

CRYSTALLINE AND AMORPHOUS SOFT MAGNETIC MATERIALS AND THEIR APPLICATIONS – STATUS OF ART AND CHALLENGES

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The paper is a survey of fundamental properties and applications of crystalline and amorphous soft magnetic materials. The materials are firstly studied in the general frame of the electrical devices. A description is given for the main magnetic properties that characterize their quality and specific use: hysteresis, losses, anisotropy and magnetostriction. Then, one describes the most important soft magnetic materials classes: electrical steels, Fe-Ni and Fe-Co alloys, soft ferites and magnetic amorphous alloys, pointing out their future evolution.

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1. Introduction

There is no doubt that the magnetic materials represent today one of the most important issues of research. With a wide variety of structures, a lot of remarkable properties and infinity of application possibilities, these materials attract both the attention of physicists and engineers [1]. Using advanced experimental techniques of remarkable precision (neutron diffraction, magnetic resonance, Mossbauer effect, force microscopy, magneto-optic effects etc.) the research field has been permanently enlarged. The study of bulk crystalline materials has been followed by the magnetism of: samples with special geometry such as disordered systems, organic molecules and ferrofluids, each category with huge application fields. From the big electrical alternators to the computer peripherals or hundreds of sensors and miniature devices of the modern vehicles, the magnetic materials are present everywhere in our life. Ten years ago, the market of magnetic materials had already exceeded 6 G\$.

Magnetic materials are usually classified in soft and hard materials; and more recently a new category was added, that of semi-hard materials used especially as magnetic recording media. The soft magnetic materials (Figure 1) received their name in regard of the correlation between the low hardness of the ordinary steels and the easiness to reverse their magnetization. They have the capacity to concentrate the magnetic flux in different parts of the magnetic circuits.

The applications using soft magnetic materials are divided in two categories:

- (i) the conversion of the electromagnetic energy into mechanic energy or inversely (electrical machines) and the modification of parameters characterizing the use of electrical energy (electrical transformers). Actually, for producing and using the electromagnetic energy – the least expensive and the most ecological form of energy produced by humans – one cannot ignore the magnetic materials. They represent the yoke of many devices and engines. For these materials, the most important parameters are the permeability, the energy losses and the magnetization at saturation. The Fe-Si alloy can definitely be considered the most representative material for this class.
- (ii) the signal processing – a more restricted domain from the viewpoint of the total amount of materials used, but essentially for informatics, control and computers, electronics, telecommunications, with a large lot of materials requested for television, telephony, micromachines and small transformers with special utilization, transducers, magnetic recording heads, computer peripherals, microwave installations etc. In most of these applications, the

quantity of material used to produce one unit is small, but the number of units is very large. Fe-Ni alloys are the most representative materials for this category.

The main directions of development for the magnetic materials have been always imposed by the industrial needs. In the last decades, a considerable progress has been obtained thanks to the new technologies, especially the rapid solidification that allowed the obtaining of materials with new compositions and amazing microstructures [4]. There is no doubt that the most important result is the production of amorphous materials. Due to their remarkable properties, the magnetic amorphous have replaced many conventional (crystalline) soft materials in all kinds of applications, from DC to high frequencies. At the same time, the nanocrystalline materials have become up to date.

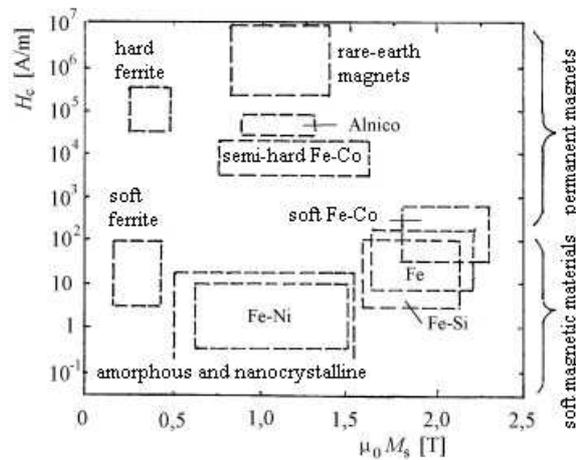


Fig. 1. Main classes of soft magnetic materials [2].

A deep theoretical knowledge of magnetic material properties represents an essential condition both in the modern approaches for designing the magnetic cores of electrical engines and devices and in the evolution of designing new materials. The problem presents interest also because nowadays one is concerned with the very efficiently use of electrical energy.

2. Fundamental properties

2.1 Characterization of soft magnetic materials

According to the usual criteria, a good soft magnetic material subjected to a low field should have the magnetization close to the saturation (about 80-90 %). This signifies a high magnetic permeability and a high flux density at saturation. If during its use the material must be magnetized along different directions, it should usually have the same behavior for each of these directions, i.e. the absence of the magnetocrystalline anisotropy. Even its magnetostriction should be not too high to avoid the induced anisotropies.

When the material is magnetized in AC, its hysteresis loop must be as narrow as possible because the losses are proportionally to the hysteresis loop area. On the other hand, a narrow B-H loop signifies a very low coercive field. In fact, the initial permeability and the coercive field are in a close relationship, the materials with high permeability having reduced coercivities. For permalloys, with an initial permeability of around 10^5 , the coercive field is as low as 0.4 A/m. The second important contribution to losses is due to the eddy currents, thus the material resistivity should be high enough.

The hysteresis loop of the material (B-H or M-H) is the best representation of all its magnetic properties. It could have a variety of forms, due to the non-linear and dissipative character of the phenomena involved. This means big difficulties to put it in an analytical form, but represents the source of the extraordinary richness and complexity of devices that use these materials. A lot of

problems that must be solved for developing the magnetic materials concern of obtaining appropriate hysteresis loops for those applications to which the materials are designed.

Obviously, no material may have all the ideal properties mentioned above. On the other hand, according to the type of the applied magnetic excitation, these properties are more or less important. The field frequency plays the main part from the point of view of the priority that must be given to one of these criteria. So, in stationary state, the best material is that having the largest magnetization at saturation. From this point of view, the best products seem to be with Fe-Co alloys, e.g. $B_s \cong 2.45$ T for 65%Fe-35%Co binary alloy. In the low frequencies domain (e.g. the industrial frequency) high permeability and magnetization at saturation and low coercivities are required. At higher frequencies, the condition of a high magnetization at saturation becomes less restrictive. Finally, at very high frequencies, the losses by eddy currents, proportionally with the square of the frequency, are more important while a very high resistivity becomes the main characteristic.

2.2 Energy losses

The reduction of energy losses is a very good criterion to illustrate the evolution of soft magnetic materials. During the last century, the energy losses of the best Fe-based soft materials having a high magnetic flux density, diminished almost exponentially. For a better understanding of this problem, one should note that the only fundamental cause of the energy dissipation in metallic magnetic materials is the induction of eddy currents due to a variable magnetic field. From the mesoscopic viewpoint (i.e., taking into account the samples domain structure), the mechanism that explain the energy losses is the movement of the domain walls.

