In-situ Observation of Transformation Behavior in High-Purity Fe–Ni Alloys

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The phase transformations from ferrite (α phase) to austenite (γ phase) and from the γ phase to martensite were experimentally examined by an *in-situ* observation technique for high-purity Fe–Ni alloy specimens with Ni concentrations between 0 and 25 mass%. The specimens were heated from room temperature to 1273 K at heating rates of 1 to 50 Ks⁻¹, held at 1273 K for 120 s, and then cooled to room temperature by furnace cooling. The cooling rate was 25 Ks⁻¹ at temperatures between 1273 and 823 K. During the heating and cooling, the microstructure of the surface was directly observed with a high-temperature optical microscope without thermal etching for each specimen. The start and finish temperatures, A_s and A_f , for the α to γ transformation during the heating slightly decrease with increasing heating rate in all the specimens. On the other hand, the start and finish temperatures, M_s and M_f , for the γ to martensite transformation as well as the A_s and A_f temperatures remarkably decrease with increasing Ni concentration of specimen.

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1. Introduction

Microstructure observation is one of the most important techniques for studies on the inherent properties of pure iron and iron-base alloys at high temperatures. It has been reported by Abiko and co-workers^{1–5)} that the behavior of phase transformation in iron considerably varies depending on the kind and amount of nonmetallic impurity like carbon and phosphorus. However, the effects of metallic impurities on the transformation behavior in iron at high temperatures have not been studied in detail. In binary Fe–Ni alloys, ferrite (α phase) is transformed to austenite (γ phase) owing to heating whereas the γ phase is transformed to martensite due to rather rapid cooling. The transformation behavior during heating and cooling in the binary Fe–Ni system has been experimentally studied by many investigators^{6–9)} using a dilatation measurement technique, an X-ray analysis technique, and so on. In these experiments, the microstructure was observed for the specimens that were annealed at high temperatures and then quenched to room temperature. Therefore, the microstructural changes during heating processes were not observed continuously and thus the transformation temperatures could not be determined with sufficient accuracy. In the present work, microstructural changes due to the phase transformations during heating and cooling in the binary Fe–Ni system were experimentally examined by an *in-situ* observation technique with a high-temperature optical microscope using high-purity Fe–Ni alloys with various compositions. The dependence of the transformation temperatures on the composition and the heating rate was evaluated on the basis of the observations on the microstructure.

2. Experimental

2.1 Specimen preparation

High-purity Fe–Ni alloys with Ni concentrations of 5, 10

and 20 mass% were prepared as ingots from electrolytic iron (MAIRON-HP) and pure nickel rods (JONSON MATTHEY) with purity of 99.99% in an ultra high vacuum furnace with a high-purity CaO crucible. On the other hand, high-purity Fe–25 mass%Ni alloy was prepared as ingot from electrolytic iron (MAIRON-HP) and pure nickel rods (JONSON MATTHEY) with purity of 99.99% in an ultra high vacuum furnace containing a copper-cold-crucible. An ultra-high-purity Fe ingot was prepared from the electrolytic iron in the ultra high vacuum furnace with a copper-cold-crucible and then purified in high-purity hydrogen gas by a floating zone refining technique. Hereafter, the ultra-high-purity Fe is called UHP-Fe whereas the high-purity Fe–Ni alloys with Ni concentrations of 5, 10, 20 and 25 mass% are designated as 5Ni, 10Ni, 20Ni, and 25Ni, respectively. The chemical compositions of UHP-Fe and Fe–Ni ingots are shown in Table 1.

2.2 In-situ observation

The Fe–Ni alloy ingots were forged, hot rolled, cold rolled and then machined to sheets with a thickness of 5.0 mm. The alloy sheets were cold rolled into thin sheet with a thickness of 0.25 mm and then mechanically polished. On the other hand, UHP-Fe disk specimens with a diameter of 10 mm and a thickness of 0.25 mm were cut from the ingot and then mechanically polished. A Pt/Pt–Rh thermocouple with a diameter of 50 μ m was spot welded at the center of a surface of each specimen. In a high-temperature optical microscope schematically shown in Fig. 1, the spot welded specimen was heated from room temperature to 1273 K at heating rates of 1, 10 and 50 Ks⁻¹ by electron bombardment, held at 1273 K for 120 s and then cooled to room temperature by furnace cooling. The cooling rate during the furnace cooling from 1273 to 823 K was 25 Ks⁻¹. All the heating processes were carried out in an ultra-high vacuum of 5.0×10^{-7} Pa without thermal etching. The transformation temperatures were determined with the thermocouple within accuracy of ± 5 K whereas the microstructure of the surface of each specimen was recorded with a digital video recorder.

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Table 1 Chemical Compositions in Ultra-High Purity Iron and High-Purity Fe–Ni Alloys.

