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INFLUENCE OF BALL TO POWDER RATIO AT MECHANICAL MILLING ON THE COERCIVITY OF SOFT MAGNETIC COMPOSITES

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Abstract: Soft magnetic composites (SMCs) represent specific and useful class of materials with expanding application range. They are intensively studied to reveal its large potential for their properties improvement. These soft magnetic compacted powdered materials are used in a variety of electromagnetic applications such as computer, relay, disk drive, printer, hearing aid devices and others. The aim of this work was determination the coercivity of iron based SMCs prepared from iron particles, which were milled with different intensity.

1 Introduction

Magnetic materials are very famous and important for engineering and technical practice. These materials have revolutionized in materials research, physics, electronics and electrical engineering. Magnetic materials and their products are used in the construction of magnetic circuits, in generators, transformers, electric motors, cores of the coils, sensors and also as storage media in IT technology. Some of magnetic materials are known for many years and some of them have only been discovered in recent decades. It can be expected that the advancing development of newly discovered magnetic materials will bring their using in areas where they have not been used before.

Physicists and engineers also take advantage of electromagnetism in the production of magnetic materials, as everything around us, from elementary particles to galaxy clusters, has more or less different electromagnetic properties.

2 Soft magnetic materials

Today, magnetic materials occupy important economic position. These materials can be distributed by various physical and magnetic properties. We know that according to the arrangement of the basic magnetic moments of the atoms of which the material is composed, we distinguish between diamagnetic, paramagnetic and ferromagnetic materials [1]. Ferromagnetic substances are usually metals or alloys based on 3-d transition elements Fe, Co, Ni and according to basic magnetic properties, for example coercivity, can be divided into soft magnetic materials and hard magnetic materials [2].

Hard magnetic materials mostly known as permanent magnets are characterized by a wider hysteresis loop and a high value of the energy product, which indicates how much energy the permanent magnet creates in its surroundings. These materials are difficult to magnetize and demagnetize, they have high values of coercivity. They include for example neodymium magnets, hexagonal ferrites, rare earth magnets (Samarium Co5) and others. They are mostly using for digital storing of information in computer industry, telecommunications, magnetic filters, but also as in measurement and control technologies [2-3].

Soft magnetic materials are characterized by a narrower hysteresis loop, high permeability value and low coercivity, they are easily magnetized and demagnetized. These include Fe, Ni, Co alloys, various amorphous and polycrystalline alloys and are used for example in the cores of coils. For soft magnetic materials the most important is to have low value of coercivity as possible to prevent energy losses and high permeability. Because of their ability to be easily to magnetize and demagnetize they are actively using in progressive brunches of technologies, starting from transformers [2] and finishing with cosmology and medicine depending on type of materials [4-6].

3 Soft magnetic composite materials

The fact that metal ferromagnets generate eddy currents perpendicular to the direction of magnetic induction when they are re-magnetized in alternating magnetic fields is well known. If the magnetic induction flux flowed in the entire cross section of the homogeneous ferromagnet, the reduction of the magnetic field by the eddy currents and thus the reduction of the magnetic induction flux would be considerable. This undesirable phenomenon is partially eliminated by filling the cross-section of the original ferromagnet with thin sheets coated with an electro-insulating coating, so that the eddy currents flow only in a smaller thickness of the ferromagnet and thus reduce the magnetic flux much less than in the previous case. A further reduction is possible if a large number of small mutually insulated particles are deposited in the cross section, in which a further reduction in the effect of eddy currents has occurred. We can say that the concept of further reduction of eddy current over magnetization losses is successfully fulfilled by soft magnetic composite materials (SMCs) [2,4-6].

SMCs are generally compacted ferromagnetic powder particles which are coated with a thin layer of inorganic insulation, because of its better temperature resistance. Those powder particles are randomly arranged with each other what forming a heterogeneous structure. SMC have lot of advantages over other soft magnetic materials. For example, SMCs combining better microstructure-operational conditions (such as isotropic 3D behaviour and mechanical stability, higher permeability and saturation induction constant for a wide range of frequencies and lower coercivity which minimizes energy loss) than the main commercial soft magnetic materials, which is the electrical steel (1.878 billion tons produced in 2020 in the world [7]). Besides, SMCs have a high potential in use also because of low-cost production and environmentally friendly recycling [8]. Therefore, developing of new technologies, especially in the electrical industry and powder metallurgy, has brought new possibilities for the use of the SMCs materials. So, today, we can see them to be used as transformer cores [5], but they can also be used in motors [9-13], inductors [14-15] or generators [16-19].

4 Preparation of samples

SMCs could be possibly made from different soft magnetic materials with adding nonmagnetic inorganic or organic layer constituents for insulation. As ferromagnetic powder particles could be used pure iron or its alloys (Permalloy, Supermalloy, Fe-Ni-P, Fe-Si, Fe-Co, Fe-Co-V, Fe-Si-B-Nb-Cu etc.) [20].

We have prepared three types of compacted (ring form) samples of iron SMCs (S1, S2, S3). We have used Alfa Aesar high-purity iron granules (99.98%, granule size 1-2 mm), which were milled for making ferromagnetic powdered composites from needed fraction particles.

