



Characterization of the Demagnetization Field in Iron-Nickel Nanopowder and Spark Plasma Sintered Samples

T. Ashokkumar¹, A. Rajadurai², A.H. Abishini³, A.H. Anchani³ and Subash C.B. Gopinath^{4,5}

¹Marthandam College of Engineering and Technology College Road, Kuttakuzhi, Veeyannoor Post, (Near Marthandam) Kanyakumari District-629 177, Tamil Nadu, India

²Madras Institute of Technology, Chromepet, Chennai 600044, Tamil Nadu, India

³St. Joseph's College of Engineering, Old Mahabalipuram Road, Chennai-600119, Tamil Nadu, India

⁴Faculty of Chemical Engineering & Technology, Universiti Malaysia Perlis (UniMAP), 02600 Arau, Perlis, Malaysia

⁵Institute of Nano Electronic Engineering, Universiti Malaysia Perlis (UniMAP), 01000 Kangar, Perlis, Malaysia
ntashoknt.1966@gmail.com

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Abstract

This study investigates the impact of alloying elements; spark plasma sintering, grain size, and particle size on the coercive field (H_c) in iron-nickel alloys. The findings reveal that, generally, an increase in nickel content leads to a reduction in H_c after 2 hours of milling. However, in these two-hour milled samples, a notable increase in H_c is observed compared to the initial values of iron and nickel powders. This H_c increase may be attributed to the weakened particle-to-particle interaction between iron and nickel. Changes in H_c are closely linked to variations in grain and particle size. H_c values decrease as the grain size diminishes, ranging from 250 nm to 75 nm. Remarkably, a high H_c value of 103 Oe is observed in the 75 wt.% Ni-Fe alloy after 100 hours of milling, indicating the formation of single-domain grains. A decrease in H_c is noted until a critical particle size is reached; beyond this point, H_c values remain relatively constant until reaching a second critical dimension, after which H_c increases with decreasing particle size. Additionally, the study reveals that the H_c values in spark plasma sintered samples are consistently lower than those in the powder samples.

Keywords: Saturation magnetization; Spark Plasma Sintering; Alloyed power; Coercive Field.

Introduction

Recent studies have employed various methods to investigate and manipulate the properties of iron-nickel (Fe-Ni) alloys. Notably, You et al. utilized a self-designed magnetron sputtering system to coat copper material onto La-Fe_{11.65}Si_{1.35} alloyed powder¹. In contrast, Shamba et al. synthesized LaFe_{11.6}Si_{1.4} alloy in a polycrystalline form through spark plasma sintering and arc melting methods². Research efforts have explored the characteristics of bulk alloys, with particular attention given to Ni-Fe alloys, as seen in the works of Qazi et al.³, Polychronis et al.⁴ and Barakat et al.⁵.

The fabrication of smaller-sized particles has also garnered interest. Techniques such as heat spraying for producing Fe-Ni nanoparticles⁶ and mechanical synthesis of iron and nickel solid solutions using high energy ball milling⁷ have been investigated. Furthermore, Du and Ramanujan⁸ demonstrated mechanical alloying of Fe-Ni based magnetic nanostructures, while Bui et al.⁹ analyzed the nanocrystalline properties of iron powders synthesized through ball milling techniques. Recent work by Ashokkumar and colleagues^{10,11} focused on the saturation magnetization and magnetic retardation analysis of iron-nickel ball-milled nanopowders and sintered samples, respectively, using the spark plasma sintering (SPS) method.

A pivotal aspect of this research pertains to the coercive field (H_c) in Fe-Ni alloys. Yelsukov et al.¹² examined the nanocrystalline state of α -Fe through ball milling, highlighting its impact on magnetic and structural properties, particularly the increase in the coercive field. Sedlacek¹³ posited that in soft magnetic materials¹⁴, H_c may be influenced by lattice defects and residual stresses, especially when domain sizes are comparable to the thickness of the Bloch walls. Conversely, in hard magnetic materials, heterogeneous particles of a size similar to the thickness of the Bloch walls tend to hinder the displacement of these walls. Additionally, in polycrystalline materials, the mobility of Bloch walls is significantly dependent on grain size, which also governs the initial permeability and coercivity. Coarse-grained materials tend to exhibit higher initial permeability and lower H_c compared to fine-grained ones, as an increased initial permeability necessitates a stronger magnetic field for remagnetization.

