Effects of cooling rate upon annealing on the microstructure and properties of the FeCo49V2Nb0.2 alloy

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ABSTRACT

In this paper, the influence of heat treatment on the magnetic properties of permendur the FeCo49V2Nb0.2 alloy used as the rotor core of a generator is investigated. The as-cast alloy is forged, hot and cold rolled to a thickness of 0.2 mm. The alloy samples were annealed in an argon gas furnace at different cooling rates. The results of measuring properties using the vibrating sample magnetometer method and measuring hardness show that the cooling rate has a significant impact on the properties of the FeCo49V2Nb0.2 alloy. Combining X-ray diffraction analysis and microstructure analysis allows us to explain the changes in properties obtained. From the results obtained, a conclusion can be drawn about the appropriate cooling rate for this alloy to achieve the best magnetic properties.

Keywords: Microstructures; Properties; FeCo49V2Nb0.2 alloy; Cooling rate; Soft magnetic alloys.

1. INTRODUCTION

Fe-Co alloys near their equiatomic composition are technologically significant due to their high saturation magnetization, low crystal magnetic anisotropy, and high permeability. Important commercial alloys based on this system include 2V-Permendur which contains approximately 49% Fe, 49% Co, and 2% V. These alloys are relatively expensive and are used in specialized applications that demand the unique magnetic properties of these materials. The high saturation magnetization, high Curie temperature, high permeability, and low coercivity values achieved in Permendur and Supermendur alloys make them suitable for use as transformer core materials requiring high magnetic flux density. The high saturation magnetization values enable a significant reduction in the mass or size of parts, which is crucial in many applications, such as generators in aircraft. These alloys are also used for generator windings, switch and storage cores, and components operating at high temperatures, such as barriers in portable phones, and they are applied in magnetostrictive transducers in sonar devices [1-4].

The decisive factors for the outstanding magnetic properties of FeCoV alloys are the deformation conditions (hot, cold) and the heat treatment process, that have significant influences on the microstructure obtained. The addition of vanadium improves the ductility of FeCo alloys in the disordered state. The ordering transformation in FeCo-V alloys leads to a loss of ductility at room temperature [5-7]. However, Kawahara [8] has shown that the ductility of FeCo-V alloys under ordered conditions can be improved by cold rolling the alloys above 70%. Alloys with a nominal composition of Fe49-Co49-V2 can be cold-worked up to 90% in the disordered state [2, 9]. Commercially cold-rolled FeCo-V alloy sheets are heat-treated, typically in the temperature range of 600 and 900 °C, to achieve a balance between mechanical and magnetic properties. On the other hand, room temperature strength and ductility can reach their maximum levels with suitable heat treatment.

In addition to alloying FeCo equiatomic alloys with V, small amounts of C, Nb, W, etc., were also studied to improve the mechanical or technological properties of these alloys [2, 3]. Although there are manufacturer’s recommendations regarding the application characteristics of these alloys, the specific influence of Nb on the heat treatment process and heat treatment characteristics of FeCo-2V alloys with Nb have not been extensively disclosed. To take proactive measures in the manufacturing technology of FeCo-2V alloy containing Nb in Vietnam, the influence of cooling rate during the heat treatment process on the microstructure and properties of FeCo-2V alloy with Nb is investigated, aiming to establish a heat treatment process for this alloy.

2. EXPERIMENTS

The Fe-Co-V-Nb alloy is manufactured from electrolytically pure metals (Fe, Co, V, and Nb). Subsequently, the alloy is melted in a vacuum induction melting furnace (VIF02) in a MgO crucible with an ultra-clean argon gas environment. A 0.6 kg batch is cast at temperatures ranging from 1520-1540 °C and poured into a water-cooled copper mold. Chemical composition analysis of the cast samples is conducted using a combination of Co background emission spectroscopy for major elements and the Fe background emission spectroscopy method for impurities such as P and S (table 1). The results of chemical composition analysis indicate that the fabricated alloy grades have compositions consistent with the 49К2ФА alloy grade (ГОСТ 10994-74) [10]. However, there is an additional component of Nb not specified in the standard. Considering the inclusion of Nb in its composition, the alloy manufactured is equivalent to VACODUR 50 grade by the Vacuumschmelze firm, Germany [11]. In the following section, the alloy fabricated for this research is denoted as FeCo49V2Nb0.2.

