$), very close to that ($214 \text{ A m}^2 \text{ kg}^{-1}$) for the original Fe particles because of the presence of 70-nm thick ferrimagnetic Fe₃O₄ shells. A relatively high M_s value reaching $201 \text{ A m}^2 \text{ kg}^{-1}$ is maintained for Fe/Fe₃O₄/resin composites due to the 0.8 wt% silicon resin coating layers. It has been reported that seriously decreased M_s values along with low compacted density would occur in conventional SMCs prepared from coating metal-based magnetic powders with some insulating organic [37] and inorganic coating layers containing MgO [38] or SiO₂ [39] because of the nonmagnetism along with low density for these coating layers. In this work, high M_s values accompanying with high compaction density are maintained in Fe/Fe₃O₄/resin SMCs due to the existence of nanoscale thick ferrimagnetic Fe₃O₄ layers and suitable amount of silicone resin layers properly coated on the surfaces of initially-oxidized Fe/Fe₃O₄ particles. From Fig. 6, the two composites show low coercivity value of 9.6 Oe for Fe/Fe₃O₄ composites and 11.4 Oe for Fe/Fe₃O₄/resin SMCs, close to that (8.9 Oe) for high-purity Fe powders. It is reported that compared with high-purity FeSi powder cores, a much higher coercivity occurs in the FeSi/SiO₂ core-shell composites prepared by a chemical deposition because of the pores and air gaps existing between SiO₂ shells and FeSi cores, which hinder the movement of magnetic domain walls in the composites under their magnetization [40]. As shown in Fig. 4e, the tightly integrated Fe/Fe₃O₄ interfaces with in situ formation of nanoscale thick Fe₃O₄ layers coated on the surfaces of unoxidized Fe parts can probably decrease some interface defects containing holes or stress, which can improve magnetic domain movement so as to result in their low coercivity in these two composites. Moreover, the possible magnetic interaction occurring between two magnetic crystallographically coherent Fe and Fe₃O₄ phases in these two composites reduces movement resistance of magnetic domains and further promote their low coercivity [9,16]. A small amount of silicone resin coating layers do not significantly increase the coercivity of the Fe/Fe₃O₄/resin SMCs, which may be ascribed to the uniform and complete resin coating upon the surfaces of the oxidized particles.

Fig. 7 presents dc magnetic induction (B) values for three toroidal samples tested with a closed-loop mode at maximum external magnetic fields (H) in a range of 1200–10000 A/m. From the inset of Fig. 7, the

Fig. 8. Frequency-dependent core loss (W , P_s , W_h and W_e) for three toroidal samples compacted from original Fe particles, the Fe/Fe₃O₄ composites and the Fe/Fe₃O₄/resin SMCs tested at an applied magnetic induction with 1 T.

turn numbers (N_1 for primary and N_2 for secondary coils) for the magnetic induction measurement are 220 and 40, respectively. The two compacted composites show slightly decreased B values tested at the same H of 1200–10000 A/m than those for high-purity Fe powder cores, which is mainly ascribed to the magnetic dilution of the coating layers. Nevertheless, high magnetic inductions varying from 0.8 to 1.6 T measured at H increased from 1200 to 10000 A/m are sustained for the Fe/Fe₃O₄/resin SMCs, which is attributed to the existence of high-purity ferrimagnetic Fe₃O₄ nanolayers. As discussed above, high compaction density for Fe/Fe₃O₄/resin SMCs also promotes their relatively high magnetic inductions. The present metal-based SMCs with decreased core loss have been prepared from coating metal-based magnetic particles using some insulating organic and inorganic layers containing resin, MgO and SiO₂. However, seriously decreased B values would occur in these conventional SMCs due to nonmagnetism of these coating layers [41–43], which always impedes their practical usages for high-power situations. In the present work, high B values are achieved in the Fe/Fe₃O₄/resin SMCs because of the presence of nanoscale thick ferrimagnetic Fe₃O₄ layers and suitable amount of resin layers can overcome the disadvantages of conventional SMCs and promise their actual applications for high-power and low-loss magnetic components.

Fig. 8 presents the frequency-dependent core loss (W - f , P_s - f , W_h - f and W_e - f curves) tested at an applied induction with 1 T for three toroidal samples compacted from the original Fe particles, the Fe/Fe₃O₄ composites and the Fe/Fe₃O₄/resin SMCs. As shown in Fig. 8a and b, in general, both of the two composites show greatly decreased W and P_s values (W is defined as total core loss, and P_s is defined as per-cycle core loss expressed as W/f) than those for high-purity Fe powder cores compressed from original Fe particles. Moreover, the resin coating leads to greatly decreased core loss in the Fe/Fe₃O₄/resin SMCs than that of the Fe/Fe₃O₄ composites. For the investigation of the loss mechanism of

the samples, the total core loss W is divided into eddy-current loss (W_e) and hysteresis loss (W_h) according to an experiential formula expressed as follows:

$$W = W_h + W_e = eB_m^2 f^2 + \eta B_m^{1.6} f \quad (2)$$

where f is applied frequencies, B_m is an applied maximum magnetic induction, η is a material constant and e is a loss coefficient. It should be pointed out that very low residual loss measured at f beyond 1000 Hz is ignored in this work. Moreover, the P_s - f curves for all the three toroidal samples as shown in Fig. 8b show an obvious linear characteristic, which indicates that the two loss terms (W_h and W_e) dominates total loss W for these samples [15]. Therefore, the loss W_h and W_e is separated from measured core loss W based on formula (2) as illustrated respectively in Fig. 8c and d.

The hysteresis loss as shown in Fig. 8c dominates total loss W for three samples measured at low frequencies, and their eddy-current loss as shown in Fig. 8d increases much more quickly than hysteresis loss does with increasing applied frequencies. Moreover, the eddy-current W_e for the compacted Fe/Fe₃O₄ composites is comparably lower than that for high-purity Fe powder cores. The obviously decreased eddy-current loss for Fe/Fe₃O₄ composites is mainly attributed to much higher resistivity for the Fe₃O₄ shells ($1 \times 10^{-2} \Omega \text{ cm}$) [23] than that ($1 \times 10^{-7} \Omega \text{ cm}$) for high-purity Fe [44], for the high resistivity for powder grains of soft magnetic materials is sure to greatly inhibit their eddy current loss [45]. However, compared with high-purity powder cores, the loss terms W_h for Fe/Fe₃O₄ composites is slightly decreased and still maintains higher loss values. As discussed in Fig. 5, the inconsistent deformation between Fe/Fe₃O₄ core-shell interfaces leads to their low bonding strength resulting in low TRS value for compressed Fe/Fe₃O₄ composites. So the relatively high W_h for Fe/Fe₃O₄ composites can be

Fig. 9. Complex permeability spectra for three toroidal samples compacted from the Fe/Fe₃O₄/resin SMCs, Fe/Fe₃O₄ composites and original high-purity Fe particles.

probably attributed to the occurrence of increased internal stress for the compressed Fe/Fe₃O₄ composites due to inconsistent deformation between the ceramic Fe₃O₄ nanolayers and unoxidized Fe parts [16]. As showed in Fig. 8c, the loss terms W_e for Fe/Fe₃O₄/resin SMCs can be further greatly reduced by coating the oxidized particles with 0.8 wt% silicone resin, which is sure to result from the increased resistivity of the composites due to the existence of resin layers. It is worth pointing out that the loss terms W_h for Fe/Fe₃O₄/resin SMCs is also greatly decreased, which can be attributed to their reduced interfacial defects such as holes and cracks between Fe cores and the coating layers in the fully compacted sample as discussed in Fig. 4.

Fig. 9 shows the complex permeability with real part of μ' accompanying with imaginary part of μ'' versus frequency for three toroidal products tested at different frequencies ranging from 40 to 200 KHz. From Fig. 9, desirable frequency-dependent soft magnetic performance is observed for the Fe/Fe₃O₄/resin SMCs due to the successive uniform resin layers coated on the surfaces of the initially oxidized particles. Moreover, the Fe/Fe₃O₄/resin SMCs exhibit decreased μ' values slightly lower than those for high-purity Fe powder cores tested at frequencies beyond 60 KHz, and however, high μ' values are maintained for the composites when measured at frequencies ranging from 60 to 200 KHz, which implies that the composites possess an improved ac permeability. A gradual descending trend can be also observed in $\mu'-f$ curve for the Fe/Fe₃O₄/resin SMCs and very low μ'' values can be simultaneously obtained at applied frequencies reaching 200 KHz, which further indicates the decreased core loss for the Fe/Fe₃O₄/resin SMCs because of the increased resistivity of coating layers. Thus, among three samples, the Fe/Fe₃O₄/resin SMCs show best frequency-dependent magnetic performance containing low μ'' but high μ' values under applied frequencies reaching 200 KHz. From the above mechanical and magnetic investigations, it is concluded that these Fe/Fe₃O₄/resin composites possess greatly improved overall performance with high density, high TRS and good soft magnetic characteristics, which can make them behave as new-type low-loss metal-based SMCs for high-power metal-based magnetic components.

4. Conclusions

A new type of Fe/Fe₃O₄/resin SMCs with reinforced structure has been prepared by properly oxidizing high-purity micron-sized Fe powders with 70-nm-thick Fe₃O₄ layers in situ formed on the surfaces of unoxidized Fe parts and then coating the initially oxidized products with 0.8 wt% silicone resin through a physical coating method. Good intrinsic magnetism containing a high saturated magnetization value (201 A m²

kg⁻¹) in company with a low coercivity value (11.4 Oe) can be achieved in Fe/Fe₃O₄/resin SMCs because of the existence of single-phase ferromagnetic Fe₃O₄ shells in company with uniform silicone resin coating layers. The fully compacted Fe/Fe₃O₄/resin SMCs possess good overall performance with high TRS value and greatly improved soft magnetic properties including high magnetic inductions, decreased core loss and desirable frequency-dependent magnetic characteristics because of the suitable coating of Fe₃O₄ and silicone resin with appropriate thickness, which is superior to that of high-purity Fe powder cores and traditional metal-based SMCs and promises their latent usages for high-power and low-loss magnetic devices.

Declaration of competing interest

All the authors declare no conflict of interest.

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