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Effect of Cold-Rolling and Annealing on Microstructural and Magnetic Properties of 79HM Soft Magnetic Alloy Manufactured by Vacuum Induction Melting

Phung Tuan Anh*, Tran Van Nghia**, Nguyen Van Hoang***and Nguyen Manh Hung***

Keywords: Soft magnetic alloy, Ni-Fe-based alloys, 79HM permalloy alloy, cold-rolling, vacuum induction melting.

ABSTRACT

In this study, the effect of cold-rolling and annealing on the microstructural and magnetic properties of 79HM permalloy alloy was investigated systematically. The alloy was fabricated in a vacuum environment using a vacuum induction furnace (VIF), and followed by homogenization and cold-rolling processes. The grain size, grain morphology, hardness, chemical composition, and magnetic properties of the resulting alloy were analyzed by various materials characterization techniques, including optical microscopy, Vickers hardness testing method, scanning electron microscopy (SEM), energy-dispersive spectroscopy (EDS) integrated with a scanning electron microscope, and vibrating-sample magnetometry (VSM) technique. The experimental data show that the hardness and coercivity of the alloy increase significantly with the increase of cold-rolling degree, and reach maximum values of 320 HV5 and 496 A/m, respectively. However, upon suitable annealing conditions, the alloy can reach a coercivity value of 7.2 A/m, while its saturation magnetization is further enhanced. In addition, the effect of the annealing treatment on the recrystallization process and the magnetic properties of the 79HM permalloy alloy were also considered.

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E-mail: hungnm@lqdtu.edu.vn
* *Associate Professor, Department of Materials Science and Engineering, Le Quy Don Technical University, Hanoi, 100000, Vietnam*
** *Lecturer, Department of Materials Science and Engineering, Le Quy Don Technical University, Hanoi, 100000, Vietnam*
*** *Doctor of Philosophy in Materials Science and Engineering, Department of Materials Science and Engineering, Le Quy Don Technical University, Hanoi, 100000, Vietnam*

INTRODUCTION

Permalloys are Ni-Fe-based soft magnetic alloys with some other elements. Permalloys are known for their high permeability, low coercivity, and near-to-zero magnetostriction (Arnold and Elmen 1923; Pfeifer and Radeloff 1980; Couderchon 1991). Due to their excellent soft magnetic properties, permalloys are used widely as magnetic core material and magnetic shielding material under a weak magnetic field (Sun et al. 2016), electrical machines (Gehrmann 2005; Narasimhan, Hanejko, and Marucci 2007), magnetic sensors (Yanai et al. 2018), magnetic amplifiers, and high-quality transformers (Akomolafe and Johnson 1989). Among the permalloys, 4-79 molybdenum permalloys are the most well-known in aerospace and other electronics industries. They are used to produce the types of equipment that possess high sensitivity, precise dimensions, small size, low high-frequency loss, good time and temperature stability as well as special functions. It is also widely used in communications, instrumentation, computers, telemetry, remote control, wireless charging and other systems (Popa et al. 2012; Olekšáková et al. 2007; Karimi et al. 2013). Permalloys, including 4-79 molybdenum permalloys, can be manufactured by powder metallurgy method (Ma et al. 2014), mechanical alloying (Marinca et al. 2022), injection molding, selective laser melting, or vacuum induction melting (VIM) (Olekšáková, Kollár, and Füzer 2017; Vahdati Yekta, Ghasemi, and Sharifi 2018; Ma et al. 2013; Chicinas, Pop, and Isnard 2004; Ghosh et al. 2014; Kaloshkin et al. 2001; Popa et al. 2012). Among these methods, the fabrication of permalloy alloys by VIM possesses lots of advantages, such as high purity and quality, rapid melting and cooling, controlled atmosphere.

