Au-Fe-Ni Permanent Magnet Alloys

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Abstract—**A gold permanent magnet for an ornamental material is developed in the Au-Fe-Ni system. It is found that the magnetic properties of the Au–12.5 wt% Fe–12.5 wt% Ni alloy aged at 450°C for seven hours and cold-swaged by 92 percent are Br = 0.53 T (5300 G), iHc = 40.8 kA/m (510 Oe), and $(BH)_{max}$ = 9.1 kJ/m^3 (1.1 MGOe). The Au–12.5 wt% Fe–10 wt% Ni–2.5 wt% Co alloy aged at 450°C for 2.5 h and swaged by 86 percent attained Br = 0.54 T (5400 G), iHc = 42.4 kA/m (530 Oe), and $(BH)_{max}$ = 9.6 kJ/m^3 (1.2 MGOe). In these alloys, the magnetic hardening is due to the fine Fe-Ni rich ferromagnetic particles precipitated in the Au matrix.**

INTRODUCTION

A UNIQUE APPLICATION of magnetically hard materials is the use of permanent magnets for ornaments such as necklaces and rings. The Au-Fe-Co permanent magnet alloys have been developed for such ornamental applications [1].

The Au-7.2% Fe–44.8% Co alloy (weight percent unless specified otherwise) was aged at 600°C for ten minutes and subsequently control-cooled to 400°C at a cooling rate of 50°C/h, it was then cold-swaged by 50 percent reduction in area. The magnetic properties of the alloy are Br = 0.34 T (3400 G), iHc = 81.2 kA/m (1015 Oe), and $(BH)_{max}$ = 8.2 kJ/m^3 (1.0 MGOe). The alloy has a microstructure consisting of ferromagnetic phase finely dispersed by precipitation in a nonferromagnetic matrix. According to the phase diagram by Köster *et al.* [2], it is anticipated that the Au-Fe-Ni alloys decompose into two phases, with a microstructure of an Fe-Ni rich ferromagnetic phase dispersed in a Au rich nonferromagnetic phase. Therefore, it is the purpose of this investigation to examine whether the Au-Fe-Ni alloys have magnetic properties suitable for permanent magnets in the same manner as the Au-Fe-Co alloys. It is found that the magnetic properties for permanent magnets are obtained with the Au-Fe-Ni alloys. The alloys containing 2–2.5 percent Co are also studied. The magnetic properties of the alloys are discussed in conjunction with the microstructural observations.

EXPERIMENTAL PROCEDURE

The alloys were prepared from commercial pure gold (99.99 percent), electrolytic iron (99.9 percent), Mond nickel (99.97 percent), and electrolytic cobalt (99.9 percent). They were melted in an argon atmosphere with a high-frequency induction furnace.

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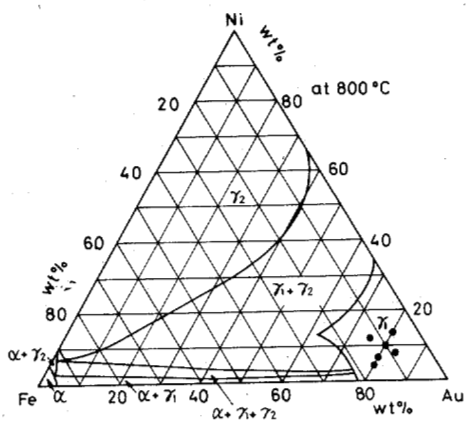

Fig. 1. Phase diagram of Au-Fe-Ni system at 800°C (after Köster *et al.* [2]). Studied alloys are shown by solid circle.

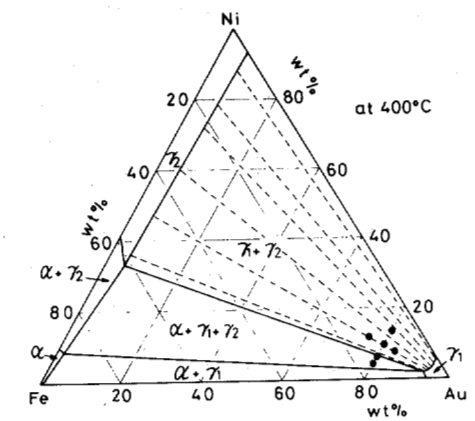

Fig. 2. Phase diagram of Au-Fe-Ni system at 400°C (after Köster *et al.* [2]). Studied alloys are shown by solid circle.

