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Properties of Fe-based nanocrystalline magnetic powder cores (MPC) and structure of particle size distribution (PSD)

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This paper discusses the influence of the Particle Size Distribution (PSD) of the nanocrystalline Fe-based granular-soft-magnetic material on the final magnetic properties of a Magnetic Powder Core (MPC). Here we show how PSD impacts the final magnetic properties. Mixing fine and coarse particles, with a dominance of coarse particles, significantly influences the magnetic permeability increase of the core. Better magnetic features are noted for MPCs constructed with certain mass ratio of fine and coarse particles due to improvement in the magnetic path in the cores. This allows to offer new induction components to industry.

K e y w o r d s: soft magnetic material, magnetic powder core, nanocrystalline strip, particle size distribution

1 Introduction

Electrical, electronic and telecommunication devices are more and more often equipped with inductive components operating at kHz or higher frequencies. Such components are with compact volume but also required cores possessing proper geometric shape universality and excellent magnetic properties. Due to these facts powdered and compacted magnetic cores applicability with Fe-Ni [1], Fe-Co [2, 3, 4] and Fe-based [5] are under very intensive research nowadays. A magnetic powder core (MPC) is a distributed-air-gap core with an arbitrary construction-for example, toroidal, race-track, block, etc. Magnetic granular material involves conventional magnetic particles-such as Fe and Fe-Co, Fe-Ni alloys-or very soft-magnetic particles such as amorphous [6], nanocrystalline [6, 7] and microcrystalline [1]. Popular types of binder include resins, polymers and mineral materials [8]. The particles are scattered by size distribution and sieve analysis [9] or a more advanced method [10] yields the particle size distribution (PSD), which is the statistical function reflecting particle size dispersions [11, 12, 13].

Due to stable permeability and low power loss at a high frequency bandwidth, MPCs are used in power circuits [14, 15], switching power supplies [15, 16], filter inductors and smoothing chokes [17], radio and communication devices [15], coupling devices [18] or electrical motors and generators [17, 19], diesel fuel injection valves or reactors in boost converters for hybrid cars [17] and many others.

There are general papers which focus on the influence of MPC preparation stages on magnetic properties [20, 21]. In [8] the authors focused on the comparison between cores made of amorphous and nanocrystalline magnetic powder material within a limited range of particle sizes from 110 μm to 300 μm. They highlighted that the magnetic permeability of both cores was stable up to a frequency of approximately 1 MHz. Some papers like [22, 23] have focused on the production process of particles from soft nanocrystalline materials. These authors compared the magnetic features of MPC made in limited PSDs, such as 10 μm to 18 μm and 109 μm to125 μm.

It should be noted that no known literature analyzes the influence of PSD shapes on the fundamental features of MPC. In order to extend the existing analysis, we focus on MPCs made of nanocrystalline magnetic particles, flakes and idles. By modifying particle size classes in PSD, we would like to demonstrate how to control and organize the magnetic features of MPCs.

2 Experimental details

Nanocrystalline soft magnetic strips are mostly recent materials which still supersede power ferrites for high frequency application [1, 5, 23]. Magnetic cores under consideration were formed by the use of magnetic powder made of commonly used nanocrystalline strips with the $Fe_{81}Si_{9.7}B_{2.5}Nb_{5.5}Cu_{1.3}$ chemical composition. The strip was heated at a temperature of 823 K for 1 hour in an argon atmosphere under technology where recuperation system is used [24]. Nanocrystalline powder was obtained by grinding the annealed nanocrystalline strips in a preliminary crushing machine with worm adjustment,

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Table 1. Characterization of particle size classes used in the analysis

Feature	$d_{min}(m)$	$d_{max}(m)$	$(d(m)$	$d_m(m)$
fine	0_{residue}	25	20	17.5
very low	25	45	20	35.0
low	45	100	55	72.5
intermediate	100	250	150	175.0
high intermediate	250	500	250	375.0
coarse	500	750	250	625.0

