Chapter 10

Hexagonal Ferrites, Synthesis, Properties and Their Applications

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Abstract

We intend to report on possible fabrication routes for all types of hexagonal ferrites which are known for their wide area of use and applications. Hexagonal ferrites have now become an intense topic of research as they are the part of most of magnetic recording and data storage applications globally. Hexagonal or popularly known as ‘Hexa-ferrites’ are known for their utilization in permanent magnets and their utilization in electrical devices being operated at high frequencies especially at GHz frequencies. We have presented in this chapter all main six types of hexagonal ferrites i.e. M Type, Z-Type, Y-type, W-type, X-Type and U-type hexa-ferrites. Hexaferrites belong to ferromagnetic class of magnetic materials and their properties are purely dependent on intrinsic structure of ferrites. In this chapter, we aim to discuss more on M-type of hexa-ferrites, their properties and their applications. Also, recent advances on M-type ferrites are also a part of this chapter.

Keywords

Hexaferrites, M-Type, Anisotropy, High Frequency Applications

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1. Introduction

Ferrites belong to the class of most widely used magnetic materials with diverse application field. The applications covers a broad spectrum of today's technological devices whether electronic, storage devices, antennas and much more for high frequency applications. Ferrites are mainly classified into spinel, garnet, hexagonal, and perovskite types. Each type of these ferrites are unique in term of properties and corresponding applications [1, 2]. With the discovery of hexagonal ferrites in 1950's, there has been a continuous completion in exploring more exotic properties of hexa-ferrites. Most of the research works on hexa-ferrites are concentrated on their complex structure, structure modified properties and applications of theses ferrites [3-6]. Growing interest in the field of hexagonal ferrites can be analysed viewing the number of papers published on hexagonal ferrites during 2010-2020.

Figure 1 shows the publications data gathered from the Scopus database. We can see the tremendous increase in paper published on hexagonal ferrites. Hexaferrites finds their space in today's electronic devices, mobile as well wireless communication at GHz frequencies, EMI shielding, multiferroics, microwave absorbers, antenna applications etc. [7,8]. Hexagonal ferrites have certain advantages over their spinel counterparts, in term of higher ferromagnetic resonance frequency due to which they become suitable for being utilize in microwave regime to the preferred plane of magnetization [9].

Hexaferrites are mainly classified into six categories named as M-type (BaM or $\text{BaFe}_{12}\text{O}_{19}$, SrM or $\text{SrFe}_{12}\text{O}_{19}$), Z-type (Ba-Sr , Co hexagonal ferrite), Y-type ($\text{Ba}_2\text{Me}_2\text{Fe}_{12}\text{O}_{22}$), W-type ($\text{BaMe}_2\text{Fe}_{16}\text{O}_{27}$), X-type ($\text{Ba}_2\text{Me}_2\text{Fe}_{28}\text{O}_{46}$), U-type ($\text{Ba}_4\text{Me}_2\text{Fe}_{36}\text{O}_{60}$).

Out of possible types of hexaferrites, BaM constitutes the most of the commercially available magnetic material for diverse technological applications. In this chapter, we have focused on technological importance of hexaferrites, their classifications and most important properties and applications of hexaferrites.

Figure 1: Ten Years data on hexaferrites publications from 2010-2020 (searched using keyword 'hexaferrites' on Scopus).

2. Classification and structure of hexaferrites

All magnetic oxides containing iron as a primary metallic element are referred to as ferrites. Due to their ferromagnetic and insulating characteristics at room temperature, they have made a significant contribution to various technological applications [10, 11]. According to chemical content and crystal structure, hexagonal ferrites are classified into six types: M ($\text{AFe}_{12}\text{O}_{19}$), W ($\text{AMe}_2\text{Fe}_{16}\text{O}_{27}$), X ($\text{A}_2\text{Me}_2\text{Fe}_{28}\text{O}_{46}$), Y ($\text{A}_2\text{Me}_2\text{Fe}_{12}\text{O}_{22}$), Z ($\text{A}_3\text{Me}_2\text{Fe}_{24}\text{O}_{41}$), and U ($\text{A}_4\text{Me}_2\text{Fe}_{36}\text{O}_{60}$); where $\text{A} = \text{Ba}^{2+}$, Sr^{2+} , La^{2+} , Pb^{2+} and $\text{Me} =$ a bivalent transition metal [12, 13]. Hexaferrites are categorized into six primary categories based on their chemical formulae, as shown in Table 1. Figure 2 shows the schematic crystal structures of each hexaferrite. The M-type (magnetoplumbite) is the very well-known hexaferrite, whose structure (Figure 2) can be built up from S blocks interspersed by R blocks and symbolically expressed as RSR^*S^* . (The * equivalent block has been rotated 180 degrees around the hexagonal c-axis). The construction of the Y-type seen in Figure 2(d) can be thought of as an alternating stacking of the S and T blocks along the c-

axis TST'S'T"S" where the apostrophe indicates that the accompanying block is rotated 120 degrees around this axis. Ferrimagnetism is one of the most essential features of hexaferrite from a technical perspective. On the other hand, not all hexaferrites have a ferrimagnetic ordered state as the ground state. Some hexaferrites, such as M-type $\text{Ba}(\text{Fe},\text{Sc})_{12}\text{O}_{19}$ [14], W-type $\text{BaNi}_2\text{Sc}_2\text{Fe}_{16}\text{O}_{27}$ [15], and Y-type $(\text{Ba},\text{Sr})_2\text{Zn}_2\text{Fe}_{12}\text{O}_{22}$, are reported to explain a non-collinear magnetic order [16, 17].