In terms of the magnetic characteristic aspect, both permeability and coercivity depend on many factors, including chemical constituents, mechanical processing, and thermal treatment after casting. A cold deformation process always leads to significant changes in the dislocation density, grain size, residual stress, and strain. Especially, when the deformation degree is large enough, the deformed material will become anisotropy and textures are formed. In the meantime, the movable ability of the magnetic domain walls is deeply affected by microstructural characteristics, such as inclusions, grain boundary, dislocation density, texture…Therefore, the effect of the deformation processing on the magnetic properties of the permalloy alloys is an attractive interest, and has been reported in the literature (Yekta, Ghasemi, and Sharifi 2018; Mazaleyrat 2021). In addition, the final annealing, which is indispensable for the manufacture of magnetic materials, also affects strongly grain size, residual stress, and strain, thereby all of the above-mentioned magnetic characteristics (Masahiro and Kazuhiro 1995; Dost et al. 2020; Li et al. 2020). To the best of our knowledge, although lots of papers have been reported up to now about the effects of cold-deformation and/or annealing on the magnetic properties of permalloy alloys, there are few papers about 79HM permalloy alloys, especially the alloys fabricated by VIM. Therefore, in this study, we synthesized 79HM permalloy alloy by VIM and investigated the effect of cold-rolling and annealing on the microstructural and magnetic properties of this alloy systematically. In addition, the discussions related to recrystallization and its effects on the magnetic performance of the alloy were also proposed.

EXPERIMENTAL SECTION

Synthesis of 79HM permalloy alloy samples

The reagents of nickel (Ni, ≥ 99.99 %, Norilco Co., Ltd, Russia), molybdenum (Mo, ≥ 99.99 %,), iron (Fe, ≥ 99.99 %), and manganese (Mn, ≥ 99.99 %) were purchased from AEM Co., Ltd (China). All the chemical reagents were used in the supplied state without further purification.

In this study, a 79HM permalloy alloy ingot was synthesized by melting a mixture of nickel, molybdenum, iron, and manganese elements in the vacuum environment, using a vacuum induction furnace (VIF, Nanjing Boyuntong Instrument, China). A brief procedure can be summarized as follows: Firstly, an ingot (~ 200 g) was prepared by melting the above mixture with a heating power of 6 kW and followed by naturally cooling to room temperature. The casting temperature was maintained at 1550 ºC during the melting process; (2) Secondly, the obtained ingot was homogenized at 1200 ºC for 4 hours in a chamber furnace; (3) Finally, the ingot was cut into 2 mm-thick sheets for further investigations.

In order to consider the effect of the deformation degree on the microstructure and magnetic properties of the 79HM permalloy alloy, a series of as-mentioned 2 mm-thick sheets were cold-rolled to reach various thickness reduction percentages, including 7 %, 15 %, 25 %, 40 %, and 60 %. The labels of all samples were denoted as in Table 1. In addition, for evaluation of the effect of the annealing temperature on the magnetic performance of 79HM alloy, the S60 sample was chosen to anneal at two temperatures: 900 ºC and 1050 ºC in various annealing temperature ranges (2 h, 4 h, and 6 h).

Table 1. Sample labels of different 79HM alloy samples in as-cast and cold-rolling states

Deformation degree	As-cast	7 %	15 %	25 %	40 %	60 %
Sample	S0	S7	S15	S25	S40	S60

Materials characterizations and magnetization measurement

The surface morphology and microstructure of the synthesized 79HM alloy samples were observed by Schottky field-emission electron scanning microscopy (FE-SEM, JSM 7610F, JEOL, Japan), which is integrated with an energy-dispersive spectroscopy (EDS) equipment for examination of the chemical composition of the obtained 79HM alloy. Besides, the changes in the grain morphology and size of the 79HM alloy samples were also investigated by optical microscopy, using an Axiovert optical microscope (Carl Zeiss Pte. Ltd, Germany). Magnetization measurement of the 79HM alloy samples was performed on a vibration sample magnetometer (VSM US/7407, Lake Shore Cryotronics, US).

RESULTS AND DISCUSSION

Effects of the cold-rolling degree on the microstructural properties and hardness

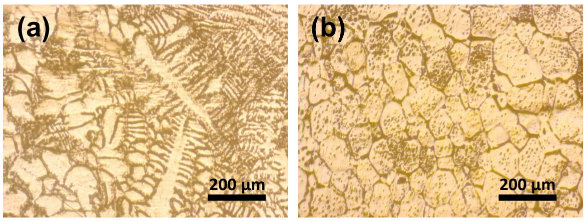

Figure 1. Microstructural images of the 79HM permalloy alloy in various states: (a) as-cast and (b) after homogenization annealing.

