The Effect of Aluminium Additions on Characteristic Properties of New High Magnetic Permeability Alloy "Nimalloy" in the System of Nickel and Manganese*

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Kobayashi and two of the present authors discovered in a previous work that nickel-manganese alloys exhibit high permeability in the concentration of less than about 24% manganese, and named them Nimalloy. Much subsequent work has been carried out on the effects of additions of various elements on the properties of nickel-manganese alloys and obtained the highest initial permeability of 76000 and the highest maximum permeability of 441000. In the present paper, the effect of aluminium additions on the characteristic properties of nickel-manganese alloys have been studied. The results show that the optimum cooling rate for attainment of the highest permeability in each alloy increases in general with increasing aluminium content. Thus the alloy containing 78.95% Ni, 19.85% Mn and 1.20% Al showed the highest initial permeability of 10780 when cooled at a rate of 400°C/hr from 900°C and the alloy containing 78.81% Ni, 19.56% Mn and 1.63% Al exhibited the highest maximum permeability of 73200 when cooled at a rate of 800°C/hr from the same temperature; the latter shows a magnetic hysteresis loss of 9.49 erg/cm³/cycle and a coercive force of 0.0121 Oe for the maximum magnetic induction of 2000 G, and a specific electrical resistivity of 61.0μΩ-cm at 20°C.

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I. Introduction

Kobayashi and two of the present authors⁽¹⁾ discovered in an earlier work that high permeability can be obtained in Ni-Mn alloys by properly adjusting the degree of order of the ferromagnetic superlattice Ni₃Mn, and termed these high permeability alloys "Nimalloy". Much subsequent work has since been conducted by Masumoto, Murakami and others on the effects of additions of various elements to Ni-Mn alloys on

their characteristic properties. At the biannual Meetings of the Japan Institute of Metals, the results of experiments have been presented for the cases of additions of Fe⁽²⁾, V⁽³⁾, Si⁽⁴⁾, Ti⁽⁵⁾, Mo⁽⁵⁾, Cr⁽⁶⁾, Al⁽⁶⁾, Sb⁽⁷⁾, Sn⁽⁷⁾, W⁽⁸⁾, Co⁽⁸⁾, Cu⁽⁸⁾, Ge⁽⁹⁾ and Nb⁽⁹⁾ to Ni-Mn alloys as third elements and additions of Cr⁽¹⁰⁾ and Mo⁽¹¹⁾ to Ni-Mn-Fe alloys as fourth elements. The results of measurements of magnetostriction λ_s ⁽¹²⁾~⁽¹⁴⁾ and crystal magnetic anisotropy energy K_1 ⁽¹⁵⁾~⁽¹⁶⁾

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- (1) H. Masumoto, T. Kobayashi and Y. Murakami : Presented at the Spring Meeting of the Japan Institute of Metals, Apr. 5, 1963, Tokyo, Japan; J. Japan Inst. Metals, **29** (1965), 228; Trans. JIM, **8** (1967), 49.
- (2) H. Masumoto, T. Kobayashi and Y. Murakami : Presented at the Spring Meeting of the Japan Institute of Metals, Apr. 5, 1963, Tokyo, Japan; J. Japan Inst. Metals, **29** (1965), 1101; Trans. JIM, **8** (1967), 54.
- (3) H. Masumoto, T. Kobayashi and Y. Murakami : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 19, 1963, Nagoya, Japan; J. Japan Inst. Metals, **30** (1966), 837.
- (4) H. Masumoto, T. Kobayashi and Y. Murakami : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 19, 1963, Nagoya, Japan; J. Japan Inst. Metals, **31** (1967), 1117.
- (5) H. Masumoto, T. Kobayashi and Y. Murakami : Presented at the Spring Meeting of the Japan Institute of Metals, Apr. 5, 1964, Tokyo, Japan.
- (6) H. Masumoto, Y. Murakami and M. Hinai : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 2, 1964, Toyama, Japan.