Ferromagnetic elements such as Fe and Ni show limited solid solubility in Au. The compositions of the studied alloys in the ternary systems were selected by the criterion that the alloys have a solid-solution region at high temperatures and a ferromagnetic phase precipitated out of a gold-rich phase at low temperatures. V. W. Köster *et al.* [2] studied the ternary Au-Fe-Ni system, the isothermal sections of which are shown in Fig. 1 at 800°C and Fig. 2 at 400°C. The solid circles in Fig. 1 and Fig. 2 show the compositions of the studied alloys.

The specimens were solution-treated at 950°C for one hour and cooled in air. They were then aged at 400–450°C. All heat treatments were carried out in an argon atmosphere or in a vacuum. In order to further improve the magnetic properties, some of the specimens were cold-

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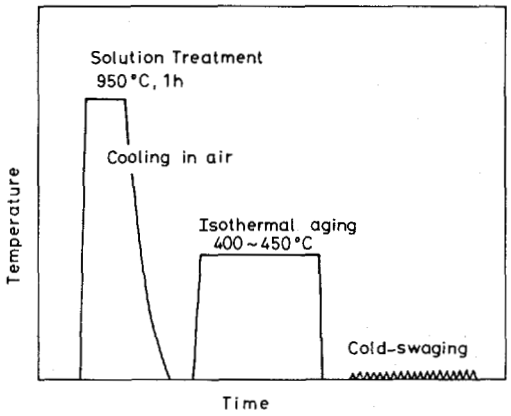

Fig. 3. Schematic diagram of heat treatment and cold-swaging.

swaged after aging. Fig. 3 shows the schematic diagram of heat treatment and cold-swaging.

Magnetic properties were measured with an automatic recording fluxmeter. The maximum applied field was 300 kA/m (3.8 kOe). In this paper the symbol *Is* represents the magnetization intensity at 300 kA/m (3.8 kOe) at room temperature.

The samples for microscopic examination were electro-etched using 10 vol% hydrochloric acid and 90 vol% ethyl alcohol mixture. The microstructures of some alloys were observed by transmission electron microscopy. Thin disk samples were prepared by electropolishing in a solution of 67 vol% hydrochloric acid and 33 vol% acetic acid.

Results and Discussion

A. Solution Treatment

The solution treatment of Au-Fe-Ni alloys can be performed after heating the alloy at 950°C for one hour, followed by 1) quenching in water, or 2) cooling in air. The two kinds of quenching after solution treatment have no influence on the magnetic properties after aging, but have a great influence on the deformability of the alloys by cold-swaging. The alloys quenched in water break up into small pieces by cold-swaging, while the alloys cooled in air are very workable. For that reason, in this work, the solution-treated specimens were obtained by cooling in air.

B. Magnetic Properties After Aging

The variations of intrinsic coercive force *iHc* and the magnetization intensity *Is* are shown in Fig. 4. The total content of iron and nickel in these studied alloys is 20 percent. The Au–16% Fe–4% Ni alloys and the Au–14% Fe–6% Ni alloys show about 0.6 T (~6000 G) magnetization intensity in the early stages of aging, but the magnetization intensity decreases to about 0.3 T(~3000 G) with increasing aging times. At the same time, the intrinsic coercive force increases with longer aging times. The Au–6% Fe–14% Ni alloy has a maximum coercivity of about 30 kA/m (~380 Oe), but has a low magnetization intensity (0.35 T). The Au–10% Fe–10% Ni alloy, on the other hand, can reach a 0.4 T (4000 G) magnetization intensity and exceed a 30 kA/m (380 Oe) intrinsic coercive force.