The AC losses depend on the maximum magnetic flux density, B_m , of the excitation field and on its frequency, f , the sample width, D , (usually a magnetic sheet), the area of the hysteresis loop and the material electrical conductivity, σ . They have two main components that are strongly related: the losses caused by hysteresis and the (dynamic) losses caused by the eddy currents. For taking into account the big distance between the theoretical computed losses and the experimental measured ones, a third component was added, named *abnormal* or *excess losses*:

$$p_{Fe} = p_h + p_F + p_{an} \quad (1)$$

The hysteresis losses are proportionally to the frequency, while the eddy current losses are proportionally to the square of frequency, thus allowing an easy separation between these three components. For a sample subjected to sinusoidal variable induction field, $B(t)$, and if take into account the reaction of induced currents, the specific losses are given by:

$$p_F = \frac{\alpha}{\sigma D} \left(\frac{B_m}{\mu} \right)^2 \frac{\sinh(\alpha D) - \sin(\alpha D)}{\cosh(\alpha D) + \cos(\alpha D)}, \quad \alpha^2 = \pi f \sigma \mu \quad (2)$$

Even this equation does not reduce the difference with respect to the measured values; the gap being the consequence of the magnetic domains structure of the sample. The irregular and non-uniform movement of the domain walls, that locally induces eddy currents, is responsible both for the energy losses and for the Barkhausen noise that accompanies the magnetization process. (The magnetization rotations are important only near saturation and at high frequencies.) The excess losses result from the non-uniform distribution of the currents induced by the wall movement and from their interactions; the maximum intensity of these losses being obtained in the wall proximity. Adopting this idea and assuming that the 180° walls are independent, plane and spaced at, $2a$, Pry and Bean [5] gave for the excess losses:

$$p_{an} = \left(1,628 \frac{2a}{D} - 1 \right) p_F \quad (3)$$

A similar result has been found assuming the walls are elastic and fixed on the sheet surface, but in low fields only [6].

The model of Pry and Bean (PB), considering the uniform and regular movements of the walls, was proved to be too simple. There is a big variety of wall motions or of their different parts, to which the nucleation process and the wall interactions that were not taken into account should be added. Moreover, the PB model represents a deterministic approach of a physics problem with a huge complexity and doesn't allow inferring any relation between the material coercivity and its losses.

In spite of its weaknesses, the PB model contains all the suggestions to reduce losses, sustained both by physics and technology. So, one can control p_h by treatments that develop an appropriate microstructure p_F and p_{an} can be decreased by reducing the electrical conductivity, by increasing the solvent content and/or by diminishing the sheet thickness. The excess losses are further reduced by refining the domain structure by technological or metallurgical procedures, capable of creating regularly disposed centers for pinning the walls. Then, the walls are more or less immobilized and the global magnetization of the sample is obtained mainly by the magnetization rotations – mechanism whose energy price is reduced only to the eddy currents losses. Most of these methods have always damaging consequences from other points of view: the diminishing of the saturation flux density, the reduction of the volume fraction of magnetic materials in electrical sheets, etc.

Nowadays, almost ideal solutions for 2 particular problems are requested: a general statistical approach, because the magnetization process is a stochastic phenomenon. From this point of view, the most promising attempt seems to be the statistic theory proposed by Bertotti [9]. According to this theory, the material is considered to be formed by several magnetic objects that are elementary regions, including one or more walls and for whom the magnetization reversal is produced in a high correlated way. The theory has the merit of giving an actual physical sense to the losses decomposition [1]. So, the losses, p_h , represent the term that corresponds to the consumed energy that do not depends on the frequency, in the domain of frequencies currently used. p_F are the losses caused directly by the movement of the walls that are supposed independent, while the term p_{an} is connected to the correlation effects between the eddy currents that are locally induced by the different Barkhausen jumps. At the same time, the theory allows a correct prediction of losses in the case of non-sin excitation, starting from a few standard measures and the integration of the Preisach hysteresis model with the concept of losses separation, that led to the so-called dynamic Preisach model (DPM), that allows the prediction of the hysteresis loop evolution depending on the excitation frequency. Nowadays, the Bertotti's theory is particularly interesting due to the strong trend for using higher and higher excitation frequencies and more and more complicated wave shapes in electrical machines, often involving a big effect in the laminations of their magnetic cores. The calculus difficulties may be easily taken over by combining the DPM with the finite element method [10, 11].

2.3 Magnetic anisotropy

The lower the crystals intrinsic anisotropy the softer the materials are. On the contrary, the crystals with a structure characterized by a low symmetry have a very strong anisotropy, the uniaxial (hexagonal) cobalt representing the classical example.

For creating new materials – appropriate to various applications imaged – it is important to know how the anisotropy can be controlled: by the material composition and by thermomechanical procedures applied to the sample.

On the other hand, for understanding the order of anisotropy constants, one should remind that in the 3d transition metals with crystalline structure, the crystalline field acts directly on the orbital angular moment and indirectly only on the spin moment. Consequently, the anisotropy constants K at room temperature have reduced values: $48 \cdot 10^3 \text{ J/m}^3$ for Fe, $4,5 \cdot 10^3 \text{ J/m}^3$ for Ni and $410 \cdot 10^3 \text{ J/m}^3$ for Co. On the contrary, in the 4f transition metals (rare earths) the orbital moment remains almost unchanged and the constant K is higher: $\sim 10^6 \text{ J/m}^3$ and even more [1].

The situation is changing completely for amorphous materials. In the 3d metals-based alloys, the anisotropy is very low with respect to the exchange energy and because of the short-range order (2 or 3 lattice constants) the magnetocrystalline anisotropy effect is almost canceled.

For soft crystalline materials whose behavior is dominated by the magnetocrystalline anisotropy, the main aim of thermo-mechanical treatments is to produce the micro-structure and the texture able to control the coercivity, the rectangularity of B-H loop and losses. On the contrary, in

amorphous materials and permalloys, where the magnetocrystalline anisotropy is very low, the stresses and the induced uniaxial anisotropies have the prevalent role for setting up the easy axis.

Usually, the uniaxial anisotropy ($K_u \sim 10^2 \dots 10^5 \text{ J/m}^3$) in permalloys or amorphous materials is induced by annealing (thermo-magnetic treatments - TTM). The TTM may be realized in longitudinal or transversal field. The former assures a rectangular B-H loop, a high permeability and the best properties for using the material in DC applications, while the latter leads to reduced eddy current losses (Fig. 2).

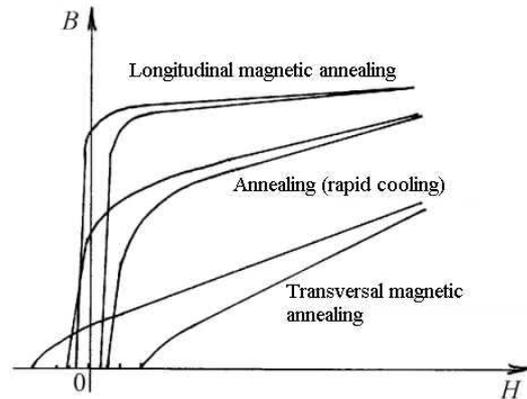


Fig. 2. Typical hysteresis loops for amorphous alloys subjected to annealing.

The shape anisotropy is predominant in magnetic circuits produced from sintered powders and those having air-gaps, because the demagnetizing fields are stronger than the coercive fields. Such circuits are used for obtaining flattened hysteresis loops or when one tries to realize devices with constant magnetic permeability.

2.4 The effect of stresses

For soft magnetic materials, it is convenient to have a magnetostriction constant λ_s as low as possible, to get rid of the stresses effect. λ_s of the best materials is around 10^{-7} . However, the permendur has $\lambda_s \cong 27 \cdot 10^{-6}$ along the axis [001] and $\lambda_s \cong -5 \cdot 10^{-6}$ along the axis [111], while in Fe-based amorphous alloys λ_s is isotropic and is around $30 \cdot 10^{-6}$. For the rare earth-based alloys λ_s is higher reaching values of 10^{-5} .

The magnetostriction is important from the point of view of producing and using soft magnetic materials. It helps to realize various types of sensors or the improvement of certain thermal treatments. However, in most cases, the magnetostriction represents a stress that has to be taken into account by the designer, especially because it may induce an important anisotropy when the material is liable to stresses.

This effect entirely justifies the annealing of magnetostrictive materials that must reduce the residual stresses in the material after its manufacturing. This is an even more important constraint in the case of materials with a low anisotropy, like permalloy and amorphous alloys, that must be annealed only after reaching their final shape.