Table 1. *Composition of alloys melted (% wt).*

Alloy	**Chemical composition (% wt)**									
	C	P	S	Nb	V	Fe	Co	Mn	Si	Ni
FeCoV1.7Nb0.2	0.01	0.0142	0.0141	0.21	1.75	47.66	49.3	0.013	0.023	0.14

The ingots of FeCo49V2Nb0.2 alloy, measuring 35 × 70 × 50 mm, are subjected to forging and hot rolling at a temperature of 1020 °C in an argon-protected furnace. The hot rolling process is controlled to achieve a plate thickness of $1{,}8_{-0{,}1}$ mm. The width of the workpiece after hot rolling is approximately 76 mm, corresponding to a length of about 120 mm. Subsequently, the billets are annealed at 840 °C and quenched in a brine solution (10% NaCl in ice water) to obtain a supersaturated solid solution with good technological properties.

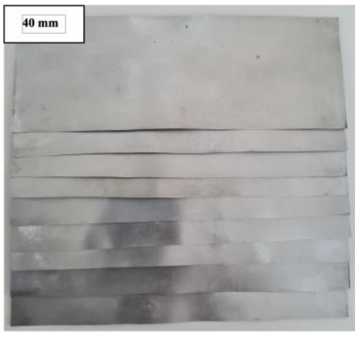

Figure 1. *FeCo49V2Nb0.2 alloy plate after fabrication.*

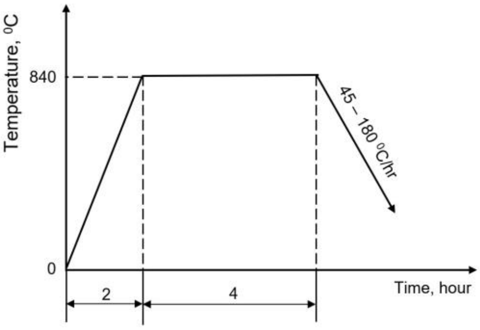

Figure 2. *The heat treatment process for the cold-rolled alloy plates.*

The alloy plates continue to be cold-rolled on a specialized four-axis rolling mill with two driving axes and two rolling axes. The rolling process is carried out in multiple steps, each step consisting of approximately 4 rolling times. The amount of deformation per rolling step is 0.1 mm and is adjusted by changing the distance between the two rolling axes. The final rolled thickness obtained is (0.2 ± 0.02) mm with a plate width of 70 mm and a length of approximately 200 mm (figure 1).

The samples, after rolling, are cut into plates with dimensions of 200 mm × 70 mm. Samples for measuring magnetic properties are cut into sizes of 4 mm × 4 mm for heat treatment along with the plates. The heat treatment process is carried out in a chamber furnace H30 with a vacuum heat treatment box. The samples are placed inside the heat treatment box, and a vacuum is applied at a pressure of 10 Pa. Then, high-purity argon gas (99.999%) is introduced into the chamber at a pressure of 1.5 atm. The vacuum evacuation and gas filling process is repeated three times before placing the box into the furnace for the heating and cooling process, as shown in figure 2. All samples are heated and held at the same temperature but cooled at different cooling rates. The samples labeled M1, M2, M3, M4, M5, and M6 are cooled at cooling rates of: 180 °C/h, 120 °C/h, 90 °C/h, 75 °C/h, 60 °C/h, and 45 °C/h respectively.

Each group of samples with the same chemical composition is selected for four samples for: measuring magnetic properties, measuring hardness, microstructure and X-ray diffraction analysis.

The magnetic properties are determined by measuring the magnetization curve using a vibrating sample magnetometer (VSM), such as the Lakeshore VSM. A maximum magnetic field of 200 kA/m is applied, and at each magnetic field intensity, the magnetization generated in the sample is recorded. From these data points, the magnetization curve is constructed, and parameters such as the remanent flux density and the coercive force, are calculated.

The samples are prepared for microstructural observation by polishing and etching in a 10% HCl + HNO_3 solution in ethanol. After etching, the microstructure is then photographed using an AxioVert A2M microscope with a magnification of 200 times. The hardness of the thin samples is determined using the Vickers hardness measurement method with a load of 0.3 kg.

After heat treatment, the phase composition of the material is determined using the X-ray diffraction (XRD) method. The X-ray diffraction patterns are scanned using a Bruker 2000 X-ray diffractometer. The main parameters of the X-ray diffraction process are a scanning angle of 20 to 80 degrees, a scanning step of 0.15 degrees, and an exposure time of 60 seconds at each scanning point.