Table 1: Hexaferrite types with their chemical formula, composition, unit cell, space group and stacking order.

Ferrite	Chemical Formula	Space Group	C(Å)	Unit Cell	Stacking order
M	$(\text{Ba},\text{Sr})\text{Fe}_{12}\text{O}_{19}$	$P6_3/mmc$	~22	2M	RSR*S*
W	$(\text{Ba},\text{Sr})\text{Me}_2\text{Fe}_{16}\text{O}_{27}$	$P6_3/mmc$	~33	2W	RS ₂ R*S* ₂
X	$(\text{Ba},\text{Sr})_2\text{Me}_2\text{Fe}_{28}\text{O}_{46}$	$R\bar{3}m$	~84	3X	(RSR*S* ₂) ₃
Y	$(\text{Ba},\text{Sr})_2\text{Me}_2\text{Fe}_{12}\text{O}_{22}$	$R\bar{3}m$	~43	3Y	TST'S'T"S"
Z	$(\text{Ba},\text{Sr})_3\text{Me}_2\text{Fe}_{24}\text{O}_{41}$	$P6_3/mmc$	~52	2Z	RSTSR*S*T*S*
U	$(\text{Ba},\text{Sr})_4\text{Me}_2\text{Fe}_{36}\text{O}_{60}$	$R\bar{3}m$	~113	U	(RSR*S*TS*) ₃

S = $\text{Fe}_6\text{O}_8^{2+}$ (spinel), R = $\text{BaFe}_6\text{O}_{11}^{2-}$ (hexagonal), and T = $\text{Ba}_2\text{Fe}_8\text{O}_{14}$ are the sub-units for stacking order (hexagonal). The asterix (*) denotes a 180° rotation of the relevant sub-unit around the hexagonal axis.

2.2 W-type hexaferrite

W-type hexaferrite is constructed of a superposition of S blocks and R blocks along the hexagonal c-axis with the structure $\text{RS}_2\text{R}^*\text{S}^*_2$, where R is a three oxygen layer block with composition $\text{BaFe}_6\text{O}_{11}$, S (Spinel block) is a two oxygen layer block with composition Fe_6O_8 , and "*" indicates that the respective block has been turned 180 degrees around the hexagonal axis. The R block still possesses a mirror plane, and the unit cell is built up of two molecular W units, yielding $\text{RS}_2\text{R}^*\text{S}^*_2$. The O^{2-} ions are found in close-packed layers, one Ba^{2+} (or Sr^{2+}) ion is located in R block replacing an O^{2-} ion, and the Fe^{3+} ions are located in five separate interstitial sublattices. The cell length of Fe_2W is 32.84 Å [12], and the unit cell is made up of two molecules of W units. It belongs to the $P6_3/mmc$ space group.

2.3 X- type hexaferrite

The crystal structure of X-type hexaferrite is made up of one M and one W unit, and it can be seen as a stacking of R and S blocks along the hexagonal c-axis in the order $(RSR^*S^*_2)_3$ with the blocks of the W segment rotated through 180 degrees relative to those of the M section. The unit cell is made up of four different layers of M and W type structures. Three chemical formula units ($Z = 3$) are contained in the elementary cell ($a = 5.88 \text{ \AA}$, $c = 84.11 \text{ \AA}$), and space group is $R\bar{3}m$ [12, 20, 21].

2.4 Y-type hexaferrite

The crystalline structure of these ferrites is made up of one T block and one S block. So because the T block lacks a mirror plane, the overlap of hexagonal and cubic close-packed layers requires three T blocks, with the relative placements of the barium atoms repeating every three T blocks. The unit cell formula is $3(ST)$, and it has hexagonal symmetry, with 18 oxygen layers and a repeat distance of only six oxygen layers, and a length of $43.56 \text{ \AA}$ on the c-axis. The crystal structure of Y-type hexaferrite is shown in Figure 2(d). This compound's fundamental structure belongs to the space group $R\bar{3}m$ and rhombohedral crystal structure, which prevents spontaneous polarisation [6].

2.5 Z-type hexaferrite

M and Y type hexaferrites make up Z-type hexaferrite, making it $ST + SR$, with a mirror plane in the R block and 11 layers of oxygen layer repeat distance. With a c-axis length of $52.30 \text{ \AA}$ [22], the crystal structure has 33 close-packed layers stacked along the hexagonal c-axis, giving order $RSTSR^*S^*T^*S^*$. A Z-type unit cell has 140 atoms; two molecular units combine to produce a single Z-type ferrite structure, which belongs to the $P6_3/mmc$ space group [6].

