

INFLUENCE OF PURITY AND FABRICATION TECHNOLOGY ON THE PROPERTIES OF SOFT MAGNETIC Fe- 50Ni ALLOY

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Aliajul Fe-50Ni aparținând grupului de aliaje Permalloy cu aplicații în electronică, automată, telefonie, instrumente electrice fine, a fost elaborat din fier extramoale și nichel de înaltă puritate (99,95Ni). La elaborare s-a folosit topirea prin inducție în aer sub protecție cu insuflare de argon, în locul topirii în vid. Aliajul a fost forjat și apoi recopt în atmosferă de H₂ pentru a obține o microstructură favorabilă și o creștere a purității chimice și fizice. Când aliajului forjat i s-a aplicat tratamentul termic de recoacere în H₂, s-a obținut o scădere a durtății Brinell de la 182 la 130 daN/mm² și a câmpului coercitiv de la 0,5 la 0,4 Oe, împreună cu o creștere a permeabilității magnetice de la 16000 la 23000 Gs/Oe și a inducției la saturație de la 12300 la 14940 Gs. Luând în considerare creșterea grăunților produsă de tratamentul termic de recoacere se poate stabili un paralelism între scăderea de duritate prezisă de teoria dislocațiilor și modificarea proprietăților magnetice prezisă de teoria câmpului coercitiv.

The soft magnetic alloy Fe-50Ni belonging to the Permalloy group that finds application in electronics, automatics, telephony and fine electrical devices has been obtained from extra-mild iron and high purity nickel (99.95). Induction melting was carried out in air under a protective argon blanket instead of vacuum melting. Forging and annealing in H₂ atmosphere have been applied for obtaining a favourable microstructure and an increase in chemical and physical purity. A decrease of the Brinell hardness from 182 to 130 daN/mm² and of the coercive force from 0.5 to 0.4Oe as well as an increase of the magnetic permeability from 16000 to 23000 Gs/Oe and of the saturation induction from 12300 to 14940 Gs have been obtained when the forged alloy was subjected to the annealing heat treatment in H₂. If the grain growth during the annealing heat treatment is taken into consideration a parallelism may be established between the hardness decrease predicted by the theory of dislocations and the magnetic properties modification according to the theory of the coercive force.

Keywords: magnetic properties, Brinell hardness, grain size, Fe-Ni alloys

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1. Introduction

Iron and nickel together with cobalt are the only non-lanthanide ferromagnetic metals. Because of their availability these ferromagnetic transition metals are preferred instead of the few scarcely available ferromagnetic metals belonging to the lanthanide group.

One may wonder at the scarcity of the ferromagnetic metals among the 80 metallic elements in the Periodic Table. This fact is related to the special conditions that have to be met for a spontaneous parallel orientation of the atomic magnetic moments. Though a large number of metals have an atomic magnetic moment $\mu_{\text{at}} \neq 0$, most of them are paramagnetic because their magnetic moments have a random orientation. The internal molecular field that makes the magnetic spin moments to adopt a parallel orientation in ferromagnetic metals is non-magnetic in its nature being produced by the quantum exchange forces. According to quantum mechanics the energy of a system of atoms is

$$E = 2E_o + (C \pm A) / 2(1 \pm S^2) \quad (1)$$

where E_o is the energy of the isolated atom, C is the energy of the Coulombian interaction, S is the overlapping integral and A is the exchange integral. The stable state of the system depends on the algebraic sign of the exchange integral A . As shown in Fig.1 the criterion for the ferromagnetic state having a parallel orientation of the magnetic spin moments requires a positive value for the exchange integral A . The reason for which only three transition metals namely Fe, Co, Ni are ferromagnetic as indicated in Fig.1 is related to several conditions:

- the atoms must have an incomplete shell of electrons (either d or f)
- the incomplete shell must have a relative low electron cloud density near the nucleus; this means that the second quantum number l should be large and so only d and f subgroups have to be considered (transition metals or lanthanides, respectively)
- the atoms must not be too close together because the electron clouds will overlap so greatly that the exchange integral A becomes negative and normal cohesion with electrons of opposite spins is encountered
- the atoms must not be too far apart because the exchange integral A becomes very small and the resultant energy is too small to stabilize the ferromagnetic state.

By taking into consideration the two last conditions it becomes clear why in the Sommerfeld-Bethe diagram in Fig.1 the criterion for a transition metal being ferromagnetic is related with the dependence of the exchange integral A on the a/r ratio, where a is the interatomic spacing and r is the radius of the incompletely filled shell. On this account the Sommerfeld-Bethe diagram in Fig.1

based on quantum theory considerations may give practical suggestions for enlarging the group of ferromagnetic metallic materials. In fact manganese is closest to iron in the Sommerfeld-Bethe diagram (Fig.1) but it is not ferromagnetic because its exchange integral A is negative. By alloying Mn with elements that increase its interatomic spacing a the exchange integral becomes positive and ferromagnetic alloys are obtained such as the Heusler alloys in the system Mn-Cu-Al as well as the ferromagnetic compounds MnSb, MnBi etc.

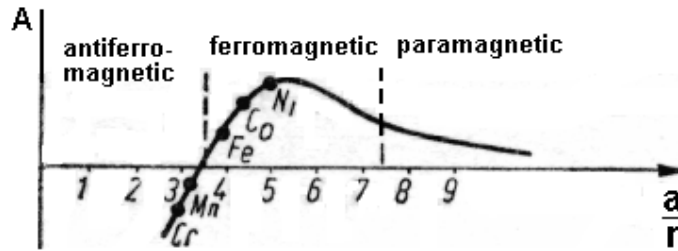


Fig. 1 The Sommerfeld - Bethe diagram indicating the location of the ferromagnetic transition metals as function of the exchange integral A versus the a/r ratio, from reference [1]

The researches in this paper are focused on ferromagnetic alloys in a system in which both components are ferromagnetic, namely the Fe-Ni system. The soft ferromagnetic alloys in the Fe-Co system have been the object of our investigations presented in previous papers [2,3]. Both systems offer opportunities to obtain soft ferromagnetic alloys with large saturation induction B_s , rectangular shape of the hysteresis loop, small coercive force H_c and high magnetic permeability μ_0 and μ_{\max} , for application in electronic and electrical devices where small currents require a large magnetic response.

2. Composition selection for the investigated Fe-Ni alloy

Fe-Ni alloys are soft magnetic alloys endowed with high magnetic permeability that allows them to give an enormous magnetic response at small currents as required in electronics, telephony, automatics, computer technology. Fig. 2 depicts the composition dependence of the main magnetic properties in the Fe-Ni system. Though the maximum magnetic permeability is located in a composition range close to 80Ni% (the classical Permalloy) we have selected a cheaper composition (50Ni%) because in spite of the magnetic permeability μ_{\max} being lower, the saturation induction is higher and the coercive force H_c keeps its value at the same low level that is specific for higher Ni contents.

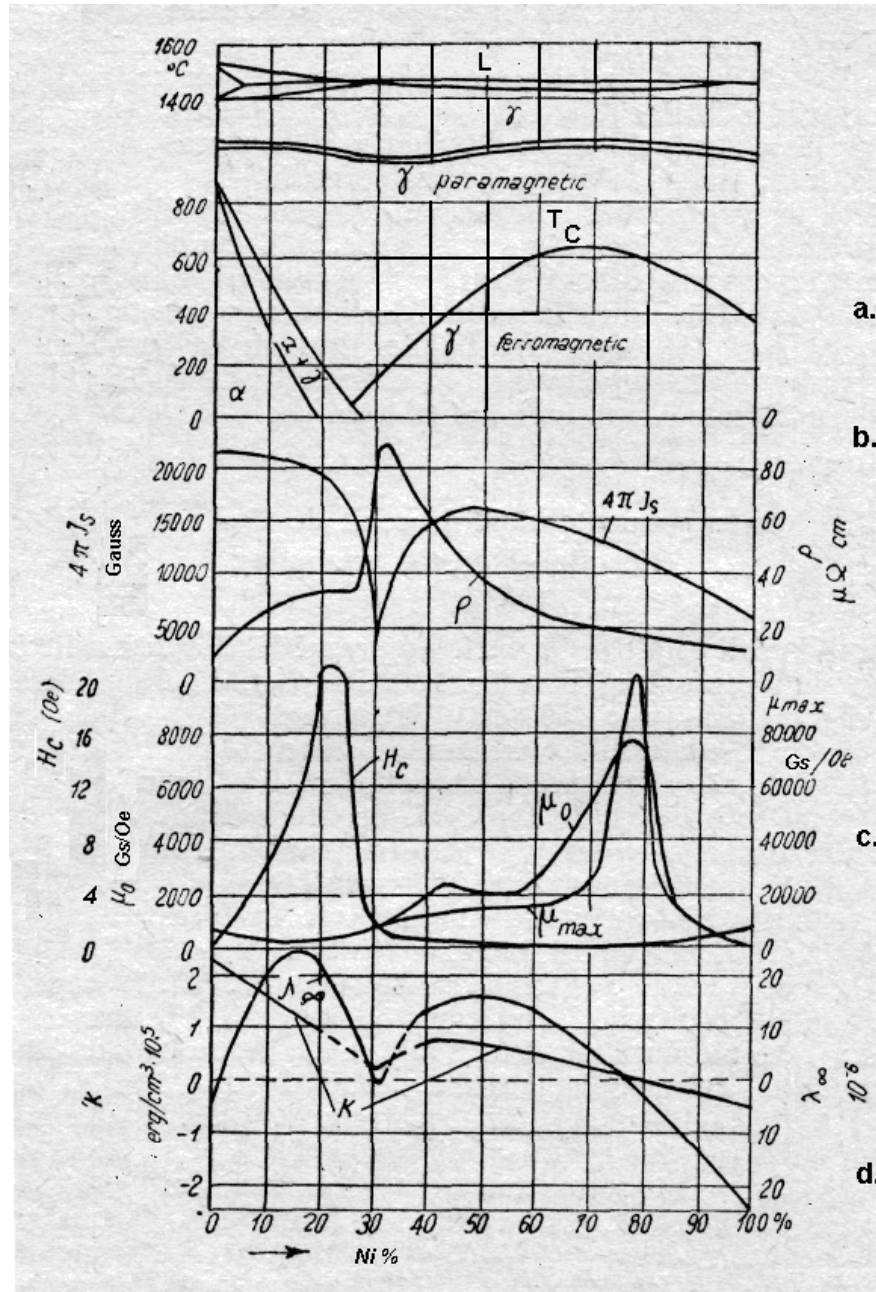


Fig. 2 Phase equilibrium diagram and composition dependence of the main magnetic properties in the Fe-Ni system reproduced from [4]

* In diagram 2b instead of the saturation induction B_s a different value has been considered namely $4\pi J_s$ where J_s is the saturation magnetization intensity according to relationship $B=4\pi J + H$

The Fe-50Ni alloys have been put on the market under various trade names: Isoperm B, Deltamax, Anhister, Permalloy P, Hypernic. The problem on which one has to focus on when fabricating a high performance Fe-50Ni soft magnetic alloy is not only to ensure the right content of the main chemical components but also to diminish the amount of the minor elements (chemical purity) and the degree of impurification with non-metallic inclusion, precipitated particles etc (physical purity). As depicted in Fig. 3 the content of minor elements such as O₂, S, P drastically deteriorates the soft magnetic properties (decrease of the magnetic permeability, increase of the coercive force).

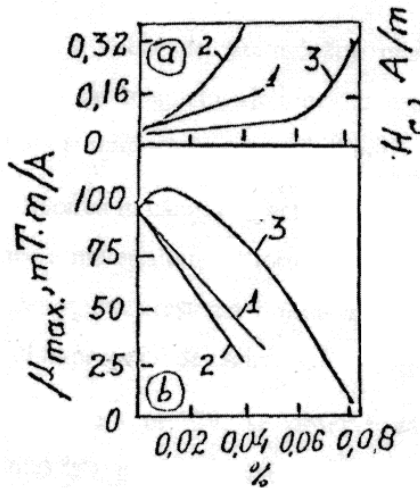


Fig. 3 Influence of oxygen (1), sulphur (2) and phosphorus (3) on the coercive force (a) and on the magnetic permeability (b) for alloy Fe-50Ni according to reference [5]

This is why in our research we have used the purest available raw materials in the alloy making process and paid attention not to deteriorate the alloy purity during casting and further processing. More specifically we have used Armco iron and high purity nickel (99.95) for obtaining the investigated alloy.

3. Results

A. Alloy making

An amount of 6 kg alloy of composition Fe-50Ni has been obtained by melting the components in an induction heated furnace and deoxidizing the melt with Si and Mn in an amount less than 0.3%. The alloy making and casting processes have been carried out under an argon protection blanket instead of making recourse to a vacuum furnace. This is a novelty that makes the alloy

melting cheaper and less pretentious; its efficiency was checked by the obtained magnetic properties. The melt was overheated at 1550 – 1600° C in view of casting and the ingot was stripped from the mould 40 minutes after casting. The chemical composition of two cast alloys is given in Table 1. It points to a Ni content very close to the value 50% we have had in view. The very low contents of the harmful elements C,S,P and deoxidizing elements Si and Mn comply with the requirements imposed by the influence of these elements on the magnetic properties reported in Fig. 3.

Table 1

Chemical composition of the investigated alloys

Code	Ni	C	Si	Mn	S	P	Fe
548	49.2	0.015	0.05	0.015	0.015	0.004	Balance
549	50.35	0.015	0.2	0.3	0.01	0.01	Balance

B. Hot working by forging

The ingots have been forged to obtain round bars $\Phi 30$ and $\Phi 40$ mm in diameter. Heating of the ingots in view of forging was carried out in a flame furnace by maintaining them for 3 hours at 1100 \pm 50° C. A 25 % cross section reduction per step was applied during the free forging process carried out by means of a 500 daN force hammer. The temperature of the forged bars at the end of the forging process was 850 \pm 50° C and air cooling was applied after forging.

C. Heat treatment of the forged alloy

An annealing heat treatment in protective H₂ atmosphere was applied in order to endow the final product with the best magnetic properties. The goals of the heat treatment were manifold: to diminish the level of the internal stresses and the content of the harmful impurities, to homogenize the chemical composition and to control the grain size [6-8]. To achieve this triple goal the following flow sheet was applied:

- (i) controlled heating inside the furnace at a heating rate not higher than 500° C /h up to the temperature 1100 \pm 25° C
- (ii) 3-5 hours maintaining in H₂ atmosphere at 1100° C
- (iii) a stepped cooling as follows:
 - slow cooling inside the furnace at a cooling rate not higher than 200° C /h in the temperature range 1100 down to 600° C
 - a faster cooling rate inside the furnace in the temperature range 600 down to 200° C at a cooling rate higher than 400° C /h, intended to avoid the appearance of the ordering transformation in the γ Fe-Ni solid solution that according to the phase equilibrium diagram starts at 503° C and is detrimental to the soft magnetic properties
 - air cooling from 200° C down to room temperature

D. Structure and properties characterization

The optical micrographs in Fig. 4 a, b, c were recorded for the alloy code 549 in Table 1, whose Ni content was very close to the nominal value 50%Ni and whose content in Si and Mn introduced as deoxidizers was at the upper limit of the admitted range.

In cast condition (Fig. 4a) a typical slow solidification structure is put in evidence. Large dendrites consisting of the face centered cubic γ Fe-Ni solid solution exhibit an almost uniform secondary arm spacing of about 40μ that point to a uniform solidification rate.

As seen in Fig. 4b forging was very efficient in uniformizing the microstructure. The dynamic recrystallization during forging has resulted in equiaxed γ grains. The temperature at the end of the forging process was low enough to produce a fine grain size.

As seen in Fig. 4c the annealing heat treatment after forging has resulted in a microstructure consisting of very large equiaxed twinned grains made up of the face centered cubic γ Fe-Ni solid solution. By comparing the micrographs in Fig. 4b and 4c it is obvious that the annealing heat treatment was very efficient in increasing the grain size that is desirable for improving the soft magnetic properties.

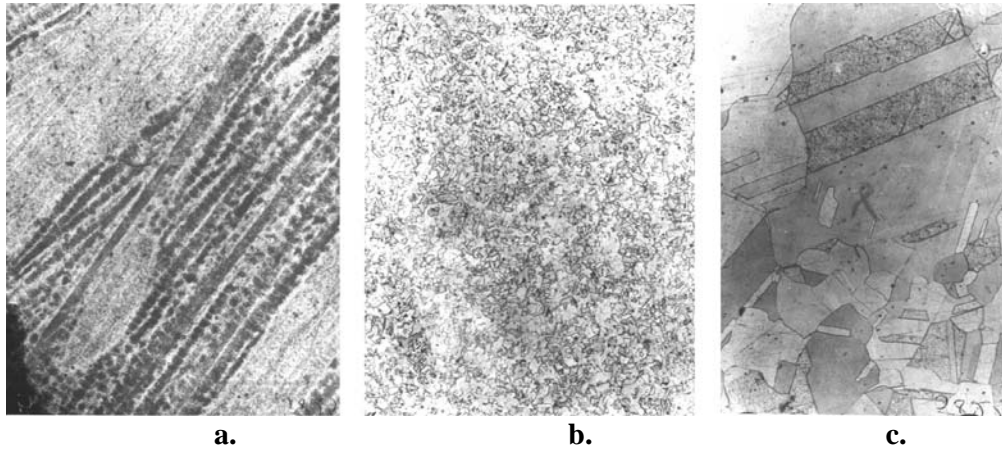


Fig. 4. Optical micrographs of the Fe-50.35Ni alloy (code number 549); etchant: FeCl_3 in aqueous solution; x50: a. cast condition; b. forged condition; c. heat treated condition

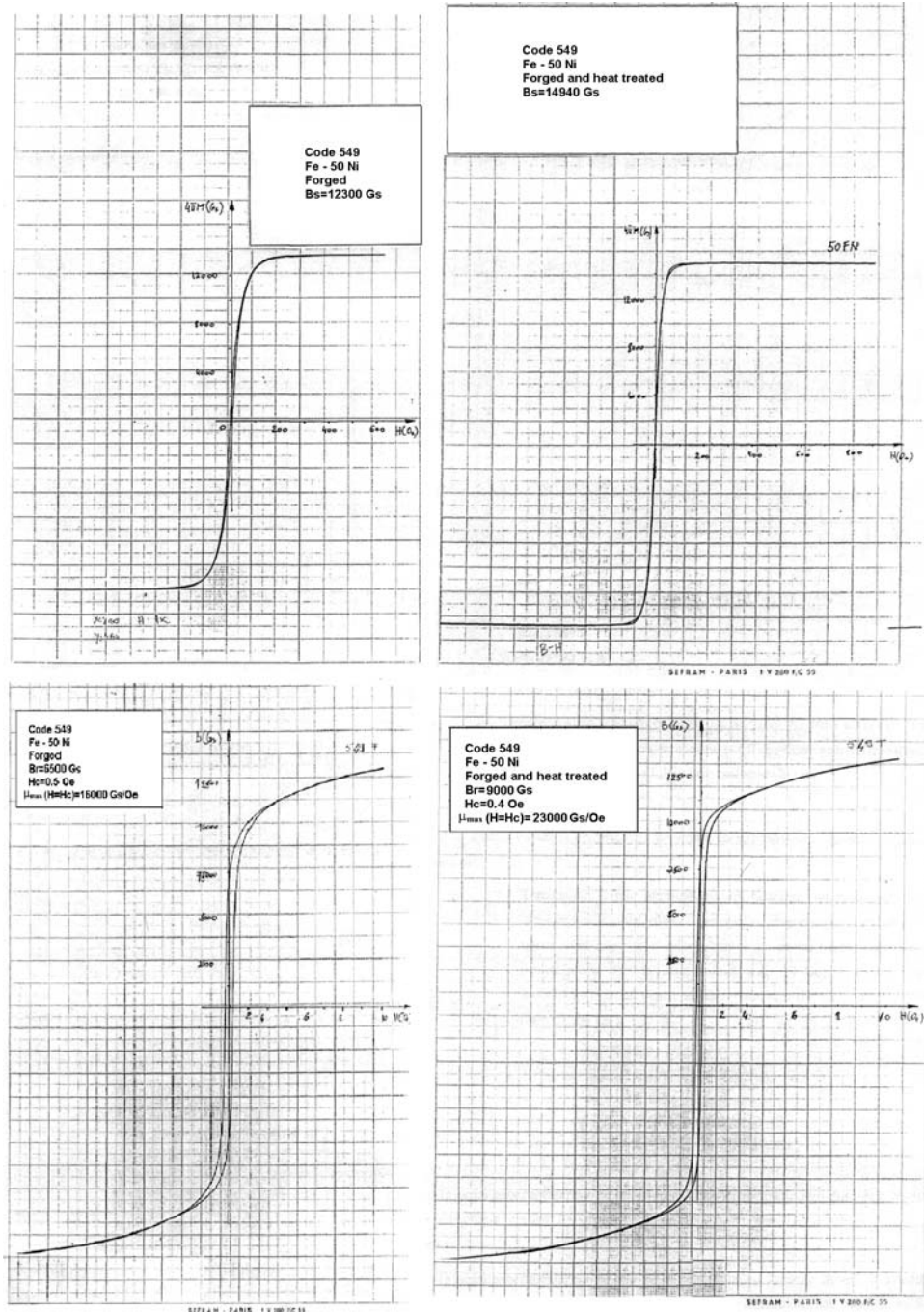


Fig. 5. Hysteresis loops and magnetic properties for the investigated alloy Fe-50.35Ni (code number 549)

The properties we have had in view to characterize the investigated alloys both in forged and heat treated condition were concerned with hardness and a set of magnetic characteristics (shape of the hysteresis loop, B_{\max} = saturation induction, B_r = remanent induction, H_c = coercive force, μ_{\max} = magnetic permeability obtained for an applied magnetic field $H = H_c$).

The Brinell test was applied to characterize the hardness H_B .

For obtaining the magnetic characteristics both cylindrical and torus shaped samples have been used. Fig.5 shows the recorded magnetization curves and points to the magnetic properties obtained for alloy Fe-50.35 Ni (code number 589 in Table 1).

Table 2 summarizes the obtained hardness and magnetic properties values. For sake of comparison the best magnetic properties reported elsewhere [9-11] for alloys in the same composition region of the Fe-Ni system are also included in Table 2.

Table 2

Brinell hardness and magnetic characteristics of the Fe-Ni investigated alloys

Code	Condition	HB [daN/mm ²]	Bs [Gs]	Br [Gs]	Br/Bs	Hc [Oe]	μ_{\max} [Gs/Oe]
548	Forged	176	-	-	-	-	-
	Heat treated	121	-	-	-	-	-
549	Forged	182	12300	6500	0.528	0.5	16000
	Heat treated	130	14940	9000	0.602	0.4	23000
Ref [9--11]	Heat treated	-	16000	10000- 20000	-	0.05-0.5	15000- 80000

4. Discussion

The values we have obtained for the magnetic properties of the alloy Fe-50.35Ni (Table 2) are close to those reported elsewhere for Fe-Ni alloys in the same composition region (48-50%Ni). Examination of Fig. 5 shows that the hysteresis loops are extremely narrow and not too far from the rectangular shape. This last feature is also to be expected from the high value of the B_r/B_s ratio. So, one may conclude that the purity of the raw materials and the technological processing conditions we have selected have been the proper ones for obtaining high soft magnetic properties for the alloys Fe-50Ni.

Some additional remarks both of practical and scientific concern are suggested by our experimental results.

The first remark consists in the fact that good magnetic properties may be obtained even if the processing of the liquid alloy is not carried out under vacuum but simply under a protective argon blanket. Of course this simplified technology

may be applied only if other conditions are observed, namely a short processing time that means induction melting and high purity of the raw materials.

The second remark is concerned with the beneficent effect of the annealing in H_2 on the magnetic properties as clearly shown by our results. Modern researches report even more spectacular increases in the magnetic properties of Fe-Ni alloys if the cooling after the annealing heat treatment is performed in a magnetic field that induces a magnetic texture in the soft magnetic material. We have not had in view such a thermomagnetic treatment because the favourable changes in the magnetic properties are at their best when the Curie temperature T_C is at its maximum and the magnetostriction coefficient λ_s and the magnetic anisotropy constant K are at their minimum. As seen in Fig. 2a and Fig. 2d this is not the case for the Fe-50Ni alloy. As a consequence as shown in Fig. 6 the efficiency of the thermomagnetic treatment is dependent on the nickel content of the Fe-Ni alloys. It is not efficient at 50%Ni content whilst its efficiency sharply increases for higher Ni contents.

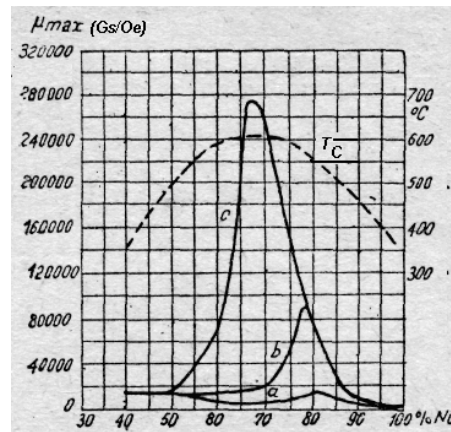


Fig.6 Influence of the cooling conditions during the annealing heat treatment on the magnetic permeability of various Fe-Ni alloys, according to reference [1]; the dotted curve for the Curie temperature is included for comparison,

a. slow cooling below 600°C; b. fast cooling below 600°C; c. slow cooling in a magnetic field

The third remark is concerned with the mechanism of improving the magnetic properties by applying the annealing heat treatment under a H_2 atmosphere. Besides its effect to diminish the content of the harmful impurities an important effect indicated by the micrographs in Fig. 4b and 4c is a considerable increase in grain size for the equiaxed grains of the ferromagnetic γ Fe-Ni solid solution. As pointed at by the results in Table 2 this grain size increase is associated both to a decrease in hardness and to a decrease in coercive force along with the increase of the remaining magnetic properties. Two theoretical relationships are available for

explaining the grain size influence on the mechanical and magnetic properties, respectively.

Indeed the theory of dislocations has established the well known Hall-Petch relationship:

$$\sigma_y = A + B / d^{1/2} \quad (2)$$

Because for ductile materials (like the face centered cubic γ Fe-Ni solid solution) the yield stress σ_y is correlated with the Brinell hardness ($H_B = 3 \sigma_y$) the Hall-Petch relationship is to be expected to be valid also for expressing the influence of the grain average diameter d on the hardness H_B .

On the other side the theory of the coercive force states that the coercive force H_c is increased by all metallurgical factors that make the displacement of the Bloch walls between the magnetic domains more difficult during the magnetization and demagnetization processes (non-metallic inclusions, precipitate particles, grain boundaries). For soft magnetic materials the expression of the coercive force

$$H_c = (\gamma / 2\mu_0 M_s) \cdot (8/\pi d^2) \cdot (\pi^2 / 4) \cdot d \quad (3)$$

may be written in a simplified form in function of the grain average diameter d :

$$H_c = a + b/d \quad (3')$$

The last relationship that has been checked with excellent results for pure iron points to a similitude in the influence exerted by the grain size both on the magnetic properties and on the mechanical hardness. Our results in Table 2 offer support to this similitude.

5. Conclusions

The Permalloy type alloys in the Fe-Ni system are recognized as excellent soft magnetic alloys of high magnetic permeability.

- Experiments carried out in this paper for obtaining and processing Fe-Ni alloys in the composition region 50Ni% have put in evidence the influence of processing conditions on the magnetic properties. Provided that the alloys are obtained from very pure raw materials (Armco iron and 99.95 pure nickel) the experiments have shown that good magnetic properties may be obtained even if induction melting under vacuum is replaced by induction melting in air under a protective argon blanket.

- Annealing in H_2 protective atmosphere in controlled cooling conditions was shown to improve the magnetic properties as compared to the forged condition both by improving the chemical and physical purity and by increasing the grain size of the ferromagnetic γ Fe-Ni solid solution.

- Improvements brought about by the heat treatment were recorded as high as 44%; 12% and 25% in the magnetic permeability, the saturation induction and the coercive force, respectively.

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