A Survey of Magnetic Materials in Relation to Structure*

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The structure dependence of magnetic characteristics of the more important magnetic materials is discussed. The natural grouping into soft magnetic materials and hard or permanent magnet materials is used. In the first group are: magnetic iron, silicon steel and the alloys of iron, nickel and cobalt. The latter group includes the well-known carbon and alloy steels, and also the newly developed precipitation hardening α -solid solution types. Examples of the latter are the iron-nickel-aluminum and the iron-cobalt-molybdenum alloys.

In discussing the properties, the relationship to structure is emphasized. With the soft magnetic materials purification and the development of proper structural configuration are important. The special metallurgical control required to produce certain magnetic materials in dust form is described. The generalization is made that permanent magnet characteristics are due to precipitation effects resulting from decomposition of supersaturated solid

solutions.

Introduction

MAGNETIC materials may be classified according to properties into two groups: (1) soft magnetic materials, and (2) hard or permanent magnet materials. The differences in properties of the two groups are illustrated in Fig. 1, which shows typical magnetization cycles for the two types. The dashed line is a normal magnetization curve while the closed full line is known as the hysteresis loop.

A soft magnetic material is, in general, characterized by a steeply ascending magnetization curve; that is, large values of flux density are produced by small magnetizing forces. For certain applications where the flux density is low, the initial portion of the curve is important. For intermediate flux density applications the steeply ascending portion is of paramount interest while for higher densities, the upper portion is of prime importance.

Another term which is widely used in discussing soft magnetic materials is permeability. The permeability at a given flux density is the slope of the straight line joining that point on the magnetization curve with the origin. Algebraically, it is the ratio of the flux density, B, to the magnetizing force, H. It varies with the flux density, and for soft magnetic materials at low magnetizing forces, is a large quantity. The permeabilities usually reported in discussing magnetic properties are the initial permeability, μ_0 , and the maximum permeability, μ_{max} . The initial permeability is the initial slope of the magnetization curve. The

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maximum permeability is the maximum value that the ratio B/H attains.

Hysteresis is also of importance, especially in alternating current applications. The phenomenon of magnetic hysteresis results in a loss of energy in a magnetic material when the material is carried through a magnetization cycle. The loss of energy is proportional to the area of the hysteresis loop. In soft magnetic materials, the aim, in practically all cases, is to keep this loss a minimum.

A hard or permanent magnet material is characterized by a gradually ascending magnetization curve; that is, the material at all magnet-

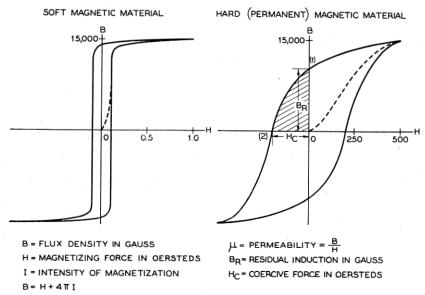


Fig. 1—Schematic representation comparing characteristics of soft and permanent magnet materials.

izing forces has a relatively low permeability. To attain practical saturation is difficult, requiring in some alloys magnetizing forces of thousands of oersteds. The important properties are associated with the demagnetization curve, that portion of the hysteresis loop between points (1) and (2) in Fig. 1. In general the hysteresis loop in its entirety is of little importance. The constants usually determined in permanent magnet investigations are the residual induction, B_r , and the coercive force, H_c . The product of these two quantities (B_r, H_c) , which is roughly proportional to $(BH)_{\rm max}$. on the demagnetization curve, has been considered by some 1 as a quality index for permanent magnet materials.

Summarizing the general properties of soft and of hard magnetic materials, in the former, high maximum permeability is associated with low coercive force and a hysteresis loop of small area. In the latter, a low permeability is associated with a high coercive force and a large hysteresis loop. The range of properties which can be obtained in magnetic materials by alloying and other metallurgical control is most remarkable. Permeabilities from 1 to over 600,000 and coercive forces from 0.012 to 600 oersteds, are attainable.

In manufacturing soft magnetic materials, the metallurgist strives constantly to eliminate those chemical elements (impurities) which broaden the hysteresis loop, while in the case of the permanent magnet materials he intentionally adds certain of the same, or other elements, and resorts to heat treatments which broaden the loop. Some details of these procedures with well known materials, and also with some more recently developed, are described in the following pages.

SOFT MAGNETIC MATERIALS

Magnetic Iron

Of the ferromagnetic elements iron, nickel, and cobalt, iron, at the present time, is the only one of industrial importance as a soft magnetic material in the unalloyed condition. Throughout the last half century, a continual improvement in the magnetic quality of iron has been effected until at present, laboratory samples have been prepared with permeabilities 2, 11 of over 200,000. Careful analysis of the data shows that this improvement has paralleled closely increases in the purity of the iron.

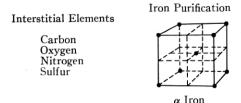
Since the production of iron of high magnetic quality is of considerable interest at this time, it seems worth while to examine, in some detail, the methods which have been used for producing high purity iron. Methods which have been developed for the purification of iron, have, in each instance, removed from the iron, elements (impurities) which

are harmful to easy magnetization.

The elements which enter into iron as impurities may be grouped, structurally, into two classes: substitutional elements and interstitial The substitutional elements exist in the iron structure at lattice points; that is, they substitute for iron atoms in the lattice shown in Fig. 2. These elements include most of the metals, for example: nickel, cobalt, manganese and silicon. The interstitial elements enter into the iron lattice at intermediate points; that is, they take Elements included positions in the structure between the iron atoms. in this classification are carbon, oxygen, and nitrogen. It is these latter elements which, although present in small quantities, cause severe strains in the lattice, and are believed, on this account, to be particularly harmful to the magnetic softness.

The solubilities of the interstitial elements at room temperature have been determined by a number of investigators. The most reliable values, which, because of the difficulties involved, probably should be considered tentative, place the room temperature solubility of carbon at approximately 0.008 per cent; ³ the solubility of oxygen at 0.01 per cent; ⁴ of nitrogen at 0.015 per cent; ⁵ and of sulfur at 0.015 per cent.

The solubilities of the interstitial elements vary with the temperature, increasing, in general, at higher temperatures. Varying solubility provides the possibility of precipitation hardening, which, in the case of iron, results in magnetic hardening. It is this latter type of



Substitutional Elements

Nickel Cobalt Silicon Manganese

Elimination of Interstitial Elements

Addition of Substitutional Element	2. Vacuum Melting	3. Hydrogen Treatment
C (Sol. in Iron) + Si \rightarrow C (Graphite) + Fe-Si O + Si \rightarrow SiO ₂	$C + O \rightarrow CO$ $Fe_x N_y \rightarrow Fe + N_2$	$\begin{array}{c} C + H \rightarrow CH_4 \\ O + H \rightarrow H_2O \\ N + H \rightarrow NH_3 \\ S + H \rightarrow H_2S \end{array}$

Fig. 2—Schematic representation of methods of purifying iron for magnetic purposes. The lattice structure shown is that for α -iron. The iron atoms are located at the cube corners and at the center and are represented by the circles shown.

hardening to which the magnetic aging of iron is attributed. This is possible because the supersaturated phase will in most of these systems precipitate the interstitial element at room temperature in a form which produces severe strains with deleterious effects on magnetic softness. Because of their harmful effects, the purification of iron for soft magnetic purposes has been in the direction of eliminating these interstitial elements. The three methods of purification which have been used are illustrated in the chart shown in Fig. 2.

