Effect of Tantalum Additions on the Properties of New High Magnetic Permeability Alloys "Nimalloy" in the Nickel and Manganese System*

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Masumoto et al. have previously discovered that alloys in composition near to Ni_3Mn exhibited a high permeability when developed to an optimum degree of order, and these alloys were named Nimalloy. In the subsequent work on the effect of various additives on the properties of Ni-Mn alloys, the highest initial permeability of 121000 and the highest maximum permeability of 643000 have been obtained with Ni-Mn-Fe-Cr-Mo alloys.

The results of Ta additions to Ni-Mn alloys show that with increasing Ta content, the permeability of Ni-Mn alloys at first increases gradually, goes through a maximum rapidly, and then decreases gradually. The highest initial permeability of 20470 is obtained on the alloy composed of 75.88% Ni, 19.47% Mn and 4.65% Ta when cooled at a rate of 240°C/hr from 900°C and baked at 440°C for 64 hr, and the highest maximum permeability of 84800 on the alloy of 75.37% Ni, 19.08% Mn and 5.55% Ta when cooled at a rate of 240°C/hr from 900°C and baked at 440°C for 65 hr. The latter alloy shows a coercive force of 0.0253 Oe and a magnetic hysteresis loss of $50.85 \text{ erg/cm}^3/\text{cycle}$ for the maximum magnetic induction of 5000 G, and its electrical resistivity being $52.7 \mu\Omega\text{-cm}$ at 20°C .

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I. Introduction

It has previously been found by Masumoto et al.⁽¹⁾ that the ferromagnetic superlattice structure Ni_3Mn in the Ni-Mn system exhibits a high magnetic permeability when it is in an appropriate degree of order, and the high permeability Ni-Mn alloys have been named Nimalloy. Following this discovery, the variation of Nimalloy characteristics were investigated with the addition of $\text{Fe}^{(2)}$, $\text{V}^{(3)}$, $\text{Si}^{(4)}$, $\text{Ti}^{(5)}$, $\text{Al}^{(6)}$, $\text{Sb}^{(7)}$, $\text{Sn}^{(8)}$, $\text{Cr}^{(9)}$, $\text{W}^{(10)}$, $\text{Ge}^{(11)}$, $\text{Mo}^{(12)}$, $\text{Co}^{(13)}$, $\text{Cu}^{(13)}$, $\text{Nb}^{(14)}$ and $\text{Ta}^{(15)}$ to the Ni-Mn system and also with a single⁽¹⁶⁾⁽¹⁷⁾ or simultaneous addition⁽¹⁸⁾ of Cr and Mo to the Ni-Mn-Fe system. In consequence, the highest value of

121000 in initial permeability and the highest value of 643000 in maximum permeability have been obtained in the Ni-Mn-Fe-Cr-Mo system. Masumoto et al. have further clarified from the measured data on some of these alloy systems that the saturation magnetostriction $\lambda_s^{(19) \sim (21)}$ and the magnetic crystal anisotropy energy $K_1^{(22)(23)}$ have very low values for the compositions at which high permeabilities are attained.

Among a series of experiments now in progress on the effect of various additive elements to the binary Ni-Mn system, this paper describes the addition of Ta to Ni-Mn alloys so as to investigate its effect on the characteristics of Nimalloy.

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II. Experimental Procedure

The alloys used were prepared from electrolytic Ni, electrolytic Mn and 99.97% pure Ta, of which the former two elements were of the same purities as reported in the preceding paper⁽¹⁾. The preparation of specimens and measurements were similar to those described previously. The specimens used were fifty kinds of alloys with compositions of 71.68~93.52% Ni, 3.80~22.56% Mn and 2.08~14.83% Ta.