3. RESULTS AND DISCUSSION

Table 2 presents the hardness values of the samples after annealing at different cooling rates. From the results in this table, it is noted that within the measurement error range, lower cooling rates lead to lower hardness of the samples. However, when the cooling rate is around 45 °C/h, the sample exhibits higher hardness compared to samples cooled at rates of 60 - 90 °C/h.

***Table 2.** The hardness of the samples after annealing with different cooling rates.*

Labels	**The annealing temperature (^oC)**	**The holding time (h)**	**Cooling Rate ($^oC/h$)**	**Hardness ($HV_{0.3}$)**
M1	840	4	180	187
M2	840	4	120	177
M3	840	4	90	154
M4	840	4	75	159
M5	840	4	60	138
M6	840	4	45	166

Table 3 summarizes the results of saturation magnetization and coercivity for the samples. According to the microstructure (figure 4), the samples can be divided into three groups: samples M1-M2 exhibit a two-phase mixed microstructure, samples M3-M5 have an almost single-phase microstructure with very few dispersed phase particles, and sample M6 exhibits a microstructure consisting of a matrix phase and a distinguishable precipitated phase. The magnetic properties of the samples also show corresponding differences with the variations in microstructure. Samples M1-M2 have a coercive force of around 340 A/m, samples M3-M5 have a coercive force of around 310 A/m, and sample M6 has a coercive force of up to 408 A/m.

Table 3. *Saturation magnetization and coercivity of the samples.*

Labels	**Cooling Rate (^{o}C/h)**	**Coercivity, H_c (A/m)**	**Saturation Magnetization, μ_0M (T)**
M1	180	344	2,31
M2	120	340	2,33
M3	90	308	2.35
M4	75	318	2.26
M5	60	309	2.36
M6	45	408	2.1

Based on the phase diagram of Fe-Co-V, the alloy with a composition of 49% Fe, 49% Co, 2% V at a temperature of 840 °C exhibits a single-phase microstructure with a body-centered cubic crystal structure, denoted as α phase. At room temperature, the equilibrium microstructure of this alloy consists of an ordered phase with a primitive cubic crystal structure, denoted as the α' phase [12]. Therefore, under the given conditions, if the alloy with the specified composition is cooled slowly enough, the α phase will transform into the α' phase. However, if the cooling rate is fast enough, a portion of the α phase will transform into the α_1 phase, which has a body-centered cubic structure. This is a metastable phase, which can be considered an excess α phase that does not undergo transformation [13]. In addition, alloy**s** with the supplementary composition of Nb under certain conditions may exhibit additional finely dispersed Nb-rich phases that can strengthen [2].

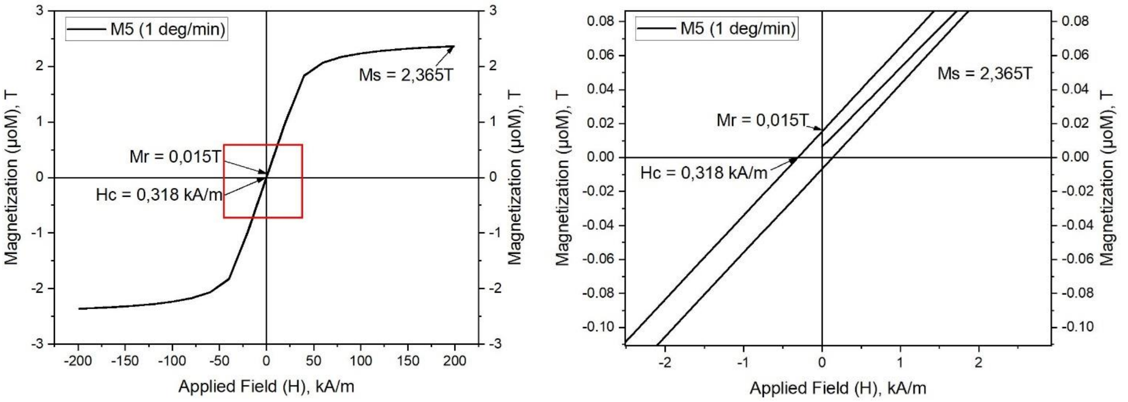

Figure 3. *Shows the hysteresis curves of the alloy samples with magnetization at different magnetic field strengths.*