Figure 1(a) displays the optical microscopy image of the as-cast 79HM permalloy alloy. The microstructure of the alloy consists of coarse grains and a typical dendrite structure. Moreover, it should be mentioned that the alloy was made from lots of elements that possess significantly different melting temperatures. While the melting temperature of molybdenum is 2620 ºC, those of nickel, iron, and manganese elements are 1453 ºC, 1539 ºC, and 1244 ºC, respectively. Therefore, the chemical composition is expected to be slightly different from the inside to the outside of each grain in the alloy. A similar phenomenon also takes place when comparing the chemical composition of the grains and the grain boundary regions. To remove these phenomena, the alloy samples were homogenized at 1200 ºC for 4 hours. The result reveals that the dendrite regions are completely replaced by spherical grains (Figure 1(b)). The size of the spherical grain is distributed in a relatively wide range of approximately 20 to 100 µm.

Figure 2(a) shows the selected area for EDS measurement integrated with SEM. According to the EDS result, the chemical constituents of the synthesized samples are in good agreement with that of standard 79HM permalloy alloy (Figure 2(b)). The strongest peaks are ascribed to nickel, iron, molybdenum, and manganese elements. A quantitative analysis of the EDS spectrum exhibits the concentration (weight percentage) of nickel, iron, molybdenum, and manganese elements corresponding to 78.38 %, 14.47 %, 5.57 %, and 0.63 %, respectively. There are no peaks of contaminants are observed from the EDS spectrum suggesting the high purity level of the fabricated sample.

In terms of morphology, after cold-rolling, the morphology of grains is changed clearly according to the deformation degree (Figure 3). The grains tend to be spread out on the rolling surface that is perpendicular to the rolling direction. In the S7 sample, the deformation degree is relatively small (7 %). Therefore, the grain boundary between initial grains still can be observed although almost all of the grains were spread out significantly. At a higher deformation degree (15 % and 25 %), the grain boundaries cannot be observed. Instead, the morphology of the sample consists of discrete small grains distributed on a uniform distorted matrix. When the deformation degree reaches 40 % and/or 60 %, the matrix of the S40 and S60 samples exhibits a strong deformation level. Furthermore, previously discrete grains are shredded and dispersed into the matrix. It is well-known that deformation always results in a significant increase in the dislocation concentration of the deformed sample. In the meantime, crystallinity is one of the key factors that play an important role in the determination of the magnetic performance. Thereby, it is expected that the magnetic properties of cold-rolled samples can be modified by controlling the deformation degree. It is worth mentioning that the hardness of metals and alloys increases when the dislocation concentration increases (Ameri et al. 2017). As a result, the dislocation density can be estimated by considering the hardness. Figure 4 displays the Vickers hardness of the homogenized 79HM permalloy alloy sample and the samples cold-rolled at various deformation degrees. The lowest hardness value is ascribed to the homogenized sample without deformation, while all of the cold-rolled samples exhibit a higher hardness. The examined hardness results show that the larger the deformation level is, the higher the hardness is. As mentioned above, the hardness of the S60 sample is the highest suggesting its highest dislocation concentration.

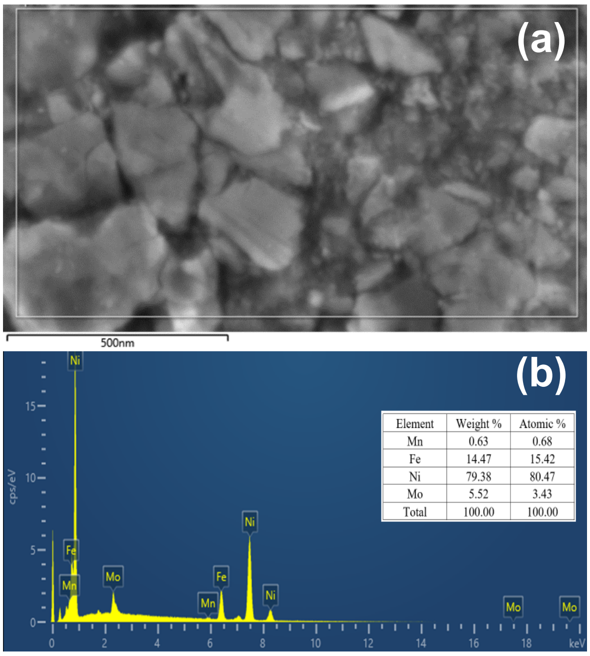

Figure 2. (a) The selected area for the EDS and (b) the corresponding resulting EDS spectrum.