2.6 U-type hexaferrite

Block structure of U-type is $(RSR^*S^*TS^*)_3$ with the space group $R\bar{3}m$, and they are a superposition of 2M and Y-blocks along the c-axis. HRTEM pictures revealed the MMY stacking sequence of U ferrite, as well as stacking defects where the crystal structure was damaged by the addition of extra Y blocks [23]. The U unit cell, which is made up of three molecular units with $a = 5.88 \text{ \AA}$ and $c = 38.16 \text{ \AA}$ [6]. $Sr_4Co_2Fe_{36}O_{60}$ has lattice parameters of $a = 5.86 \text{ \AA}$ and $c = 112.3 \text{ \AA}$ [12].

Figure 2: Schematic (a) crystal and (b-d) proposed magnetic structures of magnetoelectric Y-type hexaferrites. In the Y-type hexaferrites, (b) the proper-screw (or longitudinal-conical) magnetic structure transforms into (c) the transverse-conical one, which allows finite polarization by applying a magnetic field along the c plane. By applying higher magnetic fields (Reprinted with the permission from ref. [24]).

Figure 2: (d-f) the collinearly ferrimagnetic ordered state appears. The ratio of Sr to Ba has a significant impact on the (Fe, Me)-O-(Fe, Me) bond angles surrounded by dashed blue ellipsoids (Reprinted with the permission from ref.[24]).

3. Synthesis routes for hexaferrites

Hexagonal ferrites are highly difficult to prepare and involve much complicated steps in synthesis routes. Most of methods when utilized yield some by-products along with the required phases. A number of the researchers have opted various synthesis route like sol-gel [25], co-precipitation method [26], solution combustion method [27], hydrothermal method [28].

3.1 Traditional ceramic method

This process entails combining raw components by either grinding or melting them together, or just applying heat to a mixture of raw components. In the absence of solvents, it can happen with both wet and dry procedures. Wet mixing methods use an aqueous suspension, agitator mill, or vibration drum. This process is quite effective, but it demands a lot of energy for dewatering and drying. Dry mixing is done in a drum or ball mill, which grinds and mixes the components. Yang et al. [29] fabricated M-type Mn, Zn doped SrCaLa hexaferrite by traditional ceramic method. In the typical procedure, SrCO₃, CaCO₃, La₂O₃, ZnO, Mn₃O₄ and Fe₂O₃ were taken as raw materials. The needed weights of various starting ingredients for different stoichiometric ratio of compositions were weighted in estimate quantities and ball-mixed in a ball mill for 8 hours with distilled water. In an electric furnace, the combined powders were presintered at 1240 °C in air for 2 hours. Table 2 shows the effect of dopant on M-type SrCaLa hexaferrite.

Table 2: Magnetic parameters of different substituted SrCaLa (Reprinted with permission from ref. [29]).

MnZn content (x)	Ms (emu/g)	Mr (emu/g)	Hc (Oe)	Mr/Ms
0.0	69.15	25.32	957.0	0.366
0.1	70.52	14.32	417.6	0.203
0.2	69.10	12.97	399.2	0.188
0.3	65.69	16.11	967.5	0.356
0.4	62.43	22.25	967.5	0.356
0.5	59.62	23.45	1293.5	0.393

Figure 3: FE-SEM micrographs of the M-type $\text{Ca}_{0.4}\text{Sr}_{0.6-x}\text{Nd}_x\text{Fe}_{12.0-x}(\text{Nb}_{0.5}\text{Zn}_{0.5})_x\text{O}_{19}$ hexaferrites with Nd-NbZn content (x) of (a) $x=0.00$, (b) $x=0.08$, (c) $x=0.16$, (d) $x=0.24$, and (e) $x=0.32$ [30] (Copyright permission with the licence number: 5087660456363).

Modifications to the traditional ceramic technique yielded $\text{PbFe}_{12}\text{O}_{19}$ hexaferrite powders with strong coercivity. At this temperature (presintered at $900^\circ\text{C}/2\text{ h}$ and sintered at $1000^\circ\text{C}/2\text{ h}$), the highest parameters of saturation magnetization and coercive field were 62.0 emu/g and 4.0 kOe , respectively [31]. The traditional ceramic technique was used to make Nd-NbZn co-substituted M-type Ca-Sr hexaferrites with nominal compositions of $\text{Ca}_{0.4}\text{Sr}_{0.6-x}\text{Nd}_x\text{Fe}_{12.0-x}(\text{Nb}_{0.5}\text{Zn}_{0.5})_x\text{O}_{19}$ ($x = 0.00-0.32$) [30]. The grains have platelet-like morphologies, according to FE-SEM micrographs (Figure 3). As can be seen, following Nd-NbZn co-substitution, the average grain size does not change significantly, and the grain size is clearly greater than the crystallite size ($41.4\text{-}56.8\text{ nm}$) [30].

