TEM AND MOSSBAUER STUDIES OF AS-CAST Fe_2NiAl ALLOY DURING COOLING AT A CRITICAL RATE AFTER HOMOGENIZATION

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Abstract

The as-cast Fe_2NiAl samples were treated in accordance with the following regime: water quenching from 1240 °C (after holding for 20 min); cooling from 1240 °C (after holding for 20 min) at a critical cooling rate up to an intermediate temperature T_q = 900 - 20 °C with subsequent water quenching. Detailed studies of the solid solution decomposition, microstructure and magnetic properties of the samples are reported. TEM investigations of the solid solution decomposition ($\beta_2 \rightarrow \beta + \beta_2$) in as cast Fe_2NiAl alloy during cooling at a critical rate after homogenization showed that formation of the periodic modulated structure passes through an intermediate stage of destruction, leading to a drop of coercive force after quenching from 850 °C. It is shown that the "secondary" decomposition of the β_2-phase inside modulated structure, formed at higher temperatures, is associated with a sharp change in the volume and composition of the β and $\beta_2$phases due to the asymmetric shape of the miscibility gap in the Fe-Ni-Al system near 850 °C. Cooling of the cast Fe_2NiAl alloy at a critical rate to room temperature leads to the formation of an optimal modulated structure and obtaining the maximum value of the coercive force H_c = 51.2 kA/m.

Keywords: AlNi alloy, cooling at a critical rate,modulated microstructure, coercive force, Mössbauer spectra

1. INTRODUCTION

Nowadays, the AlNi alloys with coercive force $H_c \approx$ 56kA/m have become a less important type of permanent magnet materials in comparison to the rare-earth-based alloys. But due to a very high chemical and metallurgical stability the AlNi magnets are widely used at temperatures up to 500°C. In these alloys, the fine microstructure formsas a result of a precipitation reaction that occurs in the ingots during solidification and subsequent heat treatment. It has been contended that the β + β_2 duplex structure of Fe_2NiAl alloy is a consequence of spinodal decomposition [1, 2]. The highest$H_c$value, reached by continuously controlled cooling from high temperature (*HT*-I), was 1.5 times largerthan H_c reached by aging of previously water-quenched alloy (*HT*-II) [3, 4]. Additionally, it was observed, that during optimum continuous cooling the decomposition is a two-stage process. At relatively high temperature (≥ 850°C) the spinodal decomposition of the solid solution into β + β_2 occurs. The origin and growth of the β and β_2 particles and the formation of modulated microstructure occurs almost entirely during decomposition reaction at ≈ 850 °C [1-3]. At lower temperature (≤ 700 °C) the increase in the difference in magnetization between the phases connected with an exchange of atoms by diffusion of Fe atom from β_2 to β and Ni and Al atoms from β to β_2 resulting in an increase of the coercive force [5, 6]. The TEM results showed that the microstructure of Fe_2NiAl alloy arising from decomposition during continuous cooling is considerably different from the microstructure arising from quenching followed by aging [1, 7-9]. As mentioned above, the microstructural transformation at high temperatures has an essential effect on the magnetic properties of AlNi alloys. Therefore, it is very important to clarify the mechanism of microstructural evolution during continuous cooling of the Fe_2NiAl alloy.

The present study aims to investigate the formation and growth of the β and β_2 precipitates and the formation of modulated microstructure during cooling at an optimum rate after homogenization. Mössbauer spectrometry measurements were performed to investigate a compositional change of the β and β_2 phases during water quenching and optimum cooling to different temperatures T_q. The obtained results allow interpreting the behavior of H_c on the temperature T_q as a function of β and $β_2$ phases composition and modulated microstructure parameters.

