1.0 Summary

This chapter reviews the material properties of soft magnetic materials, in general and nickel-iron alloys, in particular, considering their physical behaviour and other basic considerations with a focus on their structure-property relationships. Their applications are also presented in this chapter. The aim and objective of the study is given at the end of the chapter.

1.1 Magnetic materials

The materials when subjected to a magnetic field (H) develop magnetisation M which is either parallel or antiparallel to H. If M is antiparallel to H the material is called a diamagnet and if M is parallel and proportional to H, it is called paramagnet. Some materials show magnetization even in the absence of an external magnetic field. These materials can be ferromagnetic or ferrimagnetic depending on the arrangement of atomic magnetic moments in the crystal. A few materials have their atomic moments so arranged that the net magnetization is zero. These are anti-ferromagnetic materials. The classification of materials based on their magnetic behaviour is shown in Table 1.1 and the arrangement of spins corresponding to these materials is given in Fig. 1.1.

Table 1.1: Classification of magnetic materials

Type	Examples
Diamagnetic	Organic solids like benzene, naphthalene; metals like Cu, Ag, Au etc.
Paramagnetic	Alkali and transition metals
Ferromagnetic	Fe, Co, Ni
Antiferromagnetic	MnO, NiO, MnS
Ferrimagnetic	Ferrites

Paramagnet Ferromagnet

Antiferromagnet Ferrimagnet

Fig. 1.1: Arrangement of spins in para, ferro, antiferro and ferrimagnetic materials

1.2 Origin of ferromagnetism

The transition metals Fe, Co and Ni are ferromagnetic at room temperature. For example, iron atom has the electron structure, [Ar] $3d^6\ 4s^2$ and an isolated iron atom has only the 3d subshell with five orbitals unfilled. By Hund's rule, the electrons try to align their spins so that the five 3d orbitals contain two paired electrons and four unpaired electrons as shown in Fig 1.2. The isolated atom has four parallel electrons spins and hence a spin magnetic moment of 4β. The Hund's rule (Fig. 1.3) shows that when the spins are parallel (same m_s), as a requirement of the Pauli exclusion principle, the electrons must occupy orbitals with different m_l and hence possess different spatial distribution. Different m_l values gives smaller coulombic repulsion energy between the electrons which is due to the exchange interaction, Pauli exclusion principle and the columbic forces.

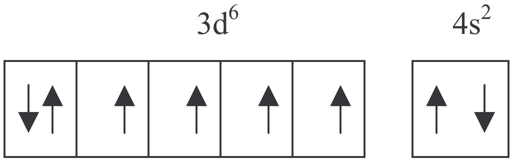

Fig. 1.2: An isolated Fe atom

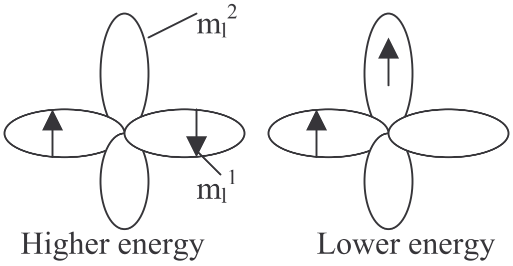

Fig. 1.3: Hund's rule for an atom with many electrons based on the exchange interaction

The parallel arrangement of spins in a ferromagnetic solid arises due to the exchange interaction. Due to this interaction, the two neighbouring spins in a solid are coupled together with an energy given by [1]

$$E_{ex} = -2 J_e S_1 . S_2 \qquad (1.1)$$

Where S_1 and S_2 are the spin angular momenta of the two electrons and J_e is the numerical quantity called the exchange integral that involves integrating the wave-functions with the various potential energy interaction terms. It therefore depends on the electrostatic interactions and hence on the interatomic distance. The exchange energy is positive or negative depending on whether spins line up parallel or antiparallel, respectively. In ferromagnetism, E_{ex} is postive; in antiferromagnetic materials like MnO, Mn, Cr, E_{ex} is negative. The correlation is given in the Fig. 1.4. It provides that for ferromagnetism to occur in metals the ratio of atomic separation distance to the radius of the 3d shell should be larger than a certain amount (~1.5). Under such conditions, E_{ex} is positive.

