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Structures, Properties and Applications of Ferrites: A Brief Review

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Abstract: *Ferrites constitue an important class of ferrimagnétique oxide materials which have been extensively investigated due to their interesting magnetic and electrical properties and wide range of technological applications. Among the different types of ferrites, hexagonal ferrites, particularly M-type magnetoplumbites such as* $BaFe_{12}O_{19}$ *and* $SrFe_{12}O_{19}$*, are of considerable importance because of their high magnetocrystalline anisotropy, large coercive force, moderate saturation magnetization and good chemical stability.*

In the present review, the crystal structures of spinel, garnet and hexagonal ferrites are discussed with emphasis on the S, R and T structural blocks and their influence on magnetic exchange interactions and anisotropy. Various synthesis techniques including conventional ceramic method, co-precipitation, sol–gel, hydrothermal, spray drying and glass crystallization methods are briefly described in relation to phase formation and microstructural development. The effect of cation substitution, particularly coupled substitutions such as Co^{2+}–Ti^{4+}, Zn^{2+}–Ti^{4+} *and* Ni^{2+}–Sn^{4+}*, on magnetic and dielectric properties has been highlighted. The importance of these materials in permanent magnets, microwave devices, electromagnetic wave absorption and other high-frequency applications is also presented.*

Key Words: *ferrites; hexaferrites; magnetic properties; dielectric properties; microwave absorption.*

I.INTRODUCTION

Oxides and semiconductors containing transition metal elements and some rare earths as dopants or constituents exhibit ferromagnetic, ferrimagnetic and even antiferromagnetic behaviour if the spins of the magnetic ions are aligned. Oxides with transition metal ions (or rare earth) as a constituent are of special interest as they exhibit large permeability. Among the subclasses of oxides using Fe as a constituent and showing antiferromagnetism is a class of materials which go by the name of ferrimagnets or ferrites.

The ferrites are usually non-conductive ferrimagnetic ceramic compounds derived from iron oxides such as hematite (Fe_2O_3) or magnetite (Fe_3O_4), as well as oxides of other metals. Like most other ceramics, they are hard and brittle, and many of them are magnetic materials[1]. 'The ferrites were developed into commercially useful magnetic materials, chiefly during the years 1933-1945, by Snoek and his associates at the Philips Research Laboratories in the Netherlands[2]. In his seminal 1948 paper, Néel[3] laid the theoretical foundation for understanding ferrites and introduced the term ferrimagnetism.

The statistics on the number of research papers published on the hexaferrites alone show that in the year 1980 about 20 research papers, in 1990 about 50, in 2000 little more than 100 and in 2011, more than 230 papers have been published[4]. In recent years, due to the rapid growth in the utilisation of electrical and electronic devices, ferrites have been extensively used in a variety of applications like motors, generators, transformers, actuators, sensors, information storage, mobile communications, etc. The most used magnetic materials are ferromagnetic metals and alloys of ferrimagnetic ceramics[4, 5, and 6]. Researchers have even synthesised nanocrystalline ferrites and hexaferrites where small particle - sized magnetic media is required[5, 6, 7, 8]. Recent advances in nanostructured hexaferrites have demonstrated improved magnetic tunability, enhanced microwave absorption bandwidth, and potential integration into 5G and electromagnetic interference (EMI) shielding technologies [5, 6,7, 8].

Despite extensive reviews on hexaferrites, a concise correlation between structural blocks (S, R, T), cation substitution, and their combined influence on magnetic and dielectric properties is still scattered across the literature. The present review attempts to provide a unified structural–property–application perspective with emphasis on M-type hexaferrites and recent substitution strategies.

II.CLASSIFICATION AND STRUCTURAL FRAMEWORK OF FERRITES

Magnetic materials are commonly classified as soft, hard, and semi-hard depending on their coercivity and magnetic response [9]. Ferrites are further categorized structurally into orthoferrites (perovskite structure), spinels (cubic), garnets (complex cubic), and magnetoplumbite-type hexaferrites (hexagonal), as described by Wyckoff[10].

The crystal chemistry of spinel, garnet, and hexaferrites can be interpreted using structural building blocks denoted as S, R, and T blocks[4]. These blocks represent specific arrangements of oxygen layers and interstitial cation sites that combine to form various ferrite structures.

- The S block corresponds to a spinel-type unit ($MeFe_2O_4$), containing both octahedral and tetrahedral cation sites.
- The R block is characteristic of hexaferrites and contains a large divalent ion (e.g., Ba^{2+}), introducing trigonal bipyramidal coordination sites unique to magnetoplumbite-type structures.
- The T block appears in certain hexaferrites and modifies the stacking sequence and coordination environment.
- Different hexaferrite families (M, Y, Z, W, X, U) arise from specific stacking sequences and combinations of these S, R, and T blocks, resulting in variations in lattice parameters, symmetry, and magnetic characteristics[4,12]. Among these structural classes, the spinel and garnet ferrites represent the fundamental cubic frameworks, while hexaferrites exhibit more complex layered architectures derived from specific S–R–T block stacking sequences.