	Alloy				
	5Ni	10Ni	20Ni	25Ni	UHPFe
Ni	5.10 mass%	9.92 mass%	20.0 mass%	24.4 mass%	<0.1 mass ppm
C	2.28	2.05	2.61	0.95	1.82
S	5.49	5.61	5.03	0.83	0.68
O	115.9	131.2	128.0	27.2	1.0
N	1.7	1.9	3.3	1.4	0.35
Al	2.5	1.8	<0.2	10	<0.1
Mn	0.15	0.26	0.06	<0.9	0.025
(mass ppm)

Fig. 1 Schematic Diagram of High-Temperature Optical Microscope.

In specimen 5Ni, the transformation from the α phase to the γ phase (α - γ transformation) occurs at temperatures between 1026 and 1053 K as shown in Figs. 3(b) and (c). On the other hand, the γ phase is transformed to massive martensite at 960 K during the cooling. The massive-martensitic transformation (γ - M^m transformation) is finished at 908 K (see Fig. 3(d)).

In the case of specimen 10Ni, the α - γ transformation occurs at 947 K as can be seen in Fig. 4(b). During the cooling, the γ - M^m transformation takes place at 823 K (see Fig. 4(c)) whereas the martensitic transformation from γ phase to lath martensite (γ - M^l transformation) starts at 817 K and finishes at 775 K. As can be seen in Fig. 4(d), the lath martensite phase is formed from the grain boundaries in the γ phase matrix.

During the heating, the α - γ transformation occurs at 818 and 755 K in specimens 20Ni and 25Ni, respectively (see Figs. 5(b) and 6(b)). On the other hand, during the cooling, the γ - M^l transformation begins at 620 and 450 K in specimens 20Ni and 25Ni, respectively, whereas it finishes at 520 and 343 K in specimens 20Ni and 25Ni, respectively (see Figs. 5(c), 5(d), 6(c) and 6(d)). The lath martensite phase is formed from the grain boundaries in the γ phase matrix also in these specimens.

3. Results and Discussion

3.1 Microstructure observation

Typical optical micrographs of specimens UHP-Fe, 5Ni, 10Ni, 20Ni and 25Ni during the heating processes at a heating rate of 1 K s^{-1} are shown in Figs. 2–6, respectively. As can be seen in Fig. 2(a), the single-phase microstructure of the α phase of the as-polished is realized for specimen UHP-Fe before the heating. In this specimen, the α phase is transformed to the γ phase at temperatures between 1185 and 1188 K (see Fig. 2(b)). In Fig. 2(c), the interface between the α and γ phases is recognized to move from the right-hand side to the left-hand side due to the transformation. During the heating and the holding, the grain size of the γ phase matrix gradually increases with increasing heating and holding times. Such grain growth of the γ phase matrix occurs in all the specimens. On the other hand, the γ phase is transformed to the α phase at temperatures between 1169 and 1166 K during the cooling and thus completely disappears at 1160 K as can be seen in Fig. 2(d).

3.2 Transformation temperature

The start and finish temperatures, A_s and A_f , for the α - γ transformation during the heating were determined for various specimens at different heating rates. The results are shown as solid and open symbols against the heating rate in Fig. 7 whereas against the Ni concentration of specimen in Fig. 8. As can be seen in these figures, the A_s and A_f temperatures slightly decrease with increasing heating rate whereas they remarkably decrease with increasing Ni concentration. Solid curves in Fig. 8 indicate the composition dependence of A_s and A_f at a heating rate of 1 K s^{-1} . Since this value of the heating rate seems sufficiently small, the solid curve for A_f may correspond to the $\gamma/(\alpha + \gamma)$ phase boundary in the equilibrium phase diagram of the binary Fe–Ni system. However, the $\gamma/(\alpha + \gamma)$ phase boundary determined by Owen and Liu⁷⁾ using an X-ray analysis technique rather corresponds to the experimental points for A_f at a heating rate of 50 K s^{-1} . On the other hand, the composition dependence of A_f experimentally determined by Pickels and Sucksmith⁸⁾ utilizing a magnetic measurement technique is fairly in agreement with

Fig. 2 Optical micrographs of surface for specimen UHP-Fe at temperatures of (a) 290, (b) 1187, (c) 1233 and (d) 1160 K during the heating and cooling.

Fig. 3 Optical micrographs of surface for specimen 5Ni at temperatures of (a) 306, (b) 1035, (c) 1052 and (d) 901 K during the heating and cooling.

that at a heating rate of 10 K s^{-1} . In the experiments by Owen and Liu and by Pickels and Sucksmith, the $\gamma/(\alpha + \gamma)$ phase boundary was estimated on the basis of the information for quenched binary Fe–Ni alloys with lower purity. On the other hand, the A_f temperature was directly determined by an *in-*

situ observation technique in the present work. Such differences in the purity and the observation technique may be the reasons why the experimental results by Owen and Liu and by Pickels and Sucksmith provide lower values for A_f .

The 10 and 90% transformed temperatures, A_{10} and A_{90} ,

Fig. 4 Optical micrographs of surface for specimen 10Ni at temperatures of (a) 303, (b) 979, (c) 825 and (d) 780 K during the heating and cooling.