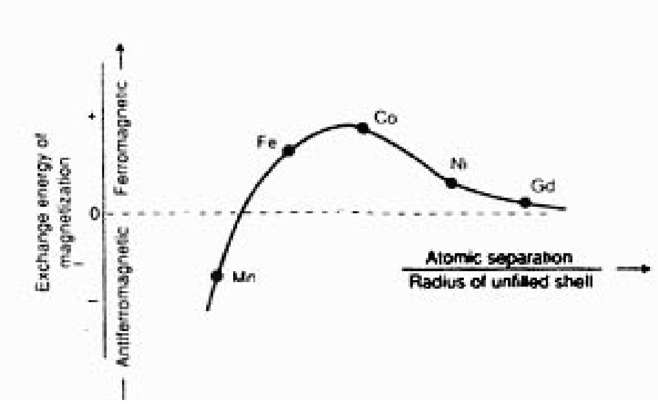

Fig. 1.4: Magnetic exchange interaction as a function of the ratio of the atomic separation to the radius of the 3d electron orbit

1.3 Curie temperature

Temperature greatly influences the magnetic characteristics of the materials. With increase in the temperature of a solid, there is an increase in the magnitude of the thermal vibrations of atoms and the atomic magnetic movements are free to rotate. Therefore, with rising temperature, the increased thermal motion of the atoms tends to randomize the directions of any moments that may aligned. For ferromagnetic materials, the atomic thermal motions counteract the coupling forces between the adjacent atomic dipole moments, causing some dipole misalignment, regardless of whether an external field is present. This results in the decrease in saturation magnetization for ferromagnets. The saturation magnetization is maximum at 0K, with increasing temperature, the saturation magnetization diminishes gradually and then abruptly drops to zero at **Curie temperature** (T_c). Above T_c the material behaves like a paramagnet. The temperature dependence of the magnetization is given in Fig. 1.5 [2].

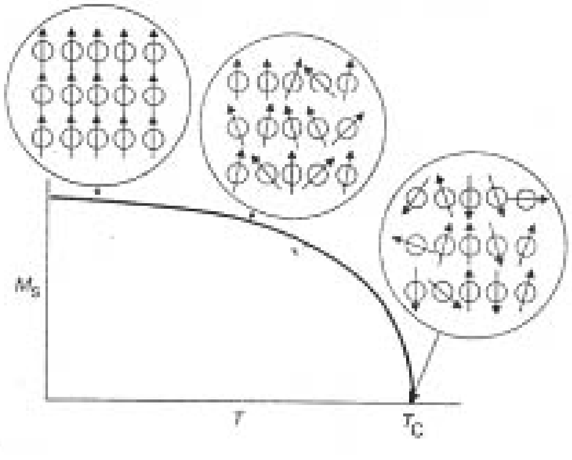

Fig. 1.5: Temperature dependence of the magnetization of a magnetic material showing mis-alignment of spins with increasing temperature

1.4 Magnetic domains

In the demagnetized state, the structure of a magnetic material consists of domains within which the magnetic moments of all the atoms are aligned. The direction of these moments, however, varies from domain to domain such that the net magnetization for the solid is zero. Domains are typically $10^{-2} – 10^{-5}$ cm in size separated by boundaries about

100 nm thick [3]. The presence of domain is due to the minimization of the exchange energy, which tends to align all magnetic moments in one direction, and the magnetostatic energy, which tends to align them in opposite directions. The magnetic moments inside each domain are directed along a ‘easy’ axis which is the direction of minimum anisotropy energy. Fig. 1.6 shows the B-H curve of the magnetic domain motion which gives the different magnetization effects depending on whether magnetic fields are increasing or decreasing.