In this context, Figure-1 illustrates a hysteresis loop showing coercivity, while Figure-2 presents the variation in coercive force (H_c) values for different Fe-Ni alloys. These disparities in H_c values may be attributed to differences in grain sizes within the same alloys.

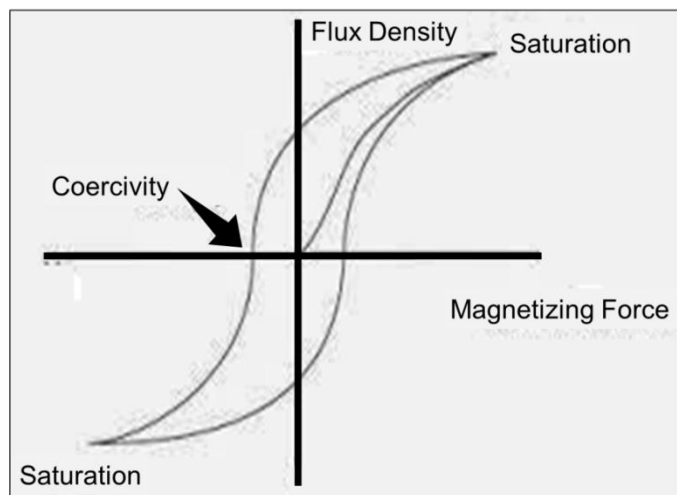


Figure-1: Hysteresis Loop Showing Coercivity.

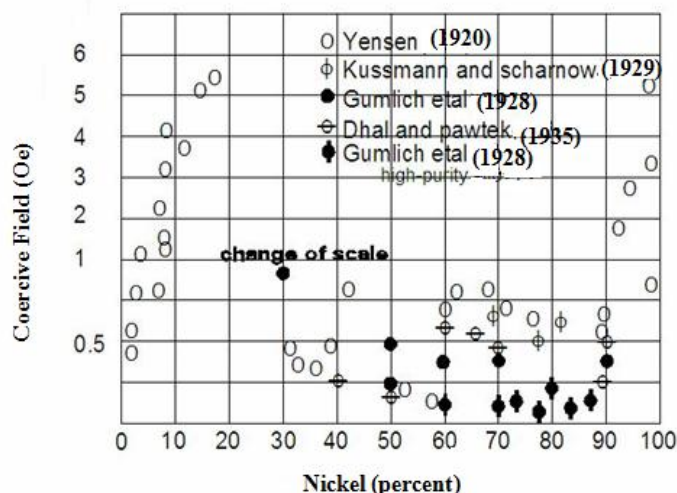


Figure-2: Coercive Force of Iron-Nickel Alloys¹⁸.

Methodology

The purities of the initial nickel and iron materials were confirmed using energy dispersive X-ray (EDX) spectroscopy, which yielded purities of 99.8 wt% for nickel and 99.5 wt% for iron.

The procedures for powder preparation, blending, characterization, morphological observations, compositional analyses, and density determination have been previously detailed in Ashokkumar et al.¹⁴.

Magnetic properties of the starting nickel and iron powders, as well as the powders obtained after different durations of ball milling and the sintered specimens produced using Spark Plasma Sintering (SPS), were examined using a vibrating sample magnetometer (VSM). In the VSM, samples were centrally positioned within a uniform magnetic field and subjected to sinusoidal motion generated by mechanical

vibrations¹⁵. This motion induced changes in magnetic flux, resulting in a potential difference in the pick-up coils, directly proportional to the magnetic moment within the sample¹⁶.

Two thermal cycles, specifically at 923 K and 1223 K, were employed for the SPS process. Both thermal cycles utilized a compaction pressure of 30 MPa and a vacuum pressure of 6 Pa. Subsequently, small sections were excised from the sintered specimens and subjected to ultrasonic cleaning using acetone as the cleaning agent.

