

The Magnetic Properties of Nanocrystalline Fe₅₀Ni₅₀ alloys Prepared by Mechanical Alloying Method

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ABSTRACT

In this study, nanocrystalline Fe₅₀Ni₅₀ alloy powders were synthesized by Mechanical alloying process using planetary high- energy ball mill (Pulverisette 5, Fritsch) for milling times: 2,5,10,30,50,70 h and for the weight ratio of balls to powder (BPR) 30:1, under argon atmosphere. The alloy formation and different physical properties were studied as a function of milling time, using X-ray diffraction (XRD) technique, Field emission scanning electron microscopy (FESEM) Transmission Electron Microscopy (TEM), vibration sample magnetometer (VSM) and Fourier transform infrared (FTIR) spectroscopy. Increase in milling time, led to reduction in crystallite size (D) in the super paramagnetic phase, thus inducing a higher magnetization to the about 120 emu/g, which is significantly higher compared with the work of others. Also reduction in crystallite size led to lower coercivity. Optical studies showed that determined grain size based on hysteresis curve for 70h of milling time is in the same order of radiation wavelength.

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INTRODUCTION

Magnetic alloys are the key to the future with high power electronics industry. The recent development of transformers, chokes, inductors, filters and the emergence of completely new technologies call for entirely new types of magnetic alloys prepared by mechanical alloying (MA) methods. The suitability of a metal or oxide to be used as a magnetic material is determined by its mean atomic moment ($\bar{\mu}_{at}$). For metals the Bethe –Slater – Pauling curves, Fig.1, indicate how $\bar{\mu}_{at}$ depends on the average number (n) of 3d and 4s electrons per atom, and on the crystal structure, i.e., face- centered-cubic (FCC) or body-centered cubic (BCC) structure.

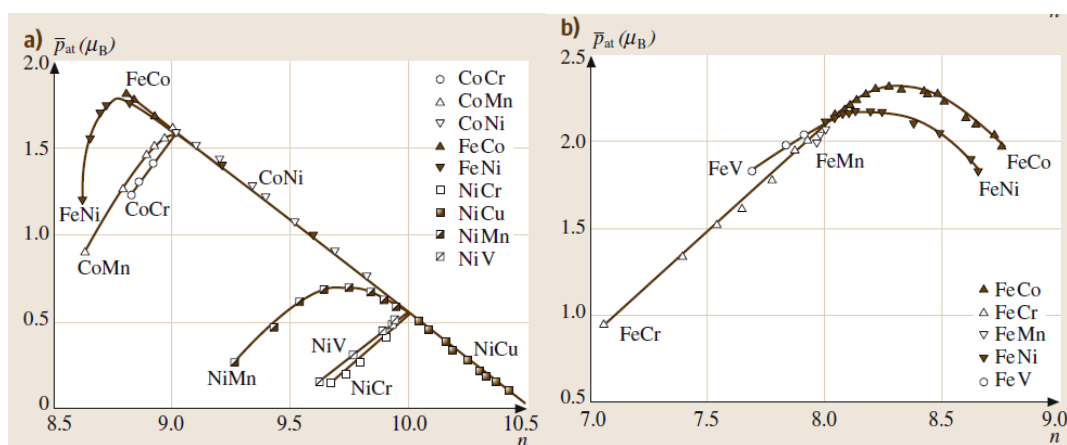


Fig. 1: a,b Bethe- Slater-Pauling relation indicating the dependence of the mean magnetic moment per atom $\bar{\mu}_{at}$ on the average number n of 3d and 4s electrons per atom for binary alloys with (a) fcc structure and (b) bcc structure [1]

Alloys based on Fe and Ni are most suitable from this point of view, corresponding to their actual use. The fcc phase in the Ni-Fe alloy system provide a wide range of structural and magnetic properties for developing soft magnetic materials with specific characteristics for different applications [2].

Nanocrystalline soft magnetic alloys are a rather recent class of soft magnetic materials with excellent magnetic properties such as low losses, high permeability, high saturation polarization up to 1.3 T, and near-zero magnetostriction. The main fields of application of high permeability Ni-Fe alloys are fault –current circuit breakers, LF and HF transformers, chokes, magnetic shielding, switching and storing cores, high sensitivity relays and for temperature compensation in permanent magnet systems [3].