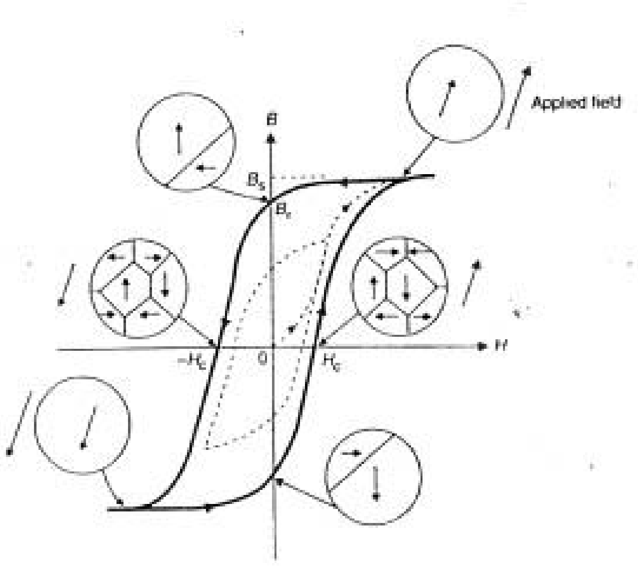

Fig. 1.6: A typical B-H hysteresis curve corresponding to magnetic domain motion in a soft magnetic material

1.5 Grain size

Grain size has an important effect on the properties of metals and therefore its measurement is of great significance [4]. The hysteresis properties of a magnetic material such as permeability, coercivity and remanence are governed by mainly magnetization process of domain nucleation, domain wall motion and domain rotation. These processes in turn are governed by the intrinsic properties such as saturation magnetization and magnetic anisotropy energy as well as microstructural details such as inclusion content and grain size. From domain theory, rotational processes give the general expression [5]

$$\mu_i \propto M_s^2 / K_{eff} \quad (1.2)$$

Where μ_i is the initial permeability, M_s the saturation magnetization and K_{eff} is an effective anisotropy constant covering all sources of anisotropy energy. A domain wall motion model taking grain size into effect the following expression [6]

$$\mu_i \propto M_s^2 \, d / (AK)^{1/2} \quad (1.3)$$

Where A is exchange constant, K the anisotropy constant and d is the grain diameter. Hence the maximization of initial permeability requires maximization of M_s, d and minimization of K. The coercive field strength H_c determined by the grain boundaries is a function of the reciprocal grain diameter [7]

$$H_c \approx 3 \, \gamma_w / J_s \, d \quad (1.4)$$

Where J_s is the saturation polarization and γ_w the wall energy which is proportional to $\sqrt{K_1}$ and K_1 is the first order magneto-crystalline anisotropy constant. The grain diameter has also great effect on magnetic losses [8-11]. When the grain diameter d increases, the hysteresis loss decreases in proportion to 1/d. The total core loss is minimized with an optimum grain diameter and this optimum grain size is obtained after the final annealing.

1.6 Soft magnetic materials

Soft magnetic materials can be easily magnetised and demagnetised. They retain their magnetization only in presence of a magnetic field. They show a narrow hysteresis loop, so that the magnetization follows the variation of the applied field nearly without hysteresis. They are used to enhance the flux produced by an electric current in them. The quality factor (figure of merit) of a soft magnetic material is to measure of its permeability with respect to the applied magnetic field. The other main parameters are the coercivity, the saturation magnetisation and the electrical conductivity.

An ideal soft magnetic material would have zero coercivity (H_c), a very large saturation magnetisation (M_s), zero remanence (B_r), zero hysteresis loss and very large permeability. The broad spectrum of soft magnetic materials includes: i) electrical steels, ii) Ni-Fe alloys, iii) Fe-Co alloys and iv) ferrites. The classification of commonly used soft magnetic materials and their magnetic properties are shown in Fig. 1.7 and Table 1.2 respectively [12, 13-18].