Prior to VSM analysis, the samples were accurately weighed using an electronic balance. The weighed powders were then folded in butter paper and placed within the VSM's powder sample holder. The magnetic field required to nullify the magnetization induced in the sample was deduced from the hysteresis loops generated with the assistance of the VSM.

Results and Discussions

Theoretical Density: The theoretical density of powder mixtures, containing 40%, 50%, and 75% Ni-Fe, was determined by considering the densities of Fe and Ni, which are $7.874 \times 10^3 \text{ kg/m}^3$ and $8.908 \times 10^3 \text{ kg/m}^3$, respectively.

SEM Micrographs: SEM micrographs provide valuable insights into the material's microstructure. Figure-3 displays an SEM micrograph of the 75% Ni-Fe sample after two hours of milling, offering a visual representation of the material's morphology at this stage.

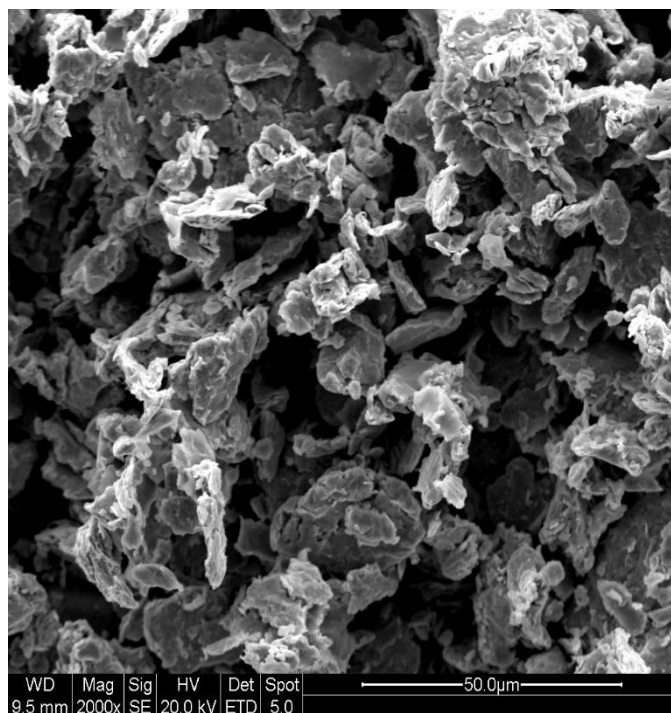


Figure-3: SEM Micrograph 75 wt% Ni-Fe after two hours of Milling.

Magnetic Behavior: Figure-4 presents the magnetic behavior of a 40% Ni-Fe powder sample after 16 hours of milling, as recorded by a vibrating sample magnetometer (VSM). This data is essential for understanding the magnetic properties of the material under various processing conditions.

XRD Diffraction Patterns: In Figure-5, we observe the X-ray diffraction (XRD) peaks of specimens sintered by spark plasma at different milling durations. The reduction in the width of XRD peaks with increased milling hours suggests enhanced grain growth in higher-hour milled spark plasma sintered samples. Moreover, spark plasma-sintered samples exhibit sharper XRD peaks compared to unsintered powders, indicating further grain growth during sintering.

Variation of Grain Size in Spark Plasma Sintered Specimens: Figure-6 depicts the relationship between grain size and milling hours in spark plasma sintered samples. The data shows that an increase in milling hours is associated with a higher rate of grain growth in smaller particle size samples. This effect results from the elevated surface and grain boundary area in higher-hour milled powders, promoting particle agglomeration and rapid grain growth during SPS.

Coercive Field (H_c): In multi-domain ferromagnetic particles, H_c is significantly influenced by grain size and particle size. Typically, H_c decreases as grain size decreases, reaching its maximum value when nanoparticles transition into single domain particles.