2. EXPERIMENTAL PROCEDURE

The $Fe_{51.1}Ni_{23.5}Al_{23.7}Si_{1.7}$AlNi alloy was studied. The as-cast alloy was prepared by melting in an induction furnace using pure metals. The cast samples were additionally treated in accordance with the following regimes: water quenching from 1240 °C (after holding for 20 min); cooling of as-cast alloy from 1240 °C (after holding for 20 min) at a critical cooling rate (V_{cr} ~ 2 K/min) up to an intermediate temperature T_q = 900 - 20 °C with subsequent water quenching. The structures of the samples were examined by X-ray diffraction (XRD), transmission electron microscopy (TEM) and scanning electron microscopy (SEM). X-ray diffraction analysis was performed using a Rigaku diffractometer with Co *Kα*radiation and a graphite monochromator. The thin foils were examined using a JEM-1400 microscope operating at accelerating voltage of 120 kV. The microstructure of samples was also analyzed using a JEOL JSM-6610LV scanning electron microscope. Magnetic properties were measured at room temperature in magnetizing fields of up to 200 kA/m using a hysteresis graph AMT-4. The Mössbauer effect measurements were carried out at room temperature by Mossbauer spectrometer MS-1104 EM usingconstant acceleration method. The 14.4 KeV radiation of ^{57}Fe from the 30 mCi source was detected by a proportional counter. Speed ranges from - 9 mm/s to 9 mm/s, to 512 channels.

3. RESULTS AND DISCUSSION

Figure 1 illustrates the behaviour of coercivity H_c of the Fe_2NiAl samples cooled at a critical rate V_{cr} from 1240 °C to temperature T_q and then quenched in water. The coercive force exhibits a nonmonotonic dependence on the temperature T_q with minimum $H_c \leq 3.2$ kA/m at T_q = 850 °C.

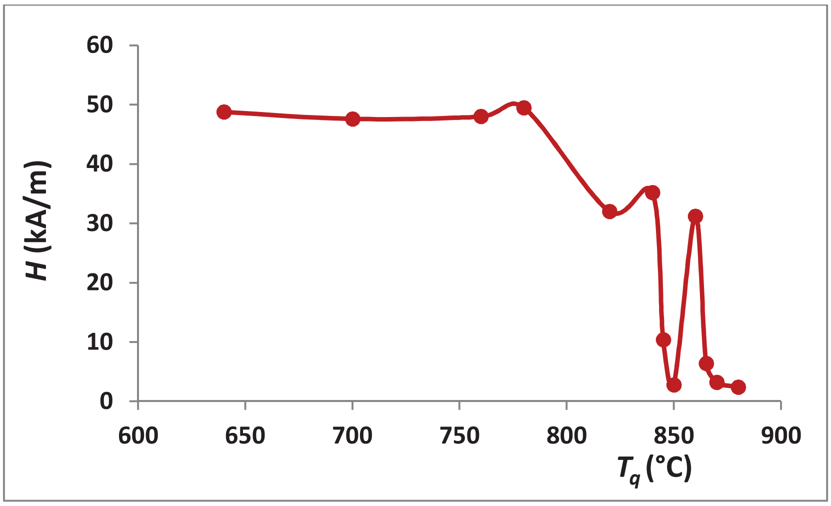
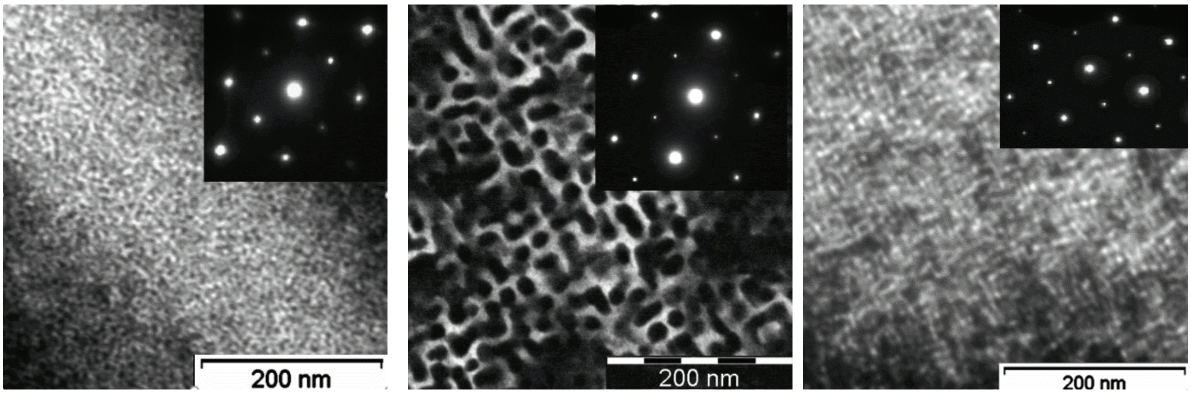