- (7) H. Masumoto, Y. Murakami and M. Hinai : Presented at the Spring Meeting of the Japan Institute of Metals, Apr. 6, 1965, Tokyo, Japan.
- (8) H. Masumoto, Y. Murakami and M. Hinai : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 18, 1965, Fukuoka, Japan.
- (9) H. Masumoto, Y. Murakami and M. Hinai : Presented at the Spring Meeting of the Japan Institute of Metals, Apr. 6, 1966, Tokyo, Japan.
- (10) H. Masumoto, Y. Murakami and M. Hinai : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 17, 1966, Amagasaki, Japan.
- (11) H. Masumoto, Y. Murakami and M. Hinai : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 1, 1967, Sapporo, Japan.
- (12) H. Masumoto, T. Kobayashi and S. Kadowaki : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 19, 1963, Nagoya, Japan.
- (13) H. Masumoto, Y. Murakami and S. Kadowaki : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 2, 1964, Toyama, Japan.
- (14) H. Masumoto, Y. Murakami and S. Kadowaki : Presented at the Spring Meeting of the Japan Institute of Metals, Apr. 6, 1965, Tokyo, Japan.
- (15) H. Masumoto, Y. Murakami and N. Nakamura : Presented at the Spring Meeting of the Japan Institute of Metals, Apr. 6, 1966, Tokyo, Japan.
- (16) H. Masumoto, Y. Murakami and N. Nakamura : Presented at the Autumn Meeting of the Japan Institute of Metals, Oct. 17, 1966, Amagasaki, Japan.

which have close bearing upon permeability have also been reported by Masumoto and others on some of these alloys at the JIM's Meetings. Among them, the cases of additions of Fe⁽²⁾, V⁽³⁾ and Si⁽⁴⁾ have already been reported in previous publications.

The highest values of initial permeability and maximum permeability obtained so far are as high as 76000 and 441000, respectively. In this paper the results obtained by additions of aluminium to Nimalloy are described.

II. Specimens and Experimental Procedure

Alloying materials used were electrolytic nickel, electrolytic manganese and aluminium, of which the former two were entirely the same as those used in a previous work⁽¹⁾ and the aluminium was 99.82% in purity. The methods of specimen preparation, heat treatments and measurements will be omitted here because of their similarity to those reported already⁽¹⁾. The specimens employed were 89 alloys with different compositions of 75.87~98.36% nickel, 0~22.75% manganese and 0.56~5.42% aluminium.

III. Results and Discussion

The experimental results are summarized in Table I and Figs. 1~8. The specimens with the compositions denoted by the closed circles were at first heated in vacuum at 900°C for 1 hr and then cooled to room temperature at ten different cooling rates of 9°C/sec, 2800°C/hr, 1400°C/hr, 800°C/hr, 400°C/hr, 240°C/hr, 100°C/hr, 50°C/hr, 10°C/hr and 5°C/hr. The equivalent curves in Fig. 1 and Fig. 2 are statistically drawn from the highest values of initial permeability

μ_0 and maximum permeability μ_m against composition, obtained for each specimen. As can be seen from the figures, with increasing aluminium content, μ_0 and μ_m generally increase comparatively rapidly and attain maximum values in the manganese-rich region, and then decrease gradually. In general, a series of maximum values runs nearly parallel to a composition line that connects Ni₃Mn and Ni₃(Mn, Al). The highest values of 10780 in μ_0 and 73200 in μ_m have been obtained in the Ni-Mn-Al system with the composition of 78.95% Ni, 19.85% Mn and 1.20% Al and of 78.81% Ni, 19.56% Mn and 1.63% Al, respectively. These values are by far larger than the highest values of 6860 in μ_0 and 20400 in μ_m of Ni-Mn binary alloys.

Magnetic and electrical properties of the typical alloys in the Ni-Mn-Al system are tabulated in Table I, where W_h is the hysteresis loss, and σ is the specific electrical resistivity and α is its temperature coefficient. As shown in the table, the addition of aluminium to Ni-Mn binary alloys improves magnetic properties of the alloys, with larger $4\pi I$ but with a little change in σ . Moreover, only the operation of slow cooling from high temperatures suffices to develop excellent magnetic properties of Ni-Mn-Al ternary alloys, and the application of re-heating, as in the case for Ni-Mn binary alloys, at temperatures below the order-disorder transformation point is not necessary. This is a great advantage from the point of view of practical applications.

Figs. 3 and 4 show the measured values of μ_0 and μ_m against the manganese content for Ni-Mn-Al alloys with the composition near 1.6% aluminium, after cooling to room temperature from 900°C at various cooling rates. Both values of μ_0 and μ_m are found generally to have very large peaks.

Figs. 5 and 6 illustrate the alloy composition

Table I The magnetic and electrical properties of typical Ni-Mn-Al alloys

Alloy No.	Mn (%)	Al (%)	Cooling rate from 900°C (°C/hr)	μ_0	μ_m	$B(G)$ for μ_m	$\frac{W_h}{(\text{erg/cm}^3/\text{cycle})}$	B_r (G)	H_c (Oe)	$4\pi I$ (G)	λ_s ($\times 10^{-6}$)	σ ($\mu\Omega\text{-cm}$, 20°C)	α ($\times 10^{-3}$, 0°~40°C)	Density (g/cm ³)
							$B_m = 2000\text{G}$			$H = 900\text{Oe}$				
33	21.63	—	10 (further heated 380°C, 50 hr)	6860	18000	891	18.30	951	0.0315	2790	—	60.7	0.93	—
35	21.80	—	5	5090	12490	626	18.50	876	0.0272	2760	+0.17	60.5	0.94	8.42
37	22.00	—	10 (further heated 380°C, 50 hr)	5880	20400	908	19.34	1052	0.0270	2804	0.00	60.7	0.93	—
7	20.83	0.71	50	10180	26500	494	* 3.30	* 499	* 0.0133	2602	—	63.9	0.79	—
14	19.85	1.20	400	6370	40000	534	* 1.31	* 833	* 0.0074	2879	+0.17	62.8	0.85	8.30
"	"	"	100	10780	32700	761	5.54	647	0.0137	4165	-0.52	56.7	1.25	—
15	20.35	1.21	400	3000	60000	801	* 3.09	* 871	* 0.0110	2735	+0.17	64.0	0.77	8.28
"	"	"	100	8330	30000	1067	12.95	711	0.0287	4576	—	55.1	1.62	—
19	19.65	1.47	1400	2940	53000	801	* 3.16	* 871	* 0.0108	3274	—	62.9	0.85	—
35	18.36	1.62	100	3430	24300	2510	19.17	1725	0.0330	—	—	—	—	—
36	18.87	1.60	1400	4660	42000	748	* 3.12	* 764	* 0.0110	3760	+0.17	61.8	1.04	8.26
37	19.16	1.64	1400	5150	65400	988	* 3.17	* 935	* 0.0106	3624	—	62.7	0.87	8.24
39	19.56	1.63	800	7350	73200	1042	9.49	1370	0.0121	3590	+0.17	61.0	1.12	8.24
"	"	"	400	10210	41200	1282	9.68	1310	0.0195	4352	-0.17	56.4	1.25	—
40	19.85	1.64	800	3700	50000	1468	14.67	1725	0.0232	3863	+0.35	60.2	1.00	8.23
44	17.97	1.84	240	4410	33000	1470	14.22	1574	0.0249	4705	0.00	57.6	1.10	8.24
48	16.35	2.11	100	1500	21800	2136	19.77	1645	0.0260	—	—	—	—	—
50	17.86	2.13	1400	5880	24000	854	19.78	881	0.0235	—	—	—	—	—
51	18.86	2.13	2800	2450	37300	1096	12.13	1362	0.0203	3567	+0.17	61.1	0.99	8.23
52	16.90	2.19	800	3430	21700	1254	19.07	1366	0.0385	3955	-0.52	57.9	1.39	8.28

* For $B_m=1000 G$.