Impact of Coercive Field with varying % of Fe-Ni: The coercive field values for the initial Fe and Ni powders were determined to be 24 Oe and 22 Oe, respectively, using VSM. Intriguingly, a two-hour milled sample exhibits a substantial increase in H_c compared to the initial values of iron and nickel (24 and 22 Oe). Specifically, H_c values are measured at 47 Oe for 40% Ni-Fe, 64 Oe for 50% Ni-Fe, and 85 Oe for 75% Ni-Fe. Moustafa¹⁷ attributes this increase in H_c to the weak particle-to-particle interaction between iron and nickel. However, for samples with higher nickel content, the experimental results reveal a reduction in H_c within the range of 8 to 64 hours of milling. The H_c values for 8, 16, 32, and 64 hours of ball milling for 40-75% Ni-Fe are 32, 17, 29, 17, 35, 22 Oe, and 41, 23 Oe, respectively. These results indicate lower coercive fields for higher nickel content alloys, consistent with Marsh's¹⁸ findings in 1938 and Qin's¹⁹ observations in 1999.

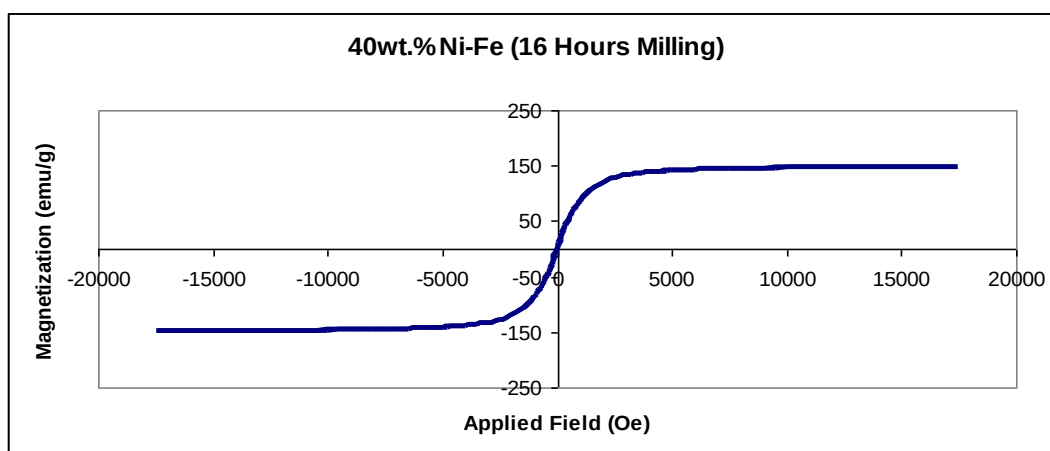


Figure-4: Magnetic Behavior of 40wt. % Ni-Fe Powder Sample.

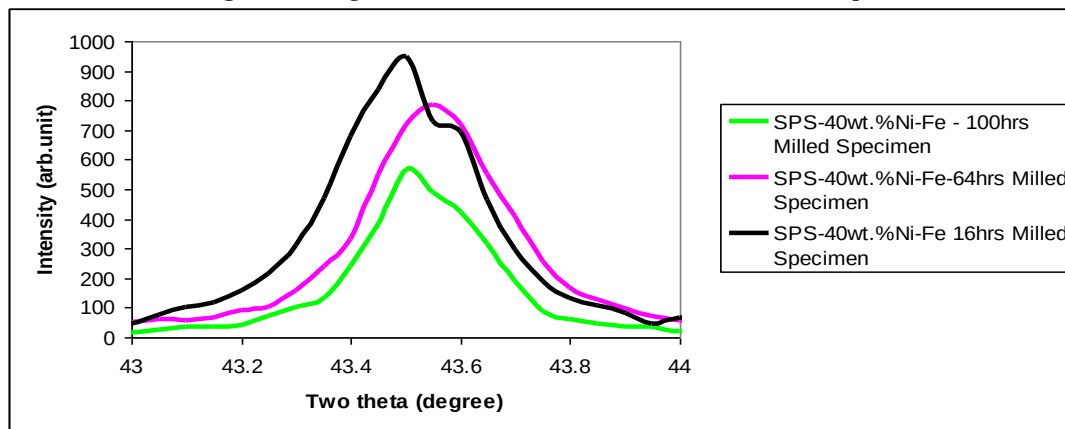


Figure-5: XRD Diffraction Patterns of SPS Sample (40wt. % Ni-Fe at Several Milling Hours).

Impact of Changes in Grain Size on Coercive Field: Figure-7 illustrates the impact of grain size on the variation in Hc. It's clear that as grain size decreases, Hc increases. For instance, in a 40% Ni-Fe alloy, Hc reaches its maximum value of 47 Oe when the grain size is 110 nm, decreasing to 29 Oe when the grain size is 73 nm, and eventually increasing to 45 Oe with a grain size of 7 nm. Similar trends are observed for 50% and 75% Ni-Fe alloys. In general, the changes in Hc values decrease within the grain size range of 250 to 75 nm. Smaller grain size samples exhibit higher lattice imperfections and internal strain, contributing to a decrease in Hc.

Impact of Changes in Particle Size on Coercive Field: The effects of particle size on Hc are shown in Figure-8. Notably, higher Hc values are observed in larger particle size samples,

and Hc decreases as particle size reduces during milling. A decrease in Hc values is noted until a critical particle size is reached, beyond which Hc values remain nearly constant until a second critical dimension is reached. Further reductions in particle size result in increased Hc. These observations are consistent with the strong particle-particle interactions that occur when particle size decreases to a certain level.

Impact of Spark Plasma Sintering on Coercive Field: Figure-9 demonstrates the impact of Spark Plasma Sintering (SPS) on Hc values. SPS samples generally exhibit increased Hc values as milling hours rise, indicating a correlation between Hc and grain size, imperfection lattice, and internal strain in higher-hour milled samples. This aligns with findings by Chicinas²⁰.

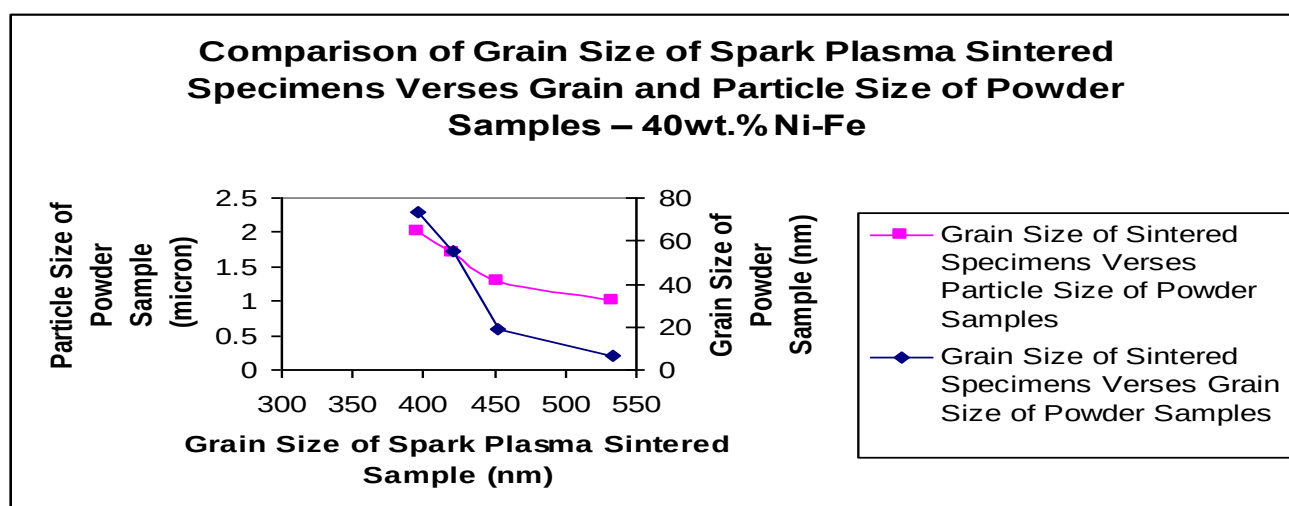


Figure-6: Comparison of Grain Size of Spark Plasma Sintered Specimens Vs Grain and Particle Size of Powder Samples.