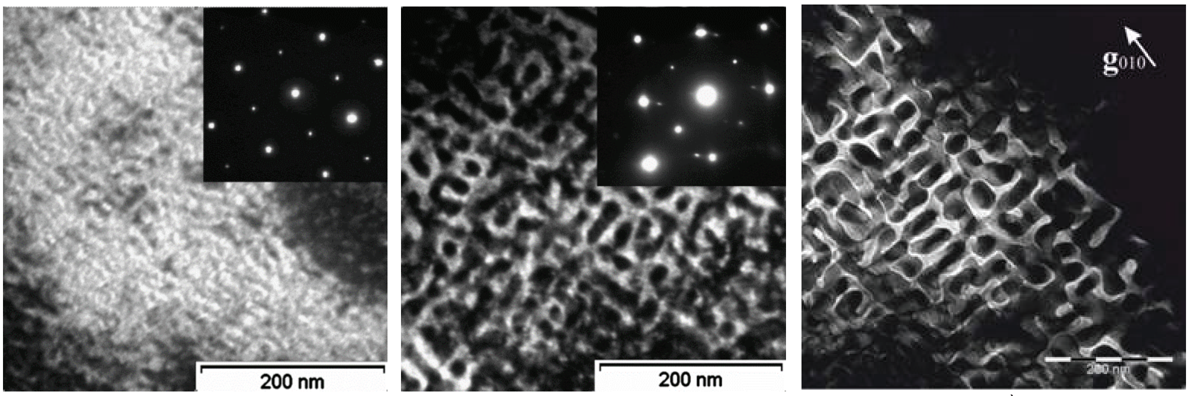
Figure 1 Dependence of H_c on the quenching temperature T_q during cooling at a critical rate from 1240 °C to T_q followed water quenching

Figure 2 shows micrographs of the as-cast samples after water quenching from 1240 °C (a) and after cooling at a critical rate from 1240 °C to T_q = 860 °C (b) and 850 °C (c). The microstructure of water quenched sample corresponds to the inhomogenies state with a zone size of <10 nm. In accordance with the electron diffraction pattern in **Figure 2(a)** (inset). X-ray data zone microstructure is in agreement with the partial decomposition of the solid solution into β (A2 type structure) and $β_2$ (B2-type structure) phases within the miscibility gap. The coercive force of water quenched sample is H_{ci} ~ 0.32 kA/m.



a) b) c)

Figure 2 Bright-field TEM micrographs of the Fe_2NiAl alloy: water quenched from 1240 °C, H_{ci} = 0.32 kA/m (a) cooled at a critical rate from 1240 °C to T_q = 860 °C, H_{ci} = 29.4 kA/m; (b) 850 °C, H_{ci} = 32 kA/m; (c) diffraction pattern (insets), zone axis [001]



a) b) c)

Figure 3 Bright-field TEM micrographs of the Fe_2NiAl alloy: cooled at a critical rate from 1240 °C to T_q = 845 °C, H_{ci} = 10.3 kA/m. (a) 840 °C, H_{ci} = 35 kA/m; (b) 20°C, H_{ci} = 52 kA/m; (c) electron diffraction pattern (insets), zone axis [001]

