—TECHNICAL PAPER*—

Studies on Industrialization of "MT" Permanet Magnets**

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1. Introduction

Today, applications of permanent magnets are increasing in various fields of industries. Along with the increase of demand in the market, the improvement of quality is of necessity required. But the magnet of excellent quality contains a great quantity of rare elements such as Ni or Co. During the World War II, research of permanent magnets was concentrated in Japan on the discovery of substitutional alloy which possessed high coercive force without containing any rare metals. MT magnets were developed in the authors' laboratory.

This new magnet steel is an Fe-Al-C ternary alloy and does not contain any of rare metals. It has magnetic properties nearly equal to a middle-cobalt magnet steel with intermediate quality between Alnico and Cr magnet steel. It must especially be noted that this magnet possess comparatively high coercive force of about 200 Oersteds though it contains no rare metals. In this report the result of basic experiments on MT magnets and the results relative to industrial aspects are reported.

II. Methods of Experiments

First, the specimens of Fe-Al-C ternary alloys containing various different quantities of Al and C were prepared to detect their optimum compositions. Further 13 kinds additional elements including Si, Mn, Cu, Sn, Co, Ni were added to the base alloy and their effect on the magnetic properties were investigated. Next, in order to obtain optimum conditions of heat treatment, effects of solution-treatment, cooling rate, aging conditions, decarburization and other treatment on the magnetic properties were studied with the specimens consisted of representative compositions. Then the relation between the phase diagram and the structure were obtained by micrography and X-ray diffraction analysis. Finally, the results of experiments are dealt with which are believed necessary to be known by customers, with emphasis on those basic magnetic properties, magnetic stability and magnetic designs in additions to industrial applications.

Raw materials used for melting were electrolytic iron or low-carbon steel, commercial aluminium and white pig iron and besides then ferromanganese was added as deoxidiser. Specimens were prepared by a high-frequency induction furnace of 35 KVA, and were cast into sand-molds in a shape of round rods (7 mm diameter) or flat bars (20×10×70 mm). Heat treatment was performed in a silicon-carbide furnace for quenching and a muffle furnace of Fe-Cr alloy heating elements for aging. Unless the condition of heat treatment specified otherwise, the experiments must be considered to have been carried out under the standard treatment by which specimens were subjected to oil-quenching after heating at 1200°C and aging for 1 hour

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at 300°C. Magnetic properties was measured by a magneto-meter (Honda method) or a permea-meter of TKS (similar to Isthmus electro-magnets).

III. Optimum Chemical Composition

Chemical composition: MT magnet alloys contained approximately 1.0-3.0% C, 4.0-12.0% Al, the remainder being Fe and few impurities which included Si, Mn and Cu. The dependence of magnetic properties on Al and C contents was studied. These specimens were subjected to aging at 300-350°C after oil-quenching. Surface-coating was used to prevent oxidation through heating at high temperature. The relations between magnetic properties and compositions are shown in Fig. 1-3.

Fig. 1 Relations between the residual induction and compositions.

Fig. 2 Relations between the coercive force and compositions.

Fig. 3 Relations between the energy product and compositions.

The experiment showed that these alloys presented the most superior magnetic properties in the region of $8.0 \pm 0.2\%$ Al, $2.0 \pm 0.1\%$ C and its residual induction became more than 6,000 Gauss, coercive force more than 200 Oersteds and energy product more than 0.45×10^6 Gauss-Oersteds.

Relation to phase diagram: The inclination of the boundary line between γ and $\gamma+K$ fields on the phase diagram seems responsible for the abovementioned phenomena. It is apparent from a phase diagram of 1200°C section⁽¹⁾ as shown in Fig. 4 that the range of optimum composition lay approximately along a "tie line" in the field.

Effect of additional elements: The dependence of magnetic properties on additional elements was examined. As to the additional elements, 13 kinds of elements as Si, Mn, Co, Ni, Cu, Sn, Cr, Ti, W, Mo, V, Be and B were chosen and these were

Fig. 4 Equilibrium diagram of Fe-Al-C ternary alloys.

(1) K. Löhberg, W. Schmidt: Arch. f. Eisenhüttenwes. 12 (1937/38) 607

added one by one to the base alloy that contained 2% C, 8% Al and the rest Fe. Some examples of these results are shown in Fig. 5 and Fig. 6.

Fig. 5 Effect of cobalt on the magnetic properties

Fig. 6 Influences of Si upon magnetic properties.

After careful observations these results were summarized as follows:—

Elements of improving effect:

- on residual induction Co, Sn.
- on coercive force Mn, Ni, Cu, Cr, Sn, B.

Elements of deteriorating effect:

- on residual induction Mn, Ni, Cu, Cr, Sn, B.
- on coercive force Si, Ti, V.

