THE STRUCTURE AND MAGNETIC PROPERTIES OF Fe-Si POWDER PREPARED BY BALL MILLING

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SUMMARY

In this work, fine Fe-Si powders with a nanocrystalline structure were prepared by mechanical alloying (high energy ball milling) of the original microcrystalline sample in the form of ribbon. The magnetization of the sample decreases with milling time due to the decrease of powder size and of the increase of the amount of superparamagnetic phase. Phases in Fe-Si (6,5 wt.% of Si) ball milled for different lengths of time have been investigated by vibrating sample magnetometer (VSM). It was found that two various phases of Fe-Si solid solution with Curie temperatures 749°C and 715°C can be formed by ball milling of Fe-Si (6,5 wt.% of Si) at room temperature. The Curie temperature of the Fe-Si (6.at%. of Si) solid solution is 749°C and the Curie temperature of the Fe-Si (12 at%. of Si) solid solution is 715°C. In the milling procedure the amorphous phase with Curie temperature 229°C were also formed.

***Keywords*:** mechanical alloying, powder, nanocrystalline, coercivity

1. INTRODUCTION

Soft magnetic materials are characterised by low coercivity and high magnetic permeability. Fe-Si alloys are widely used as transformer magnets and magnetic cores because of their excellent soft magnetic properties [1-2]. The introduction of Si into Fe can result in a decrease of magnetic anisotropy therefore leading to decrease of coercivity. The presence of Si can also enhance electrical resistivity and therefore reduce eddy current loss. High-energy milling is a novel technique to produce non-equilibrium and metastable alloys and compounds. Several investigators [3-6] have recently used this method to prepare nanocrystalline Fe-Si alloys. Nanocrystalline Fe-Si alloys prepared by mechanical alloying display structure and magnetic properties that are often not present in either single-crystalline or polycrystalline Fe-Si alloys, such as the large volume fraction of disordered grain boundaries, the extended imperfection and high coercive force, etc..

The structure and magnetic properties of the powder for compaction are determined by its chemical composition and the processing method [7,8]. We assume that non-magnetostrictive alloys (for example Fe-Si with 6.5 wt. % of Si [9]) are very suitable for preparation of bulk samples by high pressure compression, because mechanical stress does not induce magnetic anisotropy in such a ferromagnetic material. Since Si has a smaller atomic radius than that of Fe, the Fe-Si solid solutions have a smaller lattice parameter and reduced unit cell volume in comparison with those of pure Fe-bcc phase [10]. J. Ding et al. found that commercially available $Fe_{83}Si_{17}$ powders have much lower permeability than the Fe-Si powders fabricated by mechanical milling. This result indicates that soft magnetic powders fabricated by mechanical milling may be promising for microwave applications, if their magnetic and other properties can be further optimized.

In this paper the results of the mechanical milling of the Fe-Si system in argon atmosphere are reported.

2. EXPERIMENTAL

We have prepared the Fe-Si (with 6.5 wt.% of silicon) microcrystalline sample by the rapid quenching method in the form of ribbons 5 mm wide and 20-40 μm thick. The sample was milled in a vibratory micromill Pulverisette 0, Fritsch. To prevent the oxidation, the milling was done in an Ar protecting atmosphere. The milling process has been interrupted at various stages of milling to remove a part of milled material for further investigations. Magnetization and the coercivity of the powder at room temperature in magnetic field of 0.6 T were measured by a vibrating sample magnetometer (VSM) as a function of the milling time. The dependences of the magnetic moment on the temperature (thermomagnetic curves) were measured in the constant magnetic field of 300 mT in temperature range 20-800°C. The heating rate was 10 K/min.

3. RESULTS AND DISCUSSION

Our investigations have shown that the magnetic properties and structure of powder markedly depend on the milling time. During mechanical milling welding and fracture are the two major processes.

The magnetization and coercivity also significantly depend on the milling time. As is shown in Fig.1, the coercivity of the sample increases with the increase of the milling time up to

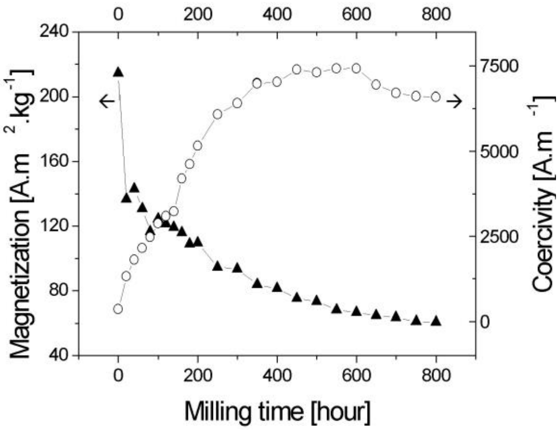

Fig. 1 Magnetization and coercivity of the Fe-Si alloy vs. milling time.

500 hours almost linearly to the value of about 7.5 kA/m. The high coercivity in the as – milled state is obviously due to a high density of internal stresses in the phases with non zero magnetostiction. We know that coercivity values of general FeSi alloys (polycrystal or single crystal in the bulk) are less than 80 A/m [3], while those of the obtained nanocrystalline FeSi alloys are more than 8000 A/m. Evidently, the coercivity values of the latter are two orders of magnitude larger than those of the corresponding polycrystal and single crystal. The increase of the coercivity at the beginning of milling is connected with the decrease of the powder size [11]. The magnetization process is realised more and more by the rotation of magnetization vectors (the domain wall motion becomes less significant). Then the coercivity of single domain particles (with coercivity $H_C=2K_1/I_S$) slowly decreases with milling time [12]. We have found that magnetization of the sample monotonously decreases with milling time. We assume that this decrease is caused by the decrease of the powder size and by the increase of the amount of superparamagnetic phase, contributing less to the total magnetization.