3.2 Co-precipitation method

Co-precipitation method is a very old method and being used for preparation of ferrites for decades. Co-precipitation method involves salts like chlorides or nitrates and a base as raw materials for co-precipitation of desired phase. A number of reports are available on M-type ferrites synthesized through co-precipitation method. However, this method is

much more sensitive to the experimental conditions involved in synthesis route. In general, it has been noticed that for the preparation of BaM ferrites, this method requires Fe deficient environment and non-stoichiometric mixture of salts [32]. In a recent work, Hg doped BaM ferrite were synthesized using co-precipitation method [33]. In the typical procedure, barium chloride, ferric chloride, and sodium hydroxide were taken as raw materials. Chlorides taken in stoichiometric ratio were dissolved in distilled water and magnetically stirred at 60 °C. pH of the solution was maintained at 12 with the drop wise addition of alkaline NaOH solution. After two hours of stirring, precipitates were collected with the help of magnet and washed a number of times till the pH of the filtrate again reached 7. After drying, the particles are sintered at variable sintering temperature. The formation of BaM takes place but with an additional phase of Fe₂O₃ at some sintering temperatures of 700, 800, 900, 1000°C. Results shows that sintering temperature must be optimized for the single phased BaM samples (Table 3). The magnetic parameters like saturation magnetization, remnant magnetization and coercive field increased with the Hg content, however intrinsic coercivity field decreased.

Table 3: Barium hexaferrite (synthesized at 700, 800, 900, and 1000 °C) and structural parameters(Reprinted with permission from ref. [33]).

Sample	BaFe ₁₂ O ₁₉ (%)	Fe ₂ O ₃ (%)	a (Å...)	c (Å...)	L (nm)
B-700	69.7	31.3	5.87	23.239	32.88
B-800	82.2	17.8	5.875	23.206	34.76
B-900	91	9	5.809	23.234	35.45
B-1000	100	0	5.893	23.132	36.08

3.3 Sol-gel route

Sol-gel method is known for mixing of particles of inorganic or metallo-organic precursors on the colloidal scale. Sol gel method involves mostly mixing of metal nitrates in desired solvent followed by continuous stirring of the solution. In general, citric acid solution is added to the above solution where citric acid works as chelating agent and helps in the formation of complex structure [34]. pH of the solution is maintained by dropwise addition of ammonia solution. Later on with heating, gel formation takes place. This gel on annealing at optimized temperature yields the desired material [35]. However, the experimental conditions like heating conditions, pH, ratio of chelating agent plays a

vital role in morphology, shape and properties of ferrites prepared through sol-gel route. Pullar et al. devised an unique aqueous sol-gel approach for the synthesis of a variety of hexagonal ferrites from stoichiometric precursors, by adding barium, strontium, and cobalt salts to an acidpeptised iron(III)hydroxide (FeOOH) sol. This was utilised to make BaM [36], SrM [37], Co₂Y [38], Co₂W [39], Co₂Z [40], and Co₂X [40] ferrites, which were then blow-spun into continuous filaments. In a recent work, BaM nano-hexaferrites are synthesized by this route at various experimental conditions. Pictorial presentation of gel conversion into nanoferrites ash (tree like) is shown in Figure 4. Particles morphology, shape and size change changes with the change in annealing temperature which shows the sensitivity of the method (Figure 5). The sol-gel auto-combustion process was used to make M-type hexaferrites with Tb³⁺ substitution and a nominal composition of Ba_{0.5}Sr_{0.5x}Tb_xAlFe₁₁O₁₉ (x = 0.0, 0.05, 0.1, 0.15, 0.2, and 0.25) reported by Ali et al. The M-H loops were used to calculate magnetic parameters such as retentivity (M_r), saturation magnetization (M_s), coercivity (H_c), and squareness ratio (M_r/M_s). The saturation magnetization (M_s) and retentivity (M_r) reduced from 48.9 to 26.9 and 36.8 to 18.1emu/g, respectively, possibly due to spin canting and hence reduced super-exchange interactions. Coercivity is being improved (H_c) Higher magneto-crystalline anisotropy, which is attributed to the Fe²⁺ ions situated on a 2a site, could explain the increase in coercivity (H_c) from 1825 to 4440 G[41]. By using the sol-gel technique at 850 °C, a series of geometrically pure Co-Zr doped ferrites of the composition BaCo_xZr_xFe_(12-2x)O₁₉ were effectively produced. On substitution, M_s decreases somewhat from 63.63 to 56.94 emu/g, whereas H_C decreases dramatically from 5428 to 630 Oe, according to the magnetization data. Substituted compounds have a high M_s and a low H_C, making them ideal for data recording[42].

Figure 4: Conversion of gel into nano-hexaferrites [43] (Copyright permission with the licence number: 5087650493817).

Figure 5: (a-l) TEM images and particle size histogram for BaM nanohexaferrites annealed at 600, 700, 800, 900, 1100 °C[43] (Copyright permission with the licence number: 5087650493817).