Fig. 7 Effect of Mn on cracking.

Even the improving elements on the magnetic energy product $(BH)_{\max}$, however, by adding more than 3%, would turn to give a bad effect on magnetic properties except Co. Addition of small amount of Cu, Sn or Ni gave a good effect on magnetic properties.

It is known from Fig. 6 that magnetic properties seem susceptible to the effect of Si content. Hence Si must be lower than 0.35%. Investigation was made to see the effect of Mn content on the frequency of cracking with the products sensible to quenching cracks. From the Fig. 7 it can be seen that cracking decreased as added Mn increased. As S gave bad effects on cracking, Mn had to be added up to 0.2%.

IV. Heat Treatment

Heat treatment for this Fe-Al-C alloy consisted in:

- a. Heating at a temperature for solution-treatment to form a single phase.

- b. Quenching at the optimum cooling rate.
- c. Tempering for converting the structure as quenched to martensite.

In the special case of chill castings, the metal mold was previously heated to about 100°C and the melt of 1530–1560°C was poured into it. The chill-cast specimens were aged without quenching operation. The sufficient hardening was obtained in the specimens as chill-cast.

Specimens showed the representative composition (8% Al, 2% C and the rest Fe) and cast in a shape of flat bar into sand mold or round rod into metal mold.

Quenching temperature and holding time: The relations between quenching temperature and some magnetic properties were shown in Fig. 8. The plotted points on the curves represent the experimental results with the specimens which were quenched in oil bath after heating at respective temperatures for 30 minutes to be followed by aging at 300°C. Consequently, it was apparent that this alloy possessed poor magnetic properties when it was quenched below 1150°C. Quenching temperature should be changed according to the different compositions and the amount of impurities. The critical quenching temperature for the alloy containing 1.95–2.05% C and 7.7–8.2% Al is 1200°C. Overheating above 1230°C caused blister on the surface.

For the purpose of finding the minimum holding time, the relations between magnetic properties and the holding time at the temperature of solution-treatment was examined. It was recognized that holding time of 30 minutes at 1200°C was most profitable.

Cooling rate: The cooling rate strongly affected the magnetic properties of this alloy. It was associated with the formation of martensite and its dispersion ratio in austenitic matrix.

Fig. 8 Effect of quenching temperature on the magnetic properties.

Fig. 9 Relations between the residual induction and aging temperature (for various cooling methods)

Fig. 10 Relations between the coercive force and aging temperature (for various cooling methods)

The variations of magnetic properties with the aging at 100°C to 600°C for 1 hour after quenching into water or oil from 1200°C (excepting the case of chill-castings) were investigated respectively. Its results were shown in Fig. 9 and Fig. 10.

The curves for specimens oil-quenched, 70°C water-quenched and chill-cast showed approximately the same features. Among them, oil quenching gave the best result. The most inferior result was obtained with the specimen quenched into water at 20°C., and the tempering-temperature curve of coercive force showed inverse tendency to that of the others. It is worthy to notice that the double hardening occurred at about 500°C.

Tempering temperature and holding time: As the matrix of this alloy is consisted of austenitic phase after quenching, it is necessary to convert its structure to ferromagnetic phase such as martensite through tempering at an appropriate temperature by which the magnetic induction will be increased. Tempering curves of magnetic properties, derived by tempering the specimens for 1 hour at each temperature after oil-quenching from 1200°C are shown in Fig. 11. It is observed that 300°C is most suitable as the tempering temperature.

The same curves on metal-mold casting, derived by tempering after chill-casting, are shown in Fig. 12.

Fig. 11 Effect of tempering temperature on the magnetic properties (oil-quenched specimen)

Fig. 12 Effect of tempering temperature on the magnetic properties and hardness (chill-casted specimen)

The energy product was superior at both 300°C and 500°C.

Tempering time-curves of the magnetic properties at various temperatures for specimens after oil-quenching are investigated. From them it is known that magnetic energy product becomes saturated by holding for 60 minutes at 300°C, a little change occurs by making the aging-time prolonged. In the case of oil-quenching, the transformations is somewhat more quickly done than in the case of chill-casting.

Decarburisation by heating: Heat treatment accompanies oxidation on the surface of alloy

Table 1 Chemical analyses of the surface layer and the interior portion.

Compositions	Interior portion	Surface layer
Carbon	1.98%	1.66%
Aluminum	8.07%	8.06%

which decreases some alloying elements near the surface layers of the specimen and probably causes the decrease of magnetic properties. Chemical analyses of the surface layer is shown in Table 1. Decarburisation is clearly observed on the surface.

The change of magnetic properties by eliminating the decarburised layer were examined for specimens heat-treated without surface-coating, and the result of these experiments are shown in Table 2. It was recognized that coercive force was unaffected by the existence of decarburized layer, but residual induction was increased by 5 to 30% by eliminating the surface layer.

