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Loss-Separation Study on Silica-insulated Gas-atomized Fe-Si-Al Soft Magnetic Composites

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Fe-Si-Al soft magnetic composites were composed of gas-atomized Fe-9.6wt.%Si-5.4wt.%Al alloy powders insulated with silica nanoparticles. The influence of silica insulation content on the core's magnetic properties was studied. It was found that increasing the silica mass ratio deteriorated the effective permeability and core loss in the frequency range of 40-120 kHz, while improved the quality factor at 100 kHz and DC-bias performance. The effective demagnetizing field reflected by density and the core's volume resistivity may cause the variations of these magnetic parameters. Loss separation fitting was performed using the Bertotti formula, indicating that silica insulation increased the hysteresis loss and reduced the eddy-current loss. The hysteresis loss took over at the frequency lower than 120 kHz in this work. With increasing the frequency, the eddy-current loss grew more quickly than the hysteresis loss. Therefore, different methods should be adopted to reduce the core loss according to the core's application frequency.

Keywords : soft magnetic powder core, Fe-Si-Al alloy, magnetic properties, silica content

1. Introduction

Soft magnetic composites (SMCs) have drawn increasing attention due to their outstanding magnetic performance at high frequency and high-power level [1]. In a typical SMC fabrication process, an insulating layer with high resistivity is firstly coated onto magnetic powder with good soft magnetic properties, which would then be pressed into a core with a specific shape. Subsequently, the core is annealed at an optimal temperature to release the internal stress introduced during the compaction process [2-4]. Due to the high saturation magnetic flux density of magnetic particles and the high resistivity of insulating materials, coupled with the special composite's microstructure of distributed air gap, the SMCs possess the advantages of less material consumption, good anti-saturation ability and low high-frequency iron loss [5-7].

As we all know, iron loss is the most important parameter of soft magnetic material. It is mainly comprised of hysteresis loss and eddy-current loss for SMCs [8]. Therefore, the physical mechanism of SMC's iron loss, depending on the application frequency, is fairly complicated. It has been generally accepted that the hysteresis loss is predominant at low frequency and the eddy-current loss takes over at high frequency. For SMC with a fixed composition of magnetic powder, the hysteresis loss is determined by the particle's morphology, the core's density, which reflect the core's effective demagnetizing field, and the residual stress in the core [9]. The eddy-current loss can be divided into two parts. One is from the eddy

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current between different magnetic particles determined by the resistivity, the thickness of the particle's insulating layer and the width of the air gap between particles. The other is from the eddy current occurred in individual particle determined by the particle's resistivity and size [10]. Obviously, making clear the proportion of hysteresis loss and eddy-current loss under actual working conditions is vital for designing an appropriate scheme to reduce the iron loss effectively. Loss separation offers a good method to understand the loss component of a core at different frequencies and different powers.

At present, the fitting model of the iron loss of soft magnetic material is mainly based on the Steinmetz equation or the Bertotti formula. The Steinmetz equation ignores the influence of the core's morphology and holds that the iron loss is determined by the core's composition, the excitation frequency and flux density. It greatly simplifies the calculation of the iron loss and has been widely used in the loss calculation for transformer, motor, reactor, etc. Unfortunately, the loss separation cannot be achieved using the Steinmetz equation [11-13]. In contrast, the Bertotti formula divides the iron loss into hysteresis loss, eddy-current loss and residual loss, which is helpful for performing the loss separation and analyzing the dependences of different components of the iron loss on the frequency and the flux density [14].

In this work, the gas-atomized Fe-9.6wt.%Si-5.4wt.%Al SMCs were prepared using the silica insulation. The influence of the silica content on the core's magnetic properties was investigated in detail. The Bertotti formula was employed to separate the core's iron loss. The dependences of hysteresis loss and eddy-current loss on the insulating silica content and the frequency were clarified accordingly. This provides the general guideline for reducing the iron loss of SMCs within desired frequency range.

2. Experimental

The silica nanoparticles and gas-atomized Fe-9.6wt.%Si-5.4wt.%Al alloy powders (−140 mesh), were commercially purchased. Silica nanoparticle size is mainly concentrated in the range of 14-20 nm, and the average size of silica nanoparticles is about 18 nm. An acetone solution of 0.5 wt.% epoxy resin (99.5 %) was firstly mixed with the Fe-9.6wt.%Si-5.4wt.%Al particles under mechanical stirring. The ethanol mixtures of silica nanoparticle with different mass ratios (0-2.4 wt.%) were then mixed homogeneously with the mixture of epoxy resin and Fe-9.6wt.%Si-5.4wt.%Al particles. After being dried at 80 °C, the insulated powders were blended with 0.4 wt.% organic silicon resin and 0.4 wt.% zinc stearate. Then the powder were compressed into toroidal cores with an outer diameter of 26.92 mm, an inner diameter of 14.73 mm and a height of 11.18 mm under the pressure of 1860 MPa. Finally, the SMCs were annealed at 760 °C for 40 min in N_2 atmosphere.

The X-ray diffraction (XRD) measurement was performed on a PANalytical X'pert PRO diffractometer with Cu-$K\alpha$ radiation to determine the sample's phase component and crystalline structure. The sample's morphology and element distribution were characterized by a scanning electron microscope (SEM, JSM-6490LV) equipped with an Oxford Instrument INCA energy-dispersive spectrometer (EDS). An LCR meter (Wayne Kerr 3260B) was used to measure the core's effective permeability (μ_e) and quality factor at the frequency of 100 kHz and percent permeability (%μ_e) under the DC magnetizing field ranging from 0 Oe to 200 Oe. A power loss analyzer (Voltech PM1000+) was employed to test the core loss at the frequency ranging from 40 kHz to 120 kHz at the maximum magnetic flux density (B_m) of 1000 Gs. The volume resistivity was measured by a DC resistance tester (TH2512). All these measurements were performed at room temperature.

3. Results and Discussion

Typical XRD patterns of the insulating silica nanoparticles, the raw Fe-Si-Al powder and the insulated Fe-Si-Al powder, have been shown in Fig. 1. It is obvious that the insulated powder has the pattern almost same with the raw powders. For one thing, the insulation coating of silica and organic resin does not damage the phase structure of the gas-atomized Fe-Si-Al powder. For another,

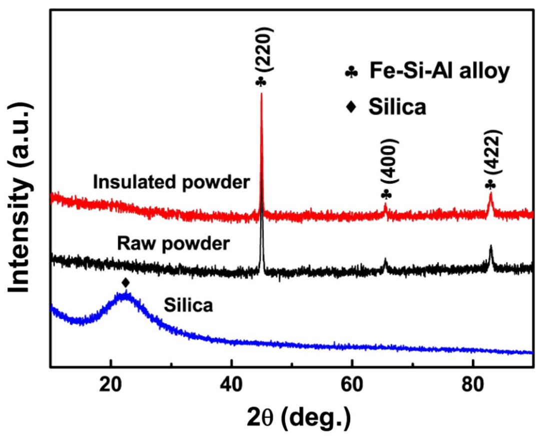

Fig. 1. (Color online) Typical XRD patterns of the insulating silica nanoparticles, the raw Fe-Si-Al powder and the insulated Fe-Si-Al powder.

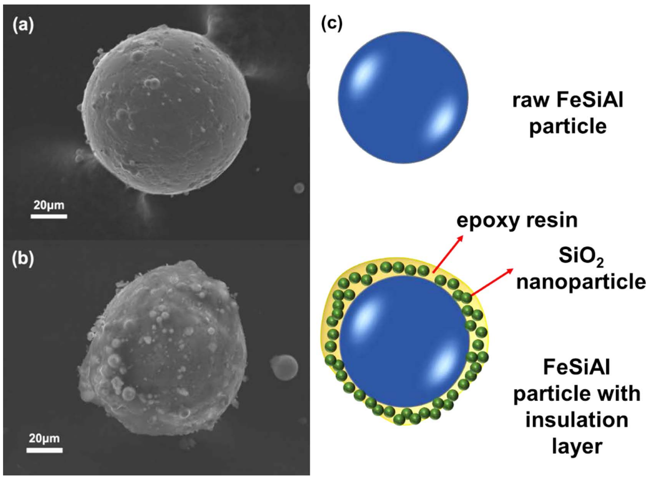

Fig. 2. (Color online) Typical SEM images of the raw Fe-Si-Al particle (a) and the insulated Fe-Si-Al particle (b). Schematic illustration of structure for Fe-Si-Al particle with and without insulation (c).