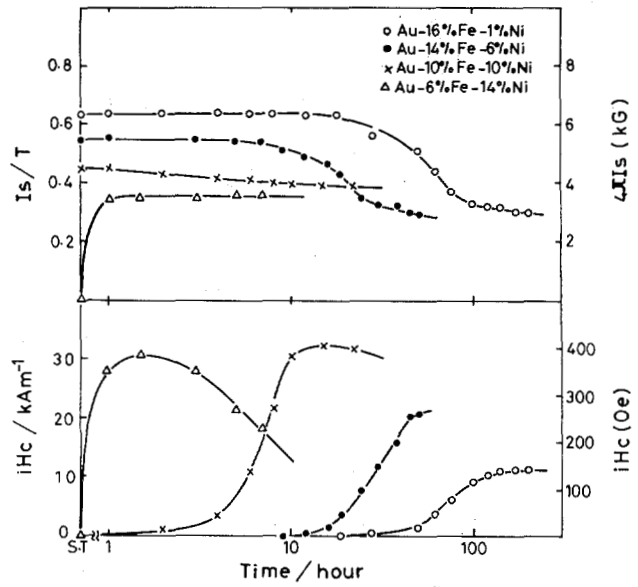

Fig. 4. Intrinsic coercive force *iHc* and magnetization intensity *Is* of Au-Fe-Ni alloys after aging at 400°C.

Consequently, it seems that the optimum Fe/Ni ratio of the Au-Fe-Ni alloys is 1 : 1 when the total content of the Fe and Ni is constant.

So far the total content of the Fe and Ni of the studied alloys has been fixed at 20 percent. In further studies the total content is fixed at 16 and 25 percent, while the ratio of Fe and Ni is maintained at 1 : 1. The Au–8% Fe–8% Ni, Au–10% Fe–10% Ni, Au–12.5% Fe–12.5% Ni alloys were solution-treated at 950°C and aged at 400–450°C. The magnetic properties of these three alloys are shown in Table I. These alloys have about 30 kA/m (~380 Oe) of intrinsic coercive force and about 1.5 kJ/m^3 (~0.2 MGOe) of $(BH)_{max}$.

C. Cold-Swaging

According to the decomposition tie-lines determined by Köster for the alloys studied here (Fig. 2), it seems that the compositions of the precipitated Fe-Ni ferromagnetic particles are 50% Fe and 50% Ni, with low magnetic crystal anisotropy [3]. The introduction of cold-swaging after aging has therefore been tried in order to increase the shape anisotropy.

The effect of the introduction of the cold-swaging on magnetic properties is shown in Fig. 5. The *iHc* and *Ir*/*Is* are improved by cold-swaging, and the obtained values of $(BH)_{max}$ increased five times by swaging. The maximum magnetic properties attained for the Au-Fe-Ni alloys after cold-swaging are summarized in Table II. The magnetic properties of the Au–12.5% Fe–12.5% Ni alloys are at their maximum after 92 percent reduction in area with values of Br = 0.53 T (5300 G), iHc = 40.0 kA/m (500 Oe), Ir/Is = 0.91, and $(BH)_{max}$ = 9.1 kJ/m^3 (1.1 MGOe). The magnetization curve of the swaged Au–12.5% Fe–12.5% Ni alloy is shown in Fig. 6 in comparison with that of the unswaged alloy.

TABLE I
MAGNETIC PROPERTIES OF AGED Au-Fe-Ni ALLOYS

	Heat Treatment °C (hours)	*Is*/T (G)	*Br*/T (G)	*iHc*/kAm^{-1} (Oe)	$(BH)_{max}$/kJm^{-3} (MGOe)
Au-8% Fe-8% Ni	400 (16)	0.28 (2800)	0.15 (1500)	35.8 (450)	1.0 (0.1)
Au-10% Fe-10% Ni	400 (20)	0.38 (3800)	0.22 (2200)	32.1 (400)	1.4 (0.2)
Au-12.5% Fe-12.5% Ni	450 (7)	0.46 (4600)	0.24 (2400)	25.6 (320)	1.5 (0.2)