Fig. 1 Highest initial permeability in each of Ni-Mn-Al alloys cooled at various rates after heating at 900°C for 1 hr

Fig. 2 Highest maximum permeability in each of Ni-Mn-Al alloys cooled at various rates after heating at 900°C for 1 hr

Fig. 3 Initial permeability of Ni-Mn-Al (about 1.6%) alloys cooled at various rates after heating at 900°C for 1 hr

Fig. 4 Maximum permeability of Ni-Mn-Al (about 1.6%) alloys cooled at various rates after heating at 900°C for 1 hr

dependence of intrinsic magnetic induction $4\pi I$ and the specific electrical resistivity σ which corresponds to the highest μ_m of each specimen obtained by heat treatment, respectively. Fig. 7 shows the same relationship for the ternary alloys with a constant content of near 1.6% aluminium. As can be seen from these figures, $4\pi I$ is large and σ is small near the composition $Ni_3(Mn, Al)$, indicating that it is in an ordered state in some extent. Lastly, magnetization and hysteresis curves of No. 39 alloy having the highest maximum permeability are shown in Fig. 8. Although it has been well known that high-permeability alloys have small values of saturation magnetostriction λ_s , it is similarly shown that Ni-Mn-Al alloys of high permeability are extremely small in λ_s , as illustrated in Table 1.

In short, as in other high-permeability alloys, high permeability is attained in Ni-Mn-Al alloys by way of reducing λ_s through a proper adjustment of the degree of order by heat treatment.

IV. Conclusions

Ring-form specimens were prepared from 89 alloys of the ferromagnetic Ni-Mn-Al system containing 0~22.75% Mn and 0.56~5.42% Al. After heating at 900°C for 1 hr, the specimens were cooled to room temperature at various cooling rates. The results of measurements of the magnetic properties, specific electrical resistivity and its temperature coefficient are summarized as follows:

(1) In the case of aluminium additions to Ni-Mn

Fig. 5 Intrinsic magnetic induction at an effective field of 900 Oe in Ni-Mn-Al alloys heat-treated to develop the highest maximum permeability

Fig. 6 Specific electrical resistivity at 20°C in Ni-Mn-Al alloys heat-treated to develop the highest maximum permeability

Fig. 7 Intrinsic magnetic induction and specific electrical resistivity at 20°C in Ni-Mn-Al (about 1.6%) alloys heat-treated to develop the highest maximum permeability for each of them

Fig. 8 Magnetization and hysteresis curves of No. 39 alloy

alloys, the cooling rate required for the attainment of the highest permeability increases gradually with increasing aluminium content.

(2) With increasing aluminium content, the highest values of μ_0 and μ_m obtained for each specimen increase to the maxima and then decrease gradually. The highest values of 10780 in μ_0 and 73200 in μ_m have been obtained in the following Ni-Mn-Al alloys: For μ_0 , composition—78.95% Ni, 19.85% Mn, 1.20% Al, cooling rate—400°C/hr; For μ_m , composition—78.81% Ni, 19.56% Mn, 1.63% Al, cooling rate—800°C/hr.

(3) The Ni-Mn-Al alloy of highest initial perme-

ability with the composition stated above shows the smallest value of 5.54 erg/cm³/cycle in hysteresis loss, while the smallest value of 0.0121 Oe in coercive force against the maximum magnetic induction of 2000 G is obtained from the alloy of highest maximum permeability in the Ni-Mn-Al system.

(4) The specific electrical resistivity is generally large. The Ni-Mn-Al alloy of highest maximum permeability shows a value of 61.0 $\mu\Omega$ -cm (20°C), with a temperature coefficient of 1.12×10^{-3} (0°~40°C).

(5) These alloys can more easily be forged and rolled at room and high temperatures than Permalloy.