During the last years the mechanical alloying (MA) technique has been found to be very effective in producing powders with interesting properties. By this means it is possible to synthesize alloys or composite materials with highly dispersed components far away from thermal equilibrium state like amorphous, magnetic or nanocrystalline materials [4-6].

The aim of the present work is first to produce the binary Fe50Ni50 system by high-energy ball alloying technique and evaluation of magnetic properties.

Experimental techniques:

Mechanical alloying as mixture of pure Fe (99.5%) powder and Ni (99.98%) powder was carried out in a commercial Fritsch Pulverisette 5 planetary ball mill. To prevent oxidation phenomena, the mixed powder was sealed in a cylindrical vial with capacity of 125 ml with stainless steel balls ($d=10$ mm) under an argon atmosphere in a glove box. The weight ratio of balls to powder was 30:1 for the 2,5,10,30,50,70 hours milling time and vial rotation speed was 300 rpm. To prevent agglomeration of powder particles, we've used toluene as process control agent (PCA). The affection of medium as PCA, which doesn't participate in any chemical reaction in MA process, has not been specially reported [7].

To avoid excessive heating during milling, each 1 h of milling was followed by a stay during 10 min under the argon atmosphere at room temperature. After different milling times, the process was interrupted and small amounts of milled powders were taken out for analysis.

X-ray diffraction patterns were recorded using D8 advanced model, product of 2002 in Bruker company (with $\text{CuK}\alpha$ radiation; $\lambda=1.54\text{\AA}$). X-ray patterns were analyzed by X'pert Highscore plus program. Scherer method is used for calculating the crystallite size of samples.

Scherer formula:

$$D = \frac{0.9 \lambda}{\beta \cos \theta} \quad (1)$$

Where D is crystallite size, λ is wavelength of used ray, β is peak wide in half of the most intensity and θ is Bragg angle.

Field emission scanning electron microscopy (FESEM) has been used for microstructure and crystallites size observations. For providing these images, very little amount of milled powder poured on carbon glue which is connected to sample vessel so that powder particles would not piled on each other. For preventing electron aggregation inside the sample, gold covering was used by DC sputtering device. Then sample-holder is placed in device and after maintaining proper vacuum, imaging is performed. FESEM device used has Hitachi label, S4160 model, product of Japan in 1996. In order to correctness of crystallite size examined using Transmission Electron Microscopy (TEM). The magnetic measurements were performed, using a vibration sample magnetometer (VSM) at room temperature. The optical measurements were performed, using a FTIR-8400-S product of Shimadzu Company for the absorption region 400 - 4000 cm^{-1} .

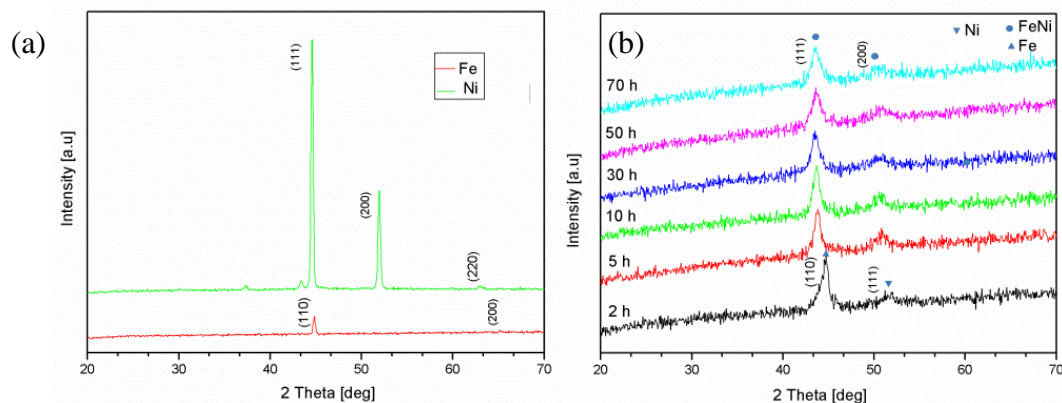


Fig. 2: X-ray diffraction patterns of (a) pure Fe and pure Ni (b) Fe50Ni50 powdered alloys synthesized in high-energy mill for various milling times.