Fig. 5 Optical micrographs of surface for specimen 20Ni at temperatures of (a) 297, (b) 869, (c) 564 and (d) 528 K during the heating and cooling.

of the α - γ transformation during the heating at a heating rate of 0.17 K s^{-1} were experimentally determined by Jones and Pumphrey⁹⁾ using a dilatation measurement technique for binary Fe–Ni alloy specimens with different compositions. Their results for A_{10} and A_{90} are shown against the Ni concentration of specimen as solid and open triangles, respectively,

in Fig. 9. In this figure, the experimental results for A_s and A_f at a heating rate of 1 K s^{-1} in Fig. 8 are also represented. As can be seen in Fig. 9, the experimental points for A_{90} rather well line on the solid curve for A_f whereas those for A_{10} are located above the solid curve for A_s . The difference between A_{10} and A_s reaches the maximum value of about 50 K at a Ni

Fig. 6 Optical micrographs of surface for specimen 25Ni at temperature of (a) 297, (b) 770, (c) 414 and (d) 362 K during the heating and cooling.

Fig. 7 The A_s and A_f temperatures versus the heating rate for various specimens.

concentration of 19 mass%.

The start temperatures, M_s^m and M_s^l , of the γ - M^m and γ - M^l transformations, respectively, and the finish temperature M_f of these transformations are shown against the Ni concentration of specimen at various heating rates in Fig. 10. In specimen 5Ni, only the γ - M^m transformation occurs at $M_s^m = 954$ K and $M_f = 915$ K during the cooling. On the other hand, only the γ - M^l transformation takes place in specimens 20Ni and 25Ni. As mentioned earlier, $M_s^l = 620$ K and $M_f = 520$ K for specimen 20Ni whereas $M_s^l = 450$ K and $M_f = 343$ K for 25Ni. In the case of specimen 10Ni, however, both the γ - M^m and γ - M^l transformations occur during the cooling. M_s^m , M_s^l and M_f are 832, 817 and 775 K, respec-

Fig. 8 The A_s and A_f temperatures versus the Ni concentration of specimen at various heating rates.

tively, for this specimen. As can be seen in Fig. 10, the values of M_s^m , M_s^l and M_f remarkably decrease with increasing Ni concentration.

Fig. 9 The A_{10} and A_{90} temperature versus the Ni concentration of specimen experimentally determined by Jones and Pumphrey.⁹⁾ The A_s and A_f temperatures at a heating rate of 1 Ks^{-1} in Fig. 8 are also represented.

Fig. 10 The M_s^m , M_s^l and M_f temperature versus the Ni concentration of specimen at various heating rates.

4. Conclusions

The microstructural changes owing to the α - γ , γ - M^m and γ - M^l transformations were experimentally examined by an *in-situ* observation technique using pure Fe specimens with ultra-high purity and binary Fe–Ni specimens with high purity. The Fe and Fe–Ni specimens were heated from room temperature to 1273 K at heating rates between 1 and 50 Ks^{-1} , held for 120 s at 1273 K, and then cooled to room temperature. The cooling rate at temperatures between 1273 and 823 K was 25 Ks^{-1} . On the basis of the *in-situ* observations, the A_s and A_f temperatures for the α - γ transformation during the heating and the M_s^m , M_s^l and M_f temperatures for the γ - M^m and γ - M^l martensitic transformations during the cooling were determined for various specimens. The values of A_s and A_f slightly decrease with increasing heating rate for each specimen whereas those of A_s , A_f , M_s^m , M_s^l , M_f remarkably decrease with increasing Ni concentration under the present experimental conditions.

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REFERENCES

- 1) K. Abiko: *Proc. Internat. Conf. Grain Boundary Structure and Related Phenomena*, JIMS-4, The Japan Institute of Metals, (1986), 633–637.
- 2) K. Abiko: *Proc. Internat. Conf. Physical Metallurgy of Thermomechanical Processing of Steels and Other Metals*, The Iron and Steel Institute of Japan, (1988), 307–314.
- 3) K. Sadamori and K. Abiko: *Proc. Internat. Conf. Recrystallization in Metallic Metals*, TMS, (1991), 491–496.
- 4) K. Sadamori and K. Abiko: *Ultra High Purity Base Metals (UHPM-94)*, The Japan Institute of Metals, (1994), 507–511.
- 5) K. Abiko and K. Sadamori: *Ultra High Purity Base Metals (UHPM-97)*, Phys. Stat. Sol., (1997), 275–287.
- 6) E. A. Wilson: *Metal Science*, **18** (1984), 471–484.
- 7) E. A. Owen and Y. H. Liu: *J. Iron Steel Inst.*, **163** (1949), 132–137.
- 8) A. T. Pickels and W. Sucksmith: *Proc. Roy. Soc. (London)*, **A175** (1940), 331–344.
- 9) F. W. Jones and W. I. Pumphrey: *J. Iron Steel Inst.*, **163** (1949), 121–131.