High Magnetic Permeability Alloys Hardperm in the Ni-Fe-Ta-Mo System*

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Ni-Fe-Ta-Mo alloys were heated in hydrogen atmosphere at 1150°C for 3 hr and then cooled at rates of 1400~0.75°C/hr from a temperature above their order-disorder transformation points. The highest initial permeability is 120000 measured on the alloy of 74.35%Ni, 11.35%Fe, 13.85%Ta and 0.45%Mo cooled at the rate of 300°C/hr, and the highest maximum permeability is 610000 on the alloy of 74.20%Ni, 11.22%Fe, 13.96%Ta and 0.62%Mo cooled at 240°C/hr. The alloy with the highest initial permeability exhibited an electrical resistivity of 69.7 $\mu\Omega$ -cm, with Vickers hardness 199. The high permeability of the Ni-Fe-Ta-Mo alloys is largely due to the effective contributions of high-temperature heat treatments in hydrogen atmosphere to the compositional homogeneity, impurity removal, together with the low values of magnetostriction and crystal magnetic anisotropy in a proper state of ordering.

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I. Introduction

Our previous studies have revealed that marked improvements of magnetic properties and considerable increases of specific electrical resistivity and hardness can be achieved in the Ni-Fe alloy system by the addition of niobium or tantalum in group V of the periodic table to the alloys which are in the composition range having the superlattice $\text{Ni}_3\text{Fe}^{(1)(2)}$.

The purpose of this report is to describe the effect of addition of molybdenum in group VI of the periodic table on magnetic and electrical properties of Ni-Fe-Ta alloys.

II. Experimental

Starting materials used were 99.8% pure nickel, 99.9% pure iron, 99.9% pure tantalum, and 99.9% pure molybdenum. The materials in a total weight of 500 g were melted with a high-frequency induction furnace in vacuum.

The melt was poured into an iron mold with an opening of 20 mm diameter and 150 mm length. After forging, the ingot was reduced to a thin plate 0.3 mm in thickness by rolling and a wire 2 mm in diameter by swaging and drawing. A ring-shaped plate 33 mm in inner diameter and 45 mm in outer diameter was stamped out from the plate. A rod 100 mm long was cut from the wire. Both the ring-shaped plate and the rod were heated at 1150°C for 3 hr under a hydrogen atmosphere with a dew point of -70°C, and then these were cooled down to room temperature from a temperature above their order-disorder transformation points at rates of 1400~0.75°C/hr. Fifty-eight alloys with compositions of 71.12~79.24%Ni, 8.80~15.77%Fe, 5.72~16.63%Ta and 0.21~4.39%Mo were used as specimens in this investigation. The magnetic properties were measured using a ballistic galvanometer and the recording fluxmeter; the magnetostriction by the optical roller method; the electrical resistivity by a potentiometer; and the hardness by a micro-Vickers hardness tester.

III. Results and Discussion

The experimental data obtained in this investigation are shown in Table 1 and Figs. 1~7. Figures 1 and 2 show the cooling rate dependence of initial permeability μ_0 and

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Table 1 Magnetic properties, electrical resistivity and Vickers hardness of high permeability Ni-Fe-Ta-Mo alloys heated at 1150°C for 3 hr.

Alloy No.	Composition (Rest Ni, wt %)			Cooling rate (°C/hr)	μ_0	μ_1	μ_m	$\frac{W_h}{\text{cycle}}$ (erg/cm ³)	B_r (kG)	H_c (mOe)	$4\pi I_s$ (kG)	λ_s ($\times 10^{-6}$)	ρ ($\mu\Omega\text{-cm}$, 20°C)	Hv (Vickers hardness)
	Fe	Ta	Mo		($\times 10^3$)				$B_m=5$ kG					
1	12.14	12.88	0.47	300	56	69	356	8.4	3.4	5.6	6.7	0.4	65.4	194
4	11.46	13.91	0.34	400	102	120	354	5.4	3.0	4.1	6.4	0.5	67.7	197
4	11.46	13.91	0.34	300	72	111	500	5.2	3.2	3.2	6.4	0.7	67.8	—
6	11.35	13.85	0.45	300	120	163	560	3.6	2.9	3.1	6.3	0	69.7	199
8	11.22	13.96	0.62	240	104	126	610	4.0	3.2	3.4	6.1	0.4	71.2	198
10	10.75	14.95	0.45	300	20	30	365	11.0	4.0	6.2	6.0	0.4	71.7	208
10	10.75	14.95	0.45	240	46	52	234	9.2	2.8	6.3	6.1	0.2	72.0	—
12	11.30	12.18	0.94	240	58	72	297	8.7	3.1	5.3	6.7	1.1	66.1	191
12	11.30	12.18	0.94	150	43	59	347	9.5	3.6	5.7	6.6	0.4	66.7	—
14	11.65	12.79	0.97	150	90	118	428	6.8	3.0	4.0	6.3	0	70.2	196
18	10.90	14.28	0.94	150	46	68	416	7.6	3.5	4.2	6.0	0.4	73.6	201
18	10.90	14.28	0.94	120	69	83	295	7.7	2.7	5.6	6.1	0.2	73.9	—
20	11.05	14.87	0.96	100	16	20	230	8.0	2.9	5.9	5.8	0.7	78.6	219
22	11.57	12.01	1.47	70	101	134	433	5.7	3.0	4.1	6.3	0.7	69.8	190
24	10.95	12.90	1.48	50	95	129	436	5.0	2.9	3.7	6.1	1.1	73.1	198
27	11.01	13.76	1.44	50	31	36	194	12.9	3.2	9.4	5.8	0.5	77.5	203
33	13.42	7.96	1.95	70	23	27	204	19.1	4.0	10.2	7.5	1.4	61.3	166
34	11.30	10.70	1.92	50	49	58	212	11.2	3.1	8.0	6.6	0.2	67.1	181
36	10.50	12.06	1.86	30	63	98	416	6.5	2.6	3.6	6.0	0.5	73.7	187
36	10.90	12.06	1.86	20	92	128	390	6.5	2.7	3.6	6.0	1.1	73.9	—
39	11.21	12.75	1.90	30	40	46	364	9.2	3.2	5.1	5.9	0.2	77.5	185
41	10.51	13.87	1.85	25	20	23	170	18.9	3.1	11.1	5.6	0.5	80.2	197
42	11.10	13.94	1.86	50	21	23	142	16.6	3.0	10.8	5.8	0.7	82.1	196
46	10.54	12.77	2.38	30	11	14	199	19.3	3.2	11.0	5.3	0.5	79.1	190
48	10.04	13.87	2.38	20	10	12	154	*21.7	*3.0	*12.0	5.2	0.5	84.5	197
51	12.29	7.81	3.01	15	50	58	201	12.0	3.2	8.6	6.7	0.7	68.7	163
53	10.85	9.98	2.98	5	103	140	390	5.1	2.6	3.6	5.9	0.2	74.4	181
56	10.54	11.94	2.92	10	14	19	281	*11.9	*3.6	*7.3	5.3	0.4	82.2	187
57	11.47	7.38	3.92	1.5	61	73	284	8.1	3.0	5.8	6.2	0.4	76.1	168

* $B_m = 4.5 \text{ kG}$

maximum permeability μ_m and of coercive force H_c and residual magnetic induction B_r , respectively, for alloys No. 6, No. 8, No. 14, No. 22, No. 36 and No. 53 in the Ni-Fe-Ta-Mo system. As can be seen from the figures, the values of μ_0 and μ_m depend strongly on cooling rate and indicate a maximum, respectively, which corresponds approximately to minima in B_r and H_c . The appearance of these minima and maxima suggests that the most easily magnetizable state of ordering was produced in the alloys by cooling at an optimum rate in a way which is similar to Ni-Fe⁽³⁾ and Ni-Fe-Ta alloys⁽²⁾. The effect of molybdenum content on the optimum cooling rate is given in Fig. 3 for Ni-Fe-Ta-Mo alloys with 73.71~74.35%

nickel and 13.76~14.28% tantalum. The tendency that the optimum cooling rate becomes slower with increase in molybdenum content has been similarly observed in the case of additions of chromium, molybdenum, copper, tantalum and niobium to Ni-Fe alloys^{(1)(2)(4)~(6)}.

In Fig. 4 the highest values of μ_0 and μ_m obtained for two series of the alloys containing, 74.14~74.67 and 73.71~74.35% nickel and (b), 12.75~12.90 and 13.76~14.28% tantalum, respectively, are plotted as a function of molybdenum content. In all cases, both μ_0 and μ_m indicate a maximum against molybdenum contents. Th maximum is shifted to the composition of lower molybdenum contents

Fig. 1 Variations in initial permeability μ_0 and maximum permeability μ_m with cooling rate for some high permeability Ni-Fe-Ta-Mo alloys. The alloy compositions were: Alloy No. 6 (74.35%Ni, 11.35%Fe, 13.85%Ta, 0.45%Mo), No. 8 (74.20%Ni, 11.22%Fe, 13.96%Ta, 0.62%Mo), No. 14 (74.59%Ni, 11.65%Fe, 12.79%Ta, 0.97%Mo), No. 22 (74.95%Ni, 11.57%Fe, 12.01%Ta, 1.47%Mo), No. 36 (75.58%Ni, 10.50%Fe, 12.06%Ta, 1.86%Mo) and No. 53 (75.82%Ni, 11.10%Fe, 10.20%Ta, 2.88%Mo).

Fig. 2 Variations in coercive force H_c and residual magnetic induction B_r with cooling rate for the same alloys as those in Fig. 1.

with an increase in tantalum content; in general, it appears at a composition in which the tantalum content is replaced by the molyb-

Fig. 3 Optimum cooling rate as function of Mo contents for Ni-Fe-Ta-Mo quaternary alloys containing 73.71~74.35%Ni and 13.76~14.28%Ta.

Fig. 4 Effect of Mo on highest initial permeability μ_0 and maximum permeability μ_m for each of Ni-Fe-Ta-Mo alloys containing (a), 74.14~74.67%Ni and 12.75~12.90%Ta and (b), 73.71~74.35%Ni and 13.76~14.28%Ta.

denum content. Figures 5 and 6 show the effects of molybdenum content on the values of H_c and B_r , which correspond to the highest values of μ_m obtained by the heat treatment, the values of the magnetic induction $4\pi I_s$ in an effective field of 1 kOe and the electrical resistivity ρ for the same alloys given in Fig. 4. The H_c and B_r exhibit their maxima which correspond approximately to the highest values of μ_0 and μ_m ; the $4\pi I_s$ decreases monotonically with an increase in molybdenum content, while the ρ shows a rapid increase

Fig. 5 Effect of Mo on coercive force H_c and residual magnetic induction B_r of the same alloys as those in Fig. 4.

Fig. 6 Effect of Mo on magnetic induction $4\pi I_s$ and electrical resistivity ρ of the same alloys those as in Fig. 4.

linearly.

In Table 1 the experimental data on the magnetic properties, electrical resistivity and hardness are listed for 24 typical alloys in the Ni-Fe-Ta-Mo system investigated, where μ_1 is the permeability in a field of 1 mOe, W_h the hysteresis loss against a maximum magnetic induction of 5 kG, λ_s a magnetostriction constant in an effective field of 1 kOe, and Hv the

Vickers hardness under a load of 500 g. The highest value of μ_0 obtained in this alloy system is 120000 measured on alloy No. 6 (74.35%Ni, 11.35%Fe, 13.85%Ta, 0.45%Mo) subjected to cooling at 300°C/hr, whereas the highest value of μ_m is 610000 on alloy No. 8 (74.20%Ni, 11.22%Fe, 13.96%Ta, 0.62%Mo) cooled at 240°C/hr. These experimental values are extremely high. In addition, the measured values of ρ and Hv for alloy No. 6 are as high as 69.7 $\mu\Omega$ -cm and 199, respectively. In consideration of their characteristics, these alloys have been named Hardperm by the present authors. The fact that the values of ρ in alloys No. 4, No. 10, No. 12, No. 18 and No. 36 become a little larger at a slow cooling rate than those at a rapid cooling rate has been thought to be due to the formation of short-range order as predicted by Nagashima et al.⁽⁷⁾ for Ni-Fe-Mo alloys. Figure 7 represents the hysteresis curves for alloy No. 6 and No. 8, where the H_c and W_h of alloy No. 6 take very small values, 3.1 mOe and 3.6 erg/cm³/cycle, respectively.

The origins of the high permeability and excellent magnetic properties in the Ni-Fe-Ta-Mo alloys ascribed chiefly to the compositional homogeneity, removal of impurities such as carbon, nitrogen and oxygen and coarsening of grains by high-temperature heat treatments in purified hydrogen⁽⁸⁾⁽⁹⁾, together with the

Fig. 7 Hysteresis curves of alloy No. 6 (74.35%Ni, 11.35%Fe, 13.85%Ta, 0.45%Mo) cooled at the rate of 300°C/hr and No. 8 (74.20%Ni, 11.22%Fe, 13.96%Ta, 0.62%Mo) cooled at 240°C/hr.

very low values of magnetostriction constants (cf. Table 1) and of crystal magnetic anisotropy constants⁽¹⁰⁾⁽¹¹⁾ in a proper state of ordering produced by use of an optimum cooling rate.

IV. Conclusions

Alloys in the composition range of 71.12~79.24%Ni, 8.80~15.77%Fe, 5.73~16.63%Ta and 0.21~4.39%Mo were vacuum melted, forged and then reduced to ring-shaped plates by rolling and rods by swaging and drawing. The plates and rods were heated at 1150°C for 3 hr under a hydrogen atmosphere with a dew point of -70°C and cooled down to room temperature from a temperature above the order-disorder transformation points at rates of 1400~0.75°C/hr. Measurements of the magnetic properties, electrical resistivity and hardness of the specimens thus prepared were carried out in this investigation.

(1) In general, the initial permeability μ_0 and the maximum permeability μ_m of Ni-Fe-Ta-Mo alloys are dependent upon cooling rate and exhibit a maximum, respectively; the optimum cooling rate at which to obtain maxima in μ_0 and μ_m becomes gradually slower with increasing molybdenum content. The highest value of μ_0 obtained in this quaternary alloy system is 120000 measured on alloy No. 6 (74.35%Ni, 11.35%Fe, 13.85%Ta, 0.45%Mo) cooled at 300°C/hr, while the highest value of μ_m is 610000 on alloy No. 8 (74.20%Ni, 11.22%Fe, 13.96%Ta, 0.62%Mo) cooled at 240°C/hr.

(2) The coercive force and hysteresis loss of the alloys against a maximum magnetic induction of 5 kG are very small. With increasing molybdenum content, the magnetic induction in an effective field of 1 kOe decreases and the electrical resistivity increases rapidly. These values for alloy No. 6 subjected to cooling at 300°C/hr are 3.1 mOe, 3.6 erg/cm³/cycle, 6.3 kG, and 69.7 $\mu\Omega$ -cm, respectively.

(3) The alloys in the Ni-Fe-Ta-Mo system have very high permeability and high values of hardness, so that these have been named Hardperm.

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