RESULTS AND DISCUSSIONS

X-ray diffraction studies (XRD):

(Fig. 2b) shows the evolution of X-ray diffraction $\text{CuK}\alpha$ patterns as a function of milling time for the Fe50Ni50 powders. XRD (marked 0 h milled) shows reflections corresponding to body-centered-cubic (BCC) Fe (Im-3m , lattice constant $a=2.8530\text{\AA}$) and face-centered-cubic (FCC) Ni (Fm-3m , $a=3.5140\text{\AA}$) metals (Fig.2a). The process continues up to 5 hours of milling time, FeNi nanocrystalline alloy with Fm-3m space groups is formed. One clearly observes that the diffraction Bragg peaks broaden, with increasing milling time. This suggests a continuous decrease of the crystallite size. Reduction of crystallite size resulted in the increase of magnetization and improved the magnetic properties because the crystallite size approaches that of a magnetic domain, offering thus the possibility to eliminate the influence of the domain walls [8, 9].

Fig. 3 shows the variation of Fe-Ni crystallite size versus milling time. As can be seen in Fig. 3, crystallite size decreases with increasing milling time and with a final value (for 70 h) of about 13nm. This value is smaller than the 45 nm [10] and 16.5 nm [11] for the iron–nickel ultra-fine particles obtained by a gas-condensation method, with the same composition.

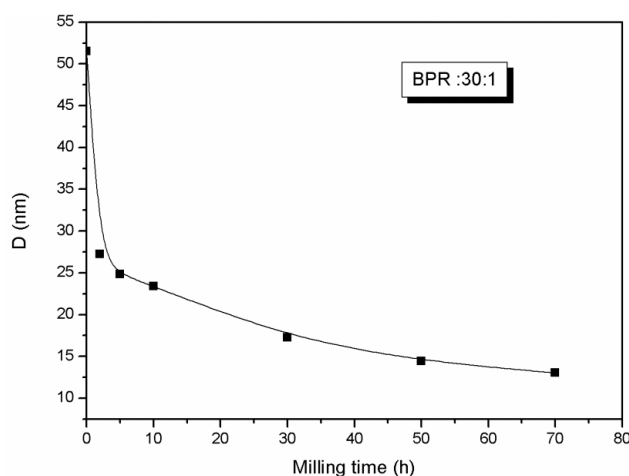


Fig. 3: Crystallite size of the (Fe, Ni) powders as a function of milling time.

Microstructure:

FESEM analysis:

Microstructural aspects of the pure Fe and pure Ni powders before milling are presented in Fig 4 (a and b, respectively). Initially, one can see that unmixed Fe and Ni powders are spherical. The morphologies of Fe–Ni powders after various milling time are shown in Fig. 5.

After 2 h of milling, bonds between nickel atoms and between iron atoms are broken, which grain size of nickel and iron become smaller in comparison with grain size of pure nickel and pure iron. In the same time, formation of FeNi nanocrystalline alloy begins. The process continues up to 5 hours of milling time, FeNi nanocrystalline alloy with Fm-3m space groups and with maximum crystallite size is formed. We can see that with increasing milling time, grain size became smaller, although size distribution is not uniform.

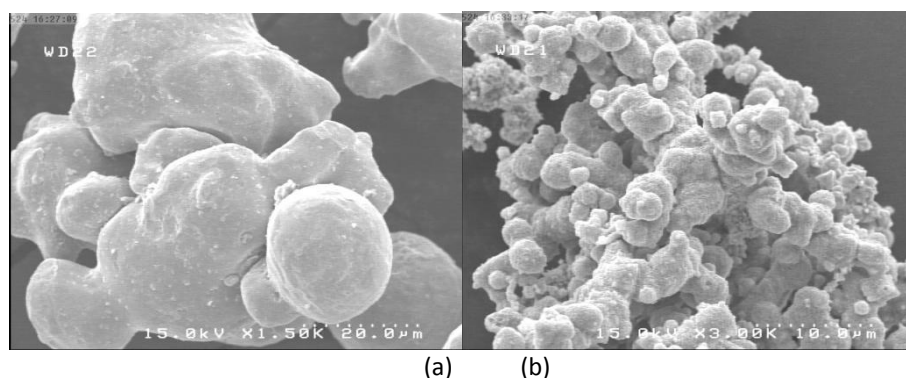


Fig. 4: FESEM micrograph of (Fe, Ni) powders before milling (a) pure Fe, (b) pure Ni.