Over the past several years the method of mechanical milling was widely spread in order to exploit it to produce a variety of equilibrium and non-equilibrium alloy phases and posses further possibility for research work and application of permalloy. The advantage of this process technology is that the powder can be produced in large quantities and the processing parameters can be easily controlled [21]. Various types of mills are used to prepare powders by mechanical milling. The mill usually consists of a milling vessel into which one or more balls are placed together with the powder. The milling vessel and balls are made of sufficiently hard materials (for example steel, agate, tungsten carbide). We know several types of mills, which differ from each other in their capacity, milling efficiency, the construction of the mill or in the principle of operation.

Mechanical milling is a process that requires the optimization of several parameters, such as type of mill, milling speed, time of milling, type and size of milling medium, ball to powder ratio (BPR), atmosphere and temperature of milling. These parameters are not completely independent of each other and their use results from the nature of the grinding process. The weight of the milling balls to the weight of the powder (BPR - Ball to Powder Ratio) has a significant effect on energy, performance and milling time. The higher this ratio, the shorter the milling time required to achieve the desired state. The mechanical milling could be possibly made in different types of mills (planetary ball mills, vibratory mills, mixer mills, rotor mills, knife mills, etc.), but for our experiments we choose planetary ball mill PM100 from Retsch (Figure 1), because of its powerful and quick milling with speed control, which makes possible to make a reproducible result for each sample.

Figure 1 Planetary ball mill Retsch PM100 (in Institute of Physics, Faculty of Science, P. J. Šafárik University in Košice)

We have prepared three types of samples of SMCs of pure iron with different BPR (Table 1).

Table 1 The conditions of mechanical milling for powdered SMCs of Fe

The name of sample	The time of milling (min.)	The rate of milling (rpm – rate per minute)	BPR
S1	120	500	3:1
S2	120	500	6:1
S3	120	500	9:1

After the preparation of powdered samples (Figure 2, a) by mechanical milling in planetary ball mill (Figure 1) we used a vibrating mill (Figure 2, b) to sieve a powder fraction and to select powder with fraction smaller than 400 µm. So we prepared powder samples with required mechanical properties (Figure 2, c).

Figure 2 a) powder after mechanical milling at millng vessel, b) vibrating mill FRITSCH with sieves , c) powder after sieving (in Institute of Physics, Faculty of Science, P. J. Šafárik University in Košice)

Next step in preparation of those samples was preparing a layer of insulation on the powder surface to prevent energy losses by minimising the eddy currents. Exist different type of insulation which could be organic, inorganic or organic-inorganic coatings and the process of coating can be made by one of the following methods “wet” or “dry”.

The dry method based on the principle of attaching smaller pieces of lubricant on ferromagnetic powder, which make it eco-friendlier method, but with less homogeny isolation of powder particles. In case of wet method arise more consistence isolation layer, because the liquid phase is used as substance for disperse organic or inorganic chemical.

We decided to choose Stöber method for insulation iron powder. The liquid phase was made from isopropyl alcohol (320 ml), distilled water (64 ml), tetraethyl orthosilicate (TEOS, 98 %, 32 ml) and ammonia (8 ml) for coating of 10 g of iron powder. The stirring was continued for 16 hours divided for two sessions by 8 hours each using a MICROSTAR 7.5 control propeller mixer from IKA (Figure 3).

One of the methods how to prepare material in bulk is to compact the powder. Powder material prepared by mechanical milling is subsequently compacted by uniaxial or multiaxial pressure hot or cold. After pouring the powder of a given weight into the compression die and closing the compression chamber, the evacuation is started by means of a rotary and turbomolecular pump. When the desired vacuum is reached, induction heating of the powder to the desired temperature begins. At this temperature (pressing temperature) the required uniaxial or multiaxial pressure (in MPa) is applied. After the set application time of the compaction, the compacting is completed and the bulk samples have the same diameter as the die in which they were compacted.