The spinel structure:

'The spinel is a close-packed cubic structure, in which the positions of the layers of four oxygen atoms repeat every three vertical layers to form an ABCABC lattice, these layers being approximately 2.30 Å apart. In the spinel structure, the interstices between these cubic layers are alternatively filled with three octahedral sites and one octahedral and two tetrahedral sites per four oxygen atoms. The spinel layers form plates either four or six oxygen layers thick, and between the outer two layers at both ends of the plates, only octahedral sites are occupied by the metal atoms. These spinel plates are joined together by two possible barium-containing layers, which both have a hexagonal close-packed structure that repeats every two vertical layers forming an ABAB lattice'[4]. The spinel has a general molecular formula $MeFe_2O_4$ (where Me is a divalent metal cation).

The garnet structure:

The garnet is a group of isomorphous minerals with a complex cubic structure with trivalent cations occupying tetrahedral (B), octahedral (A), or dodecahedral − a 12-sided distorted polyhedral − (C) sites. These three crystallographically different kinds of sites A, B, and C are occupied as follows: $16Fe^{3+}$ in A, $24Fe^{3+}$ in B, and $24Me^{3+}$ in C. The interaction between the Fe^{3+} ions in A and B sites is strongly antiparallel. In the rare-earth garnets, the moment on the rare-earth ions in C sites is antiparallel to the resultant moment of the Fe^{3+} ions. Specifically, the chemical formula for garnets is $3Me_2O_3{\cdot}5Fe_2O_3$ (alternatively dividing all by two - $Me_3Fe_2\,(FeO_4)_3$) where me represents the trivalent rare earth such as non-magnetic yttrium or a magnetic rare earth such as from lanthanum to ytterbium. The most magnetically interesting of these synthetic garnets are those in which Me is yttrium (Y) or one of the rare earths from gadolinium (Gd) to lutetium (Lu). The Y^{3+} ion in YIG has no moment, so that the net moment of YIG is solely due to an unequal distribution of the same kind of ions (Fe^{3+}) in A and B sites, as in γ-Fe_2O_3[3].

Hexagonal Ferrites: General Structural Features

Hexagonal ferrites (hexaferrites) constitute a structurally complex class of ferrimagnetic oxides characterized by layered stacking along the c-axis of the hexagonal lattice. Unlike cubic spinel or garnet ferrites, hexaferrites exhibit anisotropic crystal structures defined by lattice parameters *a* (basal plane width) and *c* (stacking height).

Their structures can be interpreted as specific stacking sequences of S, R, and T structural blocks, leading to various hexaferrite families such as M, Y, Z, W, X, and U types. These families differ primarily in the arrangement and repetition of these blocks, resulting in variations in unit cell length, symmetry, magnetic anisotropy, and magnetic sublattice interactions[4, 12].

The diversity in stacking sequences gives rise to a wide range of magnetic behaviors, from strongly uniaxial hard magnetic materials (M-type) to planar or weakly anisotropic systems (Y- and Z-types). Recent structural investigations using advanced diffraction and neutron scattering techniques have provided deeper insights into site occupancy, spin configuration, and magnetocrystalline anisotropy in various hexaferrite families[12].

Different types of Hexaferrites:

The various hexaferrite families arise from distinct stacking combinations of S, R, and T blocks, leading to differences in lattice dimensions, symmetry, and magnetic anisotropy.

M-type ferrite ($BaFe_{12}O_{19}$)

The M-type structure consists of alternating S and R blocks arranged along the c-axis and crystallizes in the space group $P6_3/mmc$. It possesses lattice parameters $a \approx 5.89$ Å and $c \approx 23.17$ Å[11]. The relatively high c/a ratio (~3.94) results in strong uniaxial magnetocrystalline anisotropy, making M-type ferrites hard magnetic materials. Recent experimental studies have demonstrated that subtle modifications in cation distribution and lattice distortion significantly influence anisotropy and coercivity in M-type hexaferrites[13].

W-type ferrite ($BaMe_2Fe_{16}O_{27}$)

The W-type structure contains additional S blocks compared to M-type, resulting in a longer unit cell ($c \approx 32.84$ Å) and modified magnetic interactions. It also crystallizes in $P6_3/mmc$ symmetry.

Y-type ferrite ($Ba_2Me_2Fe_{12}O_{22}$)

Y-type ferrites are formed by S and T block combinations and belong to the R3m space group. Their longer unit cell ($c \approx 43.56$ Å) leads to modified magnetic sublattice interactions and typically reduced anisotropy.

Z-type ferrite ($Ba_3Me_2Fe_{24}O_{41}$)

Z-type structures combine features of Y and M blocks and exhibit intermediate magnetic characteristics with $c \approx 52.30$ Å. Contemporary studies have correlated stacking periodicity and block rotation with variations in magnetic resonance frequency and microwave response[14].

X- and U-type ferrites

These structures involve extended stacking sequences and possess very large unit cells. Their complexity results in diverse magnetic behaviours, often tailored for microwave absorption applications.

Over seventy hexaferrite compounds have been identified, with increasingly complex stacking sequences leading to extraordinarily large unit cells[4].

III.CRYSTAL STRUCTURE OF M-TYPE HEXAFERRITES

In 1938, Adelskold[23] carried out a detailed investigation of the natural mineral magnetoplumbite and established that compounds such as $PbFe_{12}O_{19}$, $BaFe_{12}O_{19}$, and $SrFe_{12}O_{19}$ crystallise in a hexagonal structure derived from this mineral. The magnetoplumbite structure, commonly referred to as the M-type structure, is generalised to oxides with the formula $XY_{12}O_{19}$, where X is typically a divalent cation (Ba^{2+}, Sr^{2+}, Ca^{2+}) and Y is a trivalent cation such as Fe^{3+}.

M-type hexaferrites crystallize in the space group $P6_3/mmc$ (No. 194)[10, 24]. The oxygen ions form a hexagonal close-packed lattice with an ABAB stacking sequence along the [001] direction. Within this framework, three types of interstitial sites are available: octahedral, tetrahedral, and trigonal bipyramidal. The large Ba^{2+} ion occupies specific interlayer positions and induces local lattice distortion, giving rise to the unique five-coordinated trigonal bipyramidal site that is absent in conventional spinel ferrites.

In magnetoplumbite ($BaFe_{12}O_{19}$), Fe^{3+} ions occupy five crystallographically distinct sites: 2a, $4f_1$, $4f_2$, 12k, and 2b. Among these, 2a, $4f_2$, and 12k are octahedral sites; $4f_1$ is tetrahedral; and 2b corresponds to the trigonal bipyramidal site. Ba^{2+} occupies the 2d position, while oxygen ions are distributed over 4e, 4f, 6h, and 12k sites[10, 24]. The distribution of Fe^{3+} ions among these sublattices leads to multiple magnetic sublattices with different exchange interactions.

The magnetic ordering in M-type hexaferrites arises from superexchange interactions between Fe^{3+} ions mediated through oxygen. The magnitude of the exchange interaction depends strongly on the Fe–O–Fe bond angle (φ) and interatomic distances. Following Gorter's analysis [16], larger bond angles (≈140°) favor stronger antiferromagnetic superexchange, whereas smaller angles (≈80–90°) produce weaker interactions. In M-type ferrites, significant octahedral–octahedral interactions across relatively large bond angles contribute to strong coupling between sublattices and account for the high Curie temperature of $BaFe_{12}O_{19}$ (≈450 °C)[24].

The ferrimagnetic structure results from incomplete cancellation of magnetic moments between octahedral, tetrahedral, and bipyramidal Fe^{3+} sites, yielding a net magnetic moment per formula unit.

Hexagonal M-type ferrites are magnetically hard materials exhibiting large coercivity due to strong uniaxial magnetocrystalline anisotropy[25].

The first-order anisotropy constant K_1 can be expressed as: $K_1 = K_{1,d} + K_{1,Fe^{3+}}$

where $K_{1,d}$ arises from dipole–dipole interactions and $K_{1,Fe^{3+}}$ originates from spin–orbit coupling of Fe^{3+} ions [4, 6]. The trigonal bipyramidal (2b) site plays a particularly important role in determining the uniaxial anisotropy due to its asymmetric crystal field environment.

The electrical resistivity of magnetoplumbites varies widely ($10^{-2} - 10^{11}$ Ω·cm at room temperature) depending on composition and cation distribution[24]. Electrical conduction is generally attributed to electron hopping between Fe^{2+} and Fe^{3+} ions, and is strongly influenced by structural defects and substitution.