The cooling at a critical rate to T_q = 860 °C has caused the formation of modulated microstructure (**Figure 2b**) and increases H_c to 29.4 kA/m. However, cooling to T_q = 850 °C led to the destruction of modulated microstructure by splitting of β- and β_2-precipitates and forming the microstructure that is similar to the microstructure formed after water quenching from 1240 °C (**Figure 2b**). The coercive force of this sample shows the minimum $H_c \leq 32$ kA/m. The restoration of modulated microstructure and increase of the coercivity was observed already after cooling at a critical rate to $T_q \leq 845$ °C (**Figure 3a**). The modulated microstructure after cooling to T_q = 840 °C (**Figure 3b**) is similar to that formed after cooling from 860 °C (**Figure 2b**). The coercive force of this sample increase to H_c = 35 kA/m. The highest quality modulated microstructure giving the maximum H_c = 52 kA/m was obtained for the sample which was cooled at the critical rate to room temperature (**Figure 3c**). This structure consisting of elongated β-phase particles oriented mainly along <100> directions and separated by matrix β_2-phase areas. The bright β_2-phase forms an almost continuous net that separates dark β-phase particles. The size of β-phase particles varies from 20 to 80 nm; the degree of their aspect ratio *l/d* is from 1 to 4. It is likely that the combination of factors, such as the small size, shape anisotropy and magnetic isolation of β-phase particles ensures the high H_c = 52 kA/m of samples cooled at a critical rate to room temperature.

Figure 4 shows the Mössbauer spectrum measured at *RT* of the Fe_2NiAlsamples after water quenching from 1240 °C (a) and after cooling at a critical rate to T_q = 860 °C (b), 850 °C (c) and 840 °C (d) followed by water quenching. This is essentially a six-line spectrum typical for ferromagnetic phase and two addition peaks (doublet) for paramagnetic phase.

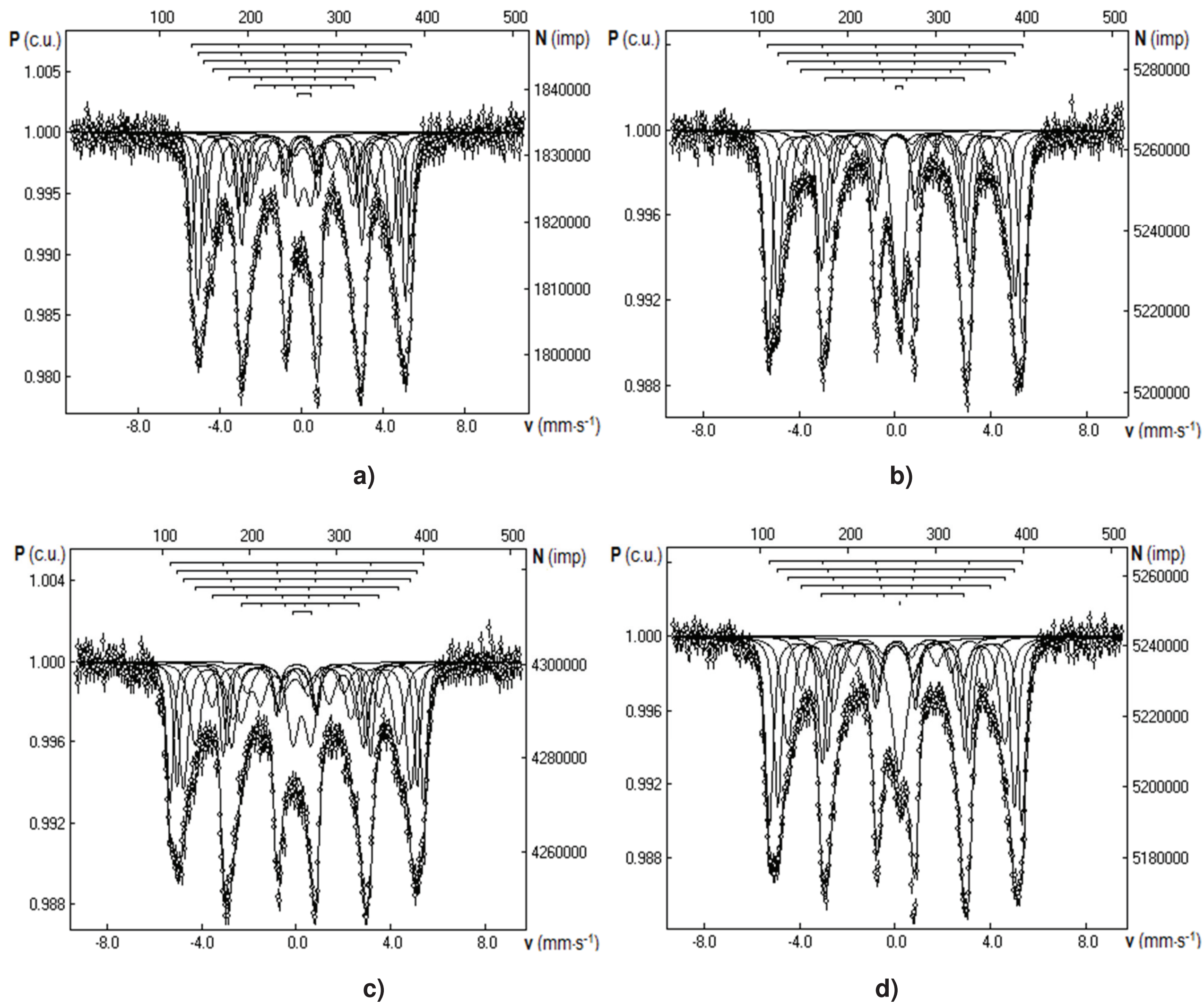


Figure 4 Mössbauer spectra of the AlNi alloy: water quenched from 1240 °C(a), cooled at a critical rate from 1240 °C to T_q = 860 °C (b), 850 °C (c) and 840 °C (d)

An approximate estimate of the ratio of the areas of the ferromagnetic and paramagnetic peaks gives about 6-8% iron in paramagnetic phase for the samples with low H_c (water quenched and cooledat a critical rate to 850 °C) and 10-12% iron in paramagnetic phase for the samples with high H_c (cooled at a critical rate to 860 °C, 840 °C and *RT*). The spectrum of the sample which was slowly cooled to T_q = 850 °C (H_c = 3.2 kA/m) is similar to that measured after quenching in water at 1240 °C (H_c = 0.32 kA/m), as well as the microstructures of these samples are similar (**Figure 2 a, c**).It is known, that due to a distinctly asymmetric formof the miscibility gap in the Fe-Ni-Al system [4], the compositions and relative amounts of β- and $β_2$-phase are very temperature dependent.Around 850 °C the fast increase of the β-phase volume due to the enrichment of the $β_2$-phase by NiAl take place. In practice, the increase of the Fe-rich β-phase may be carried out by the “secondary” decomposition of the $β_2$-phase precipitations inside them that lead to the destruction of the periodic modulated microstructure. The obtained results allow us to interpret the nonmonotonic behavior of H_c on the temperature T_q as a function of composition changes of the β- and $β_2$-phases and parameters of periodic modulated microstructure.

4. CONCLUSIONS

TEM investigations of the solid solution decomposition ($\beta_2 \rightarrow \beta + \beta_2$) in as cast Fe_2NiAl alloy during cooling at a critical rate after homogenization showed that formation of the periodic modulated structure passes through an intermediate stage of destruction, leading to a drop of coercive force after quenching from 850°C. It is shown that the "secondary" decomposition of the β_2-phase inside modulated structure, formed at higher temperatures, is associated with a sharp change in the volume and composition of the β and $\beta_2$phases due to the asymmetric shape of the miscibility gap in the Fe-Ni-Al system near 850 °C.

Cooling of the cast Fe_2NiAl alloy at a critical rate to room temperature leads to the formation of an optimal modulated structure and obtaining the maximum value of the coercive force H_c = 50.4 kA/m.

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