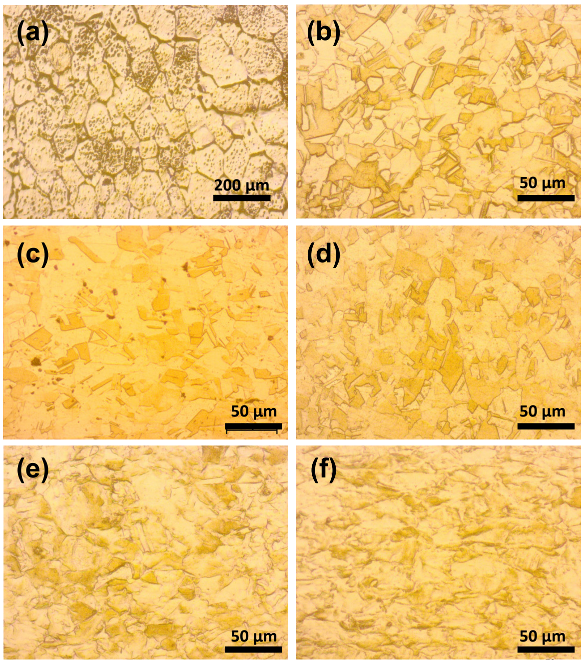

Figure 3. The microstructural images of the as-cast and cold-rolled 79HM permalloy alloy samples: (a) S0, (b) S7, (c) S15, (d) S25, (e) S40, and (f) S60.

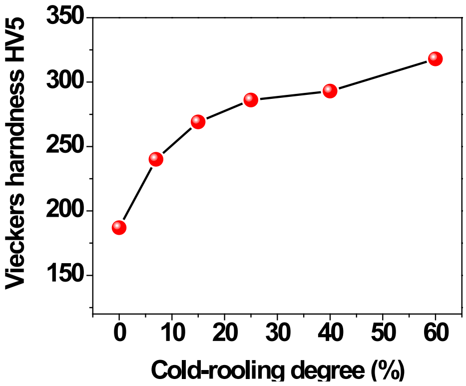

Figure 4. The Vickers hardness of 79HM permalloy alloy as a function of the deformation degree.

Effects of the cold-rolling degree and annealing temperature on the magnetic performance

In the as-cast state, the hysteresis loop of S0 exhibits a typical shape of soft magnetic materials. Therefore, it suggests that 79HM alloy was synthesized successfully. The saturation magnetization and coercivity of the alloy reach values of 68.32 emu/g and 184 A/m, respectively (Figure 5). The magnetic properties of the alloy are changed greatly after cold deformation, especially coercivity. The coercivity values of cold-rolled samples (S7, S15, S25, S40, S60) is approximately (1.5 ÷ 1.8) fold that of S0 sample (Table 2 and Figure 6). The larger the deformation degree is, the higher the coercivity of the sample is. As mentioned above, the dislocation density increases when the deformation degree increases. Moreover, the formation of new dislocations also leads to the further appearance of residual stress. These residual stress fields make the magnetic moments more difficult to rotate, thereby enhancing the coercivity of the cold-rolled 79HM alloy samples (Nagao 2004). These assessments are supported further by the magnetization measurement results of the annealed samples at 900 ºC and 1050 ºC.

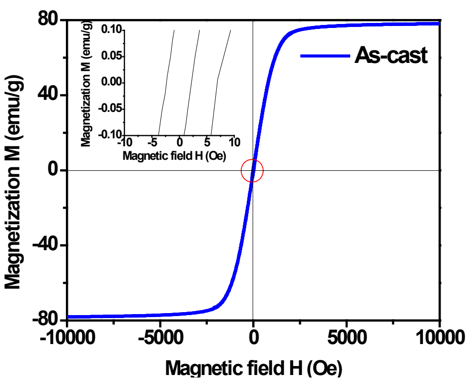

Figure 5. The hysteresis loop of the as-cast 79HM permalloy alloy specimen.