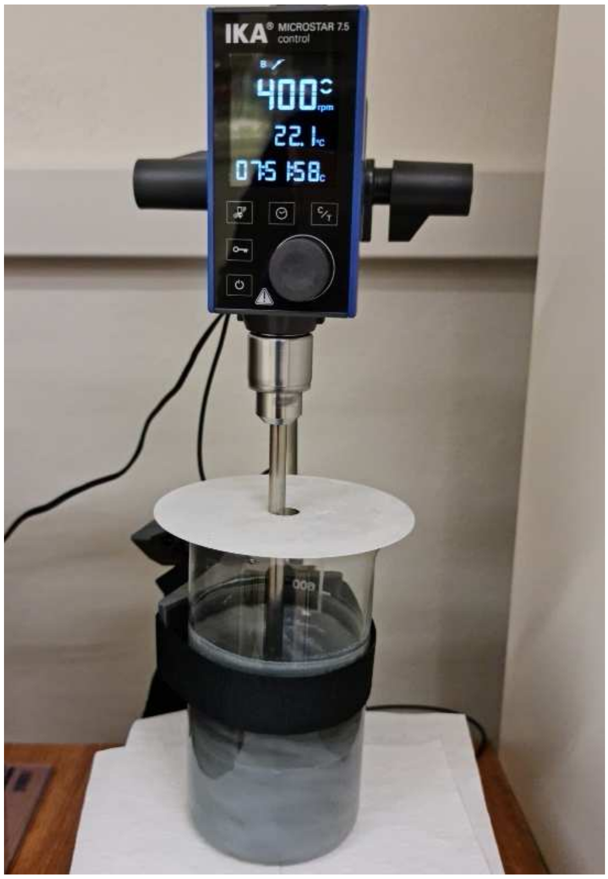

Figure 3 MICROSTAR 7.5 control propeller mixer from IKA (in Institute of Physics, Faculty of Science, P. J. Šafárik University in Košice)

After powder insulation, our samples were ready to compact into a ring shape of soft magnetic composite materials. For the compaction was used the LTVAT ZT-

25-20Y (Figure 4) pressing apparatus, which allows the compaction at temperatures up to 1000 °C.

Figure 4 LTVAT ZT-25-20Y machine for the compaction (in Institute of Physics, Faculty of Science, P. J. Šafárik University in Košice)

In our case, samples were compacted for 3 minutes at 400 °C up to 700 MPa. Thus prepared powder (Figure 2 a) and compacted SMCs iron samples (Figure 5) were used for the determination of the coercivity.

Figure 5 Compacted ring-shaped SMCs iron samples

5 Experimental part

The ring-shaped samples (S1 – S3) of SMCs Fe/SiO2, with outer diameter 24 mm and inner diameter 18 mm, were produced for analysis of influence 16 h insulation process and BPR for high-purity iron SMCs on its coercivity.

The most technical applications of magnetic materials based on revealing spontaneous magnetization. It could by possible because of magnetic hysteresis (showing changes in magnetization and energy losses in one premagnetization cycle) for which, one of the most important physical value is coercivity. In fact, it is one of the most relevant magnetic property for ferromagnetic materials in general and of SMCs materials specifically. Both hysteresis loop and coercivity are highly related to structural defects (interstitials, vacancies, dislocations, surface defects etc.), because they are the main source of the pinning centres that prevent the movement of domain walls.

In our case, we measured coercivity field strength value for our samples by using Foerster KOERZIMAT 1.097 HCJ (Figure 6) at the room temperature. We choose that measuring system because it can be applied to material in deferent shape and also to both hard and soft magnetic materials.

Results of coercivity evolution of Fe/SiO2 powders and compacted samples with increasing BPR from 3 to 9 are shown in Figure 7 and Table 2. These graph represent correlation between intensity of iron granules milling (BPR) and the compaction at high pressure to ring-shaped samples with coercivity value. This mean that after pressing we could not just have made a constant shape of SMCs material, but also removed some of inner structural defects.

Figure 6 Foerster KOERZIMAT 1.097 HCJ (in Institute of Physics, Faculty of Science, P. J. Šafárik University in Košice)

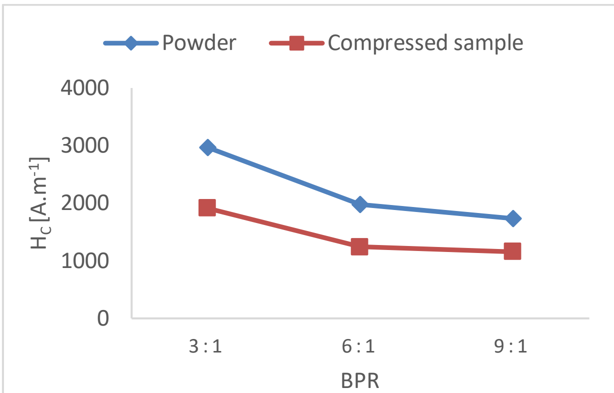

Figure 7 The values of the coercivity of powder and compacted SMCs of Fe

Table 2 The values of the coercivity of powder and compacted SMCs of Fe

Sample	Coercivity, H_c [$A.m^{-1}$]		BPR	Mass of the compressed sample, [g]
	Powder	Compacted Sample		
S1	2968	1911	3:1	3,694
S2	1979	1236	6:1	3,704
S3	1730	1156	9:1	3,794

6 Conclusions

The three compacted SMCs of Fe were prepared with the different condition of mechanical milling. Mechanical milling and subsequently of the compaction of milled powder are an alternative ways of the preparing solid materials of various shapes and sizes. In order to achieve the required physical and magnetic properties, it is necessary to study magnetization processes, select the appropriate chemical composition of the material, determine the correct procedure for preparing powder materials suitable for compaction, select the appropriate type of alloy as a milling precursor (for example BPR), know the morphology of particles and compaction properties.

The value of the coercivity decreases with the increasing of ball to powder ratio. The monotonous decrease of the coercivity with BPR for compacts can be explained as follows: the higher BPR leads to the smaller mean size of the powder particles, that are less deformed at compaction, what leads to the lower probability for creation structural defects for domain wall displacement.

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