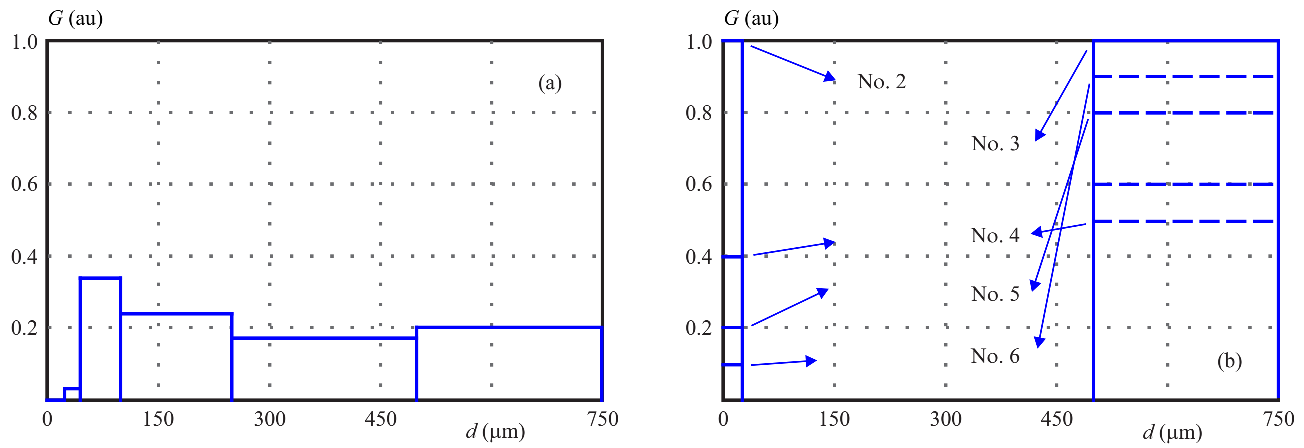


Fig. 1. Typical PSD in the form of histograms for various Samples

in air atmosphere at a speed of 150 rpm for 15 min. Next, a Vibratory Disc Mill RS 200 was used to obtain a fine magnetic powder with the friction and impact method [9]. But the research on flakes [25] obtained from nanocrystalline ribbons remains beyond the scope of the paper. This method uses the horizontal vibrations of the plate with the jar, in which the magnetic powder is inserted, and the rolling rings. The process was performed in air atmosphere at a speed of 750 rpm, two times for 4 minutes with a 250 ml powder jar. Next, the powder obtained was subjected to PSD analysis and then sorted into particle size classes [26]. Tab 1 presents the particle size classes.

In Table 1, $d_{\min}$ indicates the minimum dimension of a particle in one size class, where $d_{\min} = 0$ μm indicates the sieve residue; $d_{\max}$ is the maximum dimension of a particle in one size class while (d denotes the size range of the particles in one size class and d_m is the mean dimension of a particle in one size class.

Composite materials with different particles size classes were mixed (Fig. 1) and then bonded by using an 8.5% wt polyethylene LDPE binder. We used hand and hydraulic presses in order to compact the powder in Sample No1, Fig.1(a) under two overpressures of 200 MPa (Sample No1a) and 800 MPa (Sample No1b), as describe on Tab. 2 and in Fig. 4., respectively. Subsequently, the formed cores were submitted to thermal treatment at 190 °Cper 1.5 hour. In the way, 6 sets of Samples as toroidal cores with the following final dimensions were prepared: outer dimension 30.0±0.1 mm, inner dimension 17.7±0.1 mm and height 12.5±0.7 mm. Fig. 1 shows the construction of the histograms where the samples were distinguished and numbered. Sample No1 - presented by Fig. 1(a), involves full particle size classes.

Samples No2,3 represent individual particle size classes for fine and coarse particles, alternatively. Samples 4, 5 and 6 show mixtures of fine and coarse particles with mass proportions as shown in Fig. 1b [26]. For example, sample 5 is the mixture of 80 (coarse)/20 (fine) % and sample 4 is the mixture of 60/40 % as can be seen on Fig. 1(b).

The PSD analysis was conducted by using sets of sieves and weights, thereby establishing the density of MPC. The measurements of electrical resistivity were performed with a standard method system having separate current and potential leads [27]. Other MPC parameters were tested with a professional measurement system [24] that is suitable for collecting some magnetic properties as flux density $B(H)$, relative magnetic permeability μ_r and specific power loss $P(B)$. The measuring device encompasses National Instruments LabView software along with a high speed, and high accuracy, 16-bit NI PCI-6110 DAQ card. We determined 56 turns urns for exciting windings and 5 turns for secondary windings. The specified number of turns, referring to the frequency values, follows the voltage limitation of the measuring card (up to 10 V). The given number of turns provides the appropriate signal to noise ratio [28]. The magnetic parameters of each core are registered for 3 values of frequency: 50 Hz, 1000 Hz and 5000 Hz. For quick measurement at 50

Table 2. Basic magnetic powder cores (MPC) properties

Sample No	Mass density	Electrical resistivity (mΩcm)	Coercivity H_c (A/m)	Remanence B_r (mT)
1a	2.59	2432.4	25	1.0
1b	3.93	143.7	54	3.0
2	3.53	338.2	176	3.6
3	4.05	143.3	62	4.0
4	3.63	339.9	78	5.7
5	3.89	237.3	61	5.1
6	3,96	311.0	75	4.2