TABLE II
MAGNETIC PROPERTIES OF Au-Fe-Ni ALLOYS AGED AND COLD-SWAGED

	Heat Treatment °C (hours)	Reduction in Area (percent)	*Is*/T (G)	*Br*/T (G)	*iHc*/kAm^{-1} (Oe)	$(BH)_{max}$/kJm^{-3} (MGOe)
Au-8% Fe-8% Ni	400 (16)	96	0.38 (3800)	0.35 (3500)	55.2 (690)	7.2 (0.9)
Au-10% Fe-10% Ni	400 (20)	95	0.48 (4800)	0.43 (4300)	48.0 (600)	8.2 (1.0)
Au-12.5% Fe-12.5% Ni	450 (7)	92	0.58 (5800)	0.53 (5300)	40.0 (500)	9.1 (1.1)

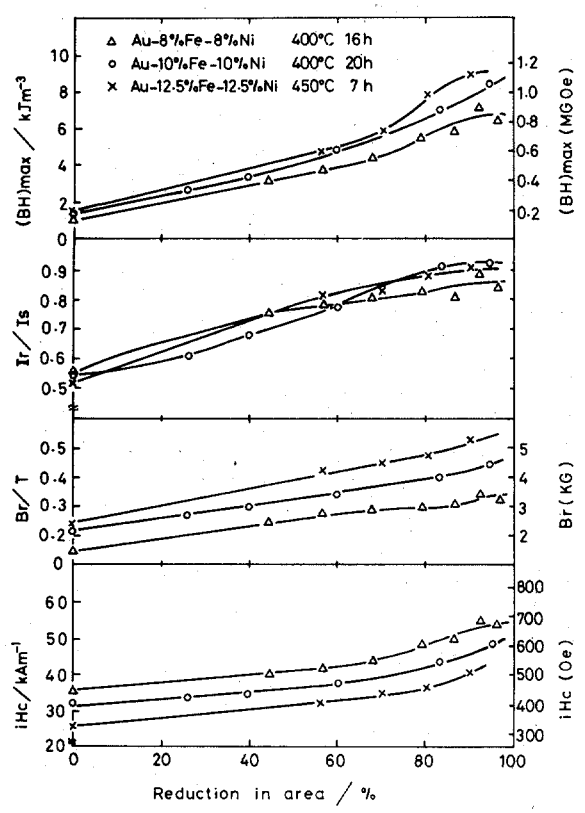

Fig. 5. Effect of cold-swaging on magnetic properties of Au-Fe-Ni alloys.

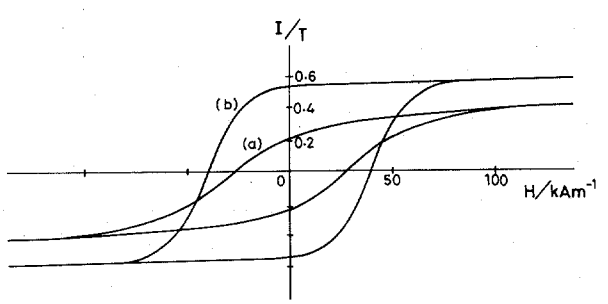

Fig. 6. Magnetization curves of Au-12.5% Fe-12.5% Ni alloy. (a) Before swaged. (b) After swaged, 92 percent reduction in area.

D. Au-Fe-Ni-Co Alloys

In order to further increase the magnetization intensity of the alloys, 2–2.5 percent Co was added in place of the Ni content. Table III summarizes the maximum magnetic properties obtained with the Au-Fe-Ni-Co alloys, and its optimum treatment conditions, which are such that the alloys are solution-treated at 950°C, aged at 400–450°C, and cold-swaged by 86–90 percent reduction in area. The maximum value of $(BH)_{max}$ obtained in the investigation of Au–12.5% Fe–10% Ni–2.5% Co alloy is 9.6 kJ/m^3 (1.2 MGOe).