Table 2 Changes of magnetic properties due to grinding-off of the decarburised layer.

Specimen numbers	Measurement after heat treatment			Measurement after grinding-off of the burrs			Measurement after grinding-off of the surface by 1 mm		
	Br (Gausses)	Hc (Oersteds)	BrHc $\times 10^6$ (G. Oe)	Br (Gausses)	Hc (Oersteds)	BrHc $\times 10^6$ (G. Oe)	Br (Gausses)	Hc (Oersteds)	BrHc $\times 10^6$ (G. Oe)
No. 1	5,750	188	1.08	6,020	198	1.19	6,510	198	1.29
No. 2	5,630	176	0.99	6,310	165	1.04	7,050	178	1.25

Deep-cooling treatment: Coercive force of MT magnets shows a considerable decrease by 38% and residual induction increases by 25% by subjecting to deep-cooling treatment at -85°C . From these, it is known that coercive force of magnet steel is not led to internal strains of martensite but to dispersion of ferromagnetic phase in non-magnetic matrix.

Cooling in magnetic field: Improvement of magnetic properties by cooling in magnetic field was examined in regard to MT magnets. The improvement of residual induction and coercive force can hardly be seen and it may be considered that the cooling in magnetic field is ineffective so far as the MT magnet is concerned.

Mass effect: The mass effect of this alloy was examined using the specimens of various thicknesses. Their magnetic properties were measured to see their relationship with thickness. From these experiments, mass effect on this alloy can hardly be seen in the specimen of thickness up to 25 mm.

V. Microstructure and Phase Diagram

Coercive force of this magnet is comparable to that of high-cobalt steel. Magnetization fluctuation or internal strains which are responsible for maintaining coercive force is obtainable through the formation of martensite and precipitation of double carbide in austenitic matrix.

When the amount of carbon content exceed 1.7%, the carbon can no longer exist as the solid solution in normal Fe-C alloy, namely carbon steel or other special steel. Accompanying with the addition of Al, however, the amount of solid solution of carbon in iron increases remarkably. For example, about 2.1% C dissolves as solid solution in iron with addition of 7% Al.

Hereon, the variation of microstructure accompanying with heat treatment was investigated by means of micrography and X ray diffraction analyses. Some examples of microstructures

is shown in Fig. 13. The changes of microstructure accompanying with heat treatment can be seen in Table 3.

(a) As quenched

(b) As tempered at 300°C

Fig. 13 Microstructure of MT magnet (C 2.0%, Al 7.99%) $\times 200$

Table 3 Microstructure of MT magnet alloy.

Heat treatment	Microstructure
Chill casting	Austenite + Martensite (body-center cubic)
Oil quenching	Austenite + Martensite (tetragonal body-center cubic)
Aging at 300°C	Martensite (body-center cubic) + Austenite (little)
" at 500°C	α -iron + double carbide
1200°C furnace-cooling	α -iron + double carbide + graphite

VI. Magnetic Properties

The demagnetization curve of MT magnets is shown in Fig. 14. According to this figure,

Fig. 14 Demagnetizing curve of MT magnets.

the residual induction is about 6,000 Gauss and the coercive force is about 200 Oersteds. The magnetic properties of MT magnets are summarized in Table 4.

Table 4 The magnetic properties of MT magnets.

Magnetic properties			Measured value
Saturation induction	Bs	(Gauss)	9,000
Residual flux density	Br	(Gauss)	6,000
Coercive force	Hc	(Oersteds)	200
Flux density, corresponding to the (BH) _{max} point			3650
	Bd	(Gauss)	
Magnetizing force, ditto	Hd	(Oersteds)	123
Energy product (BH) _{max} (G.Oe)			0.45×10^6
Fullness factor (BH) _{max} /BrHc (%)			37
Reversible permeability μ_r			8~10
Specific gravity (g/cm ³)			6.9

Thermal coefficient of MT magnets which indicates the rate of reversible change of the magnetic flux with temperature was examined. As for the specimens, braking magnets for a watt-hour meter and a magnet bar were used and the results are as shown in Table 5.

Table 5. Temperature coefficient (α) of MT magnets

Specimens	Temperature range (°C)	α (1/°C)
1. Magnets bar (L/D=6)	0~100	-6.8×10^{-4}
2. Magnets for watt-hour meters* (stabilized)	0~30	-4.1×10^{-4}

* Cross section area 3.1 cm², length 14 cm and gap length 0.2 cm.

VII. Magnetic Stability

It is important to know how the magnetic flux will decrease in practical use by the effects of various demagnetizing factors. As for the magnetic stability, it is necessary to consider commonly the following effects.