Measured at: B=0.1 T, f =1 kHz

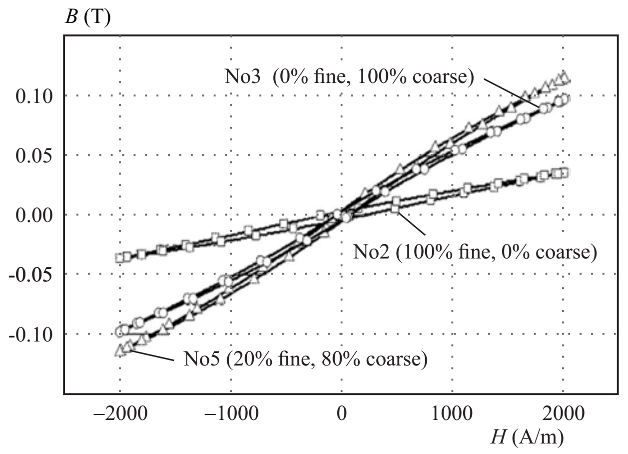


Fig. 2. Hysteresis loops for selected MPCs

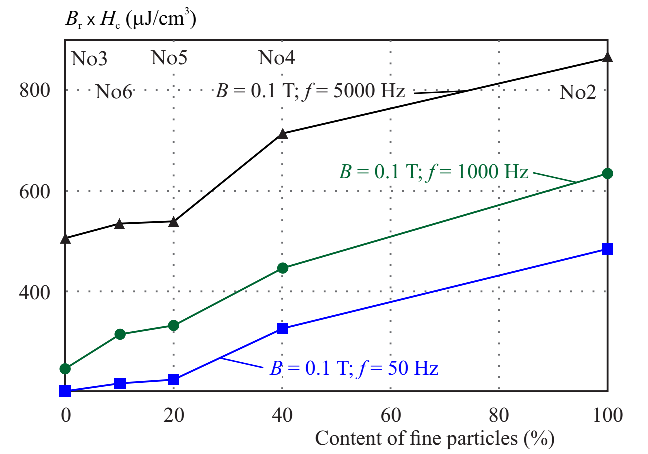


Fig. 3. Specific energy density H_c, B_r generated at the content of fine particles depending on different frequencies

Hz or for quality check-in the ring method [29, 30] was successfully used.

3 Results and discussion

3.1 Basic measurement

Density γ electrical resistivity ρ, coercivity H_c and remanence B_r of powder cores are shown in Tab. 2. It should be noted total power losses depend on electrical resistivity and coercivity but any analysis of quantities relation is above of the scope of the paper.

By comparing Samples No1a and No1b, it was observable that under a higher compaction pressure, the density of the core increases and electrical resistivity (drops because in the same volume there is more magnetic material and the insulation of the particle is reduced. Moreover, a core made exclusively from coarse flakes has the greatest density of magnetic powder and the smallest electrical resistivity. Due to the large surface of the particles, it is harder to coat a particle with an identical insulation layer than a fine one. By adding fine particles to the coarse particles, one may observe a lower density in these cores [22]. Furthermore, fine particles have higher value of coercivity H_c, which is related to tension in the powder and specific power loss P [22].

Fig. 2 shows examples of hysteresis loops.

All hysteresis loops are very narrow what is a classic feature of soft magnetic materials independent of granular structure. It is noted that the content of fine particles is important for the angle of inclination of the hysteresis loop, as magnetization shape is. Because coercivity and remanence have small values, we decided to analyze the product of these two quantities H_c B_r , called the specific energy density. Figure 3 shows such results. Here have to be noted that the specific energy density increases with the increase in the content of fine particles, as well as when the frequency increases.

3.1 Influence of compact pressure

Figure 4 compares two toroidal cores of Sample No1. The cores were formed under different levels of pressure: 200 MPa (Sample No1a) and 800 MPa (Sample No1b).

A progression in the magnetic properties is obtained by use of higher levels of compact pressure. This is due to the higher compaction of the magnetic material in a defined core volume. In this way smaller gaps are insert

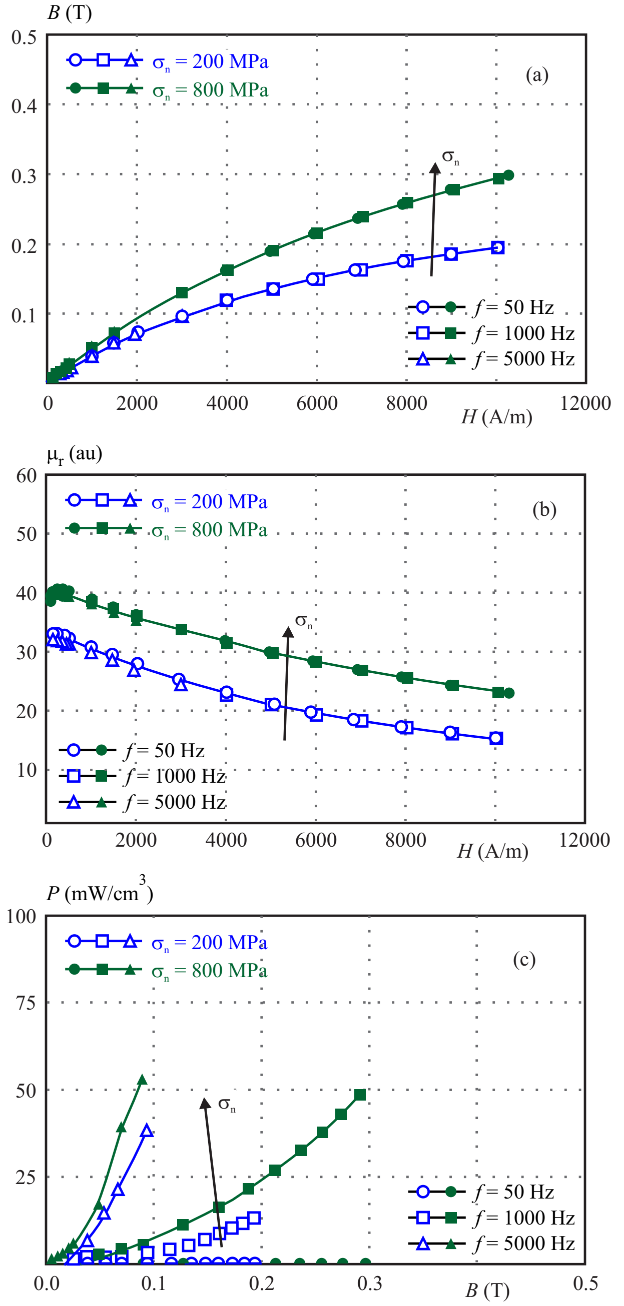


Fig. 4. Influence of Sample No1a and 1b (Tab.2) compact pressure on magnetic properties: (a) – magnetization characteristic, (b) – relative magnetic permeability, and (c) – specific power loss