The earliest method used was the addition in the molten state of a substitutional element which diminishes the harmful effect of interstitial elements in the iron lattice. Silicon* is an element beneficial in

* Messrs. Barrett, Brown, and Hadfield 7 reported the magnetic properties of iron-silicon alloys in 1900. More recent studies have been reported by Yensen. 7

this respect. Its action may be explained by postulating that its presence decreases the solubility of carbon in iron. The silicon enters the iron lattice as a substitutional element, and as such, is less harmful to the magnetic softness than the interstitial element, carbon, which is removed. Since silicon is also a strong deoxidizer, it will remove oxygen from the iron. These reactions do not completely eliminate the carbon and oxygen, and consequently, although a noticeable increase in magnetic softness is obtained, the high values of permeability achieved with the more efficient methods of purification are not

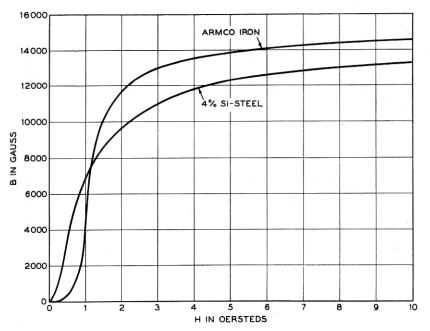


Fig. 3—A comparison of the magnetization curve of commercial 4 per cent silicon steel⁸ with that of Armco iron¹⁷ showing the superiority of the former at low induction densities.

reached. The improvement attained by this treatment is illustrated in Fig. 3 by comparison of typical magnetization curves for armco iron ¹⁷ and for a commercial 4 per cent silicon steel.⁸

The second method of producing a purer iron consists of eliminating some of the harmful elements by melting in a high vacuum. Under these conditions, the residual carbon and oxygen in the iron will combine to form carbon monoxide, which is removed by the exhausting pumps. At the same time, due to the dissociation of the nitrides at the high temperature, the nitrogen is similarly removed. Again, the

reactions do not go to completion, and residual impurities remain. Another obvious disadvantage is the fact that it would be only accidental if oxygen and carbon were present in the iron in the right proportions to eliminate one another. If the oxygen and carbon contents of the raw materials are known, adjustments can theoretically be made by adding either carbon or iron oxide, but if such a process is in commercial operation, the writers are not aware of it.

Yensen 9 reports a laboratory experiment in which this method of purification was used. A number of samples were prepared in high vacuum from electrolytic iron. To successive samples, increasing amounts of carbon were added to a maximum of 0.5 per cent. The maximum permeability increased with additions of carbon from approximately 25,000 for the sample with no addition to a maximum of 61,000 for a 0.06 per cent addition. From this value, the permeability dropped rapidly with further additions of carbon. The maximum value of 61,000 is explained as due to the fortuitous coincidence that the correct amounts of carbon and oxygen were present for optimum elimination.

Vacuum melting and vacuum purification present some interesting possibilities in connection with the preparation of magnetic materials. Commercial vacuum melting has been developed in Germany at the Heraeus plant, 10 where furnaces with capacities of 5 tons are in operation. It is stated that capacities of 20 to 30 tons can be achieved without large departures from the present designs. Vacuum melted metal is mechanically softer and works more readily than metal prepared by ordinary melting procedures. For magnetic alloys, the combination of vacuum melting and casting provides facilities for preparing material without contamination by the atmosphere. There seems to be no reason why one cannot go further and actually carry on refining operations in the furnace. The future of vacuum melting and casting appears extremely rich in possibilities, particularly for producing magnetic alloys where high purity is a primary consideration.

The third method of purification of iron is by treatment with an element which reacts with the interstitial elements to form gases which are removed. Hydrogen has been used for this purpose by Cioffi, and the method has been described recently. The treatment is carried out at high temperatures between 1300° C. and the melting point of iron. Cioffi states that ordinary amounts of carbon, oxygen, nitrogen, sulfur and phosphorus are reduced to very small quantities by hydrogen treatment. The excess hydrogen is either liberated as the metal cools, or, if it remains in the metal, is without harmful effects on the magnetic characteristics. A value of maximum permeability of 280,000

has been reported ¹¹ for iron prepared in this manner. In Fig. 4 is reproduced a curve from the unpublished work of Cioffi showing the enormous improvement in magnetic softness achieved by the hydrogen treatment.

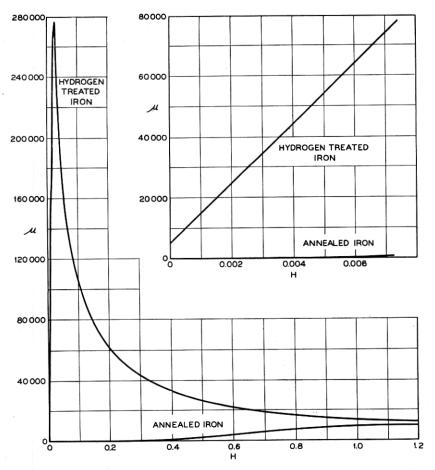


Fig. 4—The permeability of a laboratory sample of hydrogen treated iron compared with that of untreated iron—Cioffi¹¹.

The mechanism of the purification is outlined by the reactions in Fig. 2. The carbon is eliminated as a hydrocarbon gas, probably methane; the oxygen as water vapor; the nitrogen as ammonia or as nitrogen gas due to thermal dissociation of nitrides; the sulfur as hydrogen sulfide; and the phosphorus probably as phosphine. The most probable explanation to account for the purification is that, at the

high temperatures, the impurities continually diffuse to the surface of the sample where the reaction with hydrogen occurs. Another possibility is that the reactions occur within the body of the sample, in which case the hydrogen must diffuse in and the reaction products out. The first possibility appears the more promising in that the diffusion of reaction products through the iron samples would be difficult, since, in general, the products are rather large molecules.

Another type of high purity iron which is being used for magnetic purposes is carbonyl iron ¹² manufactured by the I. G. Farbenindustrie in Germany. It is prepared by first forming iron carbonyl under suitable conditions of temperature and pressure, and subsequently decomposing the carbonyl. For the best quality, the material is further purified by a low temperature hydrogen treatment. This iron is in the form of spherical particles, a few microns in diameter.

One use for this material is in cores of high frequency inductance coils. The cores of such coils are prepared by pressing the insulated magnetic particles into the desired core shape.

The material is also used in preparing iron sheet 13 and as an alloying constituent for the manufacture of iron-nickel alloy sheet. 14 The process of alloying consists of pressing the powders and sintering at a high temperature, followed by working and annealing in hydrogen or vacuum. Typical permeability values for iron sheet prepared in this manner are: $\mu_0 = 2000-3000$; $\mu_{\rm max} = 15,000$. The values are decidedly lower than those for a laboratory sample of hydrogen-treated iron shown in Fig. 4.

Iron Nickel Alloys

No alloy system has been more fruitful in yielding interesting and useful magnetic alloys than that of iron and nickel. In this system, the three regions marked with arrows on the constitutional diagram in Fig. 5 are of principal importance.

In the proximity of 25 per cent of nickel, which is the region of retarded phase change, alloys can be obtained at room temperature in the non-ferromagnetic state. How this is accomplished is evident from a consideration of the constitutional diagram. The γ solid solution above the magnetic transformation is non-ferromagnetic while the α solid solution is ferromagnetic. When alloys in the region of 25 per cent nickel are cooled, the transformation to the ferromagnetic state is not completed until temperatures below room temperature are reached. If the alloy is cooled until the ferromagnetic structure is obtained, the transformation to the non-ferromagnetic state does not occur on heating until the temperature has reached approximately 600° C.

Elmen ¹⁷ has investigated alloys containing more than 30 per cent of nickel which are characterized by great magnetic softness and have been called "permalloys," ¹⁷ the name suggesting high permeability. These alloys in the region of 40–55 per cent nickel find industrial application as soft magnetic materials. They are used in telephone and radio transformers and telephone relays. An alloy typical of this region is the one containing 45 per cent of nickel. A curve ¹⁵ showing

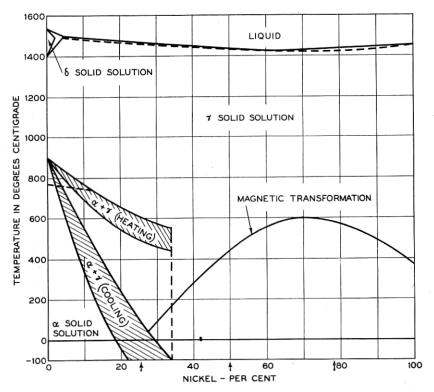


Fig. 5—The constitutional diagram of iron and nickel.

the permeability of this alloy is plotted in Fig. 6. From the data given on this curve, the material exhibits an initial permeability of 2000 and a maximum permeability of 16,000.

Another alloy in this region is the one containing 50 per cent of nickel which has been investigated by Yensen. He has applied the name "Hypernik" ¹⁶ to it. Yensen, by giving this material a special heat treatment, obtained an initial permeability of 4000 and maximum permeability of 70,000.

Arnold and Elmen ¹⁷ have investigated in great detail the alloys in the region of 70 to 80 per cent nickel. These alloys are used where extremely high initial permeability is required. One especially interesting use is in the loading of submarine telegraph cable. An alloy typical of this region is the one containing 78.5 per cent of nickel. Magnetization curves for this alloy after two different heat-treatments are compared with the curve for annealed armoo iron in Fig. 7. In the

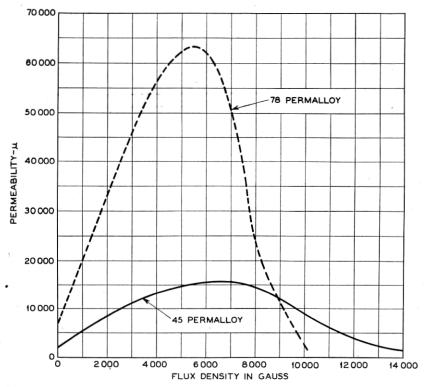


Fig. 6—The permeability of 45 permalloy compared with that of 78 permalloy— Pettit¹⁵.

air-quenched condition, in which the alloy is magnetically the softer, typical values for the initial and maximum permeability are 7000 and 65,000, respectively.

In comparing the properties of 45 per cent nickel and 78 per cent nickel alloys with reference to Fig. 6, the initial permeability is higher for the higher nickel alloy. Therefore, for very low magnetizing forces, the higher nickel alloy is characterized by higher flux densities. For higher magnetizing forces, the 45 per cent alloy has more attractive

properties in that the flux density is higher. In addition, the electrical resistivity of the alloy is approximately three times that of the 78 per cent alloy. The electrical resistivity is important in alternating current applications since the eddy current loss in sheet of the same physical dimensions varies inversely with the electrical resistivity of the material.

To the metallurgist the effects of heat treatment in establishing the magnetic properties of iron-nickel alloys possess a great fascination.

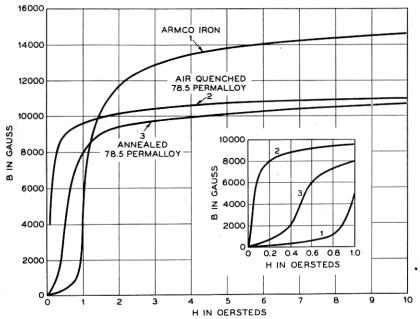


Fig. 7—Magnetization curves for 78.5 permalloy after two heat treatments—Elmen¹⁷.

The effects in question are illustrated by curves shown in Fig. 8. In the region of 78 per cent nickel, the maximum magnetic softness is exhibited after air-quenching, while increases in magnetic hardness are obtained after annealing or baking for extended times at low temperatures. The compositions near 50 per cent nickel are not as much affected by heat treatment.

Elmen ¹⁷ attributed the heat treating effect to the separation of the homogeneous phase, stable at high temperatures, into two phases on slow cooling. The two phase structure under these conditions would be less magnetic. More recently in Germany, investigators ¹⁸ have advanced the analysis by postulating that in the region of 76 per cent nickel, where the ratio of iron to nickel atoms is 1:3, the alloy can exist

in two states: one, a solid solution state in which the atoms of iron and nickel are in a disordered distribution; and two, a state in which the iron and nickel atoms are regularly distributed.

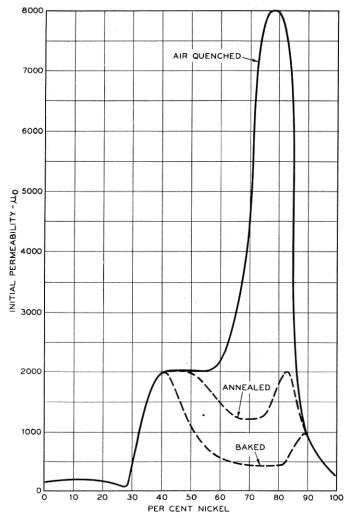


Fig. 8—Effect of heat treatment on initial permeabilities of iron-nickel alloys— Elmen¹⁷.

In the disordered distribution the lattice points are fixed but the atom positions are not; that is, any lattice point may be occupied by either an iron or nickel atom. In the regular or ordered distribution, the lattice points are fixed and also the atom positions. The second

arrangement may be clearer to some if it is considered a "compound," although strictly speaking, the arrangement does not correspond to a compound in the chemical sense. The regular arrangement is now ordinarily called a "superlattice" or a "superstructure," and as such, will be designated in this paper. If it is clearer to the reader, he may substitute for the term "superlattice" or "superstructure," the term "compound" without great sacrifice in the sense of the discussion.

With this picture in mind, the effect of heat treatment is immediately evident. Rapid cooling prevents the formation of the "superlattice" arrangement, which occurs somewhere in the region of 500° C., and a disordered distribution of atoms is retained. This is the distribution possessing the higher magnetic permeabilities. Slow cooling or baking promotes the transformation to a superlattice of lower magnetic quality.

Heat treatment effects are much less in the alloys in the region of 50 per cent nickel, and in fact would not be expected, since a face centered cubic binary alloy with the atomic ratio of 1:1 would not be expected to

have a special structure (superstructure).

The hypothesis of superlattice formation in the 75 per cent nickel region is supported further by the effect of heat treatment on other physical characteristics, for example the electrical resistivity and the tensile strength. That such special structures are formed in the solid states is well established from detailed studies of the copper-gold system.¹⁹ Unfortunately, in the nickel-iron system where the nickel and iron atoms are so near in atomic number, the detection of super-structure by x-ray methods appears, at the present time, a difficult task.

The effect of heat treatment and the general magnetic softness of iron-nickel alloys in the region of 75 per cent nickel may be explained by another hypothesis which returns for its basis to the considerations set forth in the section on high purity iron. The improvement in magnetic softness may be attributed, first, to the effect of nickel in decreasing the residual quantities of interstitial elements originally present in iron. Whether or not such an effect is present awaits a careful investigation of the interstitial element content of iron-nickel alloys. Secondly, assuming the interstitial elements the responsible factors, the effect ²⁰ of heat treatment on magnetic quality can be explained in that the quench (rapid cooling) retains the residual elements in solution, while slow cooling permits precipitation in a form more deleterious to magnetic quality.

The two hypotheses possess interesting possibilities, but it appears that further investigation will be required before the structural rela-

tionships are definitely established.

The permalloys in the region of 75 per cent nickel have been further modified by the addition of third elements ¹⁷ which are non-ferromagnetic. Molybdenum and chromium are elements of this nature which confer on the alloys certain characteristics of sufficient interest to warrant discussion. The effects of molybdenum and chromium on the magnetic quality are shown in Fig. 9, which indicates that higher initial permeabilities may be expected with the ternary alloys. The addition of a third element increases the electrical resistivity of the alloy. This is of importance in alternating current applications where eddy current losses are a consideration. In commercial manufacture the third element is particularly valuable, since it decreases the sensi-

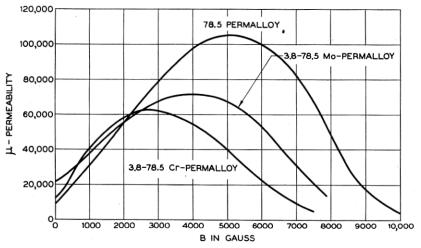


Fig. 9—Permeability curves obtained for laboratories' samples comparing the properties of chromium permalloy and molybdenum permalloy with those of standard permalloy—Elmen ¹⁷.

tivity of the magnetic properties of the alloy to the rate of cooling after annealing and, in fact, permits the attaining of high permeabilities with slow cooling. The significance of this from a structural viewpoint is that the third element keeps the iron and nickel atoms in a disordered distribution.

Iron-nickel alloys have a limited field of application in the form of thin tape in the hard rolled condition. A representative material is one containing from 30 to 70 per cent nickel and, in addition, a few per cent of aluminum or copper, which in the hard rolled condition has been called "isoperm." ²¹ The material is reported to have a low permeability of 50 to 75, which is fairly constant over a range of magnetizing forces of 0–10 oersteds. Laminated cores of this material have prop-

erties similar, but somewhat inferior to, those of magnetic dust cores ²⁷ which are described in a later section of the paper.

Iron-Cobalt Alloys

Iron-cobalt alloys in the region of 50 per cent cobalt are of importance for magnetic purposes because of the high values of flux densities obtained with medium magnetizing forces. The desirable magnetic properties of the alloy containing 50 per cent cobalt were first reported by one ²² of the authors. A very complete investigation of alloys over the whole range of cobalt contents has been described by Elmen ¹⁷ from whose paper the curves shown in Fig. 10 are taken. With

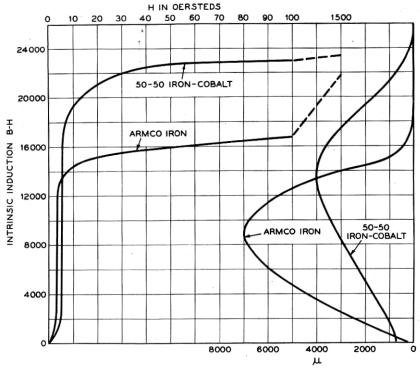


Fig. 10—Magnetization curve and permeability curve for 50–50 iron-cobalt alloy— Elmen¹⁷.

magnetizing forces from 5 to over 1500 oersteds the intrinsic induction is greater than that obtained with iron.

Because of anomalies in properties of the alloys in the region of 50 per cent cobalt, the structural features of the alloys become of interest. In addition to the maximum in intrinsic induction, the electrical con-

ductivity in this region at room temperature is greater than that of either of the components. The anomalous behavior was attributed several years ago by one of the authors,²² not to compound formation, but to a "peculiar solid solution." Kussman, Scharnow and Schulze ²² in a recent paper attribute the effects to the formation of superstructure in the body centered lattice which exists at this point. This, in a sense, is an extension of the earlier explanation. Here again, it appears that further work is warranted and is required before the structure is completely understood.

In connection with the metallurgy of the 50–50 iron-cobalt alloy, there are some interesting considerations. The binary alloy can be worked hot, but is extremely brittle when cold. This imposes definite limitations where thin sheet produced by cold rolling is required. The limitation can be overcome by adding a few percent of vanadium ²³ to the composition. This alloy can be worked hot, and after a quench from a high temperature, also can be cold-rolled to thin sheet. The magnetic characteristics are not greatly affected by the vanadium, provided the addition is small.

The structural changes resulting from the additions of vanadium are not clearly understood. It is known that vanadium retards the transformation which occurs in these alloys at approximately 900° C. and therein lies a possible explanation for its action. The high temperature modification is malleable as shown by satisfactory hot working properties. By quenching, sufficient of this modification may be preserved at room temperature, when vanadium is present, to permit cold rolling.

Iron-Cobalt-Nickel Alloys

In the ternary system, iron-cobalt-nickel, is a region in which the alloys exhibit an unusual and useful property, namely, constant permeability in low fields. Because of this characteristic, alloys in this region have been named perminvars. A typical alloy with marked perminvar characteristics contains 45 per cent nickel, 25 per cent cobalt and 30 per cent iron. The effects of heat treatment are illustrated in Fig. 11. The effect on the magnetization curve of air quenching, annealing, and baking at a low temperature is shown for this typical alloy. The extension of constant permeability to higher magnetizing forces by a low temperature bake is clearly demonstrated.

The constancy of permeability in perminvar has been explained in one hypothesis as due to the presence of two constituents in the alloy, one a soft magnetic material, the other a hard magnetic material. The experimental evidence for this hypothesis is derived largely from the constricted hysteresis loops which are obtained for these alloys after a baking treatment. A similar type of constricted loop can be obtained in a magnetic sample which is composed of thin parallel sheets of a soft and of a hard magnetic material magnetized longitudinally.

It may be that the two constituents result from a process similar to that described for the iron-nickel alloys in the region of 75 per cent

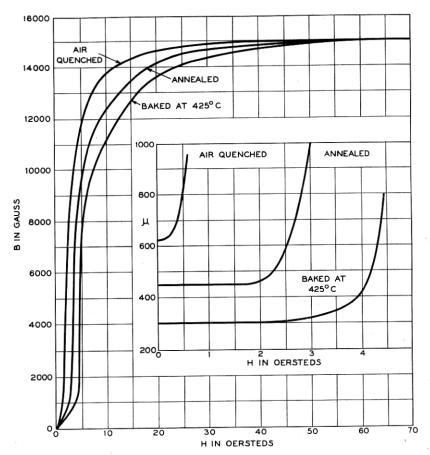


Fig. 11—Magnetization curves for a perminvar containing 45 per cent nickel, 25 per cent cobalt and 30 per cent iron after several heat treatments—Elmen ¹⁷.

nickel. In line with this, the baking treatment may form a superstructure ²⁴ which is magnetically hard, but to a greater degree, than the one in the binary iron-nickel alloys. It is probable that the similar effects of heat treatments in the binary and ternary alloys are due to similar structural transformations.

Heat Treatment of Magnetic Material in Magnetic Field 25

Recently it has been reported that radical changes have been produced in the magnetic properties of some ferro-magnetic alloys by heat treatment in a magnetic field. For example, the maximum permeability of the permalloy containing 65 per cent of nickel has been in-

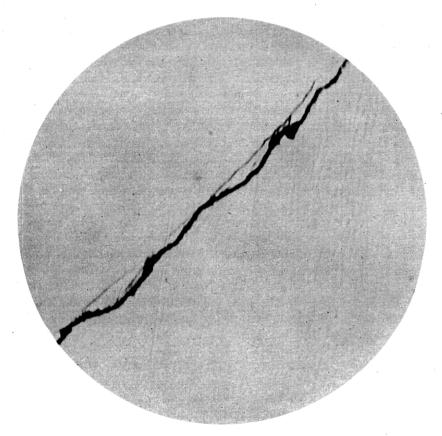


Fig. 12—Structure of an iron-nickel alloy, as cast, containing 80 per cent nickel and 0.030 per cent sulphur. The sulfide constituent exists as a brittle film surrounding the crystallites. Mag. $2000 \times$.

creased from 10,000 to 250,000 by heating to 700° C. and cooling slowly in a field of 15 oersteds. The maximum permeability has been further increased to 600,000 by so treating a specimen which had previously been treated in hydrogen at 1400° C. This value of maximum permeability is the highest which has been reported for any ferromagnetic material.

In the iron-cobalt-nickel alloys, high values of maximum permeability are also generally obtained by such a heat treatment, provided the magnetic change point is higher than about 500° C. Another feature in connection with this special heat treatment is that the heat-treated specimens have hysteresis loops which tend to have sharp corners and vertical sides.

An interesting explanation for the effect of heat treatment in a magnetic field is given by Bozorth.25 According to modern magnetic theory, when a specimen is cooled through the Curie point in the absence of an applied field, small regions or domains are magnetized to saturation in definite directions. The specimen in its entirety exhibits no external magnetic effects since the vector sum of the individual effects would be nearly zero and not detectable. The magnetization of the domains produces internal strains in the material due to lengthening in one direction and contraction at right angles (magnetostriction). By the application of an external magnetic field in the temperature region near to, but below, the Curie point, (1) the magnetic domains are oriented nearly in the direction of the applied field, and (2) the internal strains which were introduced by the local magnetization at the Curie point are removed by plastic flow of the surrounding domains. For this to occur, the temperature of Curie point must be higher than that at which plastic flow begins. Subsequent application of an external magnetic field at room temperature in the direction of, or opposite to, the direction of the magnetization at the higher temperature introduces no strains in the material tending to decrease the ease of magnetization. For that reason relatively higher permeabilities are obtained than with material heat treated in the absence of a field, in which case opposing strains are present.

The theory also offers another explanation for the effects of heat treatment on the permalloys in the region of 75 per cent nickel. This explanation will not be discussed further here except to say that it is concerned with the same magnetostrictive strains as have been mentioned above.

The fundamental idea of the theory proposed to account for the effects of heat treatment in a magnetic field, and the general effects of heat treatment on certain ferromagnetic materials, is that the magnetostriction associated with the local magnetization in the domains is sufficient to cause plastic flow. Experimental data indicate that the magnetostrictive stress is sufficient to cause such plastic flow at about the temperature which is known to be critical for the heat treatment of the permalloys.

Heusler Alloys

One of the most interesting groups of magnetic materials comprises the Heusler alloys ²⁶ which are ferromagnetic although composed of non-ferromagnetic elements. A representative alloy of this type is one containing copper, manganese, and aluminum. The magnetic properties are associated with the composition Cu₂MnAl, since the magnetic intensity of the alloys varies in proportion to the amount of this phase. These properties also depend upon structure. ²⁶ The ferromagnetic condition is obtained by quenching the alloy from approximately 800° C. This treatment produces a body centered cubic structure with a face centered superlattice. Manganese is essential to the composition; copper may be replaced by silver; ²⁶ aluminum by other trivalent or tetravalent elements—for example, tin.

Although possessing no properties of commercial utility, the alloys are extremely important in relation to magnetic theory. For those who may be particularly interested, a rather complete list ²⁶ of recent papers on the subject is given in the references.

Summary of Magnetic Characteristics of Sheet Materials

There are now available for magnetic purposes a large number of materials which are suitable for use over different ranges of magnetizing force and at different flux densities. In summarizing properties, the materials are grouped according to their adaptability at different magnetizing forces.

For low forces, where very high permeabilities are required, as in some signal apparatus, 78-permalloy is a suitable material. Molybdenum or chromium permalloy has properties similar to those of 78-permalloy but has higher initial permeability, is more readily heattreated and is to be preferred for alternating current applications because of the higher resistivity. Hydrogen-treated iron at low magnetizing forces also has properties similar to those of the 78-permalloy.

For small magnetizing forces where a constant permeability and very low hysteresis loss is required, the heat-treated perminvars are available. The flux densities in cores of this type, if constant permeability and low hysteresis loss are retained, must be kept below 1000 gauss.

For apparatus using higher magnetizing forces, iron-nickel alloys in the range of 40 to 55 per cent nickel, and the silicon steels are most suitable. These two materials are widely used in electric transformers.

In the region of fields of 10 to 50 oersteds and higher, the 50-50 iron-cobalt and vanadium-modified alloy are attractive because of the high intrinsic induction of approximately 22,000 gauss. This superiority exists even in fields of over 1000 oersteds, but in the higher

ranges, the saturation value is approached by that of the less expensive material, magnetic iron. In connection with high flux density applications, the permalloys, the perminvars, and modified alloys of this character are of little interest since they are saturated at comparatively low flux densities.

The data given here are not sufficient to select a material for a specific application since many other detailed properties must be considered in connection with each individual problem. The materials which have been described, however, cover, in a general manner, the entire range of magnetic fields.

Magnetic Alloys in Dust Form

For certain purposes where a substantially constant low permeability is desired, and in particular, for high frequency applications where eddy current losses are of consequence, it is desirable to produce the magnetic material in a fine powder, which is subsequently insulated and pressed into the desired core shape. One alloy prepared in this form is the permalloy ²⁷ containing approximately 80 per cent of nickel. This is a material suited for use at audio frequencies, for example in the cores of loading coils.

The alloy may be prepared in powdered form by a number of methods. In one method, which is essentially metallurgical in nature, advantage is taken of the effect of small amounts of added elements.²⁷ It has been found that the addition of a few thousandths of a per cent of sulfur to the iron-nickel alloy containing approximately 80 per cent of nickel produces a structure which can be hot-rolled to a small section, but which when cold, is exceedingly brittle and can be pulverized to a fine dust. The presence of other elements in small amounts also affects the properties of the alloy. One element, manganese, has an effect opposite to that of sulfur, and if present in sufficiently large amounts, nullifies the action of sulfur by producing a tough and malleable casting.

The structural behavior of sulfur and manganese in permalloy is interesting in explaining the embrittling action. Sulfur exists in the structure as microscopic films of complex sulfides at the crystallite boundaries as illustrated in Fig. 13. These films are brittle, and when they extend over the greater portion of the crystallite surface, produce an interface of weakness permitting easy pulverization. The satisfactory hot-working properties can be explained in that at the high temperature, either the sulfide film is malleable, or dissolves in the iron-nickel solid solution. If manganese is added to an alloy containing sulfur, the sulfide constituent is blackened, loses its continuous

characteristics, and becomes agglomerated as is well known in the case of iron and of nickel. In this form it has a minimum embrittling effect since contact between metallic crystallites exists over the greater proportion of the interface. The structure is illustrated in Fig. 14. If sufficient manganese is present, alloys containing fairly high percentages of sulfur can not only be hot worked, but are malleable when cold.

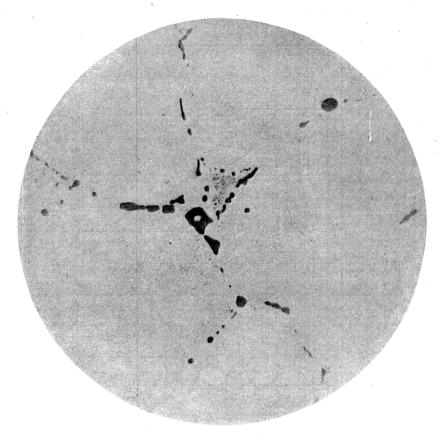


Fig. 13—Structure of an iron-nickel alloy containing 80 per cent nickel to which had been added 0.12 per cent sulphur and 1.05 per cent manganese. The sulfide constituent exists along the crystallite boundaries as grayish rounded inclusions. Mag. 2000 \times .

The production of magnetic dusts is unique as a metallurigcal process since the metallurgist is concerned with adding elements yielding a product which can be pulverized to a fine dust. Generally, the metallurgist exercises his greatest ingenuity to produce ductility and malleability in the end product.

HARD MAGNETIC MATERIALS (PERMANENT MAGNETS)

Permanent magnet materials, as pointed out in the introduction to this paper, are characterized by properties different from those of the soft magnetic alloys. With these materials, a high residual induction and high coercive force are required. Permanent magnets in use

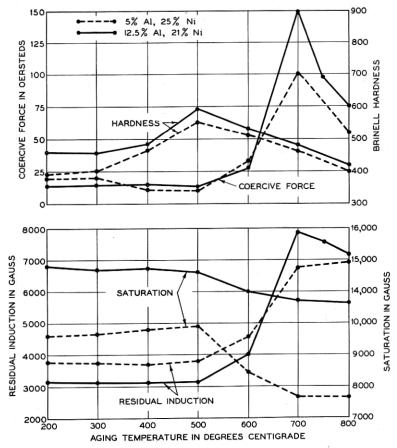


Fig. 14—Effect of aging treatment on typical iron-nickel-aluminum magnet alloys— Köster³¹.

commercially at this time are steels. High intrinsic induction is assured by a high proportion of iron in the composition; the magnetic hardness is assured by the addition of carbon, which, when the steel is suitably heat-treated, precipitates as a finely dispersed carbide throughout the matrix. This finely dispersed precipitate keys the

structure and furnishes resistance to change in magnetic condition, a property which is measured as coercive force.

Permanent Magnet Steels

Many types of steel have been developed for magnet applications.²⁸ The most important commercial ones are carbon-manganese, 1 per cent and higher chrome steels of which 3.5 chrome is typical, tungsten steel, and 35 per cent cobalt steel. Typical magnetic characteristics and required heat treatments are shown in Table I.

TABLE I
THE PROPERTIES OF PERMANENT MAGNET STEELS

Type of Steel	Typical Composition Per Cent	Heat Treatment	Magnetic Characteristics		
			H_c	B_r	$H_c \times B_r \times 10^{-3}$
Carbon-Manganese Steel	C—0.60 Mn—0.80 Si—0.20	Quenched 800° C. in water	40	9,000	360
Low Chrome Steel	Cr—0.90 C—0.60	Quenched 800° C.	50	9,500	475
3.5% Chrome Steel	Cr—3.5 C—0.90	Quenched 830° C. in oil	68	9,500	645
Tungsten Steel	W—5.0 C—0.7	Quenched 845° C. in water	60	10,500	630
35% Cobalt Steel.	Co—35 W—7 Cr—4 C—0.75	Quenched 940° C. in oil	220	9,500	2090

The heat treatment for the permanent magnet steels consists of a quench from a high temperature at which the carbon is in solution. During the period of cooling in the quench, the supersaturated solution precipitates the carbides. This is designated "quench hardening." The quenching treatment varies with the type of steel, but in general, quenching in either oil or water is used. In this connection, it is important that the austenite transformation occur during the quench, since austenite is non-ferromagnetic and its presence will result in low values of residual induction.

The carbon-manganese and 1 per cent chrome steels are the least expensive and enjoy a large use in low cost apparatus where space requirements are liberal. Typical values for coercive force and residual induction for these materials are 40–50 oersteds and 9000 to 9500

gauss. The 1 per cent chrome steel has the advantage over the carbon-manganese steel in that the desirable characteristics are produced by oil quenching while the carbon-manganese steel is ordinarily water-hardened. Water-hardening frequently results in cracked magnets and consequently a higher proportion of rejections.

Of slightly higher quality are the 3.5 per cent chrome and the 5 per cent tungsten steels.* Typical values of coercive force are from 60 to 70 oersteds, and of residual induction from 9500 to 10,500 gauss. In general, tungsten steel in the hardened condition has a higher residual induction than the other magnet steels. Tungsten steel requires water-quenching, while 3.5 chrome steel is oil-quenched. In addition, chrome steel is a somewhat lower cost material. For these reasons, in recent years it has been substituted to some extent in applications where tungsten steel was formerly used.

The best permanent magnet steel in commercial use is the 35 per cent cobalt steel, ²⁹ a complex alloy which contains, in addition to the cobalt, tungsten, chromium and carbon. Typical values of coercive force and residual induction for this material are 220 oersteds and 9500 gauss. Although this material is decidedly superior in properties to the other magnet steels, because of high cost, its use is limited to applications where space curtailment and apparatus requirements eliminate the cheaper steels.

New Developments in Permanent Magnet Alloys

Within the last five years, there have been a number of publications ^{30, 31} describing new materials which have properties of interest to engineers using permanent magnets. These materials are alloys with no intentional carbon additions, and, hence, are a radical departure in this field. The new magnet alloys solidify as alpha-solid solutions which by suitable heat treatment at a lower temperature decompose precipitating second phases. Contrary to the case with the iron-carbon alloys, the alpha-solid solution undergoes no phase change with decreasing temperature. Consequently the alloys have a coarse grain while the hardened magnet steels have a fine grained structure due to the intermediate phase change.

The permanent magnet qualities, however, result from the same type of metallurgical reaction that occurs in the carbon steels, that is, a precipitation of a second phase which is dispersed throughout the alpha-solid solution. The useful properties are secured by the usual precipitation hardening treatments; either a quench from a high

^{*}A commendable detailed discussion of tungsten magnet steels is given by Gregg in the recent book, Alloys of Iron and Tungsten, p. 212, McGraw Hill, 1934.

temperature during which precipitation occurs, or a quench followed by an aging treatment at a lower temperature to produce the precipitation. Examples of alloys of this type are the iron-nickel-aluminum alloys, which have been described by Mishima, ³⁰ and the iron-cobalt-molybdenum and iron-cobalt-tungsten alloys, which have been described by Seljesater and Rogers ³¹ and Köster. ³¹

Iron-Nickel-Aluminum Alloys

A representative composition of the alloys described by Mishima ³⁰ consists of 65 per cent iron, 25 per cent nickel and 10 per cent aluminum. The composition may be further modified by the addition of manganese, vanadium, cobalt, chromium, tungsten, molybdenum, or copper.

For the simple ternary alloy in the cast condition a coercive force of 240 oersteds and a residual induction of 9600 gauss have been reported by Mishima.³⁰ By slight modifications in compositions, coercive forces of over 500 oersteds in combination with residual inductions of approximately 9500 gauss are reported. Values for three Mishima alloys, presumably of different composition, have been reported recently by Steinhaus and Kussman ³⁰ and are given below.

Type of Mishima Alloy	Coercive Force Oersteds	Residual Induction Gauss
MK1	660	7,600
MK3	440	9,800
MK5	130	10,800

Köster ³⁰ has investigated the ternary equilibrium conditions for the iron-nickel-aluminum alloys. In the range of compositions of interest for magnet purposes, a surface of solubility varying with the temperature exists. It would be expected, therefore, that these alloys would be amenable to age hardening treatment. This has actually been demonstrated by Köster for the alloys of iron-nickel-aluminum. The curves shown in Fig. 14 are reproduced from his published data and will be recognized as demonstrating typical age hardening phenomena. The optimum aging temperature appears to be 700° C.

The fact that Mishima obtained high permanent magnet quality in specimens in the cast condition can be explained in that the precipitation of the second phase occurs during the simple cooling of the casting. It would be expected, therefore, that the magnet properties obtained would depend upon the casting dimensions and the rate of cooling. If this is true, it might be desirable in some magnet structures to subsequently heat treat the material to obtain uniform and reproducible results.

The fact that precipitation occurs is indicated more completely by examination of the photomicrographs in Figs. 15 and 16 of a typical

iron-nickel-aluminum alloy after quenching, and after quenching followed by aging at 1000° C. for 24 hours. There is no visual evidence of the second phase in the quenched specimen although some precipitation of submicroscopic particles undoubtedly has occurred. In the aged specimen, in Fig. 16, the second phase appears unmistakably.

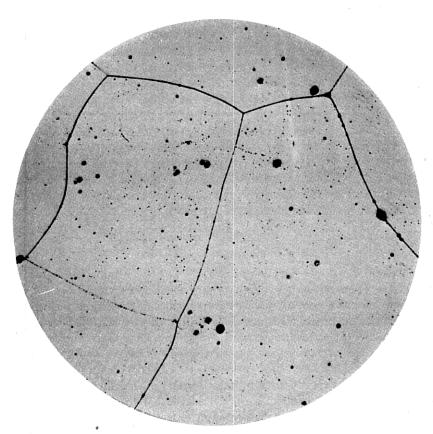


Fig. 15—Typical structure of an iron-nickel-aluminum alloy, containing 25 per cent nickel and 10 per cent aluminum, after quenching from 1200° C. in oil. In this condition the alloy has its optimum permanent magnet quality. Mag. 125 \times .

Iron-Cobalt-Molybdenum and Iron-Cobalt-Tungsten Alloys

The age hardening characteristics of alloys in these systems were established some years ago by Sykes, who developed the alloys for tools. Later, Seljesater and Rogers ³¹ reported that these alloys possess permanent magnet characteristics. A detailed report on the magnet properties has been made by Köster. ³¹ Of the two systems, it appears

that appreciably higher coercive forces are available in the one consisting of iron, cobalt and molybdenum. In Fig. 17 are shown the combinations of residual induction and coercive force obtained by Köster in the latter system with variations in molybdenum and cobalt content. Coercive forces of over 200 oersteds, associated with residual inductions of approximately 10,000 gauss, are given.



Fig. 16—Structure of the same alloy shown in Fig. 15 after a subsequent aging treatment of 24 hours at 1000° C. This treatment has resulted in the precipitation of a large amount of the second phase. Mag. $125 \times$.

The desirable properties of this class of materials are developed by a double heat treatment which consists of a quench from a high temperature followed by aging at a lower temperature. The alloys are precipitation hardening in the same manner as the non-ferrous copperberyllium alloys or lead-calcium cable sheath alloys. The heat treatment is illustrated in Fig. 18, which shows the effect of aging on the

properties of an alloy containing 15 per cent cobalt and 18 per cent tungsten reproduced from Köster's published data. The alloy was initially quenched from a temperature of 1300° C., and subsequently

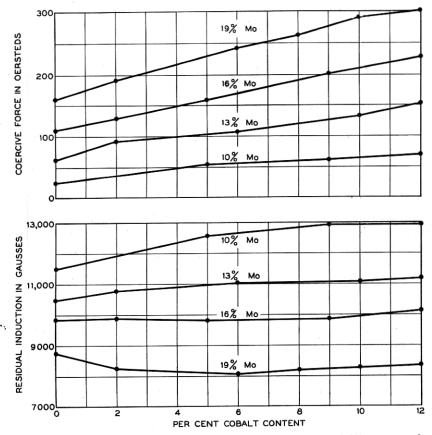


Fig. 17—Effect of different combinations of cobalt and molybdenum on the coercive force and residual induction of iron-cobalt-molybdenum alloys in the heat-treated condition—Köster³¹.

aged at temperatures from 300 to 800° C. The best results are obtained by aging at temperatures between 700° C. and 800° C.

The precipitation of the second phase in a typical iron-cobalt-molybdenum alloy is illustrated by a comparison of Figs. 19 and 20. The section in Fig. 19, which is for the alloy after quenching from 1300° C., shows a typical solid solution structure with a small number of rounded inclusions resulting either from the deoxidation of the melt or from incomplete solution at the high temperature. In Fig. 20, a section

of the same alloy after quenching and subsequent aging, the precipitate is visible as rounded particles within the grains and as elongated, needle-like structures within and along grain boundaries.

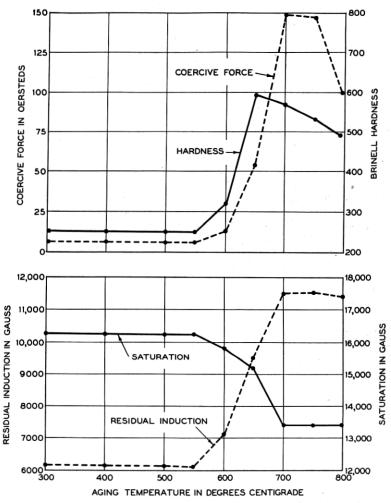


Fig. 18—Effect of aging treatment showing typical age-hardening phenomena for an iron-cobalt-tungsten magnet alloy—Köster³¹.

Köster ³¹ reports that these alloys can be rolled or otherwise shaped at sufficiently high temperatures and can be readily stamped at red heat. The alloys machine readily in the quenched condition but on account of the coarse grained structure and consequent brittleness they must be machined without shock.

Oxide Magnets

In this class of materials are included the metallic oxide magnets recently described in the literature by Kato and Tokei.³² The metallic oxide magnets open a new field of permanent magnet materials.

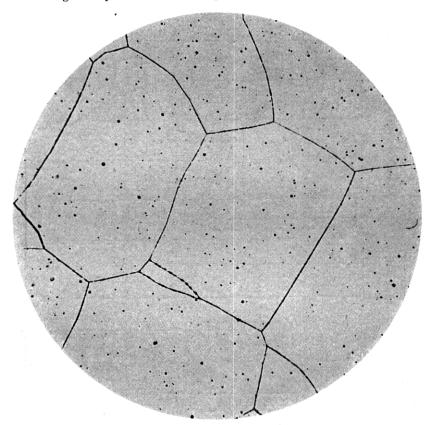


Fig. 19—Typical structure of an age-hardening iron-cobalt-molybdenum alloy, containing 12 per cent cobalt and 16 per cent molybdenum, after quenching from 1300° C. Mag. 125 \times .

These magnets are composed, according to the Japanese authors, in one instance of a solid solution of cobalt ferrite (CoFe₂O₄) in magnetic iron oxide (FeFe₂O₄). Although the method of manufacture and exact compositions are not completely disclosed, it appears that the preparation involves the powdering of the metallic oxides, compressing in a suitable die to the desired shape, and subsequent heating.

Cobalt ferrite magnets, it is reported, are not easily magnetized at room temperature; but if the temperature is raised, for example, to 300° C. the magnetization is readily accomplished. In this procedure the magnet is cooled in the magnetizing field. If the magnet has once been magnetized at the high temperature, succeeding magnetizations can be carried out at room temperature.

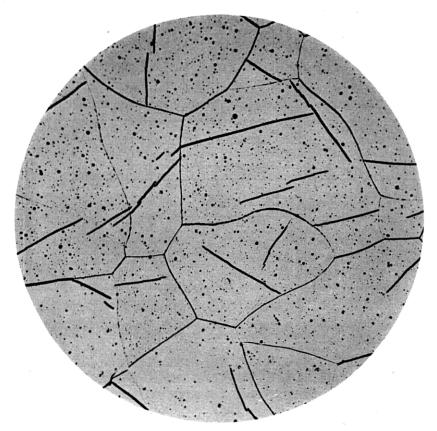


Fig. 20—The same alloy as shown in Fig. 19 after a subsequent aging treatment of 5 hours at 725° C. Mag. 125 \times .

The magnetic characteristics of the material are illustrated in Fig. 21, in which a hysteresis loop for a metallic oxide magnet is compared with one for carbon steel. The coercive force of the new material is approximately 600 oersteds and the residual intensity of magnetization, 320 units, corresponding to a residual induction of approximately 4000 gauss.

The high coercive force of the oxide magnet makes possible the more exact location of poles and permits the utilization of the material in

short magnets. Because of the lower density of the oxide compared with that of iron, the oxide magnets have approximately the same induction flux as metallic magnets on a weight basis.

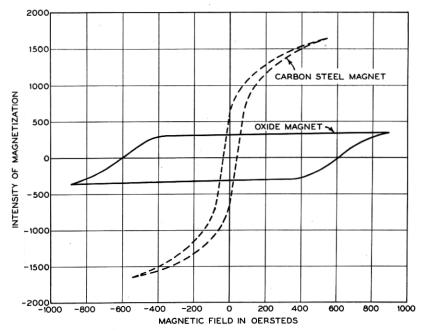


Fig. 21—Comparison of some magnetic characteristics of an oxide magnet with those of carbon steel from Kato and Tokei³².

The description of the materials given here has been obtained from the Japanese publication.³² So far as is known, no investigation of the preparation or of the properties of the material has been reported in this country.

Summary of New Permanent Magnet Materials

The properties of the new permanent magnet materials discussed are summarized in Table II.

TABLE II
THE PROPERTIES OF SOME NEW PERMANENT MAGNET MATERIALS

Materials	H_c -Oersteds	B _r -Gauss	$H_c \times B_r \times 10^{-3}$
Iron-Cobalt-Molybdenum AlloysIron-Cobalt-Tungsten AlloysIron-Nickel-Aluminum AlloysOxide Magnets	130–660	12,000-8,500 11,500 10,800-7,600 4,000	600–2,550 1,720 1,400–5,000 2,400

An important consideration in connection with the new materials is the broadening of the field of possible new magnet alloys. it had been generally considered that the field of useful permanent magnet materials was confined to the plain and alloy steels. The new metallic materials are systems which are precipitation hardening, and suggest possibilities for developing useful alloys in other similar systems of iron.

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