Research Article

Influence of Annealing Methods on the Magnetic Properties and Flexural Strength of Iron-OER Soft Magnetic Composites

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This study aims at investigating high flexural strength iron-OER soft magnetic composites (SMCs) for active magnetic bearings (AMBs). Iron powders were firstly coated with oxide layer by the passivation process at room temperature. The effect of annealing methods on the static performance, magnetic properties, and flexural strength of SMCs was studied. The results showed that the significant improvements of permeability, magnetic loss, and flexural strength were presented by increasing the bonding number between iron surface and resin and using the two-step annealing method. In addition, the effect of pore shape and angle on the behavior of material fracture has been studied by FEM and the analysis was validated by experimental results.

1. Introduction

Soft magnetic composites (SMCs) present many new functions and unique properties such as low eddy current loss, high resistance, three-dimensional (3D) isotropy, and higher thermal stability. Therefore, the research of SMCs increases year by year and has gradually become a hotspot in the field of magnetic materials. Of course, till today, SMCs have been successfully applied to the field of nonconventional electrical machines only [1–3]. In such machines topologies, the complex shapes which cannot be manufactured by traditional silicon steels are required and eddy current loss has been effectively reduced. Although the application demands for SMCs used in active magnetic bearings (AMBs) are very similar to that in electrical machines, they need higher strength without weakening the magnetic properties for AMBs, especially for thrust disc which is subjected to a centrifugal force under high-speed rotation. So it is not feasible to use the existing SMCs instead of traditional carbon steel directly.

As the named publications mainly focus on the magnetic and electrical properties, there is a little experimental

information about the strength of SMCs. To SMCs with inorganic coating, the flexural strength is performed [4–6] and the core loss of SMCs with a novel coating [5] is a little higher when the strength is 44 MPa. However, SMCs with organic coating have low eddy current loss [7], but there is a lack of studies on the flexural strength, which is influenced by many factors including coating amount, the annealing temperature and method, and the bond force between iron and OER. To enhance the experimental efficiency, some scholars have investigated that these performances can be adjusted by changing the ratio of inclusion and matrix [8] and preparation process [9–12]. And FEM analysis [12] indicates that the strength of resin layer is a quite important factor that affects strength and quality of SMCs. For the hydrophilic nature of ferromagnetic iron particles is incompatible with organic insulations, this requires the coupling agents, which can combine the inorganic and organic groups, to produce more hinge structures. Recently silane coupling agents are widely used to improve the adhesion between the metal and the polymer to increase the strength of the sample [13] and keep the insulating layer uniform. Because of the presence

of organic materials, sintering cannot be used and residual stresses induced by cold work have deleterious effects on the magnetic performance of the core material. For this reason, annealing process for the elimination of residual stresses is essential and two-step annealing contributes to the growth of the magnetic domain and domain wall motion and then reduces the magnetic losses and improves the permeability of SMCs [11].

To obtain the good thermal stability and improve the flexural strength for SMCs, organic-silicon epoxy resin (OER) was chosen. For this class of materials such as iron-OER SMCs, the optimum parameters including compaction pressure and annealing temperature have been discussed [12, 14] and the flexural strength has also improved accordingly. To improve the strength of whole composites even more without noticeable reduction in permeability, it needs to increase the numbers of sites for iron-to-OER bonding on the one hand and on the other hand eliminate residual stress and some internal defects by comparing different annealing methods.

The aim of this paper is to develop higher flexural strength iron-OER SMCs with better magnetic properties and to discuss the effects of interfacial adhesion between iron particles and OER and annealing method on magnetic properties and flexural strength of SMCs. For comparison purposes, two kinds of samples with the same composition are prepared by one- and two-step annealing, respectively. At last, to further explore the effect of annealing method on flexural strength for SMCs, finite element method (FEM) is used to analyze the damage process and fracture mechanism.

2. Experimental Procedures

2.1. Sample Preparation. Compared with traditional soft magnetic materials such as silicon steel and permalloy, iron powders provided by Fucheng Chemistry Factory were selected for the most prototypical soft magnetic material with very high saturation flux density (2 T). The purity of Fe was 98%, containing 0.1% sulfuric acid insulation, 0.06% SO_4 , 0.005% N, 0.005% Cu, and 0.03% water soluble. The iron powders were coated by organosilicon epoxy resin (OER, 0.6 wt%), silane coupling agent KH-550, and curing agent 4,4'-diaminodiphenyl sulfone (DDS) (total 2.6 wt%) which were supplied by Xi'an Resin Factory.

In order to increase the interfacial strength for organic-inorganic composites, passivation method was used to generate Fe_2O_3 film on the surface of iron powders to increase bonding numbers with the coupling agent, and then a small amount of hydroxyls was produced after being dissolved by water [15].

The powder coating process was carried out as follows. The passivated iron powders and KH-550 were mixed firstly in ethanol until the solution reaches the specified pH value. Secondly the cleaned powders were mixed with the OER in acetone by a spiral mixer for 1 hrs, and then we put DDS into the acetone solution and keep mixing until most of acetone is evaporated. At last, the coated powders were obtained and then heated at 150°C for 25 min.

FIGURE 1: pH value as a function of mixing time.

The coated powders were cold-compacted at 800 MPa in a cylindrical die with a diameter of 10 mm and 35 mm and then kept for 5 minutes. The glycerol was used as the lubricant. Finally, the specimens were subjected to one-step (200°C for 2 hrs) and two-step annealing (130°C for 1 hrs and 200°C for 1 hrs) under vacuum condition, respectively.

2.2. Electrical, Magnetic, and Flexural Strength Characterization Conditions. The electrical resistivity was measured on the polished surface of cylindrical sample using four-point probe method (CMT-SR 1000 NV). Three readings were taken on each sample (diameter 10 mm) and averaged. Magnetic properties and losses were tested by measuring system (D8 ADVANCE, Italy) for a ring-shaped core with an outer diameter of 35 mm, inner diameter of 28 mm, and height of 6 mm. Complex permeability (μ' and μ'') was calculated from $B_m \cos \delta / (\mu_0 H_m)$ and $B_m \sin \delta / (\mu_0 H_m)$, respectively, where δ is phase difference and can be obtained directly from the computer measurement and control system. Under the action of alternating magnetic field, each of the measured samples has both magnetic energy storage (μ') and consumption (μ''). The magnetic consumption described here is identified as hysteresis, eddy current, and residual losses. In the general case, for the resin-coated SMCs, the losses are mainly composed of eddy current and hysteresis losses. Flexural strength was measured by three-point bending method (Instron 5848, USA) for a bar specimen with the size $35 \text{ mm} \times 4 \text{ mm} \times 5 \text{ mm}$.

3. Results and Discussion

3.1. Surface Analysis. The variation of pH value in mixing process is presented in Figure 1. The pH value first increases before 10 min and then is saturated to a final value of 9.5 for the formation of NH_3 and NaOH [16]. Element distribution on surface of passivated irons is shown in Figure 2. It can be seen that the content of element oxygen is low and its distribution is uniform in general.

TABLE 1: Basic performance through different annealing methods.

Annealing method	Two-step annealing (130°C for 1 h + 200°C for 1 h)	One-step annealing (200°C for 2 h)
Density $\rho/\text{g}/\text{cm}^3$	5.95	5.63
HV	197.2	174.5
$R/\mu\Omega\cdot\text{m}$	668	675

FIGURE 2: Element distribution on surface of passivated iron powders.

The FTIR spectrums for two different powders at pH value about 8 are presented in Figure 3. It can be noted that $-\text{OH}$ groups at 3444 cm^{-1} (curve (a)) are generated on the surface of iron powders which indicate that the iron oxide shells have stronger absorption to the hydroxyl groups in the solution. However, the peak intensity in curve (b) corresponding to $-\text{OH}$ groups at 3444 cm^{-1} decreases, which is attributed to the reaction between silanol groups of coupling agent and the $-\text{OH}$ groups of passivated iron particles [17]. The peak at 2928 cm^{-1} is related to the stretching vibration of C-H bond in propyl groups [18]. The peak at 1272 cm^{-1} is caused by NH_2 deformation mode in coupling agent during the reaction with OER in Figure 4. The formations of Si-O-Fe and Si-O-Si are observed at 1130 cm^{-1} and 1050 cm^{-1} , respectively [19].

3.2. Basic Performance. Table 1 lists basic performance of each specimen through different annealing methods as measured in air atmosphere. All samples in Table 1 have the same

FIGURE 3: FTIR spectra (a) passivated powders and (b) insulated iron powders at pH 8.

FIGURE 4: The structure of OER and its reaction mechanism with DDS and KH-550.

FIGURE 5: SEM micrograph of SMCs: (a) 200°C for 2 h and (b) 130°C for 1 h + 200°C for 1 h.

processing conditions except annealing treatment. As seen from this table, two-step annealing is helpful to improve the density and HV but goes against the resistivity. All of these three performance parameters are functions of particle size, powder composition, resin content, internal defects, and density (porosity). In Figure 5 there are lots of pores and the OER is nonuniform for the sample at one-step annealing. The corresponding EDS spectrums at resin, pore, and iron are shown in Figure 6. Comparing Figures 6(a) and 6(b), it is clear that Fe intensity decreases while Si intensity increases. However, the two-step annealing has fewer pores and denser structure. Therefore, it can be concluded that pores in SMCs are produced by the resin layer and annealing method has significant impact on the material structure and then influences the density and HV.