The X-ray diffraction analysis results (figure 5) clearly show that the predominant structure of the alloys is the primitive cubic crystal structure Pm-3m. This corresponds to the ordered α' phase with high magnetic properties**,** as mentioned above. However, the X-ray diffraction analysis results do not indicate any diffraction peaks corresponding to the presence of secondary phases or the α_1 phase. The reason for this is that the number of these phases is small and insufficient to be detected on the X-ray diffraction diagram. To detect these phases, additional structural survey techniques need to be employed. Combining with the microstructural images, it can be inferred that the secondary phase with a small amount within the microstructure of the alloy cooled at rates of 180 °C/h and 120 °C/h (figure 4a, 4b) may be the metastable phase α_1. This metastable phase exists separately from the α matrix phase and is also a ferromagnetic phase with excellent saturation magnetization but high coercivity. Therefore, samples M1, M2 with a small amount of separated phase α_1 have good soft magnetic properties [14].

Samples M3-M5, after annealing, exhibit a single-phase microstructure. The structure consists of α phase particles with a size distribution in a wide range, so it is unable to demonstrate the differences due to the cooling rate. Therefore, the magnetic properties of these samples are similar, with lower coercive force compared to samples M1-M2, which is more significant for soft magnetic materials [15]. However, these magnetic properties could be further improved by achieving a microstructure with larger, more uniformly sized α phase particles by adjusting the annealing temperature and holding time. This would result in a microstructure with fewer grain boundaries, hindering the movement of domain walls.

Sample M6 clearly illustrates a typical precipitate microstructure with a matrix phase and finely dispersed second-phase particles within the matrix phase. The matrix phase is the solid solution α' phase with good ferromagnetic properties. The second phase is an equilibrium phase with the Nb element, requiring long holding times or very slow cooling rates [13]. This phase could be a non-magnetic or weakly magnetic phase, and its precipitation causes a reduction of the saturation magnetization of the sample to 2.1 T. Additionally, these phase particles dispersed within the α matrix phase hinder the movement of domain walls during magnetization and demagnetization processes. As a result, this microstructure exhibits higher coercivity than that of a single-phase solid solution α microstructure. The coercive force in this case is 408 A/m.

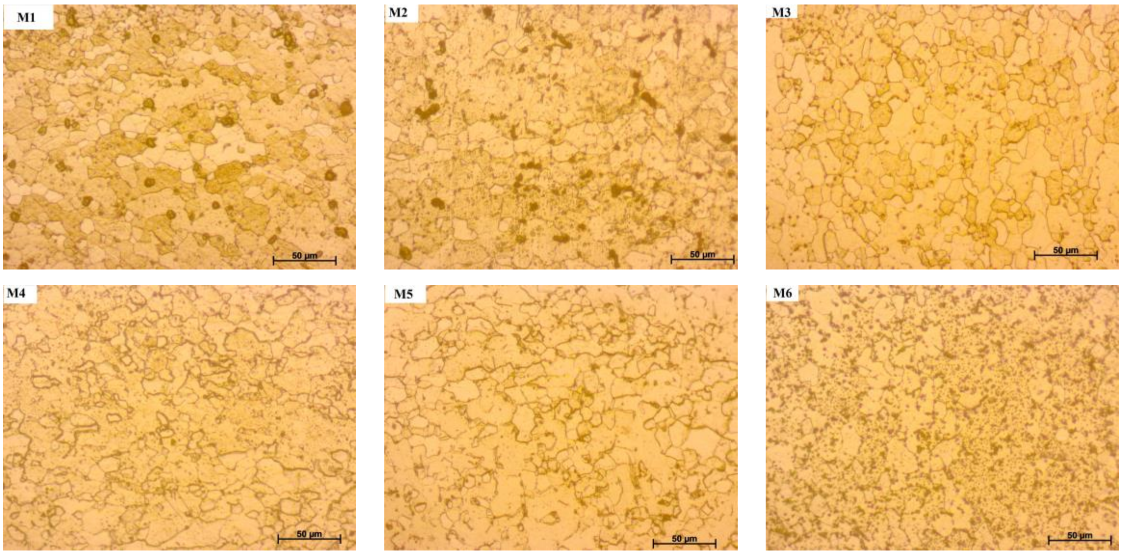

Figure 4. *Microstructure of samples cooled at different cooling rates: (M1) 180 °C/h, (M2) 120 °C/h, (M3) 90 °C/h, (M4) 75 °C/h, (M5) 60 °C/h, (M6) 45 °C/h, magnification 200 times.*

