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Investigation of the influence of mechanical milling on magnetic properties of Fe powders

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Abstract: Nowadays, there is a global search for electromagnetic gadgets that are affordable, eco-friendly, and energy-efficient. This motivates engineers and scientists to develop new materials or enhance those that already exist. Fe-based soft magnetic materials are a significant class of soft magnetic materials that are essential to many energy-related industrial applications, including motors, converters, and electric transformers. This article focuses on the general characterization of magnetic materials, their magnetic properties and the analysis of the influence of mechanical milling of Fe powders prepared by mechanical milling in two different sizes (sample 1 – smaller then < 400 μm and sample 2 greater than > 400 μm). The experimentally obtained hysterical curves of ground Fe powders are measured using the Vibratimg Sample Magnetometer (VSM).

1 Introduction

Nowadays, we observe worldwide search for electromagnetic devices that are energy efficient, environmentally friendly and economically viable. This forces scientists and engineers to improve existing materials or discover entirely new materials [1].

A new generation of energy materials is needed in order to pursue higher efficiency in energy transformation and conversion [2]. Soft magnetic materials are an essential component of human civilization and have been used in many important technical applications, including the production and conversion of electricity [3].

Soft magnetic materials can change their magnetic polarization quickly in response to mild magnetic fields. They are mostly utilized to increase or channel the flux created by an electric current and usually have low inherent coercivity [2-4].

Transformers, converters, inductors, motors, generators, and even sensors are just a few of many electromagnetic distribution, conversion, and generating devices that employ these magnetic materials [5]. The expedited development and desing of novel energy materials has drawn a lot of interest in the materials science community nowadays because of the numerous societal and environmental issues we are currently facing [2,6].

In this study, we used mechanical milling to produce microcrystalline elemental iron powders. Iron powder with purity of 99.98% and particle size less than 400 μm (sample 1) and particle size greater than 400 μm (sample 2) were prepared and used for the experiment. We used mechanical milling in our study to create microcrystalline elemental iron particles. Specifically, sample 1 (particle size less than 400 μm) and sample 2 (particle size higher than 400 μm) of 99.98% pure iron powder were used in the experiment. The purpose of this work is to present a broad review of magnetic materials with an emphasis on magnetic soft materials, and to compare the magnetic characteristics of the obtained Fe powders using primary magnetization and hysteresis loop based on their size.

2 Magnetic materials

Magnetic materials are divided into diamagnetic, paramagnetic, ferromagnetic, ferrimagnetic and antiferromagnetic on the basis of their internal structure (arrangement of electrons in atoms). Ferromagnetics are among the most widely used materials in the manufacture of various technologies because of their atypical properties such as magnetic hysteresis (hysteresis loop) [7]. The shape of the hysteresis loop indicates the magnetic substance in question. Magnetic hysteresis is the dependence of the induction $\vec{B}$ of the magnetic material from the magnetic field strength $\vec{H}$ and expresses a non-linear relationship between $\vec{B}$ and $\vec{H}$, also that induction $\vec{B}$ lags behind the magnetic field strength $\vec{H}$ [8].

Based on the hysteresis loop, with the help of which we find the dependence of the magnetic induction $\vec{B}$ on the magnetic field strength $\vec{H}$, two categories of these ferromagnetic substances are distinguished, namely soft and hard magnetic ferromagnetics [9-10].

- Soft ferromagnetic materials are usually iron-based and are obtained by pressing a soft magnetic powder with a dielectric (electroinsulating) binder (matrix). These are materials that are easily magnetized and demagnetized, making them useful when it is necessary to switch the magnetic field quickly and easily, for example, in motors [10].
- Hard (permanent) ferromagnetic materials, most commonly Nd-Fe-B, are formed by bonding hard magnetic powder to a dielectric (electrical insulating) binder. These are materials that retain their magnetisation. If these permanent magnetic materials were used in a situation where it is necessary to switch the magnetic field rapidly, they would consume a large amount of electric current [10-11].