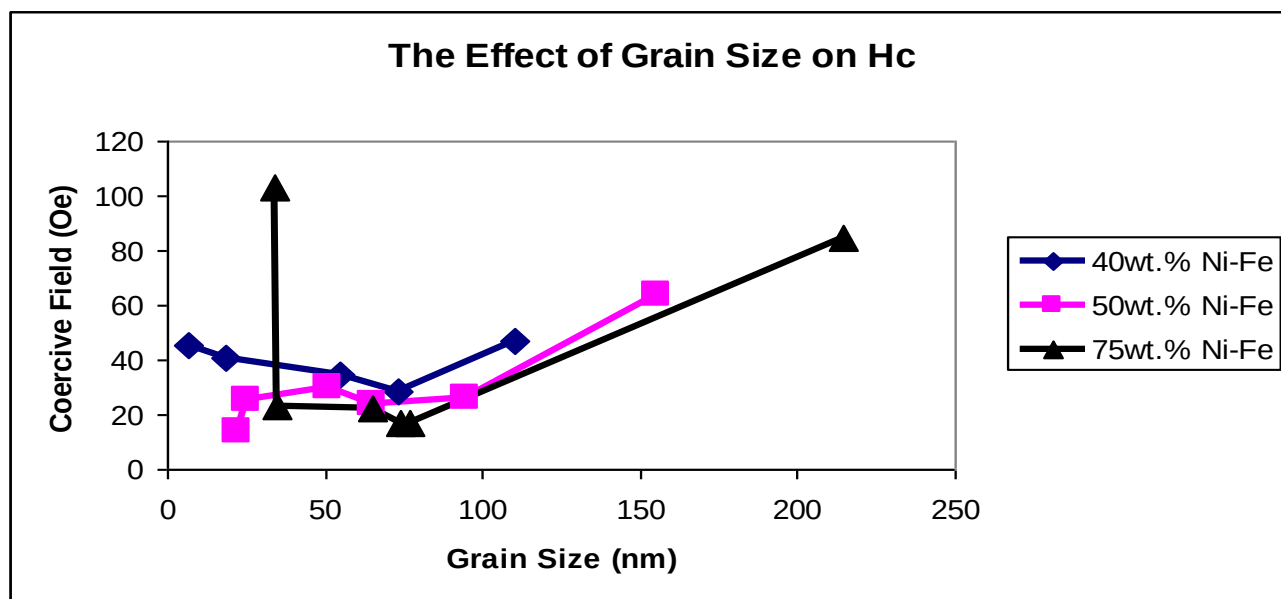


Figure-7: The Impact of Grain Size on Coercive Field.

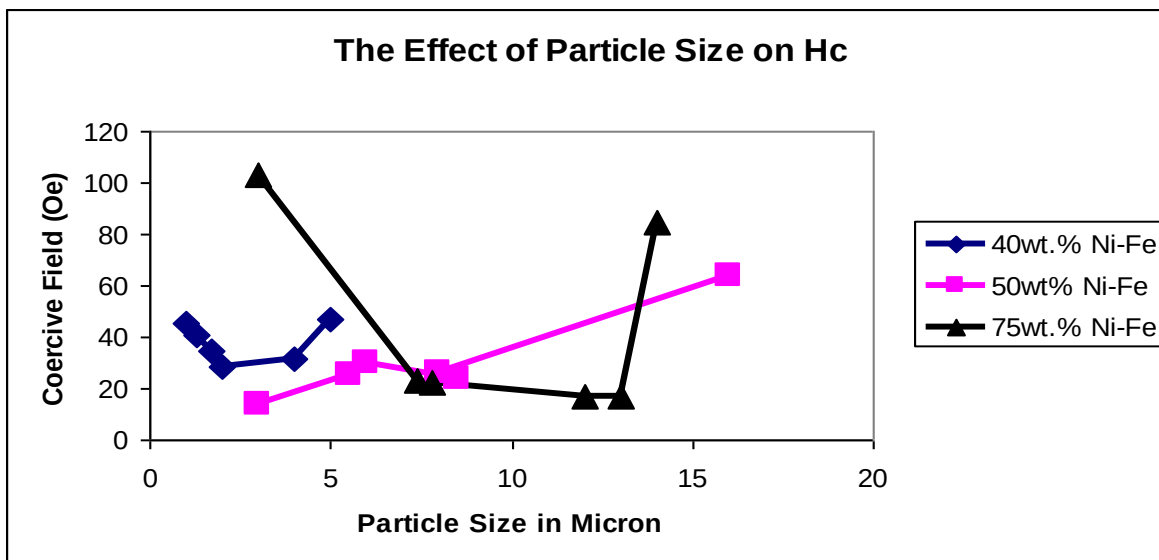


Figure-8: The Impact of Particle Size on Coercive Field.