3.4 Solution combustion method

Solution combustion method, a facile, economic and easy method which generally is used for preparation of oxide nano-particles [44]. In this method, metal nitrates are dissolved in suitable solvent with continuous stirring and heating arrangement. The fuel or reducing agent is then added for the open air combustion of the mixture at some moderate heating temperature [45, 46]. The solution combustion approach was used to make nanocrystalline M-type Al^{3+} substituted barium hexaferrite samples with the general formula $\text{BaFe}_{12-x}\text{Al}_x\text{O}_{19}$ (where $x = 0.00, 0.25, 0.50, 0.75, 1.00$). The precursors were synthesized with stoichiometric quantities of Ba^{2+} , Fe^{3+} , and Al^{3+} nitrate solutions chelated citric with. The ratio of barium nitrate to citric acid was 1:2, and the pH of the solution was maintained at 8. With a rise in Al^{3+} substitution x from $x=0.0$ to 1.0, the

saturation magnetization (M_s) and magneton number (n_B) decrease from 38.567 to 21.732 emu/g and from 7.6752 to 4.2126 μ_B , respectively[47].

Figure 6: SEM micrographs of the $\text{SrFe}_{12}\text{O}_{19-x}\text{Ni}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4$ composites; (a) $\text{SrFe}_{12}\text{O}_{19}$, (b) $x=10$ wt. %, (c) $x=20$ wt. %, (d) $x=30$ wt. % (e) Magnetization curves of the $\text{SrFe}_{12}\text{O}_{19-x}\text{Ni}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4$ composites and (f) Demagnetizing curves of the magnetic nanocomposites with different soft phase content[48] (Copyright permission with the license number: 5087630182674).

Abasht et al. prepared $\text{Ca}_{1-x}\text{La}_x\text{Fe}_{12}\text{O}_{19}$ ($x = 0$ and 0.4) by solution combustion method and calcium hexaferrite particles that were calcined at 1200°C for 1 hour produced low levels of coercive field (23.52 kA m^{-1}) and maximum magnetization ($58.75 \text{ Am}^2 \text{ kg}^{-1}$)[49]. $\text{SrFe}_{12}\text{O}_{19-x}\text{Ni}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4$ particles were synthesized using a one-pot solution combustion technique. The spherical and platelet particles in the composites exhibit two unique morphologies, which are connected to spinel and hexagonal ferrites, respectively (Figure 6(a-d)). The exchange coupling, as evidenced by its single hysteresis loop, was responsible for the increase in saturation magnetization from 54 to 56 emu/g for $x=10\%$. With the addition of more NiZn ferrite phase, a quirk in the hysteresis loop was discovered, and M_s was reduced to 52 emu/g. Furthermore, when the NiZn ferrite concentration increased, the coercivity reduced from 5143 to 1778 Oe (Figure 6 (e)) [48].

3.5 Hydrothermal method

The hydrothermal approach has piqued the interest of researchers and scientists during the last 15 years, and it is beneficial in the fabrication of nano-hybrid, mono-structural, and nano-composite materials [50,51,52]. A mixture of metal salts and a base is autoclaved under pressure to produce the product in hydrothermal synthesis. The final product is frequently a mixed phase containing unreacted precursors and occasionally α -Fe₂O₃, which can be eliminated by dilute HCl washing. One of the most appealing aspects of hydrothermal process is the variety of ways it may be coupled with some other techniques to yield benefits such as improved reaction kinetics or the possibility to create novel materials [53]. Hydrothermal method is used to successfully produce M-type hexaferrites BaMeFe₁₁O₁₉ replaced with trivalent nonmagnetic ions (Al³⁺, Bi³⁺) and magnetic ions (Cr³⁺, Mn³⁺) reported by Ghzaïel et al. [54]. In comparison to pure BaMeFe₁₁O₁₉, the magnetic characteristics of M-type hexaferrite i.e. Ms, Mr, and Hc increases with the magnetic ion (Cr³⁺, Mn³⁺) substitution. On the other hand these magnetic characteristics decrease with the non-magnetic trivalent ions (Al³⁺, Bi³⁺) substitution. Table 4 shows that the produced samples have ferromagnetic properties, which are influenced by the magnetic behaviour of trivalent ion replacements and their occupied location in the five distinct Fe³⁺ lattice positions. Non-magnetic substitution by Al³⁺ and Bi³⁺ ions results in a considerable decrease of Ms and Mr. As a result, hydrothermal synthesising affects the substitution of Fe³⁺ by these trivalent ions at various places.

Table 4: Magnetic parameters of different substituted Barium hexaferrites (Reprinted with permission from ref. [54]).

Composition	μH_c (T)	Ms (emu/g)	Mr (emu/g)	Mr/Ms
BaFe ₁₂ O ₁₉	0.241	61.84	33.95	0.567
Al900	0.335	51.89	28.86	0.535
Al950	0.329	52.86	30.05	0.538
Al1000	0.339	54.98	32.25	0.556
Bi900	0.488	46.27	24.54	0.531
Bi950	0.481	47.72	25.43	0.530
Bi1000	0.376	49.50	25.84	0.522
Cr900	0.449	36.47	19.41	0.532
Cr950	0.345	55.61	29.22	0.525
Cr1000	0.230	60.33	35.68	0.581
Mn900	0.434	50.16	27.96	0.546
Mn950	0.431	51.68	28.45	0.551
Mn1000	0.426	61.10	33.26	0.550

Figure 7: shows the SEM images of specimens hydrothermally synthesized with different atomic ratios of Fe/Sr ($R_{F/S}$). (a)–(g) $R_{F/S}$ =2–8 [55] (Copyright with the permission of license number: 5087680143574).