Another unpleasant effect of magnetostriction is the periodical variation of the core dimensions. In transformers, these variations cause vibrations that are the typical noise of all operating transformers. The magnetostrictive variations of the dimensions are produced especially when the global magnetization is realized by the rotation of the magnetization vector. This is the reason why the amorphous cores of transformers are annealed in the presence of a longitudinal field that induces an easy axis along the long axis; the magnetization rotation being then avoided. On the contrary, when the materials contain many domains with transversal magnetization, the magnetostrictive effects due to the magnetization rotation induce the resonance, which generate important energy losses.

The behavior of crystalline materials is very similar. However, the best polycrystalline alloy Fe-3%Si used for transformers, never has all the grains well oriented and so, under the bend of the B-H curve, the magnetization rotation will appear again with all associated magnetostrictive effects.

A completely different approach is to choose materials with almost zero magnetostriction: both crystalline like Permalloy 20%Fe-80%Ni and Fe-6.5%Si, and amorphous like Co-based alloys with 3-15 % Fe and/or Ni. There are also alloys like Sendust (85%Fe-10%Si-5%Al) or the modified permalloy (17%Fe-79%Ni-4%Mo), for which the magnetostriction and the intrinsic anisotropy were simultaneously minimized. One can use the same solution for amorphous materials by choosing compositions capable to assure almost zero magnetostriction and annealing the sample in the presence of a magnetic field that is rotating slowly. So, one obtains the highest possible permeability for devices like magnetic recording heads.

3. Electric steels

The electrical steels are produced in much larger quantities than all the other soft magnetic materials, because they represent the materials used for the manufacturing of the almost all the magnetic circuits from the high power devices. The non-oriented Fe-Si alloy is used for producing electrical machines and the same alloy with oriented grains is used for producing the usual transformers.

3.1 Fe-Si alloys

These alloys are mainly dedicated for the manufacturing of electrical machines and transformers that operate in alternative current of industrial frequency (50 or 60 Hz, sometimes 400 Hz). They represent the best possible combination between good magnetic and mechanical properties and a reasonable price. Despite its numerous qualities, the soft iron used in AC devices has two major drawbacks: a high electrical conductivity and its corresponding magnetic hysteresis, both of them leading to important energy losses. For reducing these losses, the iron must be alloyed with other elements, the most important being the silicon, despite of its high price.

The Fe-Si alloy is magnetic for Si contents up to 33 wt. % (50 at. %). The silicon is soluble in α Fe until 15 wt. % (25 at. %), substituting Fe atoms without notable modifications. The most important effect of the diluted alloy is the elimination of the γ Fe phase for a content of Si below 2,2 wt. %. This property allows the sheet re-crystallization at high temperatures (1000-2000°C), avoiding that the material passes through a critical point when it is cooled.

The addition of Si brings several improvements:

- (i) Increases the material resistivity and reduces the losses due to eddy currents (Fig. 3). On the same figure one illustrates the effect of alloying with other elements and one should note a similar effect for aluminum. Also note that the impurities, even in a low concentration, just like the precipitates and local stresses are able to modify dramatically the shape of the magnetization curve [13].

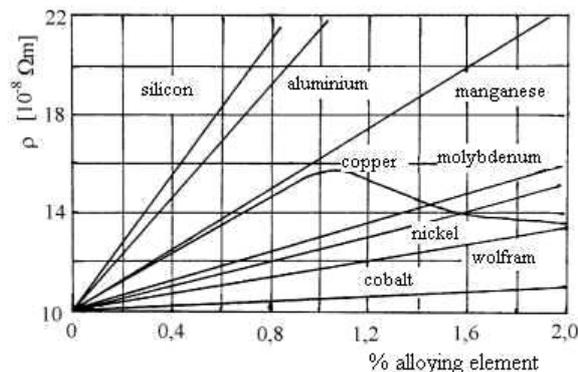


Fig. 3. Variation of the electrical resistivity of Fe-based alloys vs. addition element.

- (ii) Reduces the magnetostriction. The effect is more important when the Si content is higher (Figure 4), and involves the reduction of the coercive field and hysteresis losses, and consequently reduces the noise of operating transformers. From this point of view, the best composition is around 6 wt. %;
- (iii) Reduces the magnetocrystalline anisotropy (Figure 5) and therefore the permeability of the non-oriented alloy increases;
- (iv) Reduces the magnetic aging by caption of the interstitial atoms (carbon) assuring better stability in time;
- (v) Increases the strength and the rigidity of the considered alloys;
- (vi) Suppresses the iron crystalline phase transition from α to γ phase, allowing treatments at high temperatures ($> 900^{\circ}\text{C}$), favorable for re-crystallization and diminution of the internal stresses, while the other components (e.g. carbon) exhibit the opposite effect. This allows an easier growing of grains, producing a well-marked texture and facilitating the annealing of the alloys.

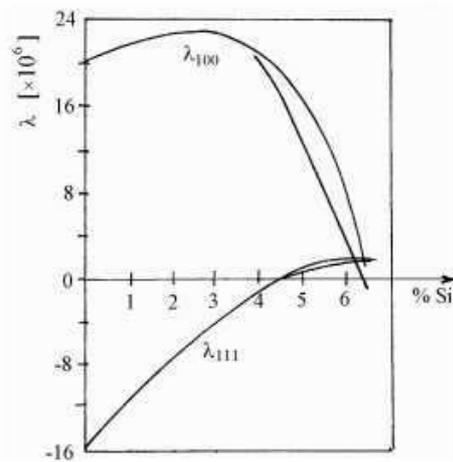


Fig. 4. The values of magnetostrictive constants λ_{001} and λ_{111} of the Fe-Si alloys versus Si content; the lower curves in the interval 4 - 7 wt. % Si correspond to a slow cooling.

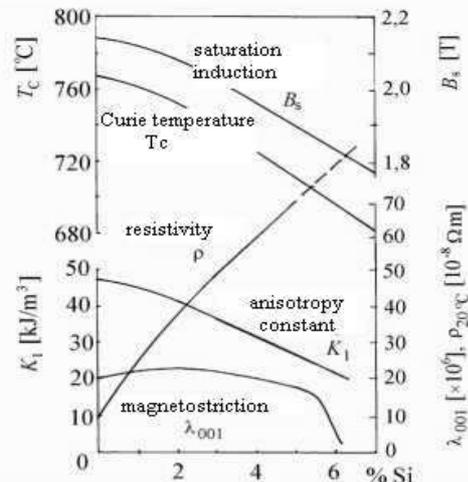


Fig. 5. The variation of the electric and magnetic properties of Fe - Si alloys in respect with their Si content (©1971 IEEE).

From Fig. 5, one can observe the harmful effects of the alloys containing Si: the diminution of the saturation flux density that, at room temperature, decreases from 2.16 T for soft (pure) iron to 2.01 T for Fe-3%Si; the reduction of the thermal conductivity; the decreasing of the Curie temperature and the reduction of the ductility of the alloy, which is fragile above 4.5%Si, fact that imposes this limit for Si content. For most of the alloys used for transformers, the content of Si is 3-4 wt. %.

An important way to reduce the losses by eddy currents is the use of smaller sheets, isolated by a mineral layer. The minimum losses are achieved for a sheet thickness comparable to the field penetration depth; at industrial frequency this is between 0.3 and 0.7 mm (see also [14,15]).

There are two big classes of magnetic sheets: the non-oriented sheets (N.O.) and the sheets with oriented grains (G.O.) that are used in different application fields and are never competing. The N.O. sheets are used for producing the magnetic cores of electrical machines and many other devices, while the G.O. sheets are used for the magnetic circuits of transformers. The N.O. sheets, representing around 80% of the market of electrical devices, are now produced almost exclusively by cold lamination, using a primary product obtained by hot rolling. They are used in two versions [2]: (i) the annealed sheets ("fully process"), that have received their cover before being delivered by the producer, usually rich in Si and Al, and with reduced losses; (ii) the non-finished sheets ("semi-process") delivered in an intermediary state and capable of a very high operating induction, less alloyed and less expensive. After the cutting up, they must be immediately annealed and covered by an electro-insulating layer.