2.1 Hysteresis loop

The hysteresis loop expresses the dependence of the magnetic induction $\vec{B}$ on the magnetic field strength $\vec{H}$. The shape of the loop characterizes the type of material. A wide hysteresis loop characterises a magnetically hard material where a high field strength is required for remagnetisation. Conversely, a narrow loop is characteristic of magnetically soft materials (Figure 1) [8-9]. In Figure 1, the magnetic moment M is a measure of the material's magnetization. It displays the amount of magnetization that a material exhibits when exposed to a magnetic field. H_k represent coercive force and H_0 frequently denotes the material's exposure to an external magnetic field.

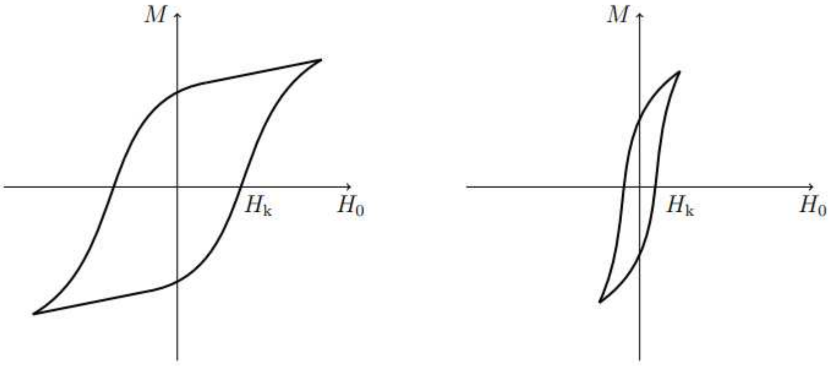

Figure 1 Hysteresis loop of magnetically hard and soft materials [8]

Figure 2 shows the hysteresis loop even with the initial magnetization. The hysteresis loop is labeled by No. 2 and in red, the initial magnetization curve is labeled by No. 1.

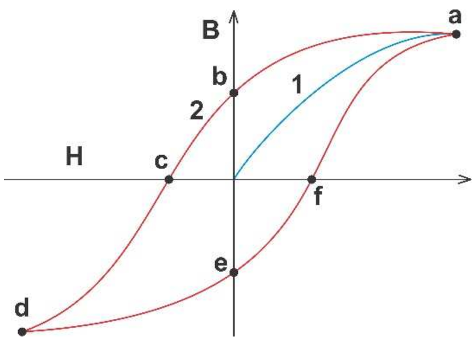

Figure 2 Hysteresis loop

The course of the magnetization curve is described by the letters *a to f*. After the initial magnetization, the *material* is saturated at point *a* . As the magnetic field strength decreases, the magnetic induction $\vec{B}$ also decreases up to point *b*. At this point, there is no magnetic field acting on the material, but the magnetic induction is not zero - that is, the material has remained partially magnetized. Such an induction is called remanent. As the strength of the magnetic field increases to negative values, the curve passes through point c. At this point, a magnetic field is applied to the material, but the magnetic induction in the material is zero - but the material still has some polarisation $\vec{I}$. The magnetic field strength required to remove the magnetic induction (magnetic field of opposite direction) in a material is called the coercivity H_c. Consequently, as the intensity increases to negative values, at point *d* the magnetic domains in the material are magnetized in the opposite direction to point *a*. Further, the magnetic field strength is varied from negative to positive values. Point *e* on the curve corresponds to point *b* and point *f* corresponds to point *c*. The initial magnetization curve (no. 1) is obtained from a completely demagnetized material which is placed in a gradually increasing magnetic field. The change in the magnetic field strength must be in one direction only to avoid the curve being depreciated [9-11].