III. Results and Discussion

The experimental results are summarized in Figs. 1~10 and Table 1. The specimens whose compositions are shown by the solid circles in Figs. 1~2 were heated in vacuum at 900°C for 1 hr and cooled to room temperature at nine different rates, 240°, 100°, 50°, 20°, 10°, 5°, 3°, 1.5° and 0.75°C/hr. Depending on the compositions the specimen subjected to the similar heat treatment were cooled at 240°C/hr and further baked at the specified temperatures below the order-disorder transformation point. The highest values of initial permeability μ_0 and maximum permeability μ_m which were measured at room temperature with each of the

ternary alloys are schematically represented in Figs. 1~2 by equi-value curves drawn statistically against composition. As shown in the figures, with the addition of Ta to Ni-Mn alloys containing more than 10% Mn, μ_0 and μ_m increase slowly at the beginning and then rapidly attain the highest values which subsequently decrease steeply and then gradually. The composition range corresponding to large values of μ_0 and μ_m lies on the higher Ni side than the composition of $Ni_3(Mn, Ta)$. The highest values of μ_0 and μ_m obtained on the ternary Ni-Mn-Ta system are 20470 in μ_0 for the composition of 75.88% Ni, 19.47% Mn and 4.65% Ta and 84800 in μ_m for the composition of 75.37% Ni, 19.08% Mn and 5.55% Ta. These values are considerably large as compared with 6860 in μ_0 and 20400 in μ_m obtained with binary Ni-Mn alloys.

Table 1 shows electrical and magnetic properties of the representative alloys in the ternary system, where W_h is the magnetic hysteresis loss, $4\pi I_{50}$ is the intrinsic magnetic induction in a field of 50 Oe, λ_s is the saturation magnetostriction, and σ is the electrical resistivity and α is its temperature coefficient. As shown in the table, the addition of Ta to the binary Ni-Mn alloys achieves a marked improvement in magnetic performance

Fig. 1 Highest initial permeability in each of Ni-Mn-Ta alloys cooled at various rates from 900°C or further baked at various temperatures.

Fig. 2 Highest maximum permeability in each of Ni-Mn-Ta alloys cooled at various rates from 900°C or further baked at various temperatures.

of Nimalloy, especially with notably increased values of μ_0 , μ_m and $4\pi I_{50}$.

Figures 3~4 show the curves for the dependence of μ_0 and μ_m on Mn concentration and cooling rate for the ternary Ni-Mn-Ta alloys containing approximately 4.6 % Ta. The values of μ_0 and μ_m indicate a marked variation with cooling rate in the composition range of 14.0~21.5% Mn. In the case of μ_0 , the curves show a first maximum in the low Mn concentration region and a second maximum in the high Mn concentration region, whereas only the second maximum is observed in the case of μ_m . The second maxima for both μ_0 and μ_m first increase with decrease in cooling rate, attaining 5040 in μ_0 at 3°C/hr for the composition of 19.47% Mn

and 58700 in μ_m at 5°C/hr for the composition of 18.72% Mn and then decrease. In the case of μ_0 , the positions of its first and second maxima shift toward smaller Mn contents with decreasing cooling rate. However, in the case of μ_m , the position of its second maximum is very slightly changed.

Figure 5 shows the variations in μ_0 and μ_m with changes in cooling rate for four kinds of the ternary alloys containing about 75.5% Ni. As can be seen from the figure, the cooling rate at which the highest value of permeability is attained becomes smaller with increasing Ta content, as evidenced by the fact that alloys No.30 and No.36 do not exhibit the highest permeability even at a very small rate such as 0.75°C/hr.