For the N.O. sheets, recent improvements have been done related to the metallurgic process that must eliminate as much as possible the interstitial impurities and the crystallographic faults, and

realize a better control of the process evolution until obtaining the appropriate grain size [16]. On the other hand, even in the range of N.O. sheets, their re-crystallization texture is significant enough [2].

In the case of transformers, because the sheets are crossed over by a magnetic flux with a well-determined direction, it is reasonable to have a higher permeability along this direction that must be an easy axis [001] of the material. This operation that involves a very strong uniaxial anisotropy is realized by hot or cold lamination, which results in oriented grains materials. Thus, for 3 wt. % Si the grains diameter is around 10 μm – value for which the losses are acceptable.

The manufacturing technology for the G.O. sheets, that have a texture (110) <001> (Gosse texture), is based on a first hot lamination, then the alternation of annealing and cold lamination, followed by a final annealing at high temperatures that produces an abnormal grains growth (secondary re-crystallization) on a direction close to (110) <001> [2]. The grain size may reach several centimeters. Orienting the texture around the above-mentioned direction more efficiently increases the permeability. The big grains offer a smaller resistance at the wall movement, reducing the coercivity and the hysteresis losses. At the same time, decreasing the wall thickness, $2a$, there are less wall nucleation centers and the eddy currents losses are also reduced.

The high permeability requested for the material is obtained by modifying the alloy composition and searching an appropriate texture characterized by a good alignment of the grains easy axes. The materials having such textures are called with oriented grains and the most performing between them are the materials with a high flux density (HB).

Recently, the research has been oriented to the reduction of eddy current losses generally obtained by refining the domain structure. The solution is to induce sites favorable for the nucleation of supplementary walls. This may be obtained in two ways: (i) covering the sheets with isolating forsterite layers that induce stresses in the magnetic material by the effect of the thermal constant different from that of the material [18] – solution which is efficient especially for the sheets with a texture similar to the Goss orientation; (ii) scratching by laser or by mechanical procedures [19]. The operation is accompanied by an important growth of the grain size and of the domain dimensions that also conforms to the Bertotti theory [15].

In the case of electrical supplying of the material that operates at 400 Hz, in order to reduce the losses, one produces very thin sheets, 0.10 and 0.15 mm, by rolling again the Fe-Si strips of standard thickness. For $D = 0.10$ mm and $f = 400$ Hz, the losses of isotropic products are around 10 W/Kg for 1T and 30 W/Kg for 1.5 T, while for the oriented products they are reduced to 6 and 15 W/Kg, respectively. Otherwise, the reduction of sheet thickness, that sometimes preserves a very good texture (usually by metallurgic procedures based on a third re-crystallization), also remains an excellent solution for diminishing the losses. So, by reducing the sheet thickness from 0.23 mm to 0.075 mm, the losses are reduced at half (from 0.8 W/Kg to 0.4 W/Kg at 1.7 T) [20]. Unfortunately, one has not succeeded yet to reduce industrially the sheet thickness under 0.18 mm.

Progresses are expected from the optimizing of the procedures concerning correlated elements – the texture, the size of grains, the domain dimensions and the sheet thickness. Other predictable evolutions concern the surface treatments, with the aim of multiplying the wall nucleation centers, without bothering their movement. These procedures allow the production of sheets reach in silicon and aluminum, for the purpose of increasing their resistivity without the diminution of the mechanical resistance. In the meantime, the evaluation of all the new technologies must always take into account the economic constraints. The table below presents several magnetic characteristics of Fe-Si alloys.

The minimum of the magnetocrystalline anisotropy of the Fe-Si alloy for a content of 6.5 % Si, as well as the fall of magnetostriction constants λ_{001} and λ_{111} towards the same composition (see Fig. 4), have predicted the possibility to realize a high permeability composite with low coercivity and hysteresis losses. Moreover, such a high value of the resistivity ($10.8 \cdot 10^{-6}$ Ωm comparatively to $0.15 \cdot 10^{-6}$ Ωm for Fe-3%Si) serves to reduce the eddy current losses, while the reduction of B_s (at about 1.81 T) and of the Curie point (at about 700°C) are not so important as to become harmful [21]. Using laboratory samples one proved that at 6.5 % Si the permeability has the highest values, $30 \cdot 10^3$, while the losses pass through a minimum of 0.4 W/Kg at 1 T and 60 Hz.

Table 1. Magnetic characteristics of several soft magnetic metallic alloys.

Type and composition	B _s [T]	T _C [°C]	$\rho (\times 10^{-8})$ [$\Omega \cdot m$]	$\mu_{rel, max}$	H _c [A/m]	Losses at 50 Hz B = 1T – B = 1,5 T
Soft iron (impurities 0,2%)	2,15	770	10	5000	80	3 – 5 ($\Delta = 0,5$ mm)
Fe-3,75% Si isotrop	1,97	690	60	7000	32	1.3 – 1.5 ($\Delta = 0,3$ mm)
Fe-3% Si oriented (110) $\langle 001 \rangle$	2,02 ⁽¹⁾	720	47	60000	8	0.49 – 1.07 ($\Delta = 0.3$ mm)
	2,02 ⁽²⁾	720	47	80000	6	0.35 – 0.74 ($\Delta = 0.23$ mm)
FeSiB amorphous	1,56	415	130	100000	3	0.2 ($\Delta = 0.02$ mm)

⁽¹⁾ C.G.O.; ⁽²⁾ H.G.O.

Unfortunately, contents of Si higher than 4.5% cannot be technologically over-passed, because of the alloy fragility, their hot rolling being difficult and their cold rolling being almost impossible. The cold reduction of the Fe–6.5%Si sheet thickness is indispensable for realizing a favorable texture. For compensating this inconvenient one produced Fe–6.5%Si alloys directly like flexible and ductile ribbons, with low thickness (30 - 100 μ m), using the technique of rapid cooling on rotating wheel [22-24]. The material is thermodynamically stable and may be treated similarly to the traditional alloys for improving its characteristics. So, one can develop appropriate magnetic properties by choosing the annealing capable to control and maintain a good equilibrium between the grain growth, texture, order of the resulted super-lattices.

One also studied the possibility to increase the silicon content through the technique of chemical deposition in the vapor phase (CVD) that takes advantage of the good chemical reactivity at high temperatures between donor and non-covered Fe–3%Si sheet, if one reduces the sheet thickness and controls the Si content during the process [27, 28]. For a sheet with D = 0.1 mm, the losses reached 9.25 W/Kg at 1T and 10 kHz [29]. The process has proved to be efficient especially in terms of its continuation with a homogenization treatment [21]. An example of the obtained gain is illustrated in Fig. 6.

The losses of these alloys, even inferior to those of the Fe-based amorphous alloys, are considerably superior comparatively to the losses of conventional alloys, especially under 50 Hz [7] (Fig. 7). For example at 2 kHz, the losses in the Fe-6.5%Si ribbons of 40 μ m represent 1/5 - 1/8 of losses in the conventional N.O. steel M-15. So, the most promising application of the Fe–6.5%Si alloy seems to be the replacement of N.O. steels in the manufacturing of electrical machines. The technique of enrichment with Si through CVD also leads to excellent results in the case of Fe-Si N.O. sheets [27].

Once again, a proper material (containing few impurities) with big grains and a texture close to the ideal case (100) $\langle 0vw \rangle$ leads to a low coercivity and consequently to low hysteresis losses. On the other hand, the excess losses, p_{an} , may be diminished with a very refined structure that always imposes finding the optimum of this structure.