2.2 Coercivity

Coercivity and remanence are plotted in Figure 2. Coercivity H_c is indicated on the loop by point *c*. This property indicates how much magnetic field strength is required to achieve zero magnetic induction $\overrightarrow{B}$ in a material that has already been magnetized. The remanence (remanent magnetization of M_r) B_r is indicated on the curve by point *b*. This property indicates how much magnetic induction remains in the material after the external magnetic field $\overrightarrow{H}$ is removed. In the case of the maximum area hysteresis loop, its ends correspond to the saturation state of the ferromagnetic substance. The values of coercivity H_c and remanence B_r are characteristic of magnetically soft and magnetically hard materials, with $H_c > 1000\ \mathrm{A}.m^{-1}$ for hard materials and $H_c < 1000\ \mathrm{A}.m^{-1}$ for soft materials [12].

3 Methodology

Ball milling is a widely used technique in the field of materials science and engineering to synthesize various advanced materials [13-15]. The process involves mechanical activation of powders through repeated collision of grinding balls with powder particles, thereby reducing the particle size and creating new interfaces. While the ball milling process is relatively simple, there are several critical processing parameters that can significantly affect the quality and properties of the final product [16]. These parameters include mill type, milling time and speed, type and viscosity of milling medium, milling temperature, Ball to Powder Ratio (BPR) and atmosphere. In our experiment, we used the PM100 planetary ball mill from Retsch (Figure 3) because it has a powerful speed-controlled grinding action, which allows for a reproducible result [17-18].

Figure 3 Planetary ball mill Retsch PM100 (in Institute of Physics, Faculty of Science, P. J. Šafárik University in Košice)

3.1 Experimental materials

To study and compare the magnetic properties, we prepared two samples (sample 1 and sample 2) of pure iron powders. One sample was prepared from pure iron granules of 1-2mm size and high purity (99.98% purity, Alfa-Aesar Co.), which was then processed by mechanical grinding in a ball mill. For the second sample we used as starting material a reduced Fe powder of high purity (99.9% purity, Centralchem), which, unlike the first sample, was passed through a reamer without processing in a mechanical mill.

We prepared two samples of pure iron powder in order to investigate and compare the magnetic properties. One sample, made of high-purity (99.98% pure, Alfa-Aesar Co.) pure iron granules with a size of 1-2 mm, was mechanically ground in a ball mill. Unlike the first sample, which was processed in a mechanical mill, the second sample was prepared without the need for a mechanical mill by passing through a reamer and using a reduced Fe powder of high purity (99.9% purity, Centralchem) as the starting material.

3.2 Preparation of Fe powders

The first sample that we weighed and prepared was iron granules of 1-2 mm in size, which were then dumped into the lubricator along with the pre-weighed steel balls. The chosen weight ratio of the beads was in the ratio of 9:1 (BPR). High-energy ball milling of iron pellets and steel balls was carried out at room temperature using a planetary ball mill which combines high friction with impact at a speed of 500 rpm. A stainless steel grinder and hardened steel balls were used. The weight of the balls was 178.28 g, the weight of the iron pellets was 19.81 g and the total weight of pellets, balls and grinding mill was 4227.4 g. The grinding process itself lasted 2 hours with 10 second breaks due to material and mill temperature stabilization and with reverse rotation in each cycle for 70 seconds for optimum mixing. One of the disadvantages of ball milling is that long grinding times are often required to achieve the desired particle size and distribution, resulting in extended processing times. In addition, friction and collisions between the balls and the material during grinding can generate a significant amount of heat which can affect the stability and properties of the material being processed, especially for heat-sensitive materials. Therefore, the selection of parameters in the ball milling process is a key factor in determining the final product properties. After the milling process was completed, we re-weighed separately the balls (179.56g) and the resulting iron powder (17.07g). We then transferred this powder into a reamer and allowed it to digest for 3 minutes.

The sample 2 was prepared from atomized iron powder with size below 400 µm whose weighed weight was 10.038g. This powder was poured into a reamer and allowed to strain for 5 minutes. Finally, we sieved the strained iron powder into a glass slide and weighed its weight (9.96g). The addition process is shown in Figure 4.