Table 1. Magnetic and electrical properties of typical Ni-Mn-Ta alloys

Alloy No.	Mn (%)	Ta (%)	Cooling rate from 900°C (°C/hr)	Baking		μ_0	μ_m	B (G) for μ_m	$\frac{W_h}{\text{erg/cm}^3/\text{cycle}}$	B_r (G)	H_c (Oe)	$4\pi I_{50}$ (G)	λ_s ($\times 10^{-6}$)	$\frac{\sigma}{\mu\Omega\text{-cm, } 20^\circ\text{C}}$	α ($\times 10^{-3}$, $0^\circ\sim 40^\circ\text{C}$)	Density (g/cm ³)
				Tem-perature (°C)	Time (hr)											
									$B_m=5000\text{ G}$			$H=500\text{e}$	$H=9000\text{e}$			
33	21.63	—	10	380	50	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	380	50	5880	20400	908	*19.34	*1052	*0.0270	**2804	0.00	60.7	0.93	—
6	19.83	2.77	20	—	—	3960	40300	1622	*18.96	*1757	*0.0241	4110	0.00	62.8	1.05	8.44
6	19.83	2.77	240	380	30	10120	23700	640	—	—	—	2300	—	—	—	—
9	19.08	3.63	50	—	—	2200	57800	1657	—	—	—	3875	—	—	—	—
12	20.43	3.18	240	460	23	5280	58800	1968	65.62	3120	0.0309	5225	0.17	60.7	1.02	8.46
12	20.43	3.18	240	460	46	16280	39400	1697	48.10	2180	0.0331	5780	0.00	57.5	1.71	—
20	18.72	4.66	240	400	90	1870	67000	1923	—	—	—	4910	0.17	60.5	1.51	—
21	18.83	4.86	5	—	—	1760	58100	2550	67.35	3230	0.0320	6340	—	—	—	8.49
21	18.83	4.86	240	440	10	2300	81600	1577	—	—	—	4870	—	—	—	—
21	18.83	4.86	240	440	86	19350	42700	2450	33.04	1272	0.0249	6420	-0.17	48.6	1.45	—
22	19.47	4.65	3	—	—	5040	48400	2794	—	—	—	—	—	54.7	1.28	8.50
22	19.47	4.65	240	440	70	12540	80200	2980	47.67	3579	0.0289	6660	-1.70	51.2	1.26	—
22	19.47	4.65	240	460	64	20470	55200	3170	53.65	2670	0.0330	6580	—	50.3	1.32	—
23	20.07	4.10	240	460	48	17390	33100	1885	46.40	2594	0.0327	5770	0.17	40.1	1.28	—
24	20.09	4.58	240	460	35	16280	81900	2741	40.83	3710	0.0242	6580	0.00	56.4	1.16	—
30	19.08	5.55	240	440	65	8360	84800	2375	50.85	4325	0.0253	6805	0.00	52.7	1.10	8.49
30	19.08	5.55	240	440	75	17280	74900	2508	57.30	4295	0.0259	7060	—	45.3	1.22	—
35	17.15	6.75	240	460	100	2750	59500	2564	77.50	4535	0.0372	6770	—	50.3	1.16	—
36	18.60	6.55	240	400	440	4180	76200	2186	63.55	4740	0.0334	7295	0.00	53.9	1.25	8.65
36	18.60	6.55	240	400	580	11000	54400	2600	60.40	3677	0.0387	7375	0.00	45.5	1.19	—
39	17.38	8.46	240	440	240	1430	54450	2865	62.74	3540	0.0376	6560	—	58.2	1.14	8.73
40	18.11	8.05	240	400	540	2420	50200	2640	96.22	4611	0.0480	6815	—	50.4	1.08	—
44	14.65	9.38	2800	440	10	880	38500	1844	—	—	—	4050	—	—	—	8.89

* $B_m=2000$ G, ** $H=900$ Oe

Fig. 3 Initial permeability of Ni-Mn- about 4.6% Ta alloys cooled at various rates from 900°C.

Fig. 4 Maximum permeability of Ni-Mn- about 4.6% Ta alloys cooled at various rates from 900°C.

Accordingly, as a condition essential to develop the highest level of permeability, the ternary alloys should be further baked at temperatures below the order-disorder transformation point in a way to advance the degree of order.

Figure 6 illustrates the relationship between the bake time and the values of μ_0 and μ_m when the same Ni-Mn-Ta alloys as in Fig. 5 were cooled from 900°C to room temperature at 240°C/hr and further baked at 440°C. In general the permeability has a maximum

Fig. 5 Effect of cooling rate on initial and maximum permeabilities of Ni-Mn-Ta alloys containing about 75.5% Ni.

Fig. 6 Effect of baking time at 440°C after cooling at a rate of 240°C/hr from 900°C on initial and maximum permeabilities of Ni-Mn-Ta alloys containing about 75.5% Ni.

against baking time, and its maximum shifts toward a longer baking time with increase in Ta content.