E. Microstructure

The intrinsic coercive force of the Au–10% Fe–10% Ni alloy aged at 400°C for various times is shown in Fig. 7. With the increase of intrinsic coercive force, the change of microstructure of the alloy can be observed by optical microscopy. The microstructures corresponding to Fig. 7(a)–(f) are shown in Fig. 8. The optical micrograph (a) shows a solid solution after the air cooling from 950°C. It was found that the two-phase decomposition of the Au-Fe-Ni alloys occurs at the grain boundary [2], [4]. As the aging times increase, the transformed region (nodule) spreads into the grain and the intrinsic coercive force increases. When the nodule covers the whole grain (the optical micrograph (f)), the Au–10% Fe–10% Ni alloy has the maximum value of intrinsic coercive force. Further aging decreases *iHc*, possibly due to the formation of new nodules which have a wider lamella spacing.

It is reported that controlled cooling is effective for the Au-Fe-Co alloys [1]. In the present investigation, the controlled cooling and the step aging were tried, but both methods were ineffective with the Au-Fe-Ni alloys. Since the lamella spacing is determined by aging temperature, the controlled cooling or the step aging forms many nodules having various lamella spacings. The remanence of the Au-Fe-Ni alloys having various lamella spacings is

TABLE III
MAGNETIC PROPERTIES OF Au–Fe–Ni–Co ALLOYS AGED AND COLD-SWAGED

	Heat Treatment °C (hours)	Reduction in Area (percent)	*Is*/T (G)	*Br*/T (G)	*iHc*/kAm^{-1} (Oe)	$(BH)_{max}$/kJm^{-3} (MGOe)
Au–8% Fe–6% Ni–2% Co	400 (4)	88	0.40 (4000)	0.35 (3500)	58.4 (730)	8.3 (1.0)
Au–10% Fe–8% Ni–2% Co	400 (15)	88	0.49 (4900)	0.44 (4400)	47.2 (590)	8.0 (1.0)
Au–9% Fe–9% Ni–2% Co	400 (8)	90	0.49 (4900)	0.45 (4500)	49.6 (620)	8.8 (1.1)
Au–12.5% Fe–10% Ni–2.5% Co	450 (3)	86	0.59 (5900)	0.54 (5400)	42.4 (530)	9.6 (1.2)
Au–11.5% Fe–11.5% Ni–2% Co	450 (2)	88	0.59 (5900)	0.53 (5300)	40.8 (510)	8.9 (1.1)

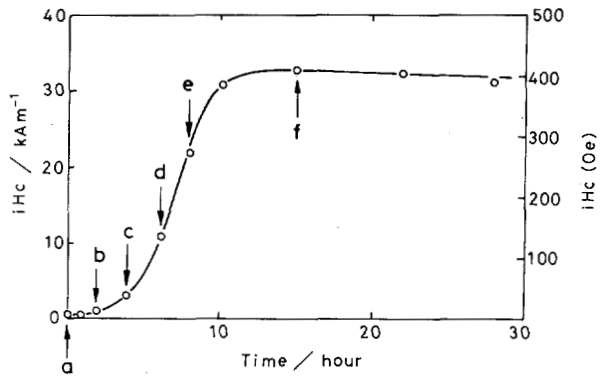

Fig. 7. Intrinsic coercive force of Au–10% Fe–10% Ni alloy aged at 400°C.

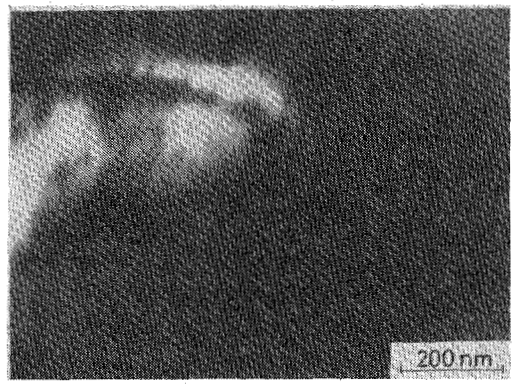

Fig. 9. Microstructure of Au–10% Fe–10% Ni alloy solution treated at 950°C and cooled in air.

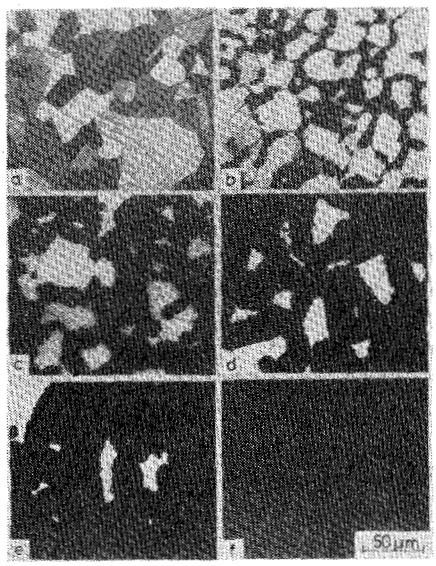

Fig. 8. Microstructures of Au–10% Fe–10% Ni alloy aged at 400°C for various times. (a) Solid solution, *iHc* = 0 kA/m. (b) 2 h, *iHc* = 1.2 kA/m. (c) 4 h, *iHc* = 3.2 kA/m. (d) 6 h, *iHc* = 11.2 kA/m. (e) 8 h, *iHc* = 21.6 kA/m. (f) 15 h, *iHc* = 32.8 kA/m.

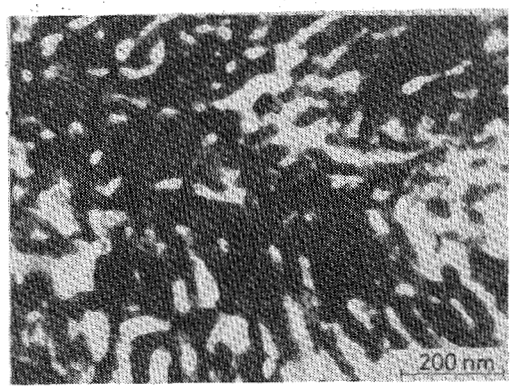

Fig. 10. Microstructure of Au–10% Fe–10% Ni alloy aged at 450°C for 7 h.

lower than that of the same alloys after isothermal aging. Consequently, the effective aging for the Au-Fe-Ni alloys is isothermal aging.

The size and shape of the precipitated ferromagnetic particles were observed by transmission electron microscopy. Fig. 9 shows the microstructure of the Au–10% Fe–10% Ni alloy solution treated at 950°C and cooled in air, while Fig. 10 shows the microstructures of the same alloy aged at 450°C for seven hours. The light contrast shows holes 15–30 nm thick and 100–150 nm long, corresponding to etched iron–nickel precipitates, and the phase with dark contrast is identified as the Au-rich phase. Fig. 11 shows the microstructures of the alloy swaged by 95 percent reduction in area, a) perpendicular and b) parallel to the direction of swaging. The micrographs indicate that the improvement of the magnetic properties of the swaged alloy would be due to the elongation of the iron–nickel particles parallel to the swaged direction marked S. The micrographs also suggest that the contributions to the coercive force may arise from the shape anisotropy of the iron–nickel precipitates in this alloy.

F. Comparison of the Au-Fe-Ni Magnet with the Au-Fe-Co Magnet

Requirements for magnetic properties, color, workability, and gold content must be fulfilled before gold-based

Fig. 11. Microstructures of Au–10% Fe–10% Ni alloy swaged to 95 percent reduction in area. (a) Perpendicular to direction of swaging. (b) Parallel to direction of swaging.

TABLE IV
COMPARISON OF Au–12.5% Fe–12.5% Ni MAGNET WITH Au–7.2% Fe–4.8% Co MAGNET

	Au–12.5% Fe–12.5% Ni	Au–7.2% Fe–4.8% Co
Magnetic properties	Br = 0.53 T (5300 G) iHc = 40.0 kA/m (500 Oe) $(BH)_{max}$ = 9.1 kJ/m^3 (1.1 MGOe)	Br = 0.34 T (3400 G) iHc = 81.2 kJ/m (1015 Oe) $(BH)_{max}$ = 8.2 kJ/m^3 (1.0 MGOe)
Color	Silver-white	gold color
Workability	very well	well
Gold content	18.0 carats	21.1 carats

permanent magnets are used for ornamental applications. Characteristics of the gold-based magnets studied here are summarized in Table IV. From these results it is concluded that the newly developed Au-Fe-Ni alloys are alternative ornamental magnets.

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