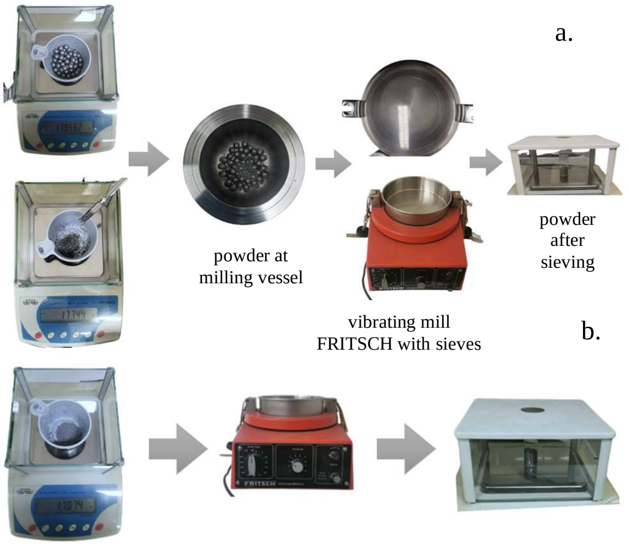

Figure 4 Process for the preparation of a. Fe granules 1-2 mm in size and b. Fe reduced powder

4 Experimental part

Magnetic properties of the sample 1 and the sample 2 were measured using a Vibrating Sample Magnetometer (VSM) on a DYNACOOL device of Quantum design in the temperature range of 200 to 390 K and in the dc applied magnetic field range of -2.39 to 2.39 MA/m. We carried out our measurements in this temperature range to approximate real situations [19].

We exposed two iron powder samples (Fe reduced powder with particles less than 400 µm - sample 1 and non – milled Fe powder with particles more than 400 µm - sample 2) to temperatures of 200 K, 300 K, and 390 K in order to study and compare the magnetic properties (primary magnetization and hysteresis loop).

Figure 5 for the sample 1 and Figure 6 for the sample 2 illustrates the experimentally measured dependence of magnetic moment on magnetic field from -2,39 MA/m to 2,39 MA/m for various temperatures (200 K, 300 K, and 390 K).

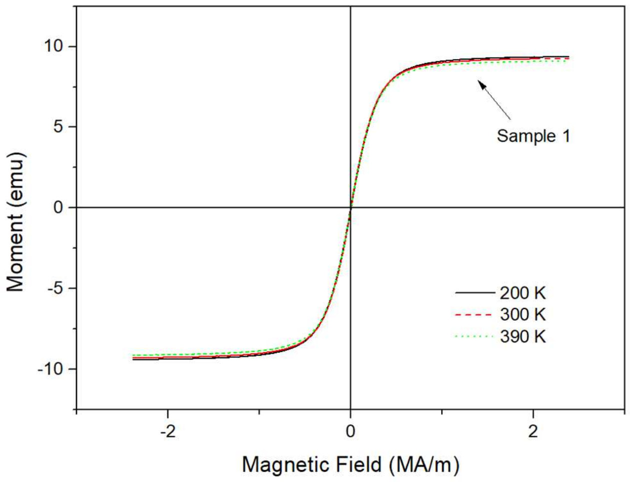

Figure 5 Effect of temperature on the magnetic hysteresis loop of the sample 1

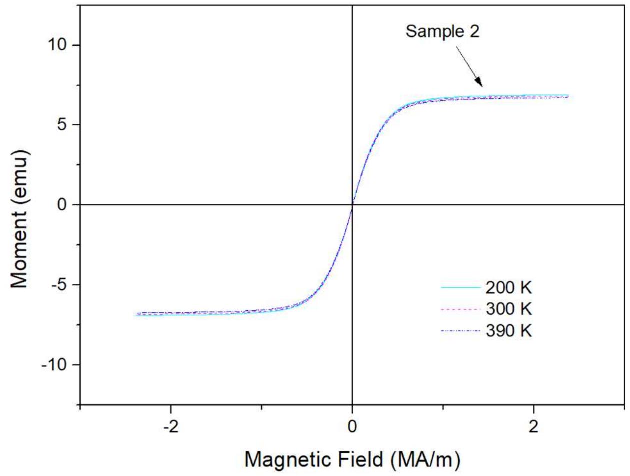

Figure 6 Effect of temperature on the magnetic hysteresis loop of the sample 2

The sample 1 (Figure 5) with a particle size less than 400 µm has larger values of the magnetic moment at the selected temperatures than the sample 2 with a particle size more than 400 µm (Figure 6), which demonstrating the clear effect of temperature on the shape of the magnetization curves. It is reasonable to assume that the mechanical milling process raises the magnetization value. Defects introduced into the sample during the milling process may be the cause. The sample 1 has higher value of coercivity. The magnetic moment decreases as the temperature increases for both samples. A clear tendency towards saturation with an applied magnetic field for sample 1 and sample 2 is around 0.9 MA/m for each temperature [20].

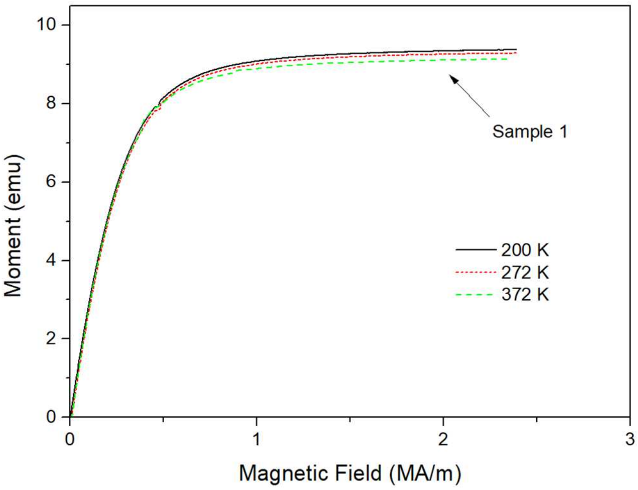

Figure 7 Initial magnetization curve of the sample 1 at different temperatures

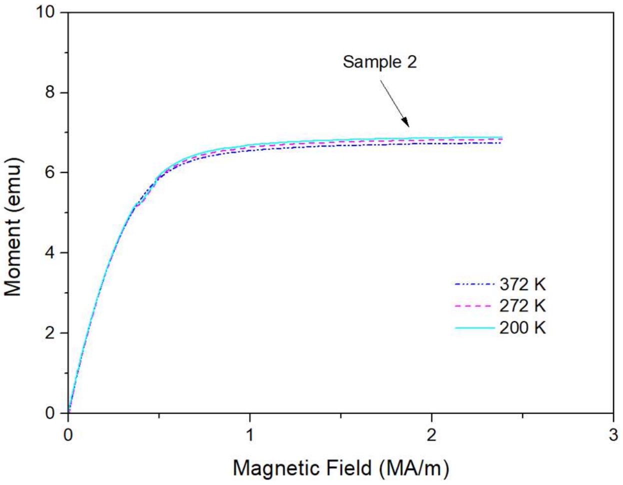

Figure 8 Initial magnetization curve of the sample 2 at different temperatures

The magnetic moment dependency on magnetic field at various temperatures for the sample 1 is shown in Figure 7 and for the sample 2 is shown in Figure 8. The initial magnetization curves (Figure 7 and Figure 8) exhibit a similar pattern to the hysteresis loops (Figure 5 and Figure 6) for both samples 1 and sample 2. It shows that the sample 1, which has smaller particles, exhibits greater magnetization values with rising magnetic field. Consequently, the sample 1 achieves higher magnetization values at lower magnetic field than the sample 2, which has particle sizes larger than 400 µm.