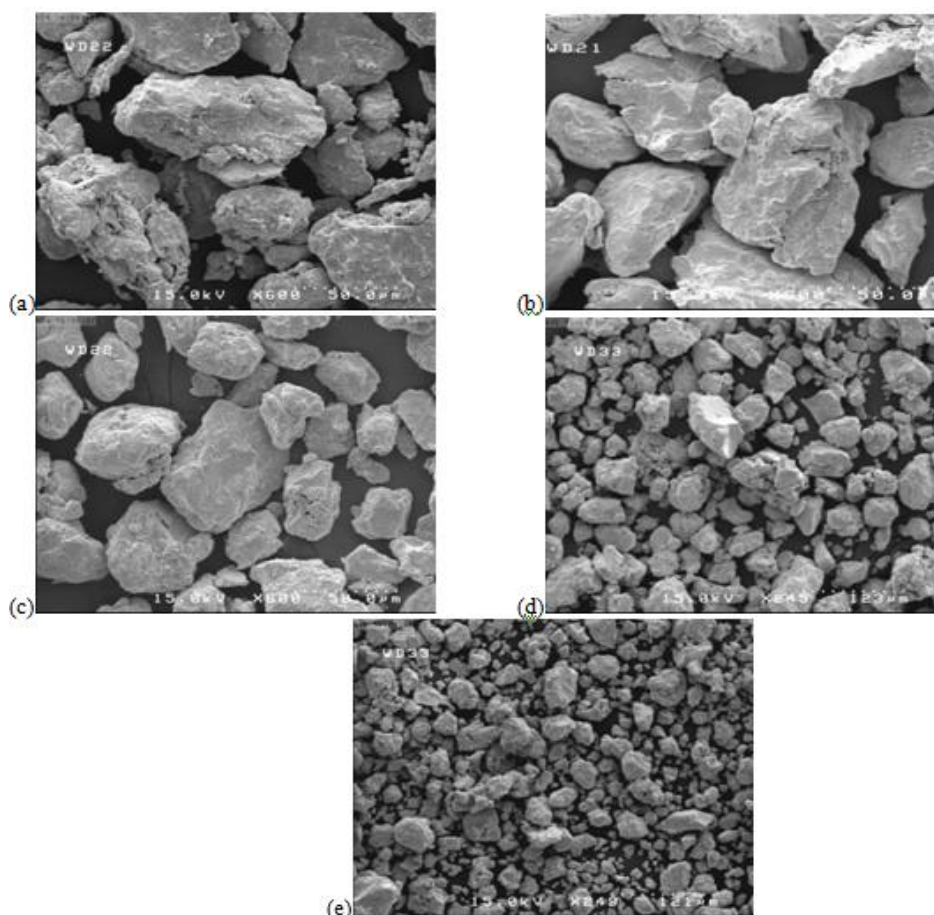


Fig. 5: FE SEM Micrograph of (Fe, Ni) powders prepared by mechanical alloying after different milling times (a) 2 h; (b) 5 h; (c) 10 h; (d) 30 h; (e) 70 h.

TEM analysis:

The TEM image of the nano- FeNi alloy particles after 70 h of milling time are shown in Fig 6. With increasing milling time for 70 h, grain size is decreased and we found that the TEM results as well as agree with the results obtained of X-ray diffraction and FE SEM Micrograph.

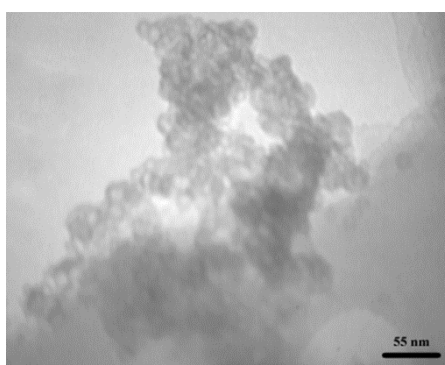


Fig. 6: TEM Micrograph of (Fe, Ni) powders prepared by mechanical alloying after 70 h of milling time

Magnetic properties:

Among all the samples, because the smallest crystallites size was suitable for experimental work, the hysteresis curve for FeNi nanoparticles synthesized after 70 hours ball milling with BPR=30:1, we examined.

Fig 7. Shows hysteresis curve (magnetization vs. applied field) for synthesized FeNi nanoparticles after 70 hours of milling with BPR=30:1.

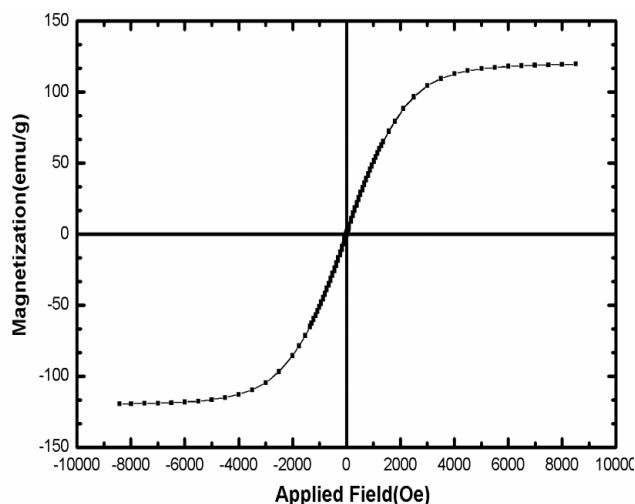


Fig. 7: Magnetization vs the field applied to milled after 70 h of milling time.

Based on this figure saturation magnetization (M_s) in applied field 8514 Oe is about 120 emu/g, which is significantly higher than that of other FeNi alloys in applied field 5000 and 10000 Oe [12]. So this is a very good super paramagnetic material. These materials may have an obvious role in creating temporary high magnetic fields. For example, super paramagnetic nanoparticles may have many potential uses in frofluid, color imaging, magnetic cooling (quenching), detoxification of biological fluids, controlled delivery of anti-cancer medications, MRI and magnetic cell isolation.

Increasing magnetic field in matter will cause alignment of magnetic domains (crystallites) and coupling between grains or magnetic particles. This will lead to surprising increase in interior mean magnetic field. This is a property of super paramagnetic material. After removing magnetic field, temperature causes disorder in domains and magnetic moments will be in random directions consequently weakens coupling between grains and decrease total field inside super paramagnetic. Fig 8. shows a schematic of a magnetic material with super paramagnetic properties, based on physical analysis of this study.

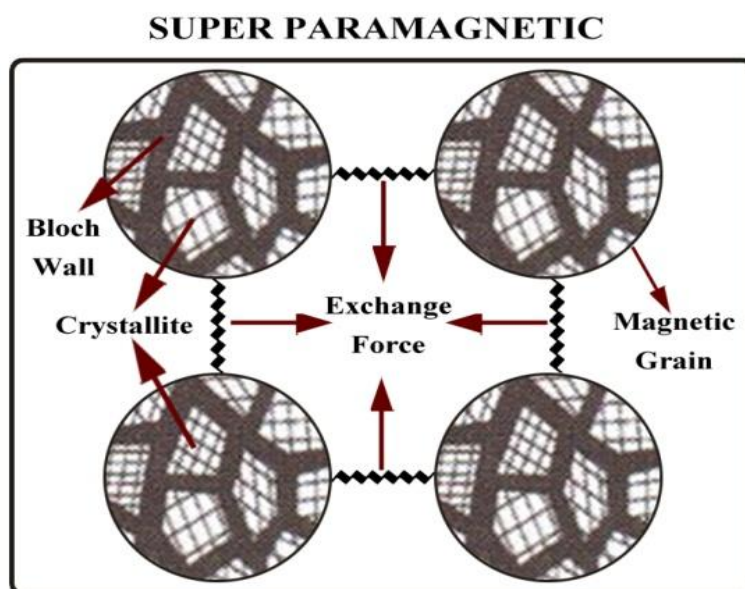


Fig. 8: Schematic of a magnetic material with super paramagnetic properties.

As shown in this figure, crystallites are divided by Bloch walls. In fact Bloch wall is a layer that separates neighbor domains with different magnetic directions.

It is expected that increase in milling time leads to increase saturation magnetization, because it will reduce the size of magnetic particles and total number of Bloch walls will also decrease. Based on [13], saturation magnetization for milling time lower than 70 hours and BPR lower than 30:1 for other FeNi alloys is lower than

saturation magnetization in this study. We can determine magnetic susceptibility in terms of gradient using hysteresis curve (eq. 2).

$$\kappa = \frac{\partial M}{\partial H} \quad (2)$$

Measured hysteresis curve has fitted with different functions, and Boltzmann equation (eq. 3) has most conformity with this curve.

$$y = \frac{A_1 - A_2}{1 + e^{\frac{x - x_0}{dx}}} + A_2 \quad (3)$$

In this equation, y is for magnetization (M) and x is for applied field (H) and constants are as follow: $A_1 = -119.178$, $A_2 = 119.111$, $x_0 = -0.156$ and $dx = 1097.850$.