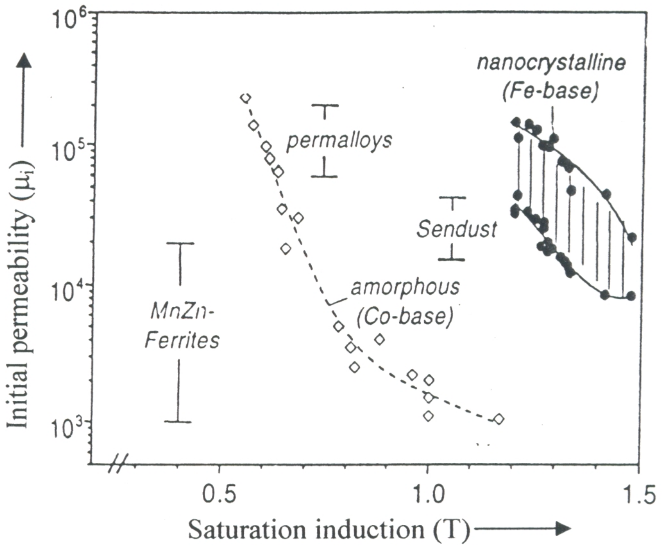

Fig. 1.7: Classes of soft magnetic materials

Table 1.2: Properties of some soft magnetic materials

Magnetic material	**B_{sat} (T)**	**ρ (μΩ-m)**	**μ_{rmax} ($X10^3$)**	**Core loss (W/kg)**
Electrical steel	2.0	0.48	5	0.9
40 -50 Ni- Fe alloy	1.6	0.48	150	110.0
79Ni-4Mo permalloy	0.8	0.58	1000	33.0
49Fe-49Co-2V	2.3	0.35	50	2.2
MnZn ferrite	0.5	$2 X10^6$	6	35

The Ni-Fe alloys are preferred over electrical steels and Fe-Co alloys since they have extraordinarily high initial and maximum permeabilities. These properties coupled with low hysteresis losses at low inductions and reasonably high electrical resistivities make some of the Ni-Fe alloys especially suitable for applications in telecommunications engineering. Another advantage is that they are relatively less brittle and therefore can be easily cold rolled into thin tapes suitable for stator, core, audio heads and laminations etc [12] to be used in magnetic devices.

1.6.1 Ni-Fe alloys

The Ni-Fe alloys constitute a class of permalloys are soft magnetic materials. They are ductile and most versatile materials for use in many industrial applications. These are based on the face-centered cubic portion of Ni-Fe system and contain about 40 to 90 % Ni at a time and up to few percent or traces of other alloying elements like Mo, Cu, Cr, Co, Mn in Fe matrix [19-21]. Their remarkable magnetic properties have made them the subject of intensive study over the last about over half a century. Typically three ranges of nickel content are used as soft magnetic alloys : 36% Ni for maximum resistivity, 50% Ni for maximum saturation magnetization and 80% Ni for optimum initial and maximum permeabilities [22-24].

With suitable alloying and proper processing, the magnetic properties of permalloys can be controlled within wide limits [25]. The magnetic properties of the alloys depend on the heat treatment under hydrogen atmosphere [7, 20-21, 26]. Their structure dependent magnetic properties such as coercivity, permeability and hysteresis loop shape depend on the basic magnetic constants like crystalline anisotropy K_1 and magnetostriction λ, which are determined by the alloy composition. The annealing treatment leads to a definite degree of Ni_3Fe order [27].

A review of historical developments that have taken place with respect of Ni-Fe alloys is summarized in Table 1.3.

Table 1.3: Milestones in the development of Ni-Fe alloys

Year	Alloys	Observations	References
1910	0, 25, 26, 28, 35, 47, 75 and 100 % Ni	B decreased with 30 % Fe addition and maximum B obtained with 47 % Fe addition	[28-29]
1910	36 % and 49 % Ni	μ_i obtained 700 and 550 respectively	[30]
1913	50 - 90 % Ni	Higher μ_i than pure Fe	[30]
1921	78 permalloy addition of Cr to Ni-Fe alloys addition of Co to Ni-Fe alloys addition of Mo, Cr and Cu to Ni-Fe alloys	μ_i = 2000-7000 Resistivity increased Low B_r and H_c obtained μ increased and heat treatment simplified	[31–33] [32–39] [40] [41–43]
1924	45 permalloy	Maximum B obtained as compared to other permalloys	[32–35]
1925 - 27	45 - 50 % Ni alloy annealed in hydrogen atmosphere at 1000°C temperature	Improvement in maximum permeability and hysteresis loss were reported	[36-38]
1950	Ni-Fe alloys	Compilation of results of Ni-Fe alloys	[26]
1966 - 71	Ni-Fe alloys with addition of non magnetic inclusions	Control on magnetic properties	[17, 21, 25, 44-45]
1980 – 97	Ni-Fe alloys (with H_2 annealing)	Kinetics of order formation	[23, 46-55]
2001	55 Ni-Fe alloys	Magnetic treatment parameters were determined	[56]
2003	Ni-Fe alloys	Recent advances and future directions were reported	[57]