It is reasonable to infer that the mechanical milling process, which produces powdered samples with varying particle sizes, modifies the magnetic properties. Therefore, one method to manufacture a magnetic material with the necessary qualities that is acceptable for further usage (compaction, preparation of magnetic composites, etc.) is through the process of powder metallurgy and mechanical milling.

5 Conclusions

In this work, we investigated how the magnetic properties of iron powders are affected by their particle size. Using a VSM system that measures the magnetic properties of substances, we obtained hysteresis curves and initial magnetization curves for both samples which show how particle size affects the magnetic properties of Fe powders. Particle size variations may not be the only factors affecting magnetic characteristics but also the morphology, purity and possible impurities in the iron powders can all have an impact. We also identified challenges associated with the milling process, such as long processing times and possible thermal effects that may affect the stability and properties of the material.

The results show a connection between magnetic behavior and particle size. In comparison to the sample containing larger particles (more than 400 µm), the sample containing smaller particles (less than 400 µm) showed higher magnetization values and stronger coercivity. This implies that the process of mechanical milling improves the magnetization, because it introduces flaws and produces smaller particles that have a higher surface area and a different domain structure.

We obtained significant knowledge of the magnetic characteristics and behavior of two pure Fe powders of varying sizes under the influence of external magnetic fields at various temperatures by comparing their hysteresis curves and initial magnetization curves. In order to enhance magnetically soft materials, the acquired results can be integrated with the findings from earlier research that examine the compactness of magnetically soft Fe materials with various mechanical milling settings.

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References

[1] SUNDAY, K., TAHERI, M.: Soft magnetic composites: recent advancements in the technology, *Metal Powder Report*, Vol. 72, No. 6, pp. 425-429, 2017. https://doi.org/10.1016/j.mprp.2016.08.003

[2] WANG, Y., TIAN, Y., KIRK, T., LARIS, O., ROSS, J. H., NOEBE, R.D., KEYLIN, V., ARRÓYAVE, R.: Accelerated design of Fe-based soft magnetic materials using machine learning and stochastic optimization, *Acta Materialia*, Vol. 194, pp. 144-155, 2020. https://doi.org/10.1016/j.actamat.2020.05.006

[3] LIU, T., LI, F., WANG, A., XIE, L., HE, Q., LUAN, J., HE, A., WANG, X., LIU C.T., YANG, Y.: High performance Fe-based nanocrystalline alloys with excellent thermal stability, *Journal of Alloys and Compounds*, Vol. 776, pp. 606-613, 2019. https://doi.org/10.1016/j.jallcom.2018.10.319

[4] KOCSIS, B., FEKETE, I., VARGA, L.K.: Metallographic and magnetic analysis of direct laser sintered soft magnetic composites, *Journal of Magnetism and Magnetic Materials*, Vol. 501, pp. 1-4, 2020. https://doi.org/10.1016/j.jmmm.2020.166425

[5] ONDERKO, F., BIRČÁKOVÁ, Z., DOBÁK, S.: Magnetic properties of soft magnetic FeSiO2/ferrite composites prepared by wet/dry method, *Journal of Magnetism and Magnetic Materials*, Vol. 543, p. 168640, 2022. https://doi.org/10.1016/j.jmmm.2021.168640

[6] CHOI, M., KIM, M., LEE. K.: The effects of Fe nano-powders on compaction behaviors and magnetic properties of SMCs, *Journal of Magnetism and Magnetic Materials*, Vol. 480, pp. 33-39, 2019. https://doi.org/10.1016/j.jmmm.2019.02.034

[7] CALLISTER, W., RETHWISCH, D.: *Materials Science and Engineering, AN INTRODUCTION*, 10th ed., Hoboken, Wiley, 2018.

[8] HOSFORD, W.F.: *Elementary Materials Science*, Ohio, ASM International, 2013.