Banihashemi et al. [56] fabricated cerium doped strontium hexaferrite by hydrothermal technique. FESEM revealed that the prepared nanoparticles are agglomerated in irregular platelets. The ferrimagnetic behaviour was confirmed by magnetic hysteresis loop experiments at room temperature, and the saturation magnetization decreased as the Ce content increased. The hydrothermal process was used to make hexagonal M type $\text{SrFe}_{12}\text{O}_{19}$ ferrites. SEM micrographs of SrM materials with various $R_{F/S}$ are shown in Figure 7. The image contains a large number of hexagonal plate-like formations, which aids in proving the synthesis of hexagonal SrM ferrites [55]. It's also been discovered that sintering specimens at high temperatures improves their magnetic characteristics.

3.6 Molten salt method

A solution of metal chlorides is coprecipitated by NaOH, Na_2CO_3 , or NaHCO_3 , and then the salts NaCl and/or KCl are added to this mixture, which is then dried and heated to a

flux between 600 and 1100 °C, yielding a mixture of ferrite material and alkaline metal salts on cooling [57]. The original procedure was simpler, requiring only a mixture of conventional ceramic precursors BaCO_3 and Fe_2O_3 warmed in a NaCl-KCl flux [57], but the co-precipitation stage produces a superior product. Although the sodium and potassium salts can be completely recovered from ferrite by washing it with water or weak acid, the ferrite product will inevitably be contaminated with the alkaline metals. BaM hexaferrite was fabricated by molten salt method and they show well crystallized powders with high anisometric morphology. The saturation magnetization of barium hexaferrite is 90emu/g at 10k and 59 emu/g at 300k[58]. Li et al.[59] prepared BaM hexaferrite by MSM as shown in figure 8.

Figure 8: shows formation mechanism of BaM ferrite via MSS with BaCO_3 and $\alpha\text{-Fe}_2\text{O}_3$ powders as raw materials and $(0.5\text{NaCl}+0.5\text{KCl})$ as molten salt[59](Copyright with the permission of license number: 5087600526257).

BaM hexaferrites with near-theoretical saturation magnetization (M_s) and large remanent magnetization (M_r) were prepared using a low-temperature MSM technique, with M_s , M_r , and H_c of 71.9 emu/g, 39.5 emu/g, 3849.1 Oe and 71.8 emu/g, 38.9 emu/g, 2890.3 Oe, respectively. The M_s value of the prepared samples BaM (MSS-S) achieved 104.8 emu/g at a low temperature of 5 K. The (0.5NaCl0.5KCl) combination salts were crucial in the establishment of hexagonal plate shape and the acquisition of considerably increased magnetic characteristics[59]. Melt-flux precipitation is a modification on this approach in which less fluxing material is introduced, resulting in only microregions of partially melted salt in which crystallisation occurs instead of a true liquid phase system. The fluxing agents $7\text{BaCl}_2 \cdot 3\text{B}_2\text{O}_3$ and $\gamma\text{-Fe}_2\text{O}_3$, BaCO_3 , CoCO_3 , TiO_2 , have been used to generate BaCoTiM [60]. Pure ferrite was generated at a lower temperature and with a grain size one third that of under 100 nm when the melt-flux was heated radio thermally by an electron beam instead of conventional thermal heating [61].

3.7 Industrial manufacture

Commercial M ferrites are made from $\alpha\text{-Fe}_2\text{O}_3$, which can be found naturally or manufactured from iron pyrite (FeS_2), iron chloride or organo-metallic pigments, and barium or strontium carbonates. Iron oxides rescued from waste steel pickling liquors, a cheap supply of raw material in sulphuric acid liquor with a high Fe^{2+} content, can be used to make ferrites. All three minerals, goethite, haematite, and magnetite, have been effectively recovered [62], and BaM with excellent magnetic characteristics has been produced from iron oxide recycled materials [63]. The vacuum hot steam process can be used to granulate ferrites, producing a material that is drier, denser, more homogeneous, and more abrasion resistant than conventional granulation methods. Shearing forces are used to homogenise and mix the ceramic in a high-speed mixer, and the material is subsequently granulated by drying under vacuum while superheated steam is pumped through it [64]. Mechanochemical processing, in which mixtures of iron and barium chlorides, in a non-stoichiometric ratio of $\text{Fe}:\text{Ba} = 10$, and a substantial excess of NaOH are milled together, is a straightforward way of producing small grain size and high coercivity ferrites. A chemical reaction is triggered, resulting in a homogenous combination of metal oxides/hydroxides and NaCl that may be washed off when the powder is annealed at 800 °C to produce BaM grains 100 nm wide and 20 nm thick [65]. M_s and H_c decrease with milling duration in dry milling of BaM due to two factors: increased lattice distortions from generated stress and development of the magnetically weak BaO and $\alpha\text{-Fe}_2\text{O}_3$ phases. As the particle size is decreased, while M_s and M_r reduced and with the increase milling time, H_c also increases. However, if surfactants are added to the liquid, a significant H_c value is still formed, but the reduction in M_r is decreased, resulting in a material with a square loop and higher energy [66].

4. Magnetic properties of BaM and substituted BaM hexaferrites

Magnetic properties of hexaferrites are sensitive to intrinsic structure and symmetries of the structure. e.g. BaM synthesized through different techniques have emerged out with different magnetic properties. In a recent reported work by Guanghui Han et al. have synthesized Al doped Ba hexaferrite through solution combustion method and the prepared materials was quite porous in nature [67]. Bare Ba ferrite exhibit significant M_r/M_s ratio and ratio increases with the Al doping. The enhanced value of coercivity with Al doping has been linked with the enhancement on the magneto-crystalline anisotropy constant and shape morphology of particles. In an earlier work, Al doped Ba ferrites are fabricated using a chemical process followed by sodium citrate as chelating agent with further thermal treatment of precursor solution [68]. The saturation magnetization value was found to be 51.43 emu/g and consequently decreases up to 28.32 emu/g with the increase in Al content. However, anisotropy field increases 16.21 kOe to 25.01 kOe. Anisotropy plays a significant role in determining the magnetic properties of such M-type hexaferrites.

Figure 9: (a) M - H loops of the $BaFe_{12-x}Al_xO_{19}$ annealed at 1000 °C (Reprinted with the permission [67]), (b) The M - H curves of $BaAl_xFe_{12-x}O_{19}$ obtained at 900 °C [68] (Copyright permission with the licence number: 5087650685018).

However, the magnetic properties of BaM ferrite have been found to vary with the sintering temperature. In a recent report, BaM ferrite has been fabricated using citrate precursor method and effect of varied sintering temperature has been studied for 1000, 1100, 1200, 1300 °C for 24, 62, 124 and 148 hours of sintering [69]. At the sintering temperature of 1100 °C, formation of single crystalline BaM take place, and for 1300 °C sintering temperature, polycrystalline BaM ferrite forms. In addition, crystallite size also varied for different sintering temperature and sintering time. Figure 10 presents the obtained structure and the corresponding magnetic properties of samples. One can easily observe the drastic changes in magnetic properties of BaM with sintering temperature and time. These results shows the sensitivity of magnetic parameters towards the crystallite structure of samples which varies with synthesis process variations.

Figure 10: (a) XRD patterns for BaM (A, B, C and D) sintered at 1000, 1100, 1200 °C for 24 h, 1300 °C for 62 h respectively and surface XRD of the two samples (E and F) sintered at 1300 °C for 124 and 148 h respectively; (b) M-H loops for A,B,C,D,E,F samples and variation of magnetic parameters with sintering temperature and sintering time [69] (Copyright permission with the licence number: 5087650894770).

Table 5: Some reported BaM type ferrites and their magnetic parameters.

Composition	Synthesis route	Annealing Temperature	Particle size	Magnetic parameters	Ref.
$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Fe}_2\text{O}_4$	sol-gel auto-combustion	700 °C	81nm (h00)	M_s -2.20emu/g M_r - 0.66 emu/g H_c -424 Oe	[70]
		1050 °C	336 nm (h00)	M_s -3.83 emu/g M_r - 1.92 emu/g H_c -3438 Oe	
$\text{BaFe}_{11.6}\text{Co}_{0.2}\text{Ni}_{0.2}\text{O}_{19}$	co-precipitation method	750 °C	29.73 nm	M_s - 34.83 emu/g M_r - 18.03 emu/g H_c -2099 Oe	[71]
$\text{BaFe}_{12-x}\text{Al}_x\text{O}_{19}(0.50)$	solution combustion technique	900 °C	70 nm	M_s - 30.874 emu/g M_r - 18.861emu/g H_c - 5169 Oe	[72]
$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_x\text{La}_x\text{Fe}_{12-2x}\text{O}_{19}(x-0.4)$	conventional ceramic method	1000 °C	47.75 nm	M_s – 59.82 emu/g H_c - 780.57 Oe	[73]
$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_x\text{W}_x\text{Fe}_{12-2x}\text{O}_{19}(x-0.2)$	ceramic method	1000 °C	21.66 ± 1.08 nm	M_s - 56.0 emu/g M_r - 17.5 emu/g H_c - 841.6Oe	[74]
$\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_x\text{Al}_x\text{Fe}_{12-2x}\text{O}_{19}(x-0.4)$	ceramic method	1000 °C	24.37 ± 1.22 nm	M_s – 49.9 emu/g M_r - 7.28 emu/g H_c - 247.6 Oe	[75]
$\text{BaCu}_x\text{Fe}_{12-x}\text{O}_{19}(x-0.8)$	co-precipitation technique	1200 °C	37.8 nm	M_s – 34.9 emu/g M_r - 26 emu/g H_c - 856.65 Oe	[76]

5. Applications of hexaferrites

Hexagonal ferrites are taking a lot scientific attention by the researchers and scientists because of its potential applications in different research areas of science and technology. Some of the applications of hexagonal ferrites are explained below.

5.1 Permanent Magnets

The finest permanent magnets are fabricated with the usage of rare earth metals-based alloys, mostly made up of neodymium. But, on the other hand, China sold 97% of the rare earth metals globally and nowadays, China raises the price of rare earth metals up to 500% which will result in the increase of shortage of production of permanent magnets. Recently, the hard ferrites are also utilized in the fabrication of permanent magnets but they are not so much superior in comparison to those permanent magnets which one gets from alloys of rare earth metals. The main advantage is that the production of permanent magnets from hexagonal ferrites is cheaper and very easy to fabricate [1]. For the production of excellent permanent magnets, magnetic materials with large retentivity and coercivity along with resistant to demagnetisation will be largely preferable. The permanent magnets of hexagonal ferrites are fabricated in two ways, first is sintered magnets which is actually a hard-pressed ceramic powder and second way is bonded magnets which is actually a moulded composite. Barium M-type hexaferrite (BaM) is used in fabricating bonded dust magnets and moreover, this type of bonded dust magnets is used to make plastoferrite. The bonded dust magnets can be easily censored into any type of form and can be used as fridge magnets. Out of all hexagonal ferrites, M-type hexagonal ferrite is considered as one of the most excellent magnetic material for making permanent magnet and it can be used as a magnet material for potential applications in loudspeakers, clocks, microwave dynamos, magnetic separators etc. The common use of hexagonal ferrites as permanent magnets in refrigerators, microphones, loud speakers and automobile applications which make the M-type hexaferrite for account over 90 percent of the fabricated material worldwide as a permanent magnet material [77]. Therefore, with the increase in the price of neodymium, hexagonal ferrites grow as an alternative valuable magnetic material for the fabrication of permanent magnets worldwide.

5.2 Electrical and microwave devices application

Today, the requirement for the signal processing devices has been increasing in radar instruments, GPS location, satellites, for wireless signals, defence system etc. and also, the frequencies at which these devices operate are increasing from microwave to millimetre wave. Therefore, for this, the demand of hexagonal ferrite will go on increasing. Moreover, the microwave dielectric losses must be lowered for their use at

microwave frequencies. For this, the hexagonal ferrite should behave like an insulator for a complete penetration of the electric field but that was a big issue with the hexagonal ferrite based magnetic materials. The solution for this issue is that the hexaferrite should be synthesized on a superconducting substratum for the elimination of conductor losses [78]. Few of the microwave applications of hexagonal ferrites are antennas, circulators and isolators. The hexagonal ferrites have high value of resistivity with low dielectric losses which makes them more suitable for high frequency and microwave devices applications. The total dielectric losses in microwave devices can be determined with the utilization of magnetic damping from ferromagnetic resonance. Thus, our main objective is to synthesize the hexagonal ferrites with minimum ferromagnetic resonance line widths to reduce the dielectric losses in microwave devices. From the last few years, Barium M-type hexagonal ferrite (BaM) has been widely utilized for microwave devices applications. The integration of ferrite passive devices (i.e., circulators, isolators, phase shifters, filters, etc.) with semiconductor active devices (i.e., amplifiers, switches, and signal processing devices) onto a single planar system platform such as a Si wafer is a current trend in microwave technology. This satisfies the need for greater system integration while simultaneously decreasing device profile, volume, and weight. This necessitates the formation of high-quality ferrite films on semiconductor substrates. It has been demonstrated that reduced microwave losses in ferrite films are caused by good crystal quality. The development and processing of highly-oriented BaM films on semiconductor substrates remains a substantial problem due to chemical reactivity, interfacial diffusion, poor lattice matching, and problems linked to the high temperatures required for ferrite deposition [6].

5.3 Data Storage and recording media application

A bit in a digital memory is preserved as 0 or 1, therefore, a square loop with a high retentivity is required to ensure the data remains preserved and is not unintentionally deleted, and moreover, a large signal to noise ratio is essential to decrease mistake rates. In case of re-recordable memory, small saturation magnetizations along with coercivity are required. This means low fields can be beneficial for rewriting the data. In both the situations, chemical stability and permanency of the magnetization are critical. Audio tape, VCR tape, Professional Hi-8 mm data tape, floppy discs, and stiff or hard discs were all traditional commercial items. Because they are chemically stable, mechanically strong, and have large saturation magnetisations, BaM and SrM are employed for high density magnetic recording and magneto-optical recording. Oriented ferrites are employed in both longitudinal and perpendicular recording, in which the material's magnetic anisotropy is aligned parallel or perpendicular to the recording media. Moreover, without any substitution, BaM or SrM films have encountered several

problems for high-density magnetic recording: they have a large positive temperature coefficient of coercivity, the grain size in such films is in the order of 102 nm, and as the grain size decreases, the magnetisation becomes thermally unstable, necessitating a larger magnetocrystalline anisotropy. The M ferrites have high coercivity for many recording applications since a significant magnetic field is required to record and re-record data, but it was discovered that in doped M ferrites, all of these characteristics may be customised by changing doping. Toshiba developed cobalt–titanium doped M-type hexagonal ferrites for recording in 1982, and they are particularly suited for use in recording media such as magnetic tape when coated as a thin film onto a substrