A Comparative Study of Insulators on Magnetic Properties of Sendust Based Nanocomposite Powder Cores

Maryam Amoohadi[*, 1]**, Morteza Mozaffari**[2]**, Abdolrasoul Gharaati**[1]**, Mohammad Rezazadeh**[3]

[1] Department of Physics, Payamenoor University, Iran

[2] Department of Physics, Faculty of Science, University of Isfahan, Isfahan, Iran

[3] Department of Materials Engineering, Isfahan University of Technology, Isfahan, Iran

Abstract: In this study, Sendust ($Fe_{85}Si_{9.6}Al_{5.4}$) nanostructure alloy was prepared by mechanical alloying and was characterized by X-ray diffraction. In addition, the alloy's morphology and its distribution particle size were investigated by scanning electron microscopy method. The alloyed powders were insulated with 4wt% of three different insulating materials including polyvinyl alcohol, epoxy resin, and sodium silicate. Then, 0.5 wt% zinc stearate was added as a lubricant and the composites were formed at a pressure of 1600 MPa in a torodial die. These cores were annealed in Ar atmosphere at 350 to 450 ℃ for 1 h. Some of the magnetic parameters of the as-milled and annealed samples were measured and compared with each other in the frequency range of 4 to 1000 kHz. The results showed that polyvinyl alcohol is not a suitable insulating agent for insulating this alloy. In addition, the real part of the initial magnetic permeability of the magnetic powder core insulated by sodium silicate in comparison with that insulated by epoxy resin is a bit smaller, but more reduction of the imaginary part of former results in an increase the Q-factor throughout measuring frequency range. The results show that annealing is more effective on improvement of the measured magnetic properties of the samples insulated with sodium silicate than that insulated with epoxy resin.

1. INTRODUCTION

Soft magnets are materials that are easily magnetized and demagnetized [1]. In general, properties of a proper soft magnet are high permeability, high

---

*Corresponding author. Email: **M.amoohadi@student.pnu.ac.ir**

saturation magnetization, and low coercivity. Permeability and coercivity are affected by materials processing and alloying methods, while saturation magnetization is less sensitive to processing variables and is primarily influenced by the composition of the alloy [2]. The application of soft magnetic materials is classified into two categories of DC and AC applications. Important parameters in DC applications are permeability, saturation magnetization, and coercivity, while in AC applications, in addition to these parameters, is the total core losses generated in the alternating magnetic field [2].

 Powder metallurgy processing can be applied to make magnetic powder cores for AC and DC applications [2]. For DC applications, cores are prepared by a shaping and sintering. In these cores, the proper choice of the alloy and the sintering process can greatly affect the final magnetic properties. sintered cores are not suitable for AC applications, due to generated heat in the alternating field on magnetic properties [3]. This heat is mainly due to the hysteresis and the eddy current losses that are proportional to f and f^2, respectively [1].

Laminated cores are usually made of electrically insulated Fe-Si alloy sheets, which are suitable for 50-60 Hz applications   [4]. Magnetic powder cores (MPCs), made with electrically insulated iron powders or its ferromagnetic alloys such as Fe-Si-Al, Fe-Ni, Fe-Ni-Mo, and Fe-Si, called as soft magnetic composite(SMC), have opened up a broad horizon for high frequency applications [5]. These cores, are usually fabricated under a high pressure of powder insulated by a heat-resistance binder without any sintering process [6]. Insulating increases electrical resistance and then reduces the eddy current and total losses. The pressed cores are usually annealed at a temperature between 400 to 700 ºC to eliminate the internal stresses introduced in the course of the cold press process [7]. Therefore, the higher the thermal stability of the adhesive and the insulating material, the more suitable they will be. Organic and inorganic insulating materials are used to insulate ferromagnetic particles [8]. Organic coating such as, epoxy resin [9], silicon resin [10] and phenolic resin [11] are used by mixing methods for insulating. These materials are used as insulators and/or binders [8]. Inorganic coating such as oxides [12], sulfates [8]and phosphates [13] with higher thermal stability than organic materials [10] are used with wet chemical methods including sol-gel [14], reverse microemulsion [12] and co-precipitation [15]. Research in this field continues to achieve easy, fast and low cost methods to create uniform and thin coating that reduce the losses along with maintenance of core permeability [13].

In general, different factors are affected the magnetic properties of the SMPCs, such as: ferromagnetic material, size of the ferromagnetic particles, type and amount of insulating material, annealing temperature, forming pressure, type of adhesive, and lubricant [16].

The Sendust alloy ($Fe_{85}Si_{9.6}Al_{5.4}$) due to infinitesimal magnetostricion and anisotropy and high resistivity [6] has been considered to fabricate SMPCs. This alloy can be prepared by various methods such as melting and casting, gas or water atomization [17], and mechanical alloying [18]. In most studies, resins has been used as insulating and adhesive, but less attention has been paid to adhesive of sodium silicate (SS) and polyvinyl alcohol (PVA) [8, 9, 19, 20].

In this study, iron, silicon, and aluminum metallic powders were weighted based on Sendust alloy composition, as 85, 9.6 and 5.4 wt%, respectively, and were mechanically alloyed by a planetary ball mill. The Nano SMCs were fabricated by mixtures of Sendust nanostructure alloy and three insulating materials including PVA, epoxy resin (ER) and SS. Then, 0.5 wt% zinc stearate as a lubricant added to the SMCs and then were formed to a torodial die by 1600 MPa. These cores were annealed in an Ar atmosphere at 350 to 450 ℃ for 1 h. Some of the magnetic parameters of cores such as real and imaginary parts of the initial permeability and Q-factor of the unannealed and annealed samples were measured and compared with each other in the frequency range of 4 to 1000 kHz.

2. EXPERIMENTAL METHODS

The used raw materials, were Fe (~ 45μ, Metal powder), Si (~44μ, Aldrich) and Al (~ 100-200μ, Fluka) powders with minimum purities 99%. These materials were weighted and mixed together based on Sendust (Fe85Si9.6Al5.4) alloy composition, as 85, 9.6 and 5.4wt%, respectively. The mixture was loaded into a hardened steel vial (250 mL), together with several hardened steel balls of different diameters (7 and 12 mm) by a ball to powder mass ratio of 10, and then, mechanically alloyed by a planetary ball mill (Fritsch Pulverisette6) at a speed of 350 rpm for 10 h, in an Ar atmosphere. Phase identification, morphology and determination of particle size of alloyed powders were performed at room temperature by X-ray diffractometery (XRD, Philips X'Pert Pro, CuKα radiation, λ=1.5406 Å), scanning electron microscopy (SEM, VEGA\\TESCAN with acceleration voltage 15.0 kV) and field emission scanning electron microscopy (FESEM, MIRA3 TESCAN), respectively. The three composites were made by Sendust alloy and 4 wt% [21] of **a)** sodium silicate (SS) **b)** polyvinyl alcohol (PVA), and **c)** epoxy resin and hardener (ER). The concentrations of solutions of these insulators are shown in table 1. Then 0.5 wt% zinc stearate as a lubricant was added to the composites.

Table 1. The concentrations of solutions of insulators

Insulator name	Solution
SS	60 wt% SiO2.3NaO + 40 wt% water
PVA	5 wt% polyvinyl alcohol + 95 wt% water
ER	67 wt% epoxy resin + 33 wt% hardener

These SMCs were formed in torodial cores (ϕ_{out} =18.7, ϕ_{in} =9.9, h≈5.5 mm) at 1600 MPa by a hydraulic press and dried at room temperature for a day. Green density was determined by the principle of Archimedes. The prepared toroids were annealed in an electrical furnace in Ar atmosphere at 350, 400 and 450 ºC for 1 h with a heating rate of 10 ºC/min and then cooled freely to room temperature. These cores were wired with 30 turns copper wire # 26. Quantities R_s, L_s, and Q-factor were measured with an LCRmeter (Fluke, PM6306) in the frequency range from 4 to 1000 kHz at a low magnetic flux density (B<3.5G).

With regard to the definition of magnetic permeability in linear environments ($\mu = \frac{B}{H}$) and phase delay of B with respect to H in alternating fields, the following equation can be written:

$$\mu = \frac{B_0 e^{i(\omega t - \delta)}}{H_0 e^{i\omega t}}, \tag{1}$$

where in (1) , ω, δ, H_0 and B_0 are the angular frequency, the phase difference between B and H, the amplitudes of magnetic induction and the magnetic intensity field respectively. In addition, $\mu = \mu' - i\mu''$, $\mu' = \frac{B_0}{H_0} \cos \delta$ and $\mu'' = \frac{B_0}{H_0} \sin \delta$. The loss per unit volume is [22]:

$$P = \frac{1}{T} \int_0^T H(t) \frac{dB(t)}{dt} dt = \frac{1}{2} \omega H_0 B_0 \sin \delta = \frac{1}{2} \omega H_0^2 \mu'', \tag{2}$$

and as can be seen the power loss is proportional to μ''. If we consider the magnetic core as a real inductor with an R-L circuit then $Z = i\omega L = R_s + i\omega L_s$, where R_s covers the effect of losses and Ls is the self-induction of core. Thus, real (μ') and imaginary (μ'') parts of the permeability and Q-factor are obtained from following equations [23].

$$\mu_r' = \frac{L_s l_m}{\mu_0 A_e N^2} \tag{3}$$

$$\mu_r'' = \frac{R_s l_m}{\mu_0 A_e N^2 \omega} \tag{4}$$

$$Q = \frac{\omega L_s}{R_s}, \tag{5}$$

where A_e, N, and l_m are the effective core cross section, number of turns and

mean magnetic flux length, respectively.

3. RESULTS AND DISCUSSION

Figure1 shows XRD patterns of the raw materials mixture (Fe, Si, and Al mixture) and 10h milled sample. As can be seen, the XRD pattern of the raw materials contained diffraction peaks of three main elements and after 10 h milling all diffraction peaks of Al and Si disappeared and formed ordered and/or disordered Sendust alloy phases. The mean crystallite size and microstrain were obtained as 19.5 nm and 0.0068, respectively, using MAUD software upon to Rietveld method [24].
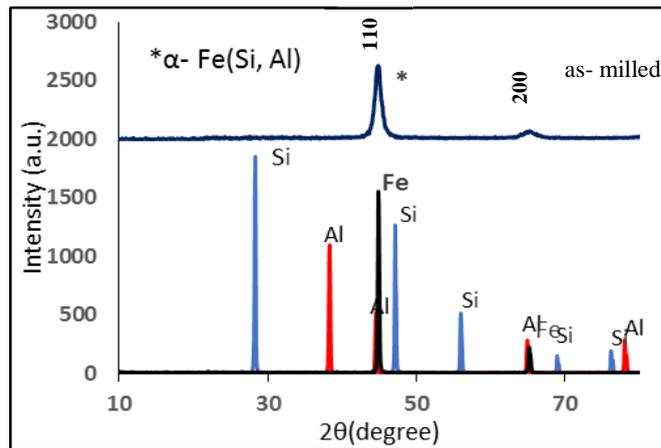
Fig. 1. Room temperature XRD patterns of the mixture of raw materials and as- milled sample.

In addition, on FESEM image mean particle size is ~53 nm, using the Image J software for ~150 particles (Fig. 2).
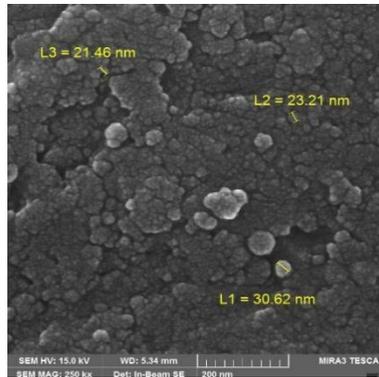
Fig. 2. FESEM image of the as-milled powder.

As the sample insulated by PVA had not sufficient strength and a lower Q-factor than others, so more characterizes had not perform on it.

Figure 3 shows the variations of the real part of initial permeability (μ_r') SSI and ERI -MPCs vs. frequency before annealing. As can be seen, the SSI –MPC has lower μ_r'.
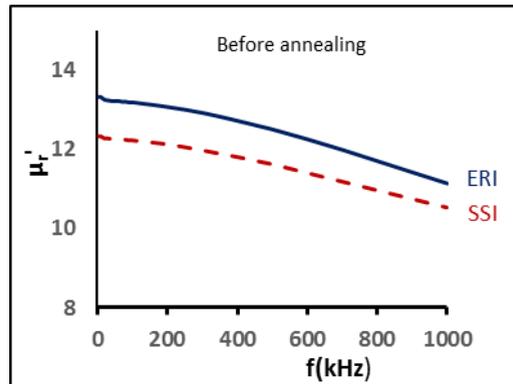
Fig. 3. Varations of μ_r' of the SSI-MPC and ERI-MPC vs. frequency before annealing.

It can be due to this fact that sodium silicate produces more air gaps around the alloy particles in comparison to the epoxy resin, which in turn results in a lower density ($\rho_{SSI\text{-}MPC} = 4.44$ and $\rho_{ERI\text{-}MPC} = 4.87$ g/cm^3) and then a lower μ_r'.

Indeed, non-magnetic impurities behave like distributed air gaps and in the

presence of an air gap, an effective permeability to the core is given by the formula $\mu_e = \mu_r / 1 + \frac{l_g}{l_m} \mu_r$ [25] , in which μ_r , l_g and l_m are relative permeability, air gap length and length of the path of mean magnetic flux, respectively. Then it concludes that an increase in air gap (l_g) results in a decrease in permeability. In fact, the demagnetization field created by the poles formed on either side of the gap, which is proportional to the gap length, reduces the applied field act on magnetic moments sites and in turns effective permeability [26].

 Figure 4 shows the variations of imaginary part of the initial permeability (μ_r'') SSI and ERI- MPCs vs. frequency before annealing (to draw these curves R_s in (5) replaced by $R_{ac}=R_s-R_{dc}$ [27]). As expected, the imaginary part of the SSI-MPC is smaller than that of the ERI-MPC, due to more air gaps.
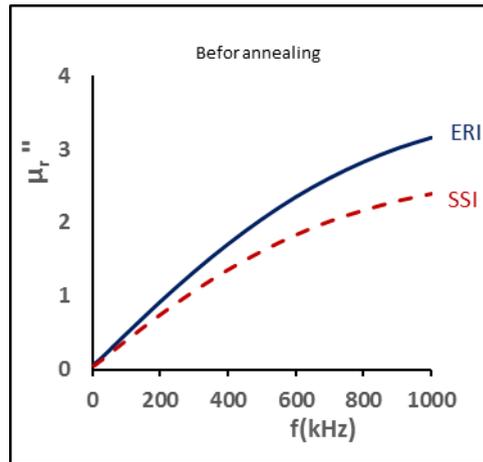
Fig. 4. Varations of μ_r'' of the SSI-MPC and ERI-MPC vs. frequency before annealing.

Figure 5 shows variation of Q-factor vs. frequency for SSI and ERI-MPCs. As can be seen, in the measured frequency range, the Q-factor of the SSI-MPC is higher than that of ERI-MPC.
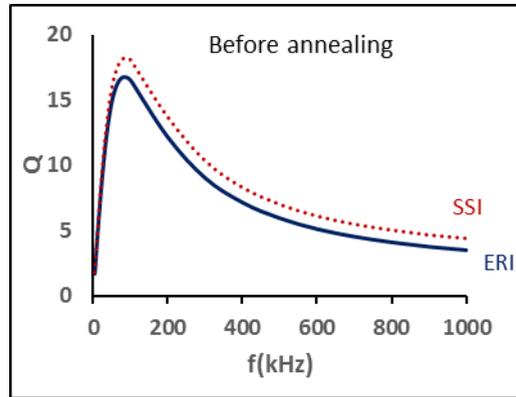
Fig. 5. Variations of Q-factor of the SSI and ERI- MPCs vs. frequency before annealing.

All cores were annealed at 350, 400 and 450 ℃. The Q-factor of cores annealed at 400 ℃ was more than that of 350 ℃. Also, the Q-factor of the core annealed at 450 ℃ and insulated by epoxy resin dropped drastically with frequency. So, cores annealed at 400 °C for further investigations.

Figure 6 illustrates variations of μ_r' vs. frequency after annealing at 400 ℃. By comparison curves data on this Fig. and Fig. 3 can be seen annealing increased μ_r' for both cores. In milling and cold press processes, some plastic deformations occur in powder particles that increase the density of dislocations. These dislocations behave like pining centers and prevent the easy movement of the wall of magnetic domains [28]. Annealing causes a decrease in dislocations and then an increase in permeability [29].

Also, for f<300 kHz, the μ_r' for an ERI-MPC is higher than that of SSI-MPC, but for f>300 kHz the SSI-MPC has a higher permeability. It can be due to more degrading insulation between powder particles for ERI-MPC and increase the amount of porosity, thus increases the presence of eddy currents for f>300 kHz and decreases permeability [29].
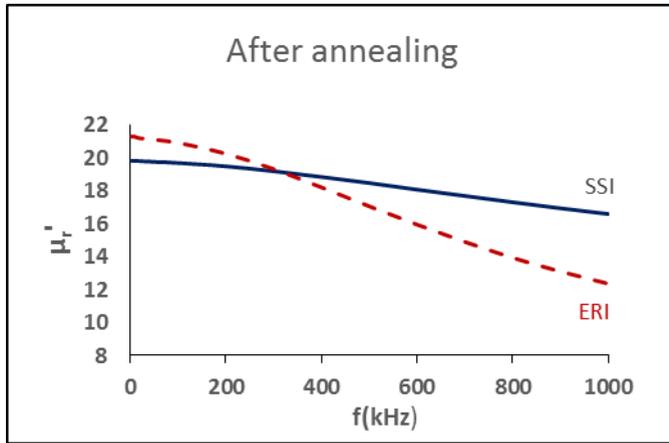
Fig. 6. Variations of μ_r' for SSI and ERI-MPCs vs. frequency after annealing at 400 ℃.

Figure 7 shows the variations of μ_r'' vs. frequency after annealing. By comparison curves data on this Fig. and Fig. 4 can be seen although μ_r'' of the both cores is increased by annealing, but that of ERI-MPC is increased not only from the view point of absolute value, but also from view point of its relative value.
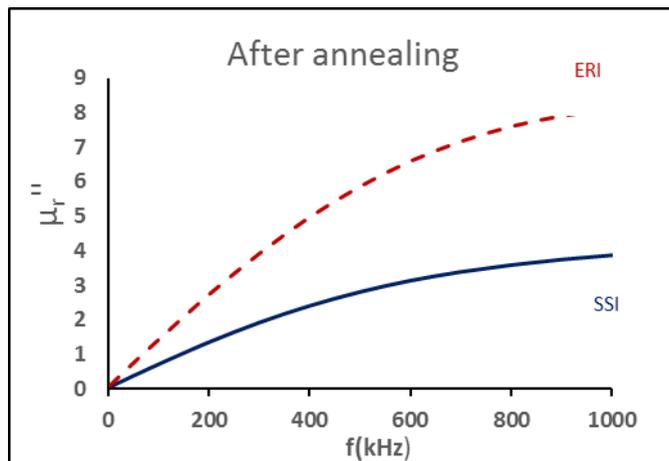
Fig. 7. Variations of μ_r'' vs. frequency after annealing at 400 ℃.

Figure 8 shows the variations of Q-factor vs. frequency after annealing. The

maximum of Q-factor of annealed SSI-MPC has a greater Q-factor in comparison with other. In fact, annealing at 400 ºC for the ERI-MPC has more degraded insulating layer, and the increased losses had reduced the Q-factor [29].
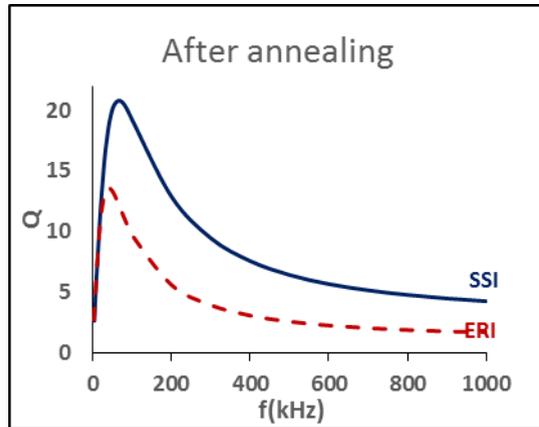
Fig. 8. Variations of Q-factor vs. frequency after annealing at 400 ºC.

4. CONCLUSIONS

In this study, soft magnetic powder cores were fabricated using, mechanical alloyed Sendust nanostructures that insulated by three different insulators and shaped at a pressure of 1600 MPa. The cores insulated by 4% sodium silicate and epoxy resin have sufficient strengths. Results indicated that annealing promote real part of the permeability remarkably. The maximum obtained real part of permeability at frequencies up to 100 kHz is greater than 21, which is related to the annealed epoxy resin-insulated magnetic powder core. But the real part of permeability drops faster than that of one, and it achieves values smaller than others at higher frequencies than 300 kHz. As, the imaginary part of permeability and its relative increase for sodium silicate insulator was is less than other one , it concluded that it has a higher thermal stability than other at 400 ºC. The Q-factor of sodium silicate-insulated magnetic powder core was higher than that of epoxy resin-insulated magnetic powder core before and after annealing. Thus, it can be concluded that for insulation of Sendust, sodium silicate is more suitable than epoxy resin.

REFERENCES

[1] B.D. Cullity, C.D. Graham, *Introduction to magnetic materials,* 2nd ed., New Jersy: John Wiley & Sons, 2011, 439-440.

[2] H. Rutz, F. Hanejko, G. Ellis, N. Riverton. *The manufacture of electromagnetic components by the powder metallurgy process*. Presented at International Conference on Powder Metallurgy and Particulate Materials. (1997, Jun) 1-26. Available: www. Yuna media. Com/ www. Hoeg test. Com /Tech Papers v2 /47. Pdf

[3] A. Schoppa, P. Delarbre, E. Holzmann, M. Sigl. *Magnetic properties of soft magnetic powder composites at higher frequencies in comparison with electrical steels*. Presented at: Electric Drives Production Conference (EDPC), 2013 3rd International, IEEE. (2013, Oct) 1-5. Available: iee explore. Iee. Org/ iel7/ 6678895/ 6689673/ 06689717. Pdf

 [4] J.F. Gieras, *Advancements in electric machines*, Germany: Springer Science & Business Media, 2008, 36-37.

[5] A.W. Chao, K.H. Mess, M. Tigner, F. Zimmermann, *Handbook of accelerator physics and engineering*, 2nd ed. New Jersy: World Scientific, 1999, 117.

[6] A. Goldman, *Handbook of modern ferromagnetic materials*, New York: Springer Science & Business Media, 2012, 197.

[7] K. Asaka, C. Ishihara. *Technical trends in soft magnetic parts and materials*. Hitachi Powdered Metals Technical Report. U.S.A., (2005, Jan.) 420053. Available: https:// www. research gate. net. / publication/287661000

[8] H. Shokrollahi, K. Janghorban. *Soft magnetic composite materials* (SMCs). Journal of Materials Processing Technology. 189 (2007, Jul.) 1-12. Available: https:// www. Science direct. Com/ science/ article/ pii/ S092401360700175

[9] L. Xiao, Y. Sun, C. Ding, L. Yang, L. Yu. A*nnealing effects on magnetic properties and strength of organic-silicon epoxy resin-coated soft magnetic composites*. Proceedings of the Institution of Mechanical Engineers. Part C: Journal of Mechanical Engineering Science. 228 (2014, Dec.) 2049-2058. Available: Journals. Sagepub. Com/ doi/ abs/10.1177/0954406213515112

[10] S. Wu, A. Sun, F. Zhai, J. Wang, Q. Zhang, W. Xu, P. Logan, A.A. Volinsky. *Annealing effects on magnetic properties of silicone-coated iron-based soft magnetic composites*. Journal of Magnetism and Magnetic Materials. 324 (2012, Mar.) 818-822. Available: https:// www. Science direct. Com/ science/ article/ pii/ S0304885311006986

[11] M. Streckova, R. Bures, M. Faberova, L. Medvecky, J. Fuzer, P. Kollar. *A comparison of soft magnetic composites designed from different ferromagnetic powders and phenolic resins*. Chinese Journal of Chemical Engineering. 23 (2015, Apr.) 736-743. Avaiable: https:// www. Science direct. Com/ science/ article/ pii/ S1004954114003280

[12] S. Wu, A. Sun, Z. Lu, C. Cheng, X. Gao. *Magnetic properties of iron-based soft magnetic composites with SiO_2 coating obtained by reverse microemulsion method*. Journal of Magnetism and Magnetic Materials. 381 (2015, May) 451-456. Available: https:// www. Science direct. Com/ science/ article/ pii/ S0304885315000414

[13] M. Huang, C. Wu, Y. Jiang, M. Yan. *Evolution of phosphate coatings during high-temperature annealing and its influence on the Fe and FeSiAl soft magnetic composites*. Journal of Alloys and Compounds. 644 (2015, Sep.) 124-130. Available: https:// www. Science direct. Com/ science/ article/ pii/ S0925838815012402

[14] M. Yaghtin, A.H. Taghvaei, B. Hashemi, K. Janghorban. *Effect of heat treatment on magnetic properties of iron-based soft magnetic composites with Al_2O_3 insulation coating produced by sol–gel method*. Journal of Alloys and Compounds. 581 (2013, Dec.) 293-297. Avaiable: https:// www. Science direct. Com/ science/ article/ pii/ S0925838813016095

[15] X. Peng, K. Peng, J. Huang. S*ynthesis and magnetic properties of core-shell structured Finemet/Ni Zn ferrite soft nanocomposites by co-precipitation* Journal of Alloys and Compounds. 691 (2017, Jan.) 165-170. Available: https:// www. Science direct. Com/ science/ article/ pii/ S0925838816324811

[16] L. Hultman. *Soft magnetic composites-properties and applications*. PM TEC 2002, in Orlando, U.S.A., 2002-6, (2002, Jul.) 1-13. Available: https:// www. Research gate. Net /.../ 4022788

[17] K.W. Bae, J., Byun, *Method for manufacturing sendust core powder,* U.S. patent 5,756,162 (1998) May. 26. Available: http://www.free patents on line. com

[18] B. Zuo, N. Saraswati, T. Sritharan, H. Hng. *Production and annealing of nanocrystalline Fe–Si and Fe–Si–Al alloy powders*. Materials Science and

Engineering: A, 371 (2004, Apr.) 210-216. Available: https:// www. Science direct. Com/ science/ article/ pii/ S0921509303014576

[19] A. Taghvaei, H. Shokrollahi, M. Ghaffari, K. Janghorban. *Influence of particle size and compaction pressure on the magnetic properties of iron-phenolic soft magnetic composites.* Journal of Physics and Chemistry of Solids, 71 (2010, Jan.) 7-11. Available: https:// www. science direct. com/ science/ article/ abs/ pii/ S0022369709002364

[20] H. Shokrollahi, K. Janghorban. *The effect of compaction parameters and particle size on magnetic properties of iron-based alloys used in soft magnetic composites*. Materials Science and Engineering: B, 134 (2006, Sep) 41-43. Available: https:// www. Science direct.Com/science/article/pii/ S0921510706004090.

[21] M. amoohadi, M. mozaffari, A.R. Gharaati, M. rezazadeh. *An Investigation of the Magnetic Properties of Iron-based Soft Magnetic Composites Coated by Sodium Silicate Insulator*. Journal of Advanced Materials In Engineering, 37 (2018, Jan) 97-104. Available: https://jame.iut.ac.ir/article-1-898-en.html

[22] F. Fiorillo, *Measurement and characterization of magnetic materials*, Amestwdam: Elsevier, 2004, 427.

[23] E. Snelling, *Ferrites for Inductors and Transformers*, London: Butter worth and Co, 1983, 47-70.

[24] L. Lutterotti. *Total pattern fitting for the combined size–strain–stress–texture determination in thin film diffraction*. Nuclear Instruments and Methods in Physics Research Section B, Beam Interactions with Materials and Atoms, 268 (2010, Feb.) 334-340. Avaiable: https:// www. Science direct. Com/ science/ article/ pii/ S0168583X09010428

[25] Y. Liu, Y. Yi, W. Shao, Y. Shao. *Microstructure and magnetic properties of soft magnetic powder cores of amorphous and nanocrystalline alloys*. Journal of Magnetism and Magnetic Materials, 330 (2013, Mar.) 119-133. Avaiable: https:// www. Science direct. Com/ science/ article/ pii/ S0304885312008657

[26] B.D. Cullity, C.D. Graham, *Introduction to magnetic materials*, 2nd ed. New Jersy: John Wiley & Sons, 2011, 73-76.

[27] A.H. Taghvaei, H. Shokrollahi, K. Janghorban, *Structural studies, magnetic properties and loss separation in iron–phenolicsilane soft magnetic composites*, Materials and Design, 31 (2010, Jan.) 142-148. Avaiable in: https:// www. Science direct. Com/ science/ article/ pii/ S0261306909003392

[28] H. Shokrollahi, K. Janghorban. *Different annealing treatments for improvement of magnetic and electrical properties of soft magnetic composites*. Journal of Magnetism and Magnetic Materials. 317 (2007, Oct.) 61-67. Avaiable: https:// www. Science direct. Com/ science/ article/ pii/ S030488530700604X

[29] Q.D. Li, X. Dong, T. Liu, J.H. You, F.Z. Lian. *Research on Preparation and Properties of Fe-Si-Al Soft Magnetic Composites*. in: Advanced Materials Research, Trans Tech Publ. (2011, Jul.) 173-178. Avaiable: https:// www. Scientific. Net/ AMR. 298.173