[9] KOLENKO, P.: *Úvod do fyziky pevných látek*, Praha, Fakulta jaderná a fyzikálně inženýrská ČVUT, 2023. (Original in Czech)

[10] KIM, Y., ZHAO, X.: Magnetic Soft Materials and Robots, *American Chemical Society*, Vol. 122, No. 5, pp. 5317-5364, 2022. https://doi.org/10.1021/acs.chemrev.1c00481

[11] COEY, J.M.D.: *Magnetism and Magnetic Materials*, Cambridge, Cambridge University Press, 2009.

[12] NGO, T.D.: *Composite and Nanocomposite Materials - From Knowledge to Industrial Applications*, Rijeka, IntechOpen, 2020.

[13] OLEKŠÁKOVÁ, D., KOLLÁR, P., FÜZER, J.: Structure and Magnetic Properties of Powdered and Compacted FeNi Alloys, *Journal of Electrical Engineering*, Vol. 68, No. 2, pp. 327-330, 2017. https://doi.org/10.1515/jee-2017-0024

[14] KUMAR, M., XIONG, X., WAN, Z.: Ball milling as a mechanochemical technology for fabrication of novel biochar nanomaterials, *Bioresource Technology*, Vol. 312, p. 123613, 2020. https://doi.org/10.1016/j.biortech.2020.123613

[15] OLEKŠÁKOVÁ, D., KOLLÁR, P., FÜZER, J., KUSÝ, M., ROTH., S., POLANSKI, K.: The influence of mechanical milling on structure and soft magnetic properties of NiFe and NiFeMo alloys, *Journal of Magnetism and Magnetic Materials*, Vol. 316, No. 2, pp. 838-841, 2007. https://doi.org/10.1016/j.jmmm.2007.03.111

[16] WEI, L.K., ABD RAHIM, S.Z., AL BAKRI ABDULLAH, M.M.: Producing Metal Powder from Machining Chips Using Ball Milling Process, *Materials*, Vol. 16, No. 13, pp. 1-43, 2023. https://doi.org/10.3390/ma16134635

[17] EL-ESKANDARANY, M.S., AL-HAZZA, A., AL-HAJJI, L.A.: Mechanical Milling: A Superior Nanotechnological Tool for Fabrication of Nanocrystalline and Nanocomposite Materials, *Nanomaterials*, Vol. 11, No. 10, p. 2484, pp. 1-35, 2021. https://doi.org/10.3390%2Fnano11102484

[18] MOTOZUKA, S., TAGAYA, M., HAYASHI, K., MORINAGA, M.: Texture formation in iron particles using mechanical milling with graphite as a milling aid, *American Institute of Physics*, Vol. 5, No. 9, pp. 2158-3226, 2015. https://doi.org/10.1063/1.4930915

[19] REIFFERS, M., VILČEK, J, MICHAELI, E., ČURLÍK, I., KLAMÁR, R., ČECH, V., IVANOVÁ, M., KOCO, Š., PASTERNÁK, T., IĽKOVIČ, S., PARMA, L., ŠEBEŇ, V., ŠTERBÁKOVÁ, K., LITECKÁ, J., CSATÁRYOVÁ, M., ZAPOTOKOVÁ, M., SOLAR, V.: Research of magnetic properties of spolic technosols (alcalic/hyperartefactic) on the landfill of industrial waste from nickel production, *Journal of Electrical Engineering*, Vol. 69, No. 6, pp. 489-492, 2018. https://doi.org/10.2478/jee-2018-0083

[20] DZUBINSKA, A., FERNÁNDEZ, J.R., KUMAR, A., VARGA, R., REIFFERS, M., GIOVANNINI, M.: YOSHIZAWA, Y.: Structural and physical properties of YbCu4Ni based heavy fermion compounds, *Journal of Magnetism and Magnetic Materials*, Vol. 588, No. December, p. 171380, 2023. https://doi.org/10.1016/j.jmmm.2023.171380

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