Investigating the structure and magnetic properties [11] of Fe-Si alloy, with 6.5 wt. % of silicon, previously, we have found that: milling leads to decrease of the amount of Fe-rich phases (in which Fe-atoms have different numbers of other Fe-atoms as their nearest neighbors). The amount of other phases, i.e. grain boundary phase, oxides and small superparamagnetic particles, increases with the milling time.

Fig.2 shows the thermomagnetic curves of the powder samples after selected stages of the milling process from 180 to 800 hours. The decrease of magnetization with the increase of the milling time becomes less significant as the milling proceeds.

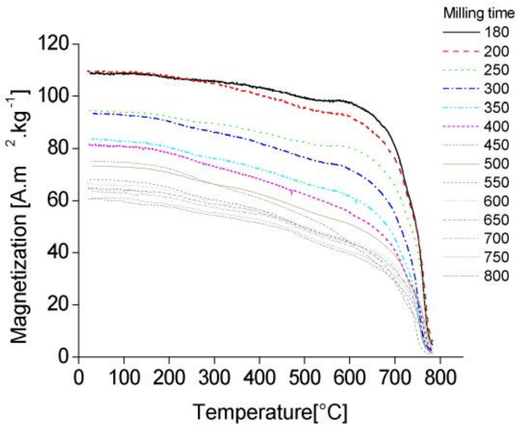

Fig. 2 Thermomagnetic curves for different stages of milling.

The "wavy" shape of the thermomagnetic curves indicates the presence of more phases. Fig. 3 gives an example of thermomagnetic curve.

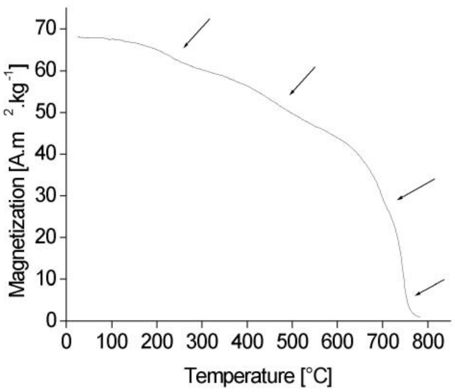

Fig. 3 Thermomagnetic curve of the sample after 550 hours of milling.

It represents the temperature dependence of the magnetic moment for the sample after 550 hours of milling. The arrows show the inflex points of this curve.

We used the Landau theory of phase transition to determine the Curie temperatures from thermomagnetic curve as its inflex points. We have identified four different Curie temperatures: 749^oC, 715^oC, 483^oC and 229^oC. The lowest Curie temperature, 229^oC, corresponds to the amorphous phase. It is known that the Curie temperature of the Fe-Si amorphous phase decreases from 330^oC to 115^oC as a function of Si concentration, x in the composition range of 0.1<x<0.2 [13]. The two highest Curie temperatures can be determined according to the known Si concentration dependence of the Curie temperature for the Fe-Si alloys [14]. The Curie temperature of the Fe-Si solid solution with 6 at. % of Si is 749°C and the Curie temperature, 715°C, corresponds to the Fe-Si solid solution with 12 at.% of Si. The phase with Curie temperature of 483°C is probably Fe_4N nitride. Fig. 4. represents the evolution of the Curie temperatures, T_C of various phases with the milling time. The phase of Fe-Si solid solution with T_C of 715°C cannot be observed after 500 hours of milling. The rest three phases are present in all the milled samples.

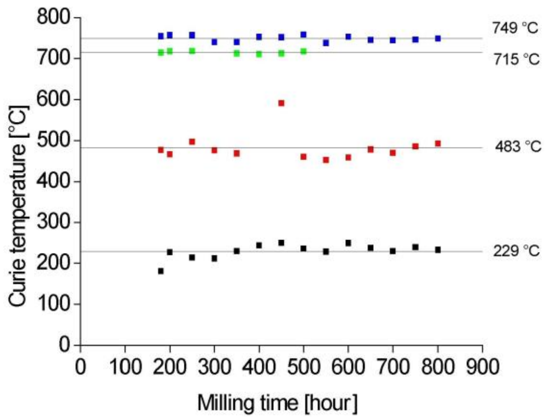

Fig. 4 Curie temperatures of the Fe-Si phases for the samples after different stages of milling.

4. CONCLUSION

The influence of milling on the magnetic properties and structure of the powdered Fe-Si (with 6.5 wt.% of silicon) alloy, with special interest in long-time milling has been investigated. The coercivity of the sample increases with milling time up to 500 hours almost linearly to the value of about 7.5 kA/m. During the last 300 hours, the coercivity slowly decreases. This decrease is connected with the decrease of the powder size and the increase of the amount of superparamagnetic phase. It can be concluded that the powder milled in argon consists of a mixture of Fe-Si solid solutions with various Si content and Fe_4N nitride. The formation of the amorphous phase in the Fe-Si system by ball milling was confirmed.

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5. REFERENCES

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BIOGRAPHY

Ján Füzer was born on 22.01.1971. In 1994 he graduated (MSc.) from the Faculty of Science at P.J.Šafárik University in Košice. He defended his PhD. in the field of physics of condensed matter and acoustics in 2000; his thesis title was "Magnetic properties of the nanocrystalline and amorphous fine grain ferromagnetic materials". Since 1995 he has been working as a researcher with the Department of Experimental Physics. His scientific research is focused on study of the magnetic properties and structure of the ferromagnetic Fe-based materials in the form of the ribbon, powder and thin films. In addition, he also investigates the possibility to prepare bulk nanocrystalline materials with very good soft magnetic properties.