3.3. Magnetic Properties. The magnetic hysteresis loop for SMCs without initial magnetization at 50 Hz, 1 kHz, and 10 kHz is shown in Figure 7. The calculated results indicate

that the area encircled by hysteresis loop at one-step annealing is larger than that at two-step annealing in the same frequency condition and the area is growing with the increasing of frequency. During magnetization, the movement of magnetic domain walls is hindered by inhomogeneous structure in SMCs as shown in Figure 5, and the pinning effects are generated and enhanced, which need to consume more energy. Therefore, it increases the area of hysteresis loop. When the frequency increases from 50 Hz and 1 kHz, the change in the loop area is minor. When the frequency continues to rise to 10 kHz, the loop area and coercivity are at least twice as large as those in the previous two figures. It is attributed to eddy current and residual losses generated under the action of the alternating magnetic field. When frequency increases, the eddy current loss and its produced reverse magnetic field also increase at the same time, which have severely weakened the magnetic properties.

The real part of permeability (μ') as a function of frequency under the magnetic field intensity 0.7 T is shown

FIGURE 6: EDS spectrums for the sample at one-step annealing: (a) resin, (b) pore, and (c) iron.

in Figure 8(a). It can be seen that this part of permeability decreases slightly with increasing the frequency. And the real part at two-step annealing is approximately 18 percent higher than that at one-step annealing, because it strongly depends on the density, the number of pores, and defects of powder products under the same components. As the numbers of the pores in SMCs increase, the domain wall displacement and spin rotations are strongly blocked, which adversely affect the magnetic properties. Since the resin has the same permeability to vacuum and the permeability is usually much smaller than that of iron, it can be equivalent to the air gap (nonmagnetic phase) and nearly no flux lines enter into this region. The pores in SMCs structure act similarly to the nonmagnetic phase and weaken the magnetic properties.

The imaginary part of permeability (μ'') as a function of frequency for SMCs under the magnetic field intensity 0.7 T is shown in Figure 8(b). This part increases sharply after 1 kHz and the sample at two-step annealing has a higher value. The reason for these behaviors could be an effect of the iron particles work hardened during compaction and compaction of incomplete coated particles to each other. The compaction

always creates both some plastic deformation to increase the dislocation density in the iron powder particles and degradation of the insulation layer. The two-step annealing can reduce distortions within the particles, lower the residual stress effectively, and then improve the magnetic properties significantly for SMCs. For the sample at one-step annealing, more pores and less volume fraction of magnetic particles reduce magnetic loss and directly lower the imaginary part.

3.4. Flexural Strength. Due to the immiscibility of iron powders and resin and different thermal expansion coefficients for both, it leads to the increase of pores in SMCs after annealing and cooling to ambient temperature. Pores in SMCs act as stress concentration centers and the load-bearing area is reduced. To improve strength of whole material, it needs to reduce the numbers of pores on the one hand and on the other hand improve the adhesion strength between iron and OER by the passivation treatment when heated in vacuum. Effect of annealing method on the porosity and flexural strength is presented in Table 2. It can be seen that the porosity at two-step annealing is lower and the corresponding flexural

TABLE 2: Effect of annealing method on the porosity and flexural strength.

Annealing method	Two-step annealing (130°C 1 h + 200°C for 1 h)	One-step annealing (200°C for 2 h)
Porosity	4.8%	13.4%
Flexural strength/MPa	44.52	40.36

FIGURE 7: Magnetic hysteresis loops of SMCs at different frequencies.

strength is higher than that at one-step annealing. This is because the residual stresses can be effectively minimized by thermal motion of the atoms for the sample at two-step annealing, and then high uniformity insulating film and low-volume fraction of pores are provided.

The iron-resin SMCs are mainly composed of iron powders, resin, pores, and defects. Under external force, some microcracks are produced at the interface between resin and iron particles as shown in Figure 9. From this figure, it can be seen that the volume of pores is smaller and the

microcracks are fewer and finer for the sample at two-step annealing comparing to that at one-step annealing. Most of these microcracks are distributed at the acute angles of pores (D) and some of them are connected with two adjacent pores (such as (A) and (B)). In Figure 9, because of irregular iron powders, sharp corners, and big curvature, it is more likely to form tip pores to exacerbate stress concentration and accelerate the formation and development process for these initial cracks under a concentrated load. When cracks propagate to the stress interface under an increasing load,

FIGURE 8: Complex permeability as a function of frequency at 0.7 T.

FIGURE 9: Fracture surfaces of SMCs at different annealing methods: (a) 200°C for 2 h and (b) 130°C for 1 h + 200°C for 1 h.

the stress is freed and corresponding bending deformation is generated.

For further analysis of the effect of pores with different shapes on the material strength, finite element method (FEM) is used to analyze the damage process for SMCs. Although flake-shaped electrolytic iron powder [20] and spherical-shaped atomized powder in Figure 10 are widely used in SMCs materials, the shape of iron powder resembles hexagon when the irregular powders are compacted together. In previous research, the experimental values for magnetic properties have a good agreement with the calculated ones between the circular and hexagon models; so in this paper those two models with different pores are also required as shown in Figure 11. In these two models, both of the hexagonal side length and the diameter of the circle are 100 μm and the smallest coating thickness is 2 μm. The porosities are calculated by the theoretical method [8], which is determined by

resin content. Material properties are then manually assigned to each region. The discretization of the three models is performed by an automatic meshing algorithm. In order to improve computing precision, finer mesh is also taken into account on the very thin insulating layer and the pores as shown in Figure 12. The mesh examples shown in Figure 3 have 9885 (a), 23757 (b), and 5843 (c) nodes, respectively.

The load, which is equal to the experiment result for SMCs with 99 wt% Fe, is applied on the center of top lines, which narrows the insulating layer on the upper half, while widening the layer on the lower half in Figure 13. The results show that the strain of insulating layer is much larger than that of the iron and the maximum strain is found to occur near the thinnest insulating layer. The strain in the model with acute pores in Figure 13(b) is larger than that in Figure 13(a) with obtuse one. Comparing the two hexagon models with and without pores [12], we can see that the model with pores

FIGURE 10: SEM micrograph of iron powders.

FIGURE 11: Simplified models: (a) circular pores; (b) circular pores with acute angle; (c) hexagon pores with acute angle.

has larger strain and the maximum strain drops almost 48 percent between two models. Magnifications of displacement vector sum for three models are shown in Figure 14. When the load is applied on the model with an obtuse pore, there is no crack generated (see Figure 14(a)) and only deformation occurred, which are consistent with Figure 9 (C). When the load is applied on the model with an acute pore, crack is generated and propagates along this angle and interface between resin and iron particles (see Figures 14(c) and 9 (D)). Especially when the acute angles of the two adjacent pores have the opposite direction and meet face to face, it can be seen from the different displacement in a vertical line that crack is generated as shown in Figure 14(b), which is corresponding to Figure 9 (A and B).

Through the above analysis, it can be concluded that the pore in SMCs plays a vital role in mechanical strength and the crack propagation depends upon the applied load as well

as the shape of pores. Regarding one-step annealing sample, there is not enough time to eliminate residual stress when the temperature reaches value as high as 200°C and some acute pores are generated, which ultimately lead to lower strength.

4. Conclusion

In this work, high flexural strength iron-OER SMCs with better magnetic properties for active magnetic bearings (AMBs) were prepared successfully. The effect of interfacial adhesion between iron particles and OER and annealing methods on the static performance, magnetic properties, and flexural strength was investigated. The results can be concluded as follows:

- (1) Coating of the iron particles was carried out successfully by passivation and modification with KH-550. EDS, SEM, and FTIR spectra proved that the iron

FIGURE 12: Mesh of models in FE environment: (a) circular pores; (b) circular pores with acute angle; (c) hexagon pores with acute angle.

FIGURE 13: Total mechanical strain intensity for three models: (a) circular pores; (b) circular pores with acute angle; (c) hexagon pores with acute angle.

FIGURE 14: Displacement vector sum for three models: (a) circular pores; (b) circular pores with acute angle; (c) hexagon pores with acute angle.

particles were uniformly covered with oxide layers and OER insulation.

- (2) Two kinds of annealing methods have different effects on the physical and magnetic properties for the samples. Two-step annealing method improved the density, HV, and permeability of SMCs, while decreasing the magnetic loss because of the low hysteresis loss and resistivity for SMCs.
- (3) The increase of the bonding number between iron surface and resin by passivation method and the choice of two-step annealing method resulted in higher flexural strength. The crack propagation analysis (FEM) shows that the flexural strength is related to the applied load as well as the shape and angle of pores.

Competing Interests

The authors declare that they have no competing interests.

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