- Natural aging (after effect)
- Temperature effect
- Mechanical shocks or vibrations
- Extraneous magnetic field
- Side shunt of the magnetic path

When the magnet is exposed to room temperature, its magnetic flux decreases gradually as shown in Fig. 15. It tends to spontaneous redistribution of domain magnetizations. Irreversible change of the magnetic flux of MT magnets by temperature effect was shown in Fig. 16.

Fig. 15. Natural aging at room temperature.

Fig. 16. Effect of heating temperature on the remanent induction.

Fig. 17. Effect of repeated impacts on the remanent induction.

Since the magnetic flux is reduced by mechanical shocks, the trend of demagnetization was examined by dropping the specimen on the brick from the height of 1m. The results are shown in Fig. 17. The effect of a stray magnetic field was examined by means of measuring the retained magnetization of magnets subjected to a 50 cycle alternative field. The results

are shown in Fig. 18. It was apparent that MT magnets exhibit the intermediate demagnetization characteristics of MK 1 (Alnico 120) and Cr steel, as were expected from those magnetic properties.

Fig. 18. Decrease of remanent induction when subjected to an alternative field.

When soft iron keeper was put and removed from the magnet repeatedly, the magnetic flux decreased gradually. The decrease of magnetic flux is as shown in Fig. 19.

Fig. 19. Effect of attraction and abstraction by ferromagnetic material.

VIII. Industrial Applications

As the production of MT magnets has been developed recently, its application to industrial field is being expanded for cheaper cost. Among its applications, some of commercial products are described in the following.

Watt-hour meters: Until late years ago, for the braking magnet on watt-hour meters, Cr steel was mostly used in Japan. Cr steel is inferior to other magnet materials in respect of its weakness against demagnetizing effects. If MT steel is substituted for Cr steel, the error in reading along with the lapse of time will be decreased and moreover the cost of watt-hour meter will be lowered.

The comparison of MT and Cr steel is shown in Table 6. It is evident that MT is more stable on demagnetization and maintains higher magnetic flux than Cr steel. The shapes of

Table 6. Comparison of Cr steel and MT steel for watt-hour meters.

	Dimension of magnets		Weight (g)	Magnetic properties after stabilization		Demagnetization percentage by an alternative field of 100 Oe.
	Area (cm ²)	Length (cm)		Total flux in air gap (Maxwells)	Braking factor (relative value)	
Cr Steel	1.8	17.5	230	10,500	14	28
MT magnet	3.2	12.9	265	14,000	29	8

Fig. 20. MT magnets used in a representative watt-hour meter.

braking magnets of watt-hour meters which are typical in markets, will be demonstrated in Fig. 20. Before the magnet is located in a watt-hour meter, it is usually subjected to stabi-

lizing treatment by means of demagnetization with an alternative field and heating. In the governmental Electric Laboratory of Japan, the demagnetizing characteristics of the braking magnets are inspected. The results of MT magnets are shown in Table 7.

Table 7. Comparison of demagnetizing properties tested by standard method of the Electric Laboratory.

Order of demagnetizing test		Test by mechanical impacts	Test by heating	Test in an alternative field
Method of testing		Dropping 30 times on wooden floor from the height of 2 m	Heating for 4 hours at 100°C	Subjecting to alternative field of 50 Oe.
Standard of Electric Laboratory		0.15%	0.75%	2.5
MT	A	0.06%	0.40%	0.14
	B	0.06%	0.24%	0.01

Magnetos: The Fe-Ni-Al alloy such as Alnico magnets normally are widely used for magnetos. To find out the possibility of substituting this magnet by the MT, various experiments were carried out. The magnet for magnetos must have the stability against these demagnetizing influences. The applicability of MT magnets to the fly wheel magnetos was valid only when ignition provided was to be comparable to Alnico in efficiency. The comparison of shape and efficiency between Fe-Ni-Al alloy and MT is shown in Table 8. Spark-gap length of more than 2 mm at 500 rpm is the required condition characteristic of this magneto. The considerable number of MT magnets have been practically used in this field.

Table 8. Performance test of MT magnets used in fly-wheel magnetos.

Material	Dimension of magnets		Air-gap length for sparking (mm)		
	Area (cm ²)	Length (cm)	200 rpm	500 rpm	1000 rpm
MT magnets	2.52	11.8	3.8	5.2	6.4
Fe-Ni-Al alloy	2.52	9.5	4.0	6.0	7.0

Other applications: At present, the demand of magnets for D.C. small motor was expanded. As the cost of MT magnets per effective magnetic energy is much cheaper, these applications are favorable. For a hysteresis motor, coercive force of magnet is necessary to be 100-200 Oersteds, and in this respect MT magnets have been used successfully.

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