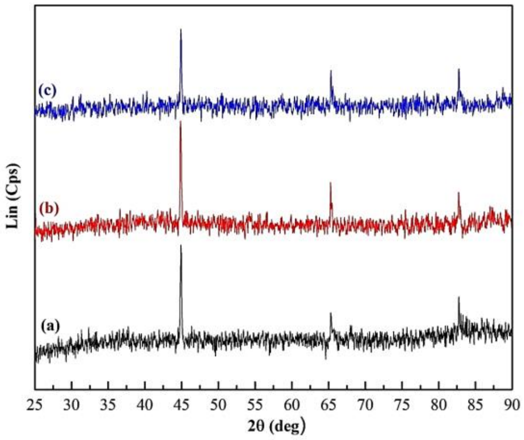

Figure 5. *X-ray diffraction diagrams of the samples cooled at different cooling rates: (a) 180 °C/h, (b) 60°C/h, (c) 45°C/h.*

From the obtained magnetic properties and structural analysis results, it can be deduced that the suitable cooling rate range for annealing the FeCo49V2Nb0.2 alloy is between 60 °C/h and 90 °C/h. Higher cooling rates may lead to the precipitation of metastable phases alongside the matrix phase, reducing saturation magnetization and increasing coercivity. On the other hand, excessively low cooling rates may result in the formation of secondary phases precipitating in the matrix phase, significantly reducing magnetic properties [16]. The microstructure achieved within the recommended cooling rate range will mostly consist of a single-phase structure with a neglectable amount of second phase, ensuring high soft magnetic properties: saturation magnetization (μ_0M) around 2.3 T and coercive force (H_c) approximately 310 A/m.

4. CONCLUSIONS

The cooling rate during annealing significantly affects the microstructure and properties of the FeCo49V2Nb0.2 alloy. Analysis of the structure by X-ray diffraction and microstructure examination reveals that the primary phase formed within the investigated cooling rate range is an ordered phase with a primitive cubic structure, exhibiting good soft magnetic properties: high saturation magnetization, low coercive force.

If annealed at high cooling rates (120 - 180 °C/h), surplus phases will appear in the microstructure, enhancing the coercive force while also improving the mechanical properties of the FeCo49V2Nb0.2 alloy. However, excessively low cooling rates (45 °C/h) also significantly degrade the magnetic properties due to the precipitation of the second phase on the matrix phase. These finely dispersed non-magnetic phases reduce saturation magnetization and increase coercivity.

The survey results indicate that the optimal cooling rate for annealing the FeCo49V2Nb0.2 alloy is between 60 °C/h and 90 °C/h. The achieved soft magnetic properties within this cooling rate range are saturation magnetization (μ_0M) approximately 2.3 T and coercivity (H_c) around 310 A/m. However, the microstructure obtained at this cooling rate range is not yet optimal, so the soft magnetic properties can still be improved by adjusting the annealing temperature and holding time.

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TÓM TẮT

Nghiên cứu ảnh hưởng của tốc độ nguội đến tổ chức và tính chất của hợp kim FeCo49V2Nb0.2

Bài báo trình bày ảnh hưởng của chế độ nhiệt luyện đến tính chất từ của hợp kim permendur FeCo49V2Nb0.2 làm lõi roto của máy phát điện. Hợp kim sau nấu luyện được rèn, cán nóng và cán nguội thành tấm đến chiều dày 0.2 mm. Các mẫu hợp kim được ủ trong lò khí argon với các tốc độ nguội khác nhau. Kết quả đo tính chất từ bằng phương pháp từ kế mẫu rung và đo độ cứng cho thấy tốc độ nguội khi ủ có ảnh hưởng đáng kể đến tính chất của hợp kim FeCo49V2Nb0.2. Kết hợp với phân tích cấu trúc bằng nhiễu xạ tia X và chụp ảnh tổ chức tế vi cho phép giải thích được sự thay đổi tính chất thu được. Từ các kết quả nhận được có thể rút ra kết luận về tốc độ nguội thích hợp trong ủ hợp kim này để đạt tính chất từ tốt nhất.

Từ khoá: Tổ chức; Tính chất; Hợp kim FeCo49V2Nb0.2; Tốc độ nguội; Hợp kim từ mềm.