3.2 Fe-Al alloys

The properties of these alloys are comparable to those of the Fe-Si alloys. If one adds the aluminum price, inferior to that of silicon, one understands that the Fe-Al alloys are in competition with Fe-Si alloys in many cases. Unfortunately, the Al₂O₃ particles contained by the alloy destroy the tools for manufacturing the sheets, fact that brakes a largely spread utilization. The alloys, which contain up to 17% Al are ferromagnetic, while under this concentration they, become paramagnetic. Aluminum is often added to Fe-Si alloys to facilitate the grains growth and so for reducing the losses. Moreover, the addition of aluminum increases the resistivity (see Fig. 3) without increasing at all the material fragility. The ternary alloys Fe-Si-Al are dedicated to some special applications.

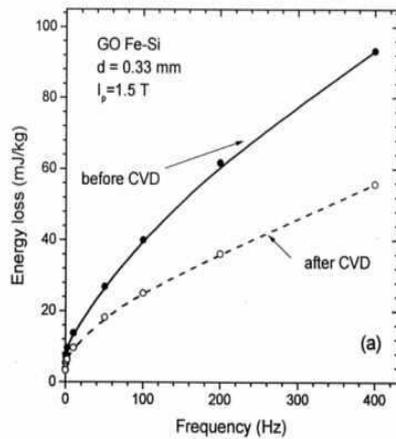


Fig. 6. Energy losses per cycle versus the excitation frequency for the G.O. sheets before and after the treatment by CVD with SiCl_4 , that leads to the Si content increase from 3% to 6,5% [15].

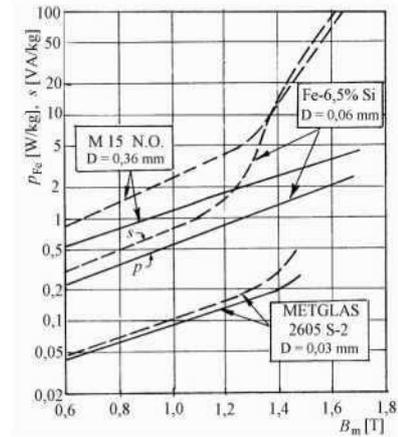


Fig. 7. Losses and excitation power at 60Hz, versus the magnetic flux, for some soft magnetic materials: Fe-Si M-15 N.O. steel, Fe-6,5%Si steel obtained by rapid cooling and amorphous METGLAS 2605S-2. Under 1,6T, the M-4 G.O. alloys and Fe-6,5%Si have almost identical losses.

3.3. Fe and steels with low carbon content (the soft iron)

The soft iron still remains the best material for DC electromagnets, because its high saturation flux density allows to obtain an important magnetic flux uniformly distributed. Consequently, in this case one should realize flux densities from 1 to 1.5 T; with the help of soft iron, the necessary excitation field descends to 200 - 700 A/m.

The initial magnetization curves for several types of soft iron are presented in Fig. 8. The iron has a low coercive field (~ 80 A/m) and a high permeability ($\sim 10^4$). An annealing in a hydrogen atmosphere eliminates the impurities decreasing the coercive field at only 4 A/m and increasing $\mu_{\text{rel, max}}$ to 10^5 (see Table 2). The highest value of $\mu_{\text{rel, max}}$ is for ultra-pure iron (around $1.5 \cdot 10^6$), but the material is too expensive for the most applications where such performances are interesting.

The material deformation causes the damaging of its magnetic properties. The stresses induced by rolling or manufacturing, are eliminated by annealing at $725 - 900^\circ\text{C}$ in hydrogen atmosphere, for avoiding the oxidation.

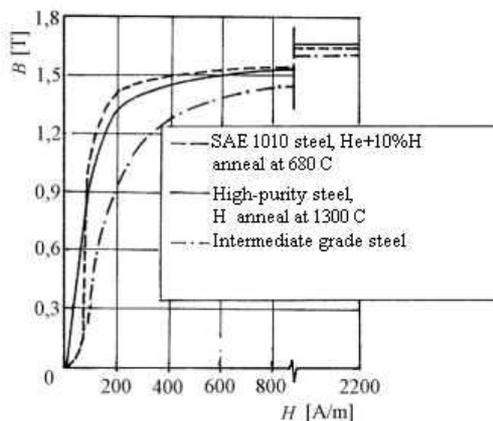


Fig. 8. Initial magnetization curves for two iron steels of high purity annealed in hydrogen atmosphere, and for usual steels [30].

Table 2. Properties of different types of high purity irons [31].

Material	B_s [T]	H_c [A/m]	μ_{rel} at 80 A/m	μ_{rel} at 800 A/m	$\mu_{rel, max}$
Magnetic iron	2.15	68.0	3500	1500	—
Fe, 0.2 mm thickness	2.15	89.0	1800	1575	—
Fe for electromagnets, 0.2 mm	2.15	81.6	2750	1575	—
Fe, casted under vacuum	—	24.8	—	—	21000
Fe, annealed	—	18.4	—	—	41500
Fe, casted under vacuum and annealed	—	7.2	—	—	61000
Puron (annealed in H_2 atmosphere)	2.16	4.0	—	—	100000

4. Iron-Nickel alloys

The Fe-Ni alloys, often called permalloys, are the most versatile from all the known soft magnetic materials. The Ni content is exceeding 30%; under this limit the temperature variation produces a structure transformation characterized by a temperature hysteresis. The alloys containing between 30 and 80 % Ni have a simple equilibrium diagram and are very ductile, allowing to obtain easily samples of reduced thicknesses that can be annealed at any temperature.

The domains where these alloys can be chosen are the electrical engines for avionics, the magnetic circuits of ultra-rapid relays and of other devices working in DC fields, the magnetic yokes of coils, the measure transformers, other transformers supplied on industrial frequency and many important applications in high frequency (e.g. different types of heads for magnetic recording and memory elements).

There are three groups of alloys that are useful from the practical point of view: the highest permeability characterizes the alloys with the nominal composition 80%Ni–20%Fe; the highest saturation flux density, with a maximum of 1.6 T, is obtained for alloys with 50%Ni–50%Fe (isoperm), while the most important resistivity is those of 30%Ni–70%Fe alloys; their magnetostriction is very low, even null for the alloy containing 81%Ni (Fig. 9) [32].

Some alloys with high permeability, like mumetal (77%Ni–16%Fe–5%Cu–2%Cr) and 78-permalloy (alloy with 78 % Ni), which have compositions situated near 80%Ni, have the permeability around $3 \cdot 10^5$ and the coercive field as low as 0.4 A/m. Their low anisotropy (the first constant K_1 is null for a content of Ni near 75 %) also contributes to the high permeability of the polycrystalline samples. So, one obtains a flux density as high as 0.6 T with the help of an excitation field of only 1.6 A/m. The magnetic properties of these two alloys are comparable, but mumetal is more ductile than Permalloy and may be used as thin ribbons. Because of their low coercivity, which may decrease even at 1 A/m, they are the ideal materials for manufacturing the rapid relays. They are also used for screening the sensible devices. Their low B_s , around 0.8 T, represents no drawback for them. By adding other elements and/or by an appropriate treatment, one can assure a large variety of properties for these alloys. Their anisotropy may be controlled by various procedures: cold rolling, magnetic annealing or thermomechanic treatment. According to the applied treatment, for the same alloy composition, one obtains various shapes of the hysteresis loops, with different functions.

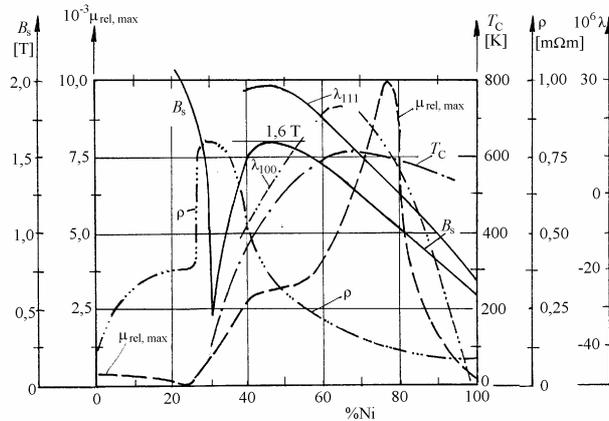


Fig. 9. Fe-Ni alloys properties versus their nickel content.

The addition of cobalt to the Ni-Fe system leads to ternary alloys from the family of permalloy, characterized by a constant permeability and practically null hysteresis losses for fields up to 200 A/m. The high quality transformers are often produced using this material. By appropriate procedures, one can realize high permeabilities of 10^5 , with various coercive fields (0.16 - 800 A/m), and a hysteresis loop of high rectangularity. The total losses of these materials are presented in Fig. 10.

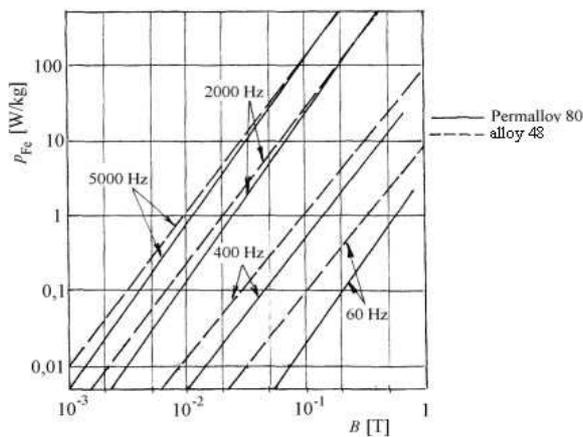


Fig. 10. Total iron losses versus magnetic flux density and its frequency for two types of sheets made from Ni-Fe alloys, $D = 0.35\text{mm}$.

Among the other useful materials from the Fe-Ni family, it is also worth mentioning: the invar (36%Ni-64%Fe) with a null coefficient of thermal dilatation, that is useful in very precise mechanisms; the 79 Mo-permalloy (79%Ni-15.5%Fe-5%Mo-0.5%Mn), with a high permeability and very reduced losses at frequencies higher than 10 kHz; the supermalloy, a more elaborated version of the previous one etc. Alloys with a composition close to 80%Ni are very much used for producing the magnetic recording heads [33].

5. Iron-Cobalt alloys

Fe-Co alloys have several applications, due to the high value of their saturation magnetization. Consequently, cobalt is the only element which, allied with iron, produces an increase of the saturation magnetization (Fig. 11) and of the Curie point, even if it is a small one. Their low

anisotropy and high permeability recommend these alloys for a wide range of applications. Unfortunately, the complicated metallurgy of these alloys and the high price of cobalt represent important constraints. Nickel and niobium are elements that are frequently added to Fe-Co alloys to improve their properties.

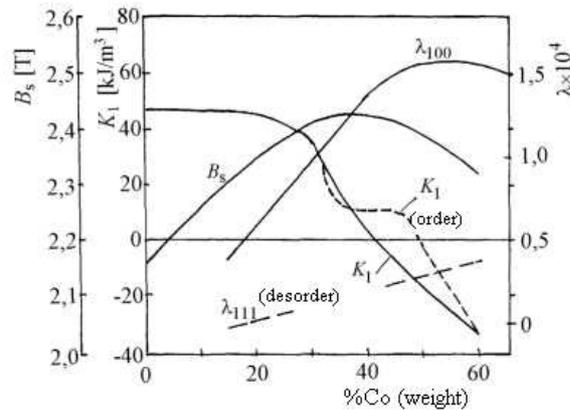


Fig. 11. Magnetic characteristics of Fe-Co alloys versus cobalt content, at room temperature [2].

The maximum of the saturation flux density ($\cong 2.45$ T) corresponds to a content of 35%Co, but this alloy is very fragile. A considerable improvement of the mechanical properties is given by the addition of vanadium or chrome. The V-permendur, 49%Fe–49%Co–2%V, has a saturation flux density $B_s \cong 2.3$ T and a constant permeability for a large range of excitation fields. The permendur will be able to represent an excellent material for relays due to its high B_s , but its strong magnetocrystalline anisotropy and its low resistivity, in addition to the prohibitive price of cobalt, eliminate it from most possible applications. However, it is still used for producing transformers and electrical machines when it is necessary to reduce as much as possible both the magnetic circuit dimensions and the polar parts of electromagnets [34].

Different other alloys of permendur type are used for the manufacturing of amplifiers, magnetic switches and of many types of memories. They are encountered in the diaphragms of the high quality telephone receivers and in the polar parts of machines used in domains where one requests specific high powers (space, aeronautics etc.) or high values of magnetic flux density at frequencies of hundreds of Hz.

Co-rich alloys (e.g. 94 %Co–6 %Fe), due to their properties close to those of pure cobalt, have a crystallographic structure with no phase change and a Curie temperature above 1000°C, which designates them for the manufacturing of devices at high temperatures, e.g. electromagnetic pumps.

Finally, the semi-remanent Fe–Co–V alloys that are situated, from the coercivity point of view, between the soft magnets and the permanent magnets, are other important alloys for applications. A well-defined coercive field and very rectangular hysteresis loops can be achieved by controlling their content of vanadium and the thermal treatments. These properties recommend them for the production of "rotors" of engines with hysteresis and for bistable relays.

6. Soft ferrites

The high conductivity of the magnetic metallic alloys forbids their use for high frequency, due to the unacceptable increase of eddy currents losses. For applications under several kHz, one should generally resort to dielectrics and magnetic semiconductors, materials with properties comparable to those of soft ferromagnets: high permeability, reduced coercive fields and high saturation flux density (see Fig. 12).

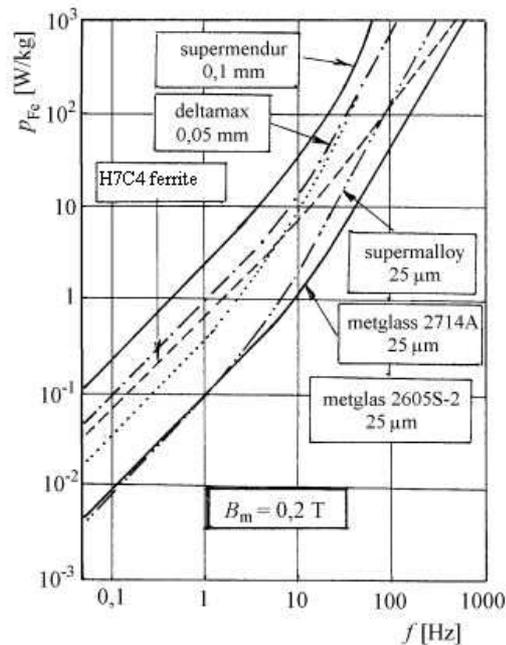


Fig. 12. Iron losses, at high frequencies, of some materials [3]. The metallic samples of reduced thicknesses can still be used.

The ferrites, ferrimagnetic ceramic materials, with the resistivity between $1 \Omega\text{m}$ (the ferrite of MnZn) and more than $10^4 \Omega\text{m}$ (the ferrite of NiZn) are particularly indicated. The rather high price of these materials is only a secondary drawback, because the high frequency is characteristic to small components, for which the performances are prevalent in respect to the economic aspects.

There are two categories of ferrites: the microwaves ferrites used in the frequency range 100 MHz - 500 GHz [35] and the ferrites for low frequencies, i.e. audio frequencies around 500 MHz [36]. The formers, among who a typical example is the grenat Y-Fe, are used for the wave-guides, particularly for systems of pulse compression. One also uses the soft ferrites to manufacture frequency filters and other electronic components as well as magnetic circuits of electronic equipments (fix and mobile telephony, radio-electricity, telecommunications, radar, and television). The soft ferrites can be found in optoelectronics and in some magneto-optical components.

The event that marked the development of the ferrite market was the appearance of semiconductors for the switch power sources, capable of good performances at frequencies situated in MHz range. The increase of the working frequency allowed the use of coils and transformers with more and more reduced dimensions, in terms of being able to limit the losses.

In the case of soft ferrites, the zinc always represents a part of their composition, despite the fact that it diminishes the saturation flux density and the Curie point, because it increases the permeability; the remainder is either the nickel or the manganese. The MnZn ferrite is used at frequencies up to 10 MHz, while under this limit the NiZn ferrite is preferred, because it has an even lower conductivity.

The permeability of these materials is not changed very much with respect to the frequency until a threshold situated between 10 and 100 MHz, but under this threshold it decreases rapidly.

The Curie points are relatively low ($\sim 150 - 300^\circ\text{C}$) because of the easy change of the Fe-O-Fe super-lattice. This fact makes all the properties to be strongly dependent on temperature, representing an important constraint for various applications. The saturation magnetization of ferrites is 0.35 - 0.5 T, considerably more reduced than those of metallic alloys of iron and of cobalt. The ferrites are produced by standard techniques used for ceramic products that allow a large variety of shapes. The realized microstructure also plays an important part, especially by the characteristic effects of the grain walls.

7. Amorphous materials

Amorphous magnetic materials are produced by rapidly cooling of the molten alloys. Fe and Co-based amorphous alloys are usually ferromagnetic. Typically, these alloys contain 80 % Fe, Ni and /or Co and approximately 20 % metalloid additions (P, Si, Al, C and B) [37]. The amorphous alloys have good magnetic properties even in the as-cast state.

Due to the rapid cooling of the material, the formation of the nucleation centres is avoided, and a short-range ordered structure is favored, so that their name of *metallic glasses*. From macroscopic point of view the material is isotropic. Therefore, the magnetization is not subject to any anisotropy. The material produced in this way is characterized by strong internal stresses, that considerably increase coercive field and decreases permeability. An appropriate annealing may partially eliminate these stresses, without material re-crystallization.

A new direction was opened with bulk metallic glasses development, for whose preparation much more reduced cooling rates are required. Due to their resistance at crystallization, one can produce massive 3D samples with dimensions of millimeter that open new application horizons. Moreover, Fe-based multicomponent amorphous alloys are magnetically soft at room temperature [38].

Fe-Si-B amorphous alloys have losses at industrial frequencies six times smaller than those of the traditional materials. From this reason, they are good competitors for traditional Fe-Si alloys. Binary Fe-B alloys were the first amorphous alloys dedicated to the manufacturing of distribution transformers and having a saturation flux density higher than 1.5 T. The METGLAS 2605 alloy (80%Fe–20%B) is the best known up to now.

A better thermal stability, with no degradation of their saturation flux density, was obtained by the addition of Si, a good example being the ternary alloy METGLAS 2605–S (82%Fe–12%B–6%Si). Unfortunately, it is difficult to reproduce, because of the air bags formed at the contact surface of the tape with the wheel; these bags allowing the material oxidation, which involves the diminishing of the magnetic flux density, more longish hysteresis loops and higher total losses. The METGLAS 2605–SC (81.5%Fe–13.5%B–3%Si–2%C) overcomes the above presented drawbacks. A better Curie temperature was obtained for METGLAS 2605–S2 (79%Fe–13%B–8%Si) without altering the saturation flux density and core losses. It competes with Fe–3%Si G.O. for the manufacturing of power transformers. At the same time, the METGLAS 2605–SC alloy has a more reduced uniaxial anisotropy and a better marked rectangularity of the hysteresis loop. At 1.4 T, the losses and the excitation power are of 0.15 - 0.25 W/kg and 0.45 VA/kg respectively.

Magnetic amorphous alloys are of particular interest due to their low coercive fields (Fig. 13), one magnitude order lower comparatively with Fe-Si steels, which correspond to higher permeabilities. The hysteresis loops are very rectangular with very low total losses (Fig. 14) – another important advantage.

The induced anisotropies play a very important role in magnetic amorphous alloys behavior, because the permeability and core losses may be controlled by the anisotropy induced by the residual and/or applied stresses. The compositions with a high content of iron (e.g. 78%Fe–13%B–9%Si or 81%Fe–13.5%B–3.5%Si–2.5%C) have an important magnetostriction $\lambda_s \sim 30 - 40 \cdot 10^{-6}$ and consequently the applied stresses may easily induce a preferential direction of magnetization. For these materials, coercive fields as low as 3 - 8 A/m are usual, but it is possible to reduce them at only 1 - 2 A/m through an annealing at 350 - 400°C [39], even if the complete elimination of the residual stresses is difficult to realize [40].

In many special applications, the requirement for low core losses comes together with that of a versatile magnetic behavior. Such requirements may be satisfied by the alloys with nearly zero magnetostriction, based on cobalt (71%Co–4%Fe–15%B–10%Si, 67%Co–3%Fe–12%B–16%Si–2%Mo etc. [41]). Additionally, these alloys exhibit reduced coercivities and a good sensibility of the B ~ H loop versus the magnetic annealing. The annealing induces a macroscopic easy magnetization axis, as a consequence of the local atomic rearrangements. The associated anisotropy may be adjusted for the specific requirements of the applications.

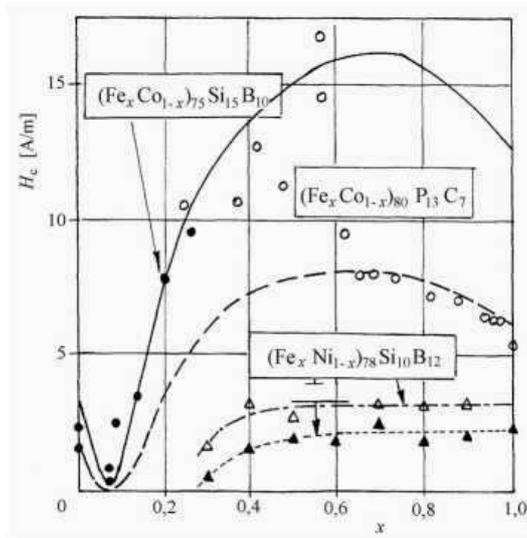


Fig. 13. Dependence of the coercive field on the composition, for several magnetic amorphous alloys.

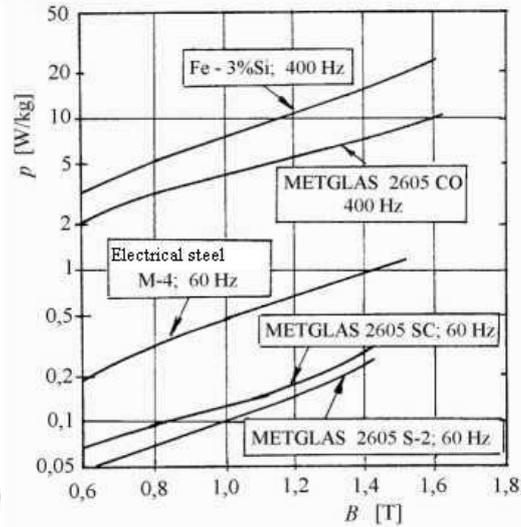


Fig. 14. Total losses for several soft magnetic materials versus magnetic flux density [31].

Fig. 15 presents the effect of magnetic annealing on the DC hysteresis loops. For 71%Co–4%Fe–15%B–10%Si amorphous strip, the rectangular cycle ($H_c \sim 0.5$ A/m) obtained after annealing at about 300°C in a longitudinal field, close to saturation, changes into an almost linear, longish curve when the field is transversally oriented. The change is due to the fact that the domain structure passes from the longitudinal to the transversal shape, and so the coherent rotations of magnetization replace the wall displacement in magnetization process with considerable effect on the core losses [42]. The variation of the total energy losses per cycle versus frequency, for a strip with the above-indicated composition and a thickness of $20\ \mu\text{m}$ is represented in Fig. 16.

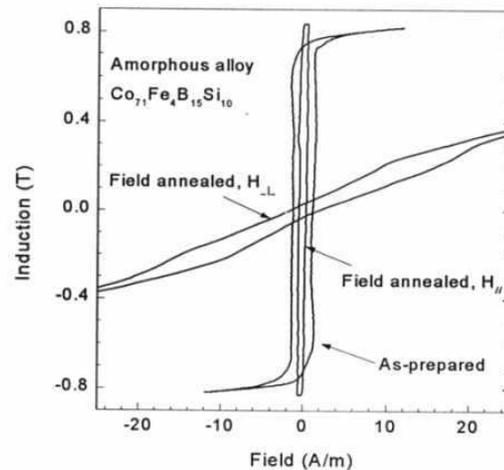


Fig. 15. DC hysteresis loops for an amorphous strip 71%Co-4%Fe-15%B-10%Si, before and after annealing in longitudinal and transversal fields close to saturation value.

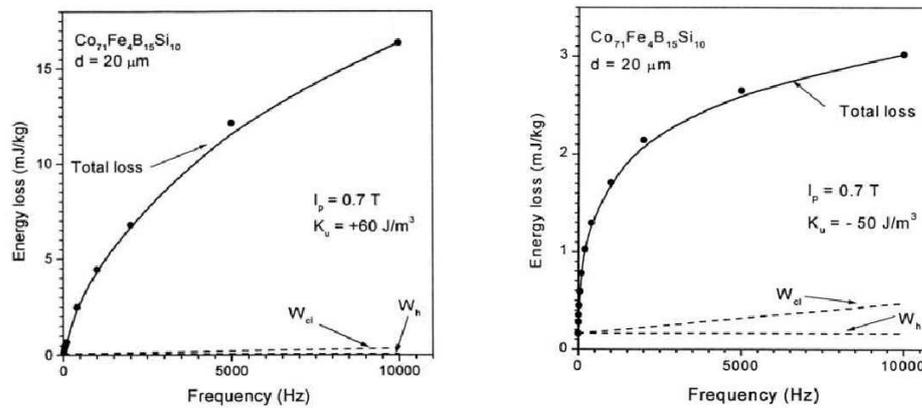


Fig. 16. Energy losses per cycle versus frequency, for amorphous Co based amorphous strips, annealed in longitudinal (a) and transversal field (b), close to saturation: (a) $K_u = 60 \text{ J/m}^3$; (b) $K_u = -50 \text{ J/m}^3$. The points correspond to the experimental measurements [15], the continuous lines represent the total losses and the dotted lines the classical and the hysteresis losses, respectively.

However, the amorphous alloys present some drawbacks: low saturation magnetization and losses that seriously increase at high excitation magnetic fields. The consequence is that the transformer cores are more sizable than those made of Fe-Si, the losses in the windings rise and the dimensions becoming clumsy. Moreover, the price of transformers manufactured with amorphous magnetic cores is higher and so the choice remains a matter of economic optimum.

In amorphous magnetic materials the effect of the domain structure is more important than in Fe-3%Si steels. But the amorphous alloys exhibit electrical resistivities of $1.2 - 1.4 \mu\Omega\text{m}$, usually bigger than the values corresponding to the magnetic crystalline materials ($0.2 - 0.5 \mu\Omega\text{m}$). However, the absence of magnetocrystalline anisotropy and the big metallurgic faults lead to the formation of magnetic domains with big dimensions. Most of the losses in amorphous alloys are due to the terms, p_h and p_{an} , (Fig. 16) and solutions for reducing them are investigated.

On the other hand, because of the pronounced rectangularity of their hysteresis loops, the operational magnetic flux density of the amorphous alloys is close to the saturation value B_s , without encountering the drawbacks of the ordinary steels [43, 44]. This fact attenuates the disadvantage of the lower values of B_s for amorphous alloys; the operational magnetic flux may be increased up to 1.4 T. To all these, one adds the fragility of the annealed alloys and the fact that the stresses alter their behavior due to their low anisotropy. This is the explanation for the recommendation to anneal the magnetic cores made from amorphous materials after all the requested manufacturing operations.

From this point of view, improvements have been obtained for rapidly quenched materials passing current impulses through the strips [45]. The best characteristics are obtained after annealing at a temperature lower than the crystallization one. Allowing atomic arrangements on a short distance, this anneal induces a uniaxial anisotropy decisive for obtaining a good permeability and low losses. As presented above the same effect is obtained through magnetic annealing.

Another choice domain for the amorphous materials is that of mean frequencies, 10 kHz - 10 MHz. For such applications, ribbons with thicknesses between 15 and 100 μm are usually used. Their resistivity ($1.2 - 1.5 \mu\Omega\text{m}$) is higher than those of Ni-Fe alloys ($0.45 - 0.65 \mu\Omega\text{m}$). In the frequency range 10 - 100 kHz, Co-rich amorphous alloys with $\lambda_s \approx 0$ exhibit the best magnetic properties among all the soft magnetic materials.

While the saturation magnetic flux density for the ordinary amorphous alloys based on cobalt is only 1.0 - 1.2 T, they have other qualities: insensibility at the applied stresses, an excellent permeability and a remarkable easiness to modify the shape of their hysteresis loop by an appropriate magnetic annealing, that recommend these alloys for the applications where the price is less important than the operation conditions. The amorphous alloys based on iron are used especially for the cores of induction coils.

The fact that plastic deformations lead to the deterioration of the magnetic properties of amorphous materials, besides their high price, explain why their applicability is much under expectations. At mean frequencies, the usual application field is that of low currents and small specialized devices: converters, pulse transformers, magnetic actuators and magnetoresistive transducers.

Table 3 indicates the main magnetic properties of several types of amorphous alloys used in DC applications.

Table 3. Magnetic properties of several amorphous alloys used in DC field applications [31].

Alloy	Shape	As-cast			Annealed		
		H_c [A/m]	M_r/M_s	$\mu_{rel, max}$	H_c [A/m]	M_r/M_s	$\mu_{rel, max}$
METGLAS #2605 80% Fe-20% B	toroid	6.40	0.51	100,000	3.20	0.77	300,000
METGLAS #2826 40% Fe-40% Ni-14% P-6% B	toroid	4.80	0.45	58,000	1.60	0.71	275,000
METGLAS #2826 29% Fe-44% Ni-14% P- -6% B-2% Si	toroid	4.60	0.54	46,000	0.88	0.70	310,000
4,7% Fe-70,3% Co- -15% Si-10% B	sheet	1.04	0.36	190,000	0.48	0.63	700,000
78% (0,8Fe-0,2Ni)- -8% Si-14% B	sheet	1.44	0.41	300,000	0.48	0.95	2,000,000
METGLAS #2615 80% Fe-16% P-3% C-1% B	toroid	4.96	0.40	96,000	4.00	0.42	130,000

The research field for a better exploitation of the amorphous magnetic alloys has known remarkable advancements. Notable progress was obtained in the production of materials for high frequencies. The most important are probably the *nanocrystalline* alloys. For example, the structure of 73.5%Fe-1%Cu-3%Nb-9%B-13.5%Si amorphous alloy is completely changed by an appropriate thermal treatment; consequently the specific magnetic properties are essentially improved [46]. Its microstructure consists of nanocrystalline grains with dimensions of 10 - 50 nm. The simultaneous presence of Cu and Nb is indispensable to inhibit the formation of borides and to limit the grains sizes. The losses and the coercive field of the annealed material are comparable to those of the best Co-based amorphous alloys. For obtaining various shapes of the hysteresis loops, the material must be annealed in the presence of a magnetic field [47].

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