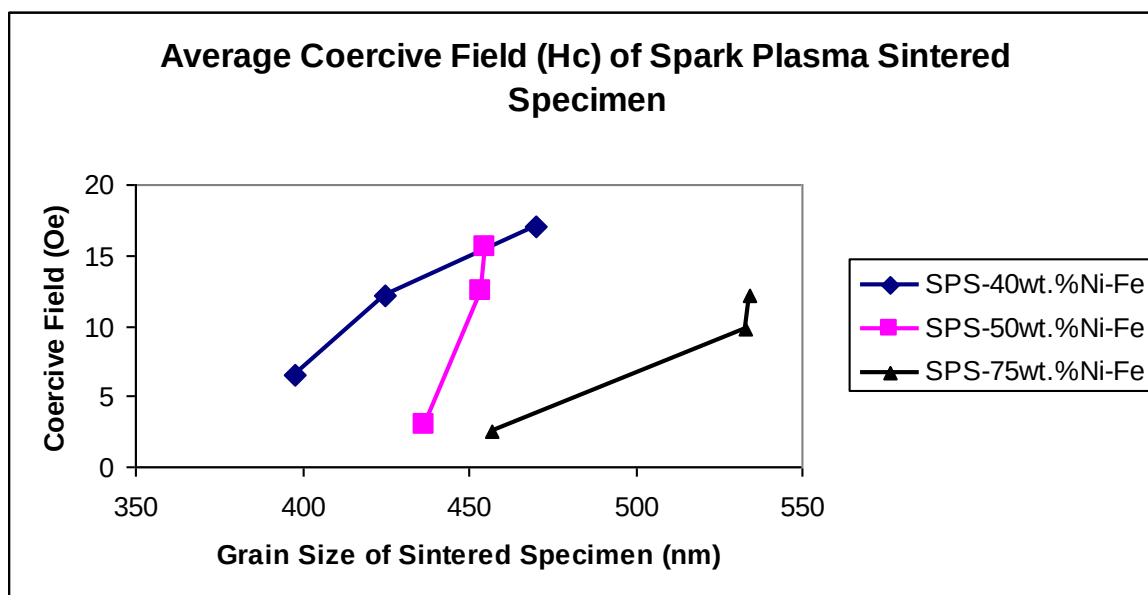


Figure-9: The Impact of SPS on Coercive Field.

Conclusion

In summary, the following key findings emerge from this study: Coercive Field and Grain Size: The coercive field (H_c) values exhibit a noticeable reduction with decreasing grain size in the range of 250 to 75 nm. Notably, a particularly high H_c value of 103 Oe is observed in the 75 wt. % Ni-Fe alloy after a 100-hour milling process. This exceptional H_c value serves as a strong indicator of the formation of single domain grains in this specific sample.

Coercive Field and Particle Size: A decrease in H_c values is evident until a critical particle size is reached. Subsequently, H_c

values remain nearly constant until a second critical dimension is achieved. Beyond this point, further reductions in particle size lead to an increase in H_c . This observed behavior underlines the intricate relationship between particle size and coercivity.

Spark Plasma Sintering (SPS) Impact: It is noteworthy that the coercive field values of all SPS samples are consistently lower than those of the powder samples. This effect indicates that the sintering process influences the magnetic properties of the material, with reduced coercivity in the sintered specimens.

These findings collectively emphasize the intricate interplay between grain size, particle size, and the sintering process in

determining the coercive field of Fe-Ni alloys. Understanding these relationships is crucial for tailoring the magnetic properties of such materials for specific applications. Further research in this area promises valuable insights into the optimization of magnetic materials for various technological and industrial uses.

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