Finally, Susceptibility value (κ) in terms of applied field is determined as equation (eq.4), in which $C_1 = 0.214$, $C_2 = 0.0009$ and $C_3 = 0.156$.

$$\kappa = c_1 (1 + \exp(c_2(H + c_3)))^{-2} \times \exp(c_2(H + c_3)) \quad (4)$$

To calculate maximum size of magnetic grain, we can assume particles as spheres with radius (r) and use (eq. 5) [14]. In this equation, K is Boltzmann constant ($1.38 \times 10^{-16} \frac{erg}{K}$), T is temperature (300 K), $\frac{dM}{dH}$ is gradient near zero field ($=0.02$ in fig. 7.), ρ is nanoparticles density for FeNi ($=8.02 \frac{gr}{cm^3}$), M_s is magnetic ($=120$ emu/g). Based on these values, particle size is about (3×10^4) nm.

$$d_{max} = \left[\frac{18KT \left(\frac{dM}{dH} \right)_{H=0}}{\pi \rho M_s^2} \right]^{\frac{1}{3}} \quad (5)$$

Estimating magnetic grain size based on (FTIR) spectroscopy:

Fig. 9 shows the room temperature transmittance of synthesized FeNi nano powders for various milling times versus wave number, that was investigated by Fourier transform infrared (FTIR) spectrum.

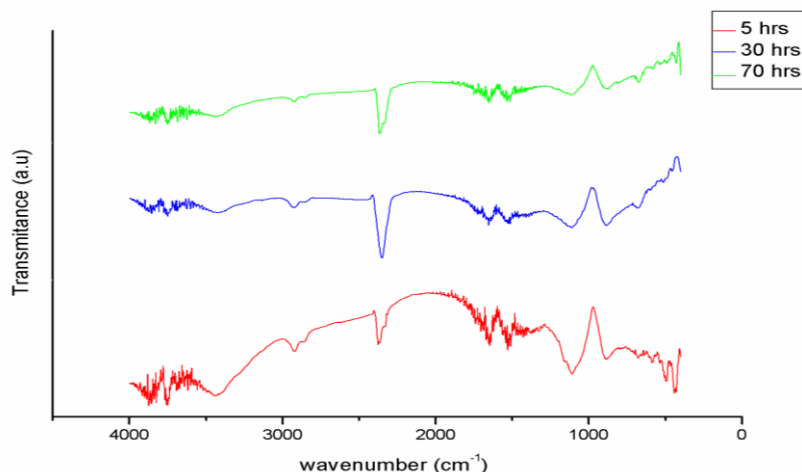


Fig. 9: Transmittance of the synthesized FeNi nano powders for various milling times versus wave number.

Based on Fig. 9 we can see that for wave number of about 2400 cm^{-1} and for 70 hours of milling, wavelength will be about 26.4 μm , which is in good agreement with calculated grain size base on hysteresis curve. As is obvious from this figure, and assuming one valley or one peak for each wave number, it is evident that increasing milling time will cause shifting these valleys and peaks to left (lower wavelength), so it is expected that increasing milling time cause smaller grain size. Thus we can suggest that after 30 hours, magnetic grains size is about 26.5 μm and after 5 hours it is about 26.7 μm . This is also in agreement with X-ray and Electron Microscopy results.

Conclusion:

In this work, we have prepared Fe50Ni50 nanocrystalline alloy by mechanical alloying technique in a high-energy planetary ball mill. A nanocrystalline state (crystallite size about 13 nm) was achieved. The saturation magnetization (M_s) has a increase when the crystallite size decreases which indicates that Fe-Ni system possesses an excellent intrinsic magnetic property. M_s value is about 120 emu/gr for Fe50Ni50 mechanically alloyed powders. In addition, we found that the coercivity force H_c presents a minimum value. Optical studies showed that determined grain size based on hysteresis curve for 70h of milling time is in the same order of radiation wavelength.

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