IV.SYNTHESIS ROUTES AND MICROSTRUCTURAL EVOLUTION

The formation of the hexagonal ferrites is a complicated process, and the mechanisms involved are not yet fully understood despite of the fact that having been investigated by many researchers since their discovery. Starting from the conventional ceramic method, a number of synthesis routes have been developed by researchers. It is well established that structural and thus other properties of the hexaferrites strongly depend upon the sintering temperature as well as the method of synthesis. Some of the methods that are frequently used to synthesise the hexaferrites are briefly discussed below.

a) **Conventional ceramic technique** - The conventional ceramic technique involves the heating of the mixtures of oxides and barium carbonate powders in stoichiometric proportions in a furnace (which is well temperature-controlled), at a high temperature ranging from 1000 °C to 1400 °C to produce the desired phase of the ferrite[15]. 'Hexaferrites assembled using ceramic method are from pre-existing crystallite blocks via topographic reactions', and the 'ceramic technique is ideal for synthesis, especially for complex compounds'[4]. Pullar[4] reviewed that the other methods, like the chemical coprecipitation method, salt-melt method, ion exchange process, sol-gel method, etc. have a varying degree of success.

b) **Coprecipitation method-** In the coprecipitation method, the chemical coprecipitation of salts with base has been used to produce ferrites. Using this process, BaM can be formed at temperatures between 750 °C to 900 °C with a submicron grain size[16]. To improve the homogeneity, a wet chemical process was also developed in which an aqueous solution of metal salts was precipitated with a strong base and then oxidised by bubbling air to get the desired sample[17].

c) **Sol- gel method -** In the sol-gel method, precursor particles, which can be inorganic or metallo-organic on the colloidal scale, are mixed.–This homogeneous mixing is maintained until the material condenses into a gel, resulting in fine-grained polycrystalline ferrites.

d) **Aqueous sol - gel method -** In aqueous sol-gel synthesis, an aqueous solution of metal salts is coprecipitated by a base, but instead of them drying and firing, the precipitates are treated to form a colloidal sol, which can then be concentrated to gel and subsequently fired to give the ferrite[18].

e) **Spray drying method -** To synthesise the ferrite by the spray drying method, fine hollow spheres of BaM are made from aerosolisation of a solution of metal nitrates. It is atomised and spread in a nitrogen gas flow into a heated zone for a very short time. After this, amorphous spheres are collected, and pure BaM is formed with further annealing, but has poor magnetic properties[4, 19]. Ultrafine particles can be synthesised at low temperatures from the decomposed citrates. The decomposition reaction is exothermic. In this reaction, CO_2 is generated, which results in a very porous product[20]. This is also known as the Pecchini process.

f) **Glass crystallization method -** In the glass crystallisation method, the ferrite is produced from raw materials mixed with a glass flux, which is then cooled and heated to a temperature below the melting point of the glass to produce the ferrite. The glassy matrix is then removed with acid to leave the polycrystalline ferrite product[21].

g) **Hydrothermal synthesis method -** In the hydrothermal synthesis method, a solution of metal salts and a base is autoclaved under pressure to give the product, which is often a mixed phase containing unreacted precursors. They are removed by washing with dilute HCl[22]. In recent years, advanced techniques such as microwave-assisted synthesis, spark plasma sintering (SPS), and molten salt methods have been employed to achieve phase purity at lower temperatures and controlled nanostructures, leading to improved magnetic performance and reduced energy consumption.

V.MAGNETIC AND ELECTRICAL PROPERTIES

Intrinsic Properties

All hexagonal ferrites contain at least one large divalent metal ion (usually Ba^{2+} or Sr^{2+}), which causes a slight perturbation in the lattice due to size differences and is responsible for the magnetocrystalline anisotropy in the hexaferrites. Further, the dielectric losses occur in materials due to the damping of the vibrations of the electrical dipoles as well as intrinsic losses due to crystal structure, extrinsic losses due to impurities, porosity and grain boundaries. Many attempts have been made to study the magnetic and electrical characteristics of BaM. In addition to the method of synthesis, these characteristics depend upon the type and concentration of substitution. In the subsequent paragraphs, some examples of the M-ferrites with interesting magnetic and electrical properties have been given.

Pullar et al. [27] manufactured aligned and random fibers of SrM and BaM by blow spinning from an aqueous inorganic sol-gel precursor, where enhanced magnetisation along the axis of alignment with respect to perpendicular to the axis was observed. These BaM random fibers are reported to have M_s = 63.8 emu g^{-1} and H_c = 428.1 kA/m, and SrM random fibres with M_s = 63.3 emu g^{-1} and H_c = 452.8 kA/m. The T_c values for BaM and SrM were observed to be 450 °C and 470 °C, respectively. A comparative analysis of the magnetic properties of $BaFe_{12-x}Me_xO_{19}$ (Me = Al, Ga, Sc, In) was carried out by Albanese et al.[28], where the influenceof the substitution of iron ions on the magnetic ordering and on the magnetic properties has been discussed. It is observed that the occupation by non-magnetic ions of lattice sites inside the R-block leads to peculiar helical spin structures, and further, the relevant contribution of iron ions in the 12k sublattice to the single-ion uniaxial anisotropy has been confirmed. BaM was synthesised by Janasi et al.[29] with the co-precipitation method by changing the sequence of addition of the reagents, the calcination temperature to 800 °C and 1100 °C and the time of calcination 1.5 and 4.0 h. The BaM so obtained by calcinating at 1.5 h shows H_c = 440 kA/m, whereas for samples calcinated at 1100 °C, H_c reduces to 162 kA/m. It is further reported that the method of preparation leads to the formation of different crystalline phases, thereby affecting the magnetic properties of the samples and especially the value of M_s. Out of the effect of calcination time and temperature on the values of H_c, calcination temperature has a significant effect.

'The CoTiM is a very important class of ferrites which was first synthesized in the 1960s as a way of lowering grain size and coercivity considerably without much loss of M_s[30] and was commercially developed by Toshiba in the 1980s to give particles 50 nm wide and 15 nm thick with a coercivity of 159 kA/m[31]. Since then, coercivity has been lowered considerably with further substitution to give a series of ferrites with the formula $BaCo_xTi_xFe_{12-2x}O_{19}$, which can be tuned with a variation in x to give the desired properties, reducing H_c to a minimum value at $x = 1.2$ [26]. The axial anisotropy also reduces with substitution until it becomes in-plane at $x = 1.3$[32]. This is because although the Ti^{4+} ion only substitutes in octahedral sites, the Co^{2+} ion substitutes in tetrahedral sites and the trigonal bipyramidal site, which is the major contributor to the uniaxial anisotropy[33]. The Co^{2+} ions were shown to be in tetrahedral positions with a value of $x < 0.8$ [34], and beyond this substitution level, they begin to occupy the five-coordinate sites as well. A cone of magnetisation begins to develop as a result of this non-colinearity. This also has an effect of reducing H_c [35].

A detailed study of non-stoichiometric $BaCo_xTi_xFe_{11.6-2x}O_{19}$ made by the citrate route produced sub-domain-sized particles with good magnetic properties below 1000 °C. Grain size was found to decrease with x, and H_c decreased linearly from 378 kA/m for $x = 0.2$ to 80 kA/m for $x = 0.8$, and a very soft ferrite was achieved for $x = 1$, while M_s remained reasonably stable around 60–55 A m^2 kg^{-1}[36].

BaCoTiM has been made by melt-flux precipitation at 980 °C with a high M_s of 58 Am^2/kg and a coercivity of 64 kA/m [37]. When made from a mixture of FeOOH, $Ba(OH)_2$ and cobalt and titanium alkoxides, the M phase always existed with some minor haematite impurity. Nonetheless, a good quality magnetic product was produced, with M_s = 49 A m^2/kg and H_c = 135 kA/m at $x = 0.5$; M_s = 48 Am^2/kg and H_c = 88 kA/m at $x = 0.7$ and M_s = 45 Am^2/kg and H_c = 16 kA/m at $x = 1$[38].

A nanosized BaCoTiM ferrite was made from the stearic acid gel method, with a grain size of only 30 nm but a coercivity of 88 kA/m for a substitution level of $x = 0.8$ [39]. The glass crystallisation method resulted in more loss of H_c than in coprecipitated samples, probably through increased lattice diffusion, so that at $x = 0.65$, the value of M_s is 60 Am^2/kg and H_c = 125 kA/m; at $x =$

0.85, M_s = 54 Am^2/kg and H_c = 50 kA/m and at x = 0.9 H_c = 41 kA/m [4, 40, 41].

'The dielectric properties as well as the magnetic properties are very important for many high-frequency applications of hexaferrites, particularly if they are to become integrated chip components. The key properties of interest for many applications are resistivity and permittivity (dielectric constant, ε, or relative permittivity, ε_r, is a measure of how easy it is to establish, or ''permit'', an electric flux in a material), which in general should both be as high as possible at as higher frequency as possible. As with permeability, a complex permittivity can be measured, consisting of real (ε') and imaginary (ε'') components, from which dielectric resonance and losses can be calculated. The metal cations and the oxygen anions form dipoles, creating an intrinsic dielectric polarisation. The dominant conduction mechanism in ferrites is electron hopping between Fe^{2+} and Fe^{3+}. Well-sintered samples of BaM, SrM and CaM (the later doped with 4 mol% La_2O_3) were found to have ε_r of ~10, ~20 and ~15, respectively, when measured at 10 GHz, and the Co_xTi_x - substituted versions had similar values, with ε_r decreasing slightly as x increases. From the measurements of ε' and ε'', $CaCo_xTi_xFe_{12-2x}O_{19}$ (with 4 mol% La_2O_3) seemed to show a clear dielectric resonance at 10 GHz for x = 0.8 and at 8.75 GHz for x = 1 [4, 42]'. Recent studies have focused on broadband microwave absorbers based on doped M-type hexaferrites, reporting reflection losses below −40 dB and effective absorption bandwidths exceeding 5 GHz in the X- and Ku-bands.

Dielectric properties of specimens with compositions $BaO.(6\text{-}x)Fe_2O_3.\ x\ Sb_2O_3$ with x = 0, 0.025, 0.1 and 0.3 were studied by Brahma et al. [43] in the frequency range 1.5kHz to 10MHz. It was observed that the doping of Sb_2O_3 increased the dielectric permittivity drastically. Two loss peaks of similar activation energies were observed due to the presence of Fe^{2+}- Fe^{3+} and Sb^{3+}-Sb^{5+} pairs in the system. The authors have also carried out direct current electrical resistivity measurements on these specimens and found to have reduced resistivities. The analysis of the data shows that the electrical conductivity in this system arises due to a combined contribution of both small polaron hopping between Fe^{2+} and Fe^{3+} sites and bipolaron transport between Sb^{3+} and Sb^{5+} sites. A simple and economical route has been proposed by Iqbal et al.[44] for producing highly homogenous Zr-Cu substituted strontium hexaferrite nanoparticles by the co-precipitation method. The DC electrical resistivity measurements of $SrZr_xCu_xFe_{12-2x}O_{19}$ (x = 0.0 - 0.8) in the temperature range 27 °C to 527 °C indicates a significant increase in the room temperature resistivity with the addition of Zr-Cu up to $x \leq 0.4$ as well as the increase in activation energy is observed till $x \leq 0.4$.

Effect of Cation Substitution:

Most of the M compounds exhibit ferrimagnetic behavior which is due to the fact that the net magnetic moment results from an incomplete cancellation of moments of ferrite ions. The moment from the octahedrally surrounded ferric ions in the spinel blocks and those in the trigonal bipyramidal sites are opposed by a minority of ferric ions in tetrahedral sites in spinel block, along with those in octahedral sites near the barium plane. The replacement of ferric ions by non-magnetic ions, for example, in the tetrahedral sites in the spinel blocks, would lead to an increase in the magnetic moment of the compound, since the dominance of the moments from the octahedral sites of spinel and trigonal sites would thereby be enhanced. Similarly, replacement in the latter by non-magnetic cations would reduce the magnetic intensity of the compound. The resistivity of oxides can also be altered by suitable introduction of impurities in small quantity (doping) or by mixing oxides (of the same structure) of high and low resistivities. For example, the room temperature resistivity of calcium ferrite ($CaO_2Fe_2O_3$) is usually greater than 10^6 ohm-cm but it is reduced remarkably to 10^3 ohm-cm when doped with yttrium oxide. The variation in the resistivity is explained on the basis of the mechanism proposed by Verwey et al. [52]. The properties of the ferrites can be modified according to the requirements in a particular application by changing the method of synthesis, sintering conditions, sintering temperature and most effectively by substituting suitable cations in suitable concentration.

As a part of a constant search for new materials with modified and improved properties of BaM, Ba^{2+}/Ba^{2+}-Fe^{3+} have been replaced by Pb^{2+}, La^{2+}, Sr^{2+}, Ca^{2+}, etc. [53-56]. Also Fe^{3+} ions may be substituted by any other trivalent ion like Al^{3+}, Ga^{3+}, Sc^{3+}, In^{3+}, etc. [28] or by combination of divalent and tetravalent ions like Zn^{2+}-Ti^{4+}, Co^{2+}-Ti^{4+}, Mn^{2+}-Ti^{4+}, Zn^{2+}-Sn^{4+}, Ni^{2+}-Sn^{4+}, etc. [57]. The researchers found that due to the addition of the substituents in different concentrations, there is a change in the properties of the BaM. This is attributed to the fact that the substituted ions have different site occupancies and show different behavior for applied electric and magnetic fields. Dho et al. [58] studied the effects of grain boundaries on coercivity of $BaFe_{12}O_{19}$ where the large coercivity observed in $BaFe_{12}O_{19}$ was mainly due to pinning of magnetization at the grain boundaries.

Among the various coupled substitutions like Zn^{2+}-Ti^{4+}, Co^{2+}-Ti^{4+}, Mn^{2+}-Ti^{4+}, Ni^{2+}-Sn^{4+}, etc. the substitution of Co^{2+}-Ti^{4+} is a popular combination which replaces the Fe^{3+} ions to reduce the magnetocrystalline anisotropy and enhance the electrical resistivity to meet wide range of industrial applications[44]. Rewatkar et al. [59] tried to explain the dc conductivity of $CaFe_xAl_x(CuTi)_{6-x}O_{19}$ using Verwey model.

Tehrani et al. [60] synthesised a composite of substituted hexaferrite $BaMg_{0.25}Mn_{0.25}Co_{0.5}Ti_{1.0}O_{19}$ mixed with polyvinyl chloride (PVC), as a single-layer electromagnetic wave absorber in which a bandwidth of 4.5 GHz has been covered by this composition at a thickness of 2.7mm. The authors have proposed that the material has an attractive potential in microwave applications as a radar absorbing media.

'The substitution of part of the Fe^{3+} ions by other magnetic or diamagnetic cations leads to both changes in the exchange interactions between the magnetic sublattices and to the appearance of new positions of the iron ions. Therefore, as x increases, the 12k site can split into two subpatterns, 12k and 12k'. This may be the result of changes in the environments or neighbors of the Fe^{3+} ions on the 12k site, when substitution takes place at R blocks. The 12k' site increases at the expense of the disturbed 12k sites. According to ligand field theory, ions with d^1, d^2, d^3 and d^4 orbitals prefer tetrahedral coordination and ions with d^6, d^7, d^8 and d^9 orbitals occupy octahedral positions. Ions with d^0, d^5 and d^{10} orbitals have no site preference. It is also known that less electronegative ions prefer tetrahedral coordination. However, the tendency to occupy a particular site depends also on the partner cation. Taking these facts into account Angeles et al. [61] believed that as Ni^{2+} and Sn^{4+} ions possess nearly the same electronegativity

(1.91 and 1.96, respectively), Ni^{2+} (d^8) ions would go to octahedral (12k, 2a, $4f_2$) sites, and Sn^{4+} (d^{10}) ions would go to the tetrahedral ($4f_1$) and bipyramidal (2b) ones. They studied $BaFe_{12-2x}(NiSn)_xO_{19}$ compounds using attrition milling and characterized it by VSM, TGA, XRD and Mossbauer spectroscopy. 'The substitution x was changed from 0 to 0.3. The experimental results suggest that Ni–Sn cationic mixtures produce similar effects on the magnetic properties, to those observed for Ir–Co and Ir–Zn substitutions. The Ni^{2+}–Sn^{4+} substitution reduced the uniaxial magnetic anisotropy field, H_a rather fast, becoming planar at low rates. Mossbauer studies revealed that Sn^{4+} ions replace Fe^{3+} ions on 2b and $4f_1$ sites, while Ni^{2+} ions prefer $4f_2$ and 2a sites. A large variation of the intrinsic coercivity, H_c (381.9–93.9 kA/m) was obtained as a function of the substitution' [61].

A series of $BaCr_xGa_xFe_{12-2x}O_{19}$ (x = 0.0–0.4) were synthesized by sol–gel auto-combustion route by Ali et al. [62] to study the effects of Ga–Cr substitution on structural and magnetic properties of BaM. The crystallite size calculated by the Scherer equation was found to be in the range of 30–48 nm, which is small enough to obtain a suitable signal-to-noise ratio in the high-density recording media. The saturation magnetization and retentivity were observed to increase from 2.078×10^3 to 2.385×10^3 Gauss and 1.286×10^3 to 1.677×10^3 Gauss, respectively. The enhanced coercivity is attributed to an increase in the magnetocrystalline anisotropy. The authors propose that due to the high values of retentivity and coercivity, these materials to be best candidates for high-density recording media and permanent magnets. Tyagi et al. [63] developed a nanocomposite of $SrFe_{11}Zn_{0.5}Ni_{0.5}O_{19}$ of hard/soft ferrite for enhanced microwave absorption by the co-precipitation method. The sample has super paramagnetic nature. The hysteresis loops show an increase in saturation magnetisation from 1.042 emu/g to 59.789 emu/g with an increase in temperature. The 'as-synthesised' particles with spherical and needle shapes have size in the range of 20– 25 nm. Further, these spherical and needle-shaped nanoparticles tend to change their morphology to hexagonal plate and pyramidal shapes with an increase in HT temperatures.

The effect of such a systematic morphological transformation of nanoparticles on dielectric (complex permittivity and permeability) and microwave absorption properties are estimated in X band (8.2–12.2 GHz).The maximum reflection loss of the composite reaches -29.62 dB (99% power attenuation) at 10.21 GHz which is a suitable application in RADAR absorbing materials.

There are a few reports on Cu^{2+}-Ti^{4+} co-substitution in the ferrites, such as Hsiang et al. [64] investigated structural and dielectric properties of $(Zn)(Cu_xTi_xFe_{1.98-2x})O_{3.97}$. Mazen et al. [65] studied thermoelectric power and electrical conductivity of $Cu_{1+x}Ti_xFe_{2-2x}O_4$. Jazirehpour et al. [66] synthesized nanosized $BaFe_{12-x}Mg_{0.5x}Ti_{0.5x}O_{19}$ (x = 0.0-5.0) ferrites using a 'modified sol-gel process which could provide a reliable capability for efficient substitution of elements like Ti in high substitution levels such as x = 5. Only one concentration of Mg^{2+}-Ti^{4+} substitution in BaM i.e. $BaFe_9Mg_{1.5}Ti_{1.5}O_{19}$ has been studied by Shams et al.[67] with reference to their magnetic and electromagnetic wave absorption properties and have reported a wide bandwidth absorption in the frequency range 13 to 17GHz with reflection losses less than -20 dB. The magnetic properties of the barium hexaferrite series $BaFe_{12-2x}Zn_xTi_xO_{19}$ (for $0 \leq x \leq 2.0$) by means of magnetization (SQUID), neutron diffraction and Mossbauer measurements studied by Wartewig et al. [68] suggest that for $x \leq 0.8$, an increase of the low-temperature net magnetization relatively to the unsubstituted sample. For larger substitution, the studies indicate a deviation of the Fe spins from the collinear structure which leads to a decrease of the net magnetization. The authors further investigated to what extent and accuracy the molecular field approximation describes the observed magnetic properties for the x = 0 and 0.4 samples, taking into account the exchange integrals between different magnetic sublattices. Dube et al. [69] carried out complex permittivity and permeability measurements of $BaFe_{12-2x}Zn_xTi_xO_{19}$ ($0.2 \leq x \leq 0.6$) in the frequency range 8-13GHz and reported an abnormal dielectric behavior i.e. an increase in dielectric constant with an increase in frequency. Results on magnetic studies of $Ba_{0.5}Sr_{0.5}Fe_{12-2x}(ZnTi)_xO_{19}$ and $Ba_{0.75}Ca_{0.25}Fe_{12-2x}(ZnTi)_xO_{19}$ (x = 0.2 to 0.6) prepared by mechanical alloying by Angeles et al.[70] show that T_c decreased dramatically (drop ~ 39%) for BaCa samples, whilst for BaSr samples remained almost without change (diminution ~ 2%) at $x \leq 0.2$. In a recent study, the structural, magnetic and dielectric properties of $BaFe_{12-2x}Mg_xTi_xO_{19}$, $BaFe_{12-2x}Cu_xTi_xO_{19}$ and $BaFe_{12-2x}Zn_xTi_xO_{19}$ (x = 1, 1.5, 2) have been reported[15, 71, 72].

VI. APPLICATIONS

The magnetic materials with high M_s and H_c show a large square loop and thus large energy is required to demagnetize these materials. Most of the ferrites have high resistance and very low eddy current losses. Their high resistivity and low eddy current losses make ferrites ideal materials for microwave components, radar detection systems, satellite communication equipment, and related high-frequency technologies. The constant use of microwave devices, radar, and mobile communications has produced EM interference which have harmful effects on the biological tissues. Therefore, the ferrites are used as EM wave absorbers. The use of X and U ferrites as MW absorbers in the X band has been thoroughly investigated by researchers[17, 45]. The Li ferrites and substituted Li ferrites are used for microwave applications and as cathode material in Li batteries [46-48]. Magnesium-substituted batteries are used in electronic devices [49]. A powder of ceramic ferrite incorporated in a thermoplastic matrix forms plasto-ferrites. They are used in magnetic sealing strips inside the rim of the door of the refrigerator [50]. Soft ferrites are used wherever effective coupling between electric current and magnetic flux is required. They form an essential part of inductors and transformers in today's main application areas of telecommunication, power conversion and interference suppression. Heating (41–46°C) of specific tissues or organs for the treatment of cancer, known as hyperthermia, has attracted a lot of attention in recent years. For this purpose, superparamagnetic– ferrimagnetic Fe_3O_4 or α-Fe_2O_3 particles have been extensively studied as they are biocompatible. The feasibility of other ferrites is also being explored [51]. The soft ferrites have applications in electronics, for television, micromachines, small transformers with special utilisation, transducers, magnetic recording heads, computer peripherals, MW installations, etc.

VII.CONCLUSIONS

It can be concluded that hexaferrites, particularly M-type ferrites, have been extensively investigated due to their layered crystal structure, high uniaxial magnetocrystalline anisotropy, and useful magnetic and electrical properties. The structural arrangement based on S, R, and T block stacking plays an important role in determining cation distribution, magnetic interactions, and anisotropy, which in turn influence parameters such as coercivity, saturation magnetization, and high-frequency behavior.

Considerable efforts have been made to modify the properties of BaM through different synthesis routes and controlled cation substitution. Substitution at specific crystallographic sites allows tuning of magnetocrystalline anisotropy, Curie temperature, electrical resistivity, and microwave absorption characteristics. Recent studies on nanostructured hexaferrites and substituted systems have further enhanced their potential for applications in high-frequency devices and electromagnetic wave absorption.

Further work is required to achieve better control over site occupancy, defect structure, and multi-ion substitution in order to optimize performance for specific applications. The close relationship between crystal structure and magnetic properties continues to provide guidance for the development of improved hexaferrite materials.

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