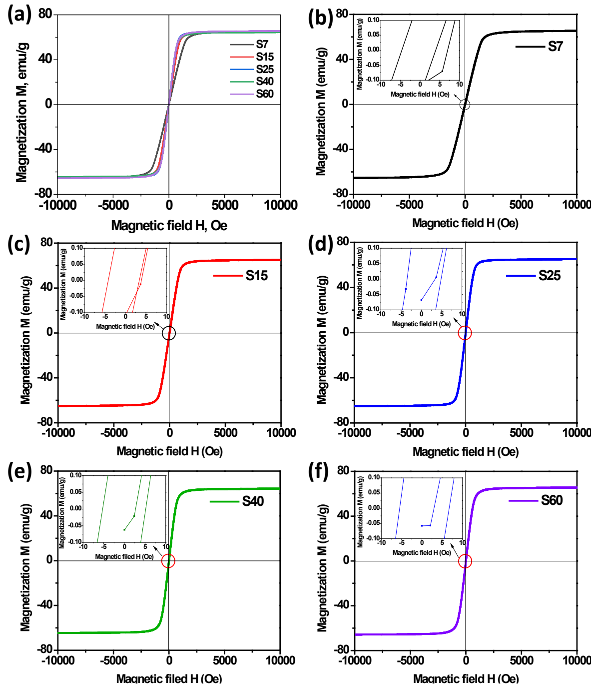

Figure 6. (a) The hysteresis loops of the 79HM permalloy alloy specimens cold-rolled at various deformation degrees and (b-f) the corresponding separated hysteresis loops: (b) S7, (c) S15, (d) S25, (e) S40, and (f) S60. The insets show the enlarged parts of the selected areas of each corresponding hysteresis loop.

Table 2. Summaries of the saturation magnetization and coercivity values of 79HM alloy samples

Sample	Saturation magnetization (emu/g)	Coercivity (A/m)
S0	68.32	184
S7	64.26	328.8
S15	64.43	304.4
S25	64.56	332.2
S40	63.95	415.6
S60	65.10	489.2

Figure 7 and Figure 8 show the hysteresis loops of the S60 sample annealed at 900 ºC and 1050 ºC for 2 h, 4 h, and 6 h. All of the samples annealed reveal a strongly reducing trend in the coercivity value. The experimental data show that when the annealing temperature increases from 900 ºC to 1050 ºC, the coercivity value decreases significantly. Especially, at an annealing time of 6 h, the coercivity of the sample annealed at 1050 ºC is reduced approximately 5 times compared to that the sample annealed at 900 ºC. This ratio is only approximately 1.74 and almost unchanged for the annealing times of 2h and 4h, respectively (Table 3 and Table 4).

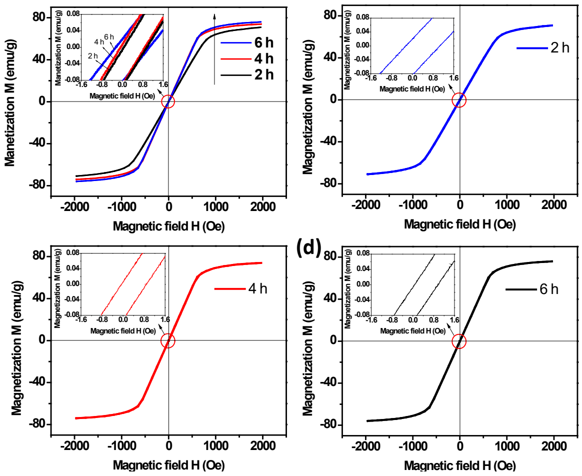

Figure 7. (a) The hysteresis loops of the S60 sample annealed at 900 ºC with various annealing times and (b-d) the corresponding separated hysteresis loops: (b) 2 h, (c) 4 h, and (d) 6 h. The insets show the enlarged parts of the selected areas of each corresponding hysteresis loop.

Table 3. Summaries of the saturation magnetization and coercivity values of S60 samples annealed at 900 ºC for various annealing times.

Annealing time	Saturation magnetization (emu/g)	Coercivity (A/m)
2 h	71.05	52.4
4 h	74.34	37.6
6 h	76.10	35.2

It should be noted that the samples were cold-rolled and followed by annealing at high temperature that is higher than the recrystallization temperature will take place the recrystallization phenomenon. Since the annealing time is relatively long, the recrystallization process of all samples transformed into a stage of grain growth. In this stage, the effects of the previous deformation process are almost removed, especially the dislocation density. Therefore, the interaction between magnetic moment with stress and strain around the dislocations becomes insignificant. This thing is suitable for the sharp reduction in the coercivity of the annealed sample compared to the pristine S60 sample. The effect of annealing time on the magnetic properties should be considered by the reduction of the grain boundary owing to the grain growth process. In terms of magnetic properties, the grain boundary is one of the factors that hinder the rotation of the domain wall upon the external magnetic field, thereby affecting the coercivity value of the magnetic materials. As a result, the coercivities of all of the samples annealed for 6h are the smallest for both 900 ºC and 1050 ºC annealing temperatures.

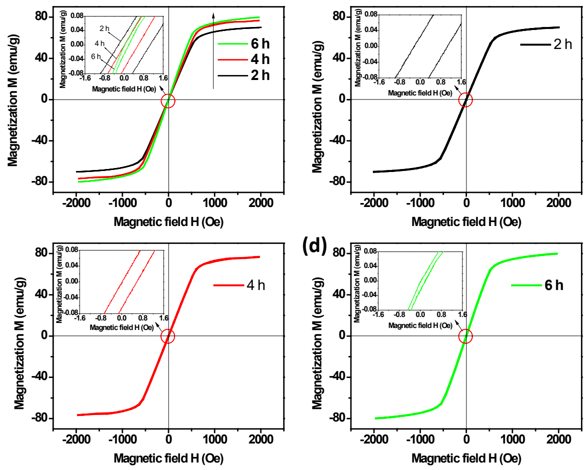

Figure 8. (a) The hysteresis loops of the S60 sample annealed at 1050 ºC with various annealing times and (b-d) the corresponding separated hysteresis loops: (b) 2 h, (c) 4 h, and (d) 6 h. The insets show the enlarged parts of the selected areas of each corresponding hysteresis loop.

Table 4. Summaries of the saturation magnetization and coercivity values of S60 samples annealed at 1050 ºC for various annealing times.

Annealing time	Saturation magnetization (emu/g)	Coercivity (A/m)
2 h	70.32	51.2
4 h	76.97	21.6
6 h	80.15	7.2

CONCLUSION

The effects of the deformation degree and heat treatment condition on the microstructure and magnetic performances of 79HM permalloy alloy were investigated thoroughly. The results suggest that effective control of the deformation degree and recrystallization process will allow for the achievement of desired magnetic properties through the adjustment of the dislocation density. Some key results can be summarized as follows:

(i) The microstructure of the alloy consists of spread-out grains on the planes that is perpendicular to the rolling direction. The grain shape is modified strongly when the deformation degree is over 25%.

(ii) The cold-rolling leads to an increase of the dislocation density in the 79HM permalloy alloy, thereby enhancing the hardness and the coercivity significantly. The maximum hardness and coercivity values of the alloy reach 320 HV5 and 496 A/m corresponding to a deformation degree of 60%, respectively.

(iii) Annealing can be used as an effective solution to obtain a relatively small coercivity value without resulting in a notable decrease in the saturation magnetization. The smallest coercivity value of 7.2 A/m was reached when the deformation degree was 60% and the alloy was annealed at 1050 ºC for 6 h.

(iv) The changes in the hardness and coercivity of the 79HM permalloy alloy are originated from the changes in the dislocation density after cold-rolling and/or annealing treatment as well as the interaction between the stress and strain around dislocations and magnetic moments in the materials.

The obtained results show that the fabricated 79HM permalloy alloy satisfies the specifications for a typical soft magnetic material. These results are the basis for further research to improve not only magnetic performance but also the microstructural and mechanical properties of 79HM permalloy alloy. However, further investigations need to be conducted to intend real applications, such as large-scale production ability, repeatability, and microstructural and magnetic properties at larger deformation levels.

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