we may safely conclude that the insulating silica is amorphous.

SEM images and schematic illustration were further provided as Fig. 2. Clearly, the spherical raw particle has a relatively smooth surface with a scaly structure which may be explained by the metal solidification shrinkage in the gas-atomization process. After insulation, a continuous insulating layer can be observed around the Fe-Si-Al particle. It is generally accepted that nanoparticles would aggregate due to their high surface energy. Fine Fe-Si-Al particles would also aggregate on the surface of large particles to some extent. Considering the stickiness of epoxy resin, it is easy to imagine that composites coating may be rough, as displayed in Fig. 2. However, it is worth mentioning that the flexibility of composites coating would ensure uniform coating state after pressing.

To analyze the insulation coating quality, the SEM image and the corresponding EDS spatial elemental mapping for the insulated Fe-Si-Al particles were measured. Fig. 3(a) shows the images of a particle insulated with 1.2 wt.% silica. Strong and uniform signals of the Si and O elements, overlapping with those of Fe and Al elements, can be detected in the insulated Fe-Si-Al particle, which may provide direct proof that a continuous and uniform insulating layer of silica has been formed on the surface of the Fe-Si-Al particle. The weak signal of C element, coming from the organic resins, can also be observed on the surface of the Fe-Si-Al particle.

Figure 4 shows the effective permeability μ_e and the quality factor of the Fe-Si-Al SMCs prepared with silica of different mass ratios. Obviously, the silica mass ratio has a direct influence on the effective permeability and

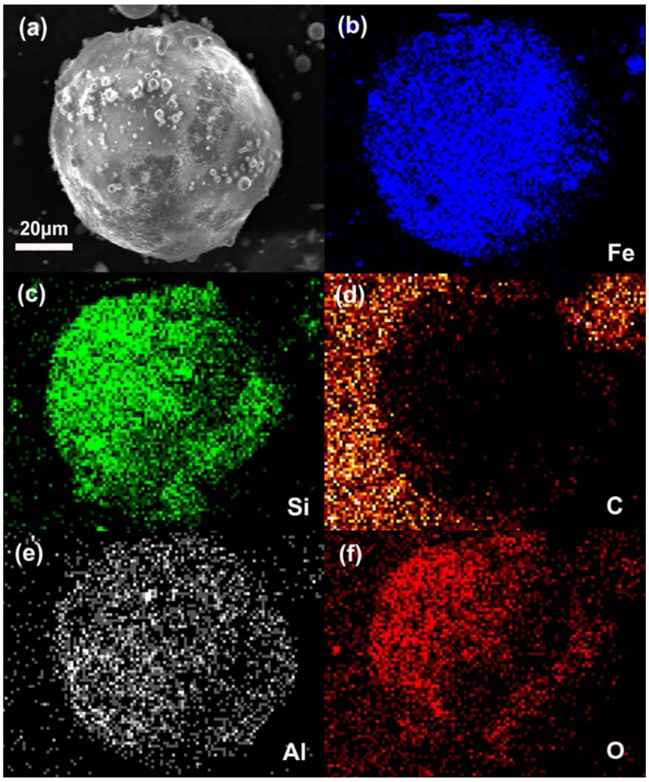

Fig. 3. (Color online) SEM image (a) and the corresponding EDS spatial elemental mapping of Fe (b), Si (c), C (d), Al (e) and O (f) for the Fe-Si-Al particle insulated with 1.2 wt.% silica.

the quality factor. The μ_e decreases monotonically with increasing silica mass ratio from 0 wt.% to 2.4 wt.%, while the quality factor exhibits an increasing tendency. For SMCs, the μ_e is generally related with the thickness and uniformity of the insulation layer coated on the magnetic particles. Thick and uniform coating layer may result in a low core density and thus leads to a strong effective demagnetizing field within the core. Thus, a remarkable decrease of the μ_e can be understood [15-17].

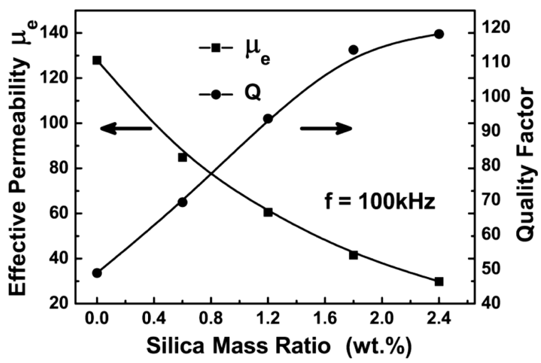

Fig. 4. Dependences of the effective permeability and the quality factor for the Fe-Si-Al SMC on the silica mass ratio.

Similarly, it can be deduced that uniform silica insulation layers have been successfully prepared, which led to homogeneous intervals among the magnetic particles. Meanwhile, the promotion of quality factor can also be ascribed to the thickness and uniformity of the insulation layers. The thicker the coating layer is, the higher quality factor would be achieved. The maximum quality factor can be reached for the core prepared with the silica mass ratio of 2.4 wt.%. The enhancement of quality factor suggests the decrease of the core loss. But the decrease of the μ_e usually implies the deterioration of the hysteresis loss. Therefore, we should point out that the silica insulation layer has effectively blocked the conducting path between the magnetic particles, which weakens the eddy-current loss of the Fe-Si-Al SMCs with increasing the insulating silica content.

The dependences of the density and the resistivity of the Fe-Si-Al SMC on the silica mass ratio have also been investigated as given in Fig. 5. Clearly, the core's density exhibits the same variation tendency with the core's μ_e and the core insulated with 2.4 wt.% silica possesses the lowest density. This is in line with the above-mentioned deduction that thick coating layer results in a low core density. It can be inferred that increasing the silica content may make the surface of the insulated magnetic particles much rougher and deteriorate their fluidity. Therefore, the core's density decreases, which may enhance the effective demagnetizing field and cause the deterioration of the μ_e and the hysteresis loss. Nevertheless when investigating the volume resistivity of all the cores, a totally opposite tendency can be observed. With the silica mass ratio increasing, the coating layer on the surface of the magnetic particle would become thicker and thicker. Thick silica coatings would effectively avoid the direct contact of conductive Fe-Si-Al particles, resulting in a high volume resistivity. It is of great significance to achieve lower eddy-current loss at higher frequency [18]. The core

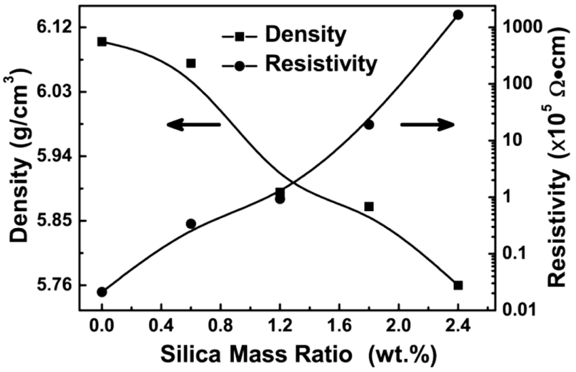

Fig. 5. Dependences of the core's density and resistivity on the silica mass ratio.

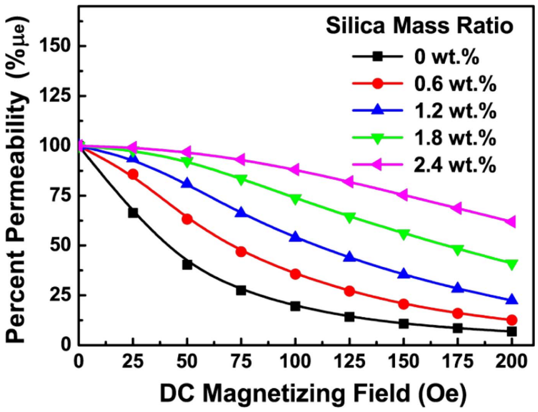

Fig. 6. (Color online) Percent permeability $\%\mu_e$ of the cores prepared with silica of different mass ratios as functions of the DC magnetizing field.

insulated with 2.4 wt.% silica possesses the relatively highest volume resistivity.

Figure 6 shows the $\%\mu_e$ of the cores prepared with silica of different mass ratios as functions of the DC magnetizing field. The $\%\mu_e$ of all the cores decreases with the applied DC magnetizing field. This is due to the core approaching the magnetic saturation gradually with the increase of the DC magnetizing field. Comparing the DC-bias performances of all the cores, it can be found that their dependences on the silica mass ratio are opposite to that of the μ_e. The core prepared with 2.4 wt.% silica whose $\%\mu_e$ at 100 Oe is 88.02 % has the relatively best DC-bias performance. These are all determined by the effective demagnetizing field which can be reflected by the core's density [19].

Most importantly, the core loss at the frequency ranging from 40 kHz to 120 kHz for the Fe-Si-Al SMCs prepared

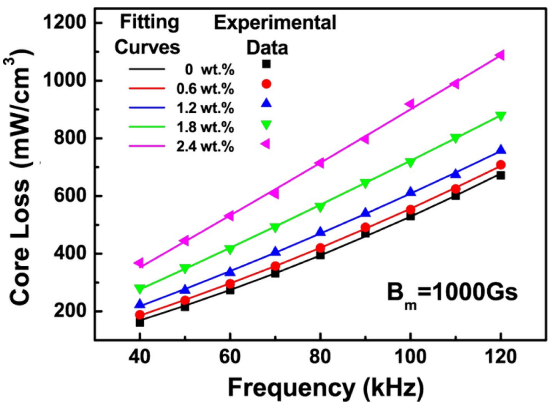

Fig. 7. (Color online) Core loss at the B_m of 1000 Gs in the frequency range of 40-120 kHz for the Fe-Si-Al SMCs prepared with silica of different mass ratios. The scattered symbols and the solid curves represent the experimental data and the fitting results, respectively.

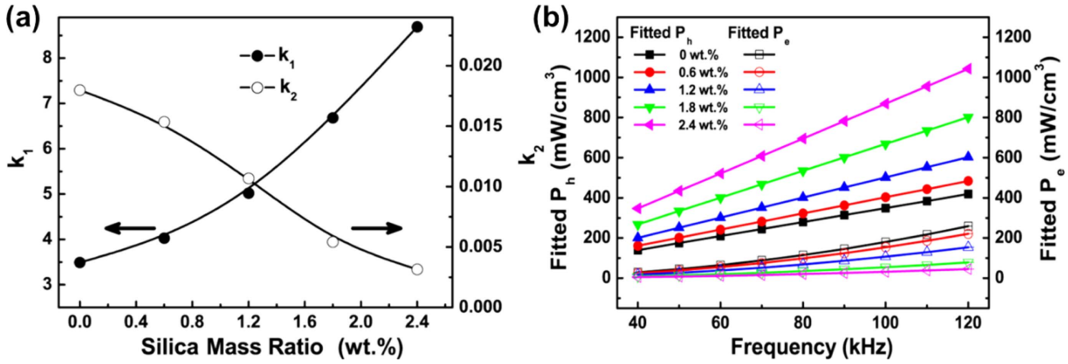

Fig. 8. (Color online) Dependences of the coefficients (a) of the core's hysteresis loss (k_1) and eddy-current loss (k_2) on the silica mass ratio. Fitted hysteresis loss and fitted eddy-current loss (b) in the frequency range of 40-120 kHz for the Fe-Si-Al SMCs prepared with silica of different mass ratios.

with silica of different mass ratios should be studied. As exhibited in Fig. 7, the loss for all the cores increases significantly with the frequency. This can be attributed to both the hysteresis loss and the eddy-current loss increasing with the frequency. It can be seen clearly that with the increase of the silica mass ratio, the core loss increases gradually in the measuring frequency range of 40 to 120 kHz. Within such frequency range, the increase of total loss is mainly originated in the increase of hysteresis loss, which is obviously affected by the effective demagnetizing field [19]. As mentioned above, the effective demagnetizing field for the SMC with fixed particle size distribution is mainly affected by the density, which has the tendency same as the core loss and opposite to those of the μ_e.

To further explore the physical mechanism of the core loss, the dependence of the core loss on the frequency at the B_m of 1000 Gs was studied by the loss separation method. Generally, the core loss (P_{cv}) of soft magnetic material can be separated into three parts including hysteresis loss (P_h), eddy-current loss (P_e) and residual loss (P_r). Hereinto, the P_r is a combination of relaxation and resonant losses of the core. It is important only at very low induction levels and at very high frequencies and can be excluded in power applications of the SMC [20]. Therefore, the core loss of the SMC mainly comprises the P_h and the P_e and is generally expressed by the Bertotti formula:

$$P_{cv} = P_h + P_e = k_1 f + k_2 f^2 \quad (1)$$

where f is the frequency and k_1 and k_2 are the coefficients of the hysteresis loss and the eddy-current loss, respectively [8, 14, 21]. Based on this equation, the loss-separation fitting, as depicted by the solid curves in Fig. 7, was performed, from which k_1 and k_2 can be achieved. Obviously, the fitting curves conform to the experimental data (scatted symbols in Fig. 7) very well. Fig. 8(a) shows the frequency dependences of k_1 and k_2. They exhibit the completely opposite variation tendencies. With the silica mass ratio increasing from 0 wt.% to 2.4 wt.%, the core's density keeps decreasing. This leads to the enhancement of both the effective demagnetizing field and the core's resistivity, thus results in the increase of the hysteresis loss and the decrease of the eddy-current loss, respectively.

Based on the fitted k_1 and k_2, we can roughly understand the variation trend of hysteresis loss and eddy-current loss with silica mass ratio. According to the Bertotti formula mentioned above, we can calculate the fitted P_h and P_e. Fig. 8(b) compares the fitted P_h and P_e in the frequency range of 40-120 kHz for the Fe-Si-Al SMCs prepared with silica of different mass ratios. It is obvious that in the frequency range of 40-120 kHz, the proportion of hysteresis loss is much higher than that of the eddy-current loss. This indicates hysteresis loss contributes more in this frequency range. Therefore, it is especially important to reduce the effective demagnetizing field to reduce the total loss at the frequency lower than 120 kHz. With the increase of the silica mass ratio, the eddy-current loss tends to decrease gradually. It can be speculated that the amount of silica, as discussed above, has a positive effect on reducing the eddy-current loss. Considering that P_h and P_e are, respectively, proportional to f and f^2, as the frequency increases, the eddy-current loss will increase more significantly and overwhelm the hysteresis loss at specific higher frequency. Increasing the insulation coating amount to achieve a large thickness of coating layer and a high core's volume resistivity is an effective route for reducing the eddy-current loss at high frequency. However, an excessively high amount of insulating substance would also cause a great decrease of the core's density and result in a sharp increase in the effective demagnetizing field and the hysteresis loss. Therefore, the balance between increasing the volume resistivity and keeping the effective demagnetizing field in a low level, depending on the core's applied frequency, should be considered for reducing the loss of the SMC in the future research and device design.

4. Conclusion

The Fe-Si-Al SMCs were prepared successfully by coating a continuous and compact silica insulation layer onto the surface of the gas-atomized Fe-Si-Al particles. The dependence of the core's magnetic properties on the addition amount of the silica nanoparticles was investigated. The density was found to decrease with the silica mass ratio, which was proposed to enhance the effective demagnetizing field, thus results in the decrease of the μ_e and the deterioration of the P_h. Meanwhile, the volume resistivity displayed a tendency of increasing with the silica content. This caused the reduction of the P_e. Loss separation based on the Bertotti formula confirmed the above-mentioned variation tendencies of the P_h and P_e. Although P_e increases with the frequency more rapidly than the P_h, the P_e was found to be much lower than the P_h in the whole measuring frequency range of 40-120 kHz. This can be a guideline, that keeping the effective demagnetizing field in a low level is the top priority to reduce the SMC's loss at low frequency, while increasing the volume resistivity is the key at high frequency.

Acknowledgments

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References

[1] J. M. Silveyra, E. Ferrara, D. L. Huber, and T. C. Monson, Sci. **362**, 418 (2018).
[2] J. Füzer, M. Strečková, S. Dobák, L. Ďáková, and M. Vojtko, J. Alloys Compd. **753**, 219 (2018).
[3] D. N. Chen, L. Huang, H. Y. Yu, X. C. Zhong, and Z. W. Liu, J. Magn. **24**, 485 (2019).
[4] A. H. Taghvaei, H. Shokrollahi, and K. Janghorban, J. Alloys Compd. **481**, 681 (2009).
[5] W. Ding, L. T. Jiang, Y. Q. Liao, J. B. Song, B. Q. Li, and G. H. Wu, J. Magn. Magn. Mater. **378**, 232 (2015).
[6] L. Y. Li, Z. Gao, A. K. Li, J. H. Yi, and Y. C. Ge, J. Magn. Magn. Mater. **464**, 161 (2018).
[7] S. H. Lee, Y. J. Kim, K. S. Lee, and S. J. Kim, J. Magn. **20**, 444 (2015).
[8] G. L. Zhao, C. Wu, and M. Yan, J. Magn. Magn. Mater. **399**, 51 (2016).
[9] M. Yaghtin, A. H. Taghvaei, B. Hashemi, and K. Janghorban, J. Alloys Compd. **581**, 293 (2013).
[10] A. H. Taghvaei, H. Shokrollahi, K. Janghorban, and H. Abiri, Mater. Des. **30**, 3989 (2009).
[11] J. Muhlethaler, J. Biela, and J. W. Kolar, IEEE Trans. Power Elec. **27**, 953 (2012).
[12] H. N. Ji, Z. W. Lan, Z. Y. Xu, H. W. Zhang, J. X. Yu, and M. Q. Li, IEEE. Trans. Appl. Supercond. **24**, 1 (2014).
[13] G. Bertotti, IEEE Trans. Magn. **24**, 621 (1988).
[14] D. Z. Xie, K. H. Lin, and S. T. Lin, J. Magn. Magn. Mater. **353**, 34 (2014).
[15] G. Q. Lin, Z. W. Li, L. F. Chen, Y. P. Wu, and C. K. Ong, J. Magn. Magn. Mater. **305**, 291 (2006).
[16] Z. M. Zhang, W. Xu, T. Guo, Y. Z. Jiang, and M. Yan, J. Alloys Compd. **594**, 153 (2014).
[17] H. Kronmüller, Phys. Stat. Sol. (B) **144**, 385 (1987).
[18] L. P. Lefebvre, S. Pelletier, and C. Gélinas, J. Magn. Magn. Mater. **176**, 93 (1997).
[19] M. M. Zhou, Y. Han, W. W. Guan, S. J. Han, Q. S. Meng, T. T. Xu, H. L. Su, X. Guo, Z. Q. Zou, F. Y. Yang, and Y. W. Du, J. Magn. Magn. Mater. **482**, 148 (2019).
[20] L. Chang, L. Xie, M. Liu, Q. Li, Y. Q. Dong, and C. T. Chang, J. Magn. Magn. Mater. **452**, 442 (2018).
[21] P. Kollár, Z. Bircáková, J. Füzer, R. Bures, and M. Fáberová, J. Magn. Magn. Mater. **327**, 146 (2013).