1.7 Magnetic recording

Since the 1940s, magnetic recording has been the universal technology for electronic information mass storage. Its presence is everywhere as audio tape, VCRs, floppy disks, computer tapes, hard disks, credit cards and many others. Sales of magnetic recording products exceeded US \$ 50 X 10^9 in 1990 with projected strong growth [58]. Magnetic recording is a process whereby information is stored in the form of tiny magnetic patterns in a flat medium. This is accomplished by moving the medium near or in contact with a recording head. The stored information can be retrieved by passing the medium near or in contact with a playback head. The information can be erased if necessary but it is otherwise a permanent record. The medium can be either a flat disk that rotates past the heads or tape that can be moved past a head either from reels, in a cassette, or on a card [59-61]. Magnetic recording involves encoding information into time varying electrical signals, which drive a magnetic recording head, and thus creating a spatially varying pattern of magnetisation on a moving storage medium. The reading process uses another (or the same) head to reconvert the pattern of magnetisation into time varying electrical signals. The magnetic storage medium provides a reversible means of storing audio, video and data signals and actually belongs to the class of permanent magnetic materials. The measure of the recording efficiency of a disk drive is its areal density-the number of bits that can be stored per square inch [62].

World market status

Magnetic recording of the human voice was first carried out in 1898 by a Danish engineer, named Poulsen. The recording was made on a ferromagnetic wire, but due to a lack of amplification the recording quality was very poor. In 1927, magnetic tape was simultaneously invented in the USA and Germany. The growth of the market for magnetic recording was assured by the efforts of the American firm Minnesota, Minning and Manufacturing (3M). As a result of the efforts of all these firms, 3M, United Kingdom, United States, magnetic recording was well established as an industry by 1950. Recorders were being sold for both consumer and professional voice and music recording [63]. The remarkable progress was made in magnetic recording and is shown in Fig. 1.8 [64-65]. The first hard disk drive, known as the “RAMAC”, was introduced by IBM in

1957, and had a storage capacity of 2000 bits in^{-2}. The storage capacity has rapidly and steadily increased since the "RAMAC" and in the year 2000 reached approximately 10 Gbits in^{-2}, i.e. an increase by a factor of five million.

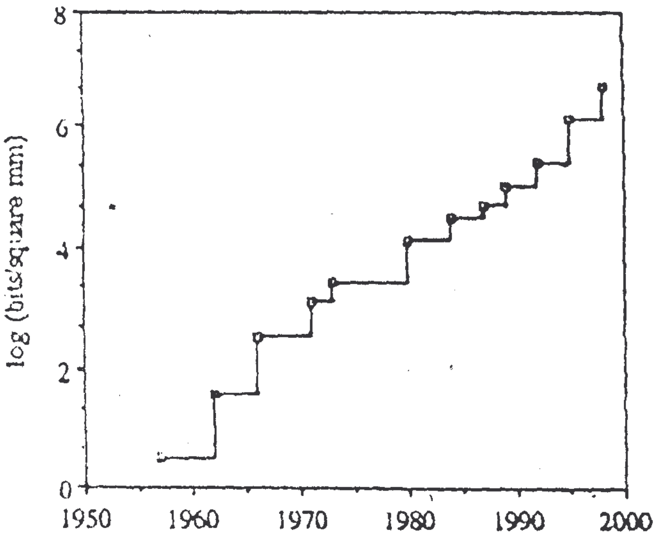

Fig. 1.8: Progress in recording density since 1957

Recording head

Magnetic Recording heads are divided into two major categories: inductive and magneto-resistive (MR) head. The inductive head can perform three functions: write, read and erase, while the MR head is read only head. The proposed work is based on the playback inductive head. In the inductive head, the fringing field produced in the gap of the core creates a magnetic state in the medium that corresponds to signal currents in the head coil. During the read process, the head senses the states by intercepting magnetic flux that emanates from the magnetic medium. The inductive head senses dB/dt and its output is media speed dependent [66-68]. The schematic diagram of a magnetic recording head is given in Fig. 1.9. The efficiency of the head measures how much the magnetomotive force i.e. the number of turns times the current, appears across the gap. If the permeability was infinite, so that the magnetic field inside the core, $H = B/\mu$ is zero, then the efficiency would be unity. So the larger permeability is better.

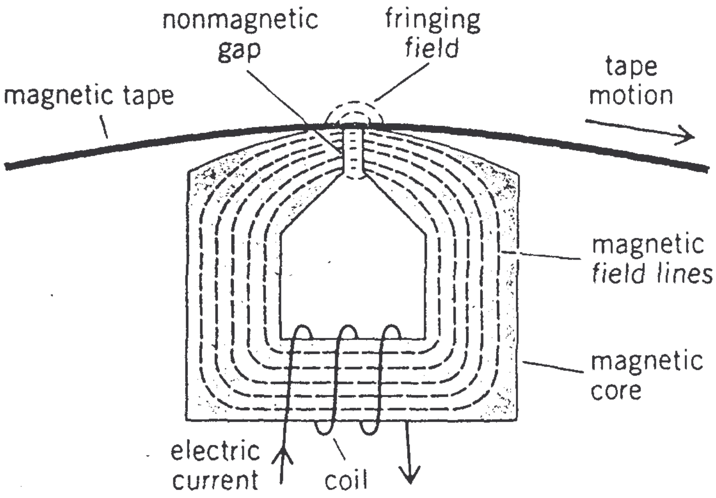

Fig. 1.9: Schematic diagram of magnetic recording head

The desirable characteristics for magnetic recording head materials are [69]:

- Magnetically soft i.e. their coercive force H_c should be very low
- For low H_c, high saturation flux density, B_s, is required for write and erase head functions.
- High effective permeability μ_e is desired over the operating frequency range during read operations.
- Low hysteresis loss from domain wall movement.
- High electrical resistivity to minimise induced eddy current loss.
- Curie temperature high enough for operating environment.

1.8 Application range of permalloys

Permalloys have found applications in many fields of science and technology. Their usage in electrical / electronic devices have made them the most sought after. e.g. in DC based electrical applications, the main consideration in them is the higher permeability. Similarly, for AC applications, the important consideration is energy loss in the system. The energy loss can originate from three different sources: i) hysteresis loss, which is related to the area contained within the hysteresis loop; ii) eddy current loss, which is related to the generation of electric currents in the magnetic material and the associated resistive losses and iii) anomalous loss, which is related to the movement of domain walls within the material. The broad applications of Ni-Fe alloys are [70];

- Magnetic recording heads
- Magnetic shielding
- Rotor and stator laminations
- Stepping motors
- Sensitive relays
- Current transformers
- LF power transducers
- Chokes
- Magnetic valves
- And others.

1.9 Aim of the present work

Permalloys in which Ni-Fe systems largely dominate, have found usage in many electrical /electronics consumable goods. The subject is vast enough to spell out all useful applications therefore we have focused our work aiming at the recording head and watch component materials. This is due to the fact that these devices consume large amounts of such alloys.

The work carried out in this thesis comprises of the study of the influence of various processing parameters such as annealing temperature, holding time and cooling rate under hydrogen atmosphere on the magnetic properties of the Ni-Fe alloys. The three samples adopted for the present investigation includes 82.13 % Ni (sample A), 79.90 % Ni (sample B) and 47.01 % Ni (sample C). The materials were characterised for their chemical composition by wet chemical analysis and atomic absorption spectrometer. The first step involved was the preparation of ring samples. Die and punch was designed and fabricated especially to punch the ring of desired shape and size. To anneal the samples in hydrogen atmosphere one lab model hydrogen annealing furnace was designed and developed upto measuring temperature range of 1200°C. The samples prepared under different conditions were analysed with optical microscope and X-ray diffractograph (XRD). The magnetic properties were studied with a standard B-H analyser. The prepared samples were tested for industrial applications such as watch and audio recording applications. We compared the performance of the material/device developed through the proposed route with the existing ones by checking the electro-mechanical characteristics of watch movement and also audio recording characteristics of audio head. The results have been discussed and deliberated in the forthcoming chapters of the thesis.

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