Figure 7 shows the variations in μ_0 and μ_m with baking time for alloy No. 30 (75.37% Ni, 19.08% Mn and 5.55% Ta) with the highest value of μ_m in the Ni-Mn-Ta system which was baked at various temperatures after cooling from 900°C to room temperature at 240°C/hr. All the curves in the figure indicate a maximum which appears at a shorter baking time with increase in baking temperature. This implies that the rate of

Fig. 7 Effect of baking temperature and time after cooling at a rate of 240°C/hr from 900°C on initial and maximum permeabilities of No. 30 alloy (75.37% Ni-19.08% Mn-5.55% Ta).

superlattice structure formation becomes faster with the rise in baking temperature. Regardless of the baking temperature, the maximum of μ_m appears at a shorter baking time than in the case of the maximum of μ_0 . The highest value of μ_0 is 17280 when baked at 440°C for 75hr and the highest value of μ_m is 84800 when baked at the same temperature for 65hr. It will be noted that although a high level of permeability can be obtained with the ternary compositions near to about 5% Ta by slow cooling from 900°C, the highest permeability can be achieved with the compositions of more than 5% Ta by baking at temperatures below the order-disorder transformation point for an appropriate baking time.

Figures 8 and 9 illustrate the composition dependence of $4\pi I_{50}$ and σ at 20°C for each of the Ni-Mn-Ta alloys which were subjected to heat treatment to develop the highest maximum permeability. In general the ternary alloys which exhibit a high permeability at compositions near to about 74~78% Ni are considered in an advanced rate of ordering in view of the fact that their values are relatively large in $4\pi I_{50}$ and somewhat low in σ . In Fig. 10 are shown the magnetization and

Fig. 8 Intrinsic magnetic induction at a magnetic field of 50 Oe in Ni-Mn-Ta alloys heat-treated to develop the highest maximum permeability.

Fig. 9 Electrical resistivity at 20°C in Ni-Mn-Ta alloys heat-treated to develop the highest maximum permeability.

Fig. 10 Magnetization and hysteresis curves of No. 30 alloy baked at 440°C for 65 hr after cooling at a rate of 240°C/hr from 900°C.

hysteresis curves for alloy No. 30 with the highest value of μ_m in the Ni-Mn-Ta system when baked at 440°C for 65 hr after cooling from 900°C to room temperature

at 240°C/hr. From these curves it is evident that the alloy has excellent magnetic properties.

As high-permeability alloys have been known to be characterized by a very low value of saturation magnetostriction, high permeabilities are shown in the alloys of the Ni-Mn-Ta system which have low values of magnetostriction as shown in Table 1.

IV. Conclusions

The magnetic properties, electrical resistivity and temperature coefficient of resistivity at room temperature were measured on fifty kinds of ferromagnetic Ni-Mn-Ta alloy, with composition of 71.68~93.52% Ni, 3.80~22.56% Mn and 2.68~14.83% Ta. The alloys measured were those heated in vacuum at 900°C for 1 hr and then cooled at various rates or further baked at various temperatures below the order-disorder transformation point after cooling at 240°C/hr. The results obtained are as follows:

(1) The highest values of initial permeability and maximum permeability obtained on each of Ni-Mn-Ta alloys increase gradually at the beginning and later rapidly with increasing Ta content, and after attaining a maximum, the values decrease gradually. The highest

value of initial permeability is 20470 for the composition of 75.88% Ni, 19.47% Mn and 4.65% Ta when baked at 460°C for 64 hr after cooling from 900°C to room temperature at 240°C/hr, and the highest value of maximum permeability is 84800 for the composition of 75.37% Ni, 19.08% Mn and 5.55% Ta when baked at 440°C for 65 hr after cooling from 900°C at 240°C/hr.

(2) The coercivity for the maximum magnetic induction of 5000 G shows a low value in general. The lowest value is 0.0242 Oe for the composition of 75.33% Ni,

20.09% Mn and 4.58% Ta, the corresponding hysteresis loss being 40.83 erg/cm³/cycle.

(3) The intrinsic magnetic induction in an effective field of 50 Oe is relatively large. The alloy composed of 75.37% Ni, 19.08% Mn and 5.55% Ta which exhibits the highest maximum permeability in the ternary system shows the magnetic induction of 6805 G and the electrical resistivity of 52.7 $\mu\Omega$ -cm at 20°C.

(5) At room and high temperatures, these Ni-Mn-Ta alloys can more easily be forged and rolled than Permalloy.