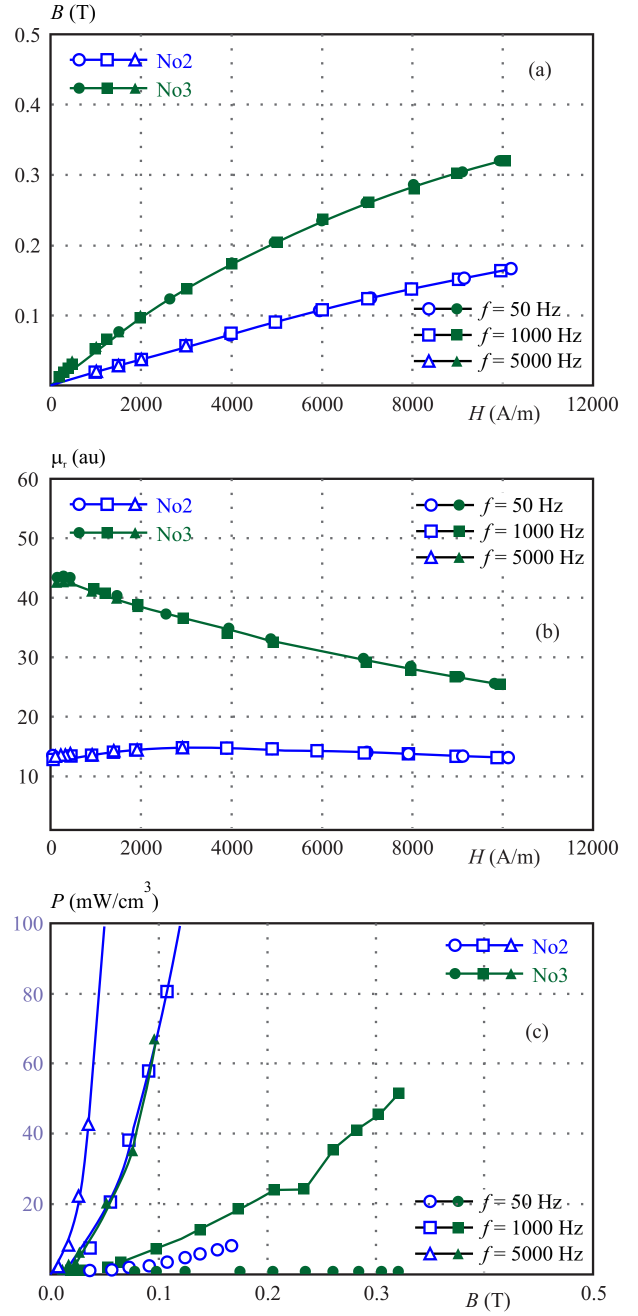


Fig. 5. Magnetic properties for individual size classes of fine and coarse particles made under compact pressure 800 MPa: (a) – magnetization characteristic, (b) – relative magnetic permeability, and (c) – specific power loss (c)

between the particle-particle contact points. On the other hand, the strong compaction process worsens the electrical insulation between the magnetic particles and increases eddy current loss, Fig. 4(c). For H =10 kA/m the flux density B rises from 0.2 T in the hand press core to 0.3 T in the core made by hydraulic press, Fig. 4(a). One may note the same behavior for the initial permeability, which increases from 32 to 40. The specific power loss in MPC most often is divided into two components [31]: a static element dependent linearly on the frequency and a dynamic one varying with the second power of the frequency. The first component is associated with hysteresis and the second with eddy currents flowing between and inside the powder particle and, therefore, strongly dependent on electrical resistivity. Figure 4(c) presents the increase in the specific power loss with frequency and with the compaction method/pressure. However, an increase in pressure causes a reduction in the insulation between the particles and an increase in eddy currents. Furthermore, higher level of pressure augments the internal stress in the particles, contributing to hysteresis loss.

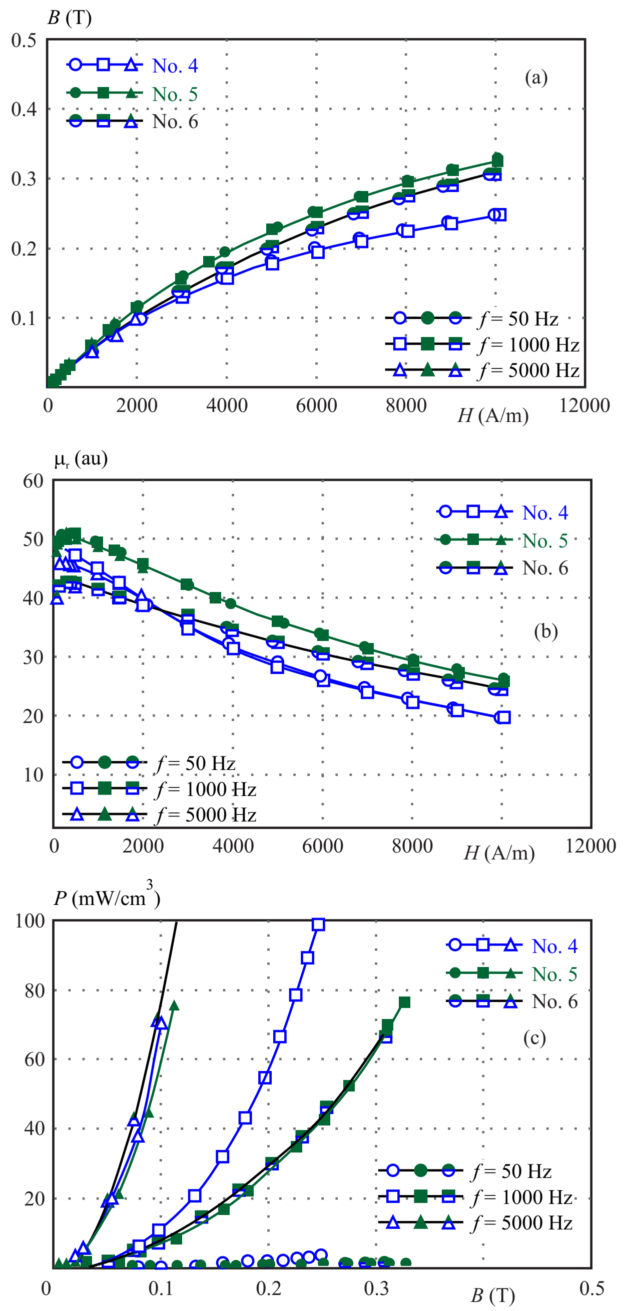


Fig. 6. Magnetic features for Samples No4,5,6 made under compact pressure 800 MPa: (a)-magnetization characteristic, (b)-relative magnetic permeability, and (c)-specific power loss

3.1 Fine vs q coarse magnetic particles

The different magnetic properties that are dependent on the particle size class are noticed. Figure 5 shows the magnetic features of individual particle sizes- *ie*Sample No2 for fine particles and Sample No3 for coarse particles, respectively.

MPC made of fine magnetic particles exhibits lower flux density B, Fig. 5(a) and relative permeability μ_r, Fig. 5(b) at the same magnetic field strength H than the core made of coarse magnetic particles. Here, we note particle-particle contacts between coarse particles. The relative permeability r starts from a high value of 44 and drops to 25 for $H = 10000$ A/m. Thus, the distributed-air-gaps between the coarse particles are less than in the fine particle ones. However, the core made of fine magnetic particles has a flat or more linear $B(H)$ curve, Fig. 5(a). The relative permeability μ_r is steady at value 13 and is practically independent from the magnetic field strength H . Furthermore, specific power loss is much higher for fine magnetic particles at all frequencies, Fig. 5(c). This can be due to the hysteresis loss component as a result of domain walls movement in the particles as coeversities Hc are very low. This is also because magnetic domain walls encounter less structural defects as particle boundaries in coarse particles (Sample No3) in comparison with fine ones (Sample No2).

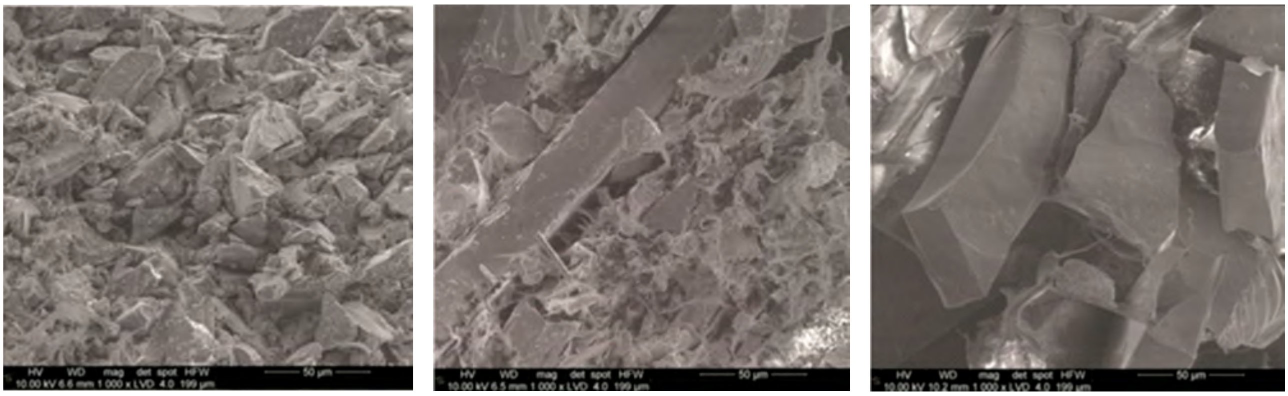
3.4 Mixture of fine and coarse particles

Figure 6 presents magnetic data for MPCs featuring mixtures of fine and coarse particles for Sample No4,5 and 6, Fig. 1(b).

Some samples reflect the mass ratios of fine to coarse particles as presented by Fig. 1(b). In direct comparison with previous and present results, we noted better magnetic properties for Sample No5, where the mass ratio was 20(fine)/80(coarse)%. The reason for the improvement in such properties involves more accurate and better filling of random air-gaps between coarse particles by fine ones [26]. In cores made of coarse powder, there are some air spaces between particles which are too small to fill with coarse particles but there is enough space for fine particles. This procedure improves the properties of the magnetic path in the powder core and increases relative magnetic permeability μ_r, Fig. 6(b). Thus, flux density B, Fig. 6(a), increases along the whole range of magnetic field strength H and the initial values of relative permeability increases by approximately 10 in comparison with the permeability in Sample No3. (Fig. 5.) However, the relative permeability in Fig. 6(b) drops more sharply and at $H = 10000$ A/m and attains a level of 25 as in Sample No3. In all 3 samples presented above, the specific power loss, Fig. 6(c), increases proportionally to the amount of fine particle contents in MPC. The biggest differences are for the 1000 Hz frequency in Sample No4. Here, we still confirm the superior properties of Sample No5, which exhibited an optimal composition of fine and coarse particles during this experiment. However, the power loss is insignificantly higher for Sample No5 than that of Sample No6, where coarse particles dominate, Fig. 6(b).

3.5 Internal structure of MPCs

Figure 7 presents the Scanning Electron Microscope (SEM) images of MPCs made of different granular magnetic materials. As noted in Fig. 7(a), Sample No2 (100% of fine particles) has lowered the distributed air gap but individual fine particles have lower relative permeability. Opposite direction is represented by Fig. 7(c), where Sample No3 (0% of fine and 100% of coarse particles) has higher the distributed air gap in comparison to previous.



(a) (b) (c)

Fig. 7. SEM images for MPCS at magnitude 1000 *times*: (a) – Sample No2 (100 % of fine particles), m (b) – Sample No5 (20 % of fine and 80 % of coarse particles); and (c) – Sample No3 (0 % of fine particles)

However, the individual particles (flakes) have higher relative permeability. Fig. 7(b) (Sample No5) is a compromise between coarse and fine particles. Here we noted a drop in the distributed air gap by filling the gap with fine particles. Therefore, macroscopic parameters such as relative permeability and power loss have been improved.

4 Conclusions

MPCs, made of nanocrystalline soft-magnetic materials, are appropriate not only for compact pressures forming the core but also for the shape of particle size distribution (PSD). During the research we implemented an additional step in MPC production-*ie* the mixing of fine and coarse particles with different mass ratios. This has a significant practical application [26]. Here, better magnetic features are noted for MPCs constructed with fine and coarse particles with a mass ratio of 20(fine)/80(coarse) % due to improvement in the magnetic path in the cores. The paper demonstrates the production technology may be replicated and due to this fact, new induction components are recently offered to industry.

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References

[1] D. Oleksakova, P. Kollar and J. Fuzer, "Structure and magnetic properties of powdered and compacted FeNi alloys", *Journal of Electrical Engineering (2017)*, vol. 68, no. 2, pp. 163-166.

[2] I. Janotova, P. Svec, Sr., I. Matko, P. Svec, Jr., J. Zigo and D. Janickovic, "Structure of rapidly quenched Fe-Co-Sn-B system with varying Fe/Co ratio", *Journal of Electrical Engineering (2015)*, vol. 66, no. 5, pp. 297-300.

[3] K. Zabaransky and S. Oldrich, "Properties of FeCo nanopowder prepared by chemical synthesis", *Journal of Electrical Engineering (2010)*, vol. 61, no. 5, pp. 299-301.

[4] I. Hegudus, Z. Birackova, P. Kollar, B. Weidenfeller, J. Fuzer, P. Kurek, R. Bures, M. Faberova and W. Kavecansky, "Energy Losses Composite Materials Based on Two Ferromagnets", *Journal of Electrical Engineering (2010)*, vol. 61, no. 5, pp. 299-301.

[5] J. Fuzer, P. Kollar, J. Fuzerova and S. Roth, "Soft Magnetic Properties of Nanostructured Vitroperm Alloy Powder Cores", *IEEE Transaction on Magnetics (2017)*, vol. 53, no. 12,.

[6] R. Hilzinger and W. Rodewald, "Magnetic Materials. Fundamentals, Products, Properties, Applications", *Publics Publishing (2013)*, Erlangen, Zweigniederlassung der PWW GmbH.

[7] G. T. Niklova and V. C. Velchleva, "Nanocrystalline magnetic materials versus ferrites power", *The 6th International Conference on Mining Science and Technology (2009)*, no. 1, pp. 1357-1361.

[8] Y. Liu, Y. Yi, W. Shao and Y. Shao, "Microstructure and magnetic properties of soft magnetic powder cores of amorphous and nanocrystalline alloys", *Journal of Magnetism and Magnetic Materials (2013)*, vol. 330, pp. 119-133.

[9] "RETSCH GMBH, General catalogue (2016)", Haan.

[10] "MALVERNS INTRUMENT LTD.", Product support (2017), United Kingdom.

[11] Y. Dong, Q. Man, J. Zhang and C. Chang, "Fabrication of FePBNbCr Glassy Cores With Good Soft", *IEEE Transactions on Magnetics (2015)*, vol. 51, no. 11, Article no. 2006704.

[12] P. Gomert, H. Heiffer, E. Sinn, R. Muller and W. Schippell, "Nanocrystalline M-Type Hexaferrite Powders: Preparation, Geometric and Magnetic Properties", *IEEE Transactions on Magnetics (1994)*, vol. 30, no. 2, pp. 714 - 716.

[13] L. K. Varga, D. Kaptas and T. Kemeny, "Soft Magnetic Nanocrystalline Powders Obtained by Mechanical Grinding", *Materials Science Forum (1998)*, vol. 262-272, pp. 731-736.

[14] Y. Igbal, H. Davies, M. Gibbs and T. Woodcock, "Nanocrystalline powder cores for high frequency applications", *Journal of Magnetism and Magnetic Materials (2002)*, vol. 242-245, no. 1, pp. 282-284.

[15] Y. Zhang, P. Sharma and A. Makino, "Production and Properties of Soft Magnetic Cores Made From Fe-Rich FeSiBPCu Powders", *IEEE Transactions on Magnetics (2015)*, vol. 51, no. 1, Article no. 22800104.

[16] J. Petzold, "Advantages of softmagnetic nanocrystalline materials for modern electronic applications", *Journal of Magnetism and Magnetic Materials (2002)*, vol. 242-245, no. 1, pp. 84-89.

[17] T. Ueno, H. Tsurata, A. Watanabe, T. Ischmine and K. Yamada, "Practical and Potential Application of Soft Magnetic Powder Cores with Superior Magnetic Properties", *SEI Technical Review (2016)*, no. 82, pp. 9-15.

[18] P. Gramatyka, A. Kolano, R. Kolano-Burian and M. Polak, "Nanocrystalline iron-based powder cores for high frequency applications", *Journal of Achievements Materials and Manufacturing Engineering (2006)*, vol. 18, no. 1-2, pp. 99-102.

[19] M. Manivel Raja, N. Ponpandian, B. Majmudar, A. Narayanasami and K. Chattopadhyay, "Soft Magnetic properties of nanostructured FINEMET alloy powder cores", *Materials Science and Engineering (2001)*, vol. 304-306, pp. 1062-1065.

[20] Y. Liu, F. Min, T. Qiu, J. Zhu and M. Zhang, "Effect of the grain size on magnetic properties of nanocrystalline", *Advanced Materials Research (2011)*, vol. 308-310, pp. 685-688.

[21] X. L. Zhou, D. F. Liang and X. Wang, "Studies on Magnetic Properties and Ball-Milling", *IEEE Transactions on Magnetics (2015)*, vol. 51, no. 11, Article no. 15303367.

[22] C. Huang, T. Liu, X. Wang, C. Lu, D. Li and Z. Lu, "Magnetic Properties of Nanocrystalline Powder Cores Fabricated by Mechanically Crushed Powders, ", *ournal of Iron and Steel Research International (2015)*, vol. 22, no. 1, pp. 67-71.

[23] M. Muller, A. Novy, M. Brunner and R. Hilzinger, "Powder composite cores of nanocrystalline soft magnetic FeSiB-CuNb alloys", *Journal of Magnetism and Magnetic Materials (1999)*, vol. 196197, pp. 357-358.

[24] M. Soinski, J. Leszczynski, C. Swieboda and M. Kwiecen, "Nanocrystalline Block Cores for High-Frequency Chokes", *IEEE Transactions on Magnetics (2014)*, vol. 50, no. 11, Article no. 2801904.

[25] F. Mazaleyrat, V. Leger, R. Lebourgeoise and R. Barrues, "Permeability of Soft Magnetic Composite From Flakes of Nanocrystalline Ribbon", *IEEE Transactions on Magnetics (2002)*, vol. 38, no. 5, Article no. 7451148.

[26] J. Leszczynski, M. Soinski, R. Rygal *et al*, "Manufacturing method for magnetic powder cores made of nanocrystalline magnetic materials", *patent pending no. P. 420752*, Polish Pending Patent Office (2017), Warsaw, Poland, vol. 15/2017, pp. 35.

[27] ASTM and A712, "Standard test method for electrical resistivity of soft magnetic alloys", , 2007.

[28] F. Fiorillo, "Characterization and Measurement of Magnetic Materials", , Academic Press (2004).

[29] IEC and 60404-6, "Methods of measurement of the magnetic properties of magnetically soft metallic and powder materials at frequencies the range 20 Hz to 200 kHz", ,.

[30] R. Pytelch, R. Rygal, W. Pluta and M. Soinski, "Quick Measurment Procedure for Quality Flux Density Checking at 0. 3 A/m Toroidal Cores", *Journal of Electrical Engineering (2004)*, vol. 55, no. 10, pp. 95-96.

[31] J. Fuzer, P. Kollar, J. Fuzerova and S. Roth, "Properties of Nanostructured Vitroperm Alloy Powder Cores", *IEEE Transactions on Magnetics (2010)*, vol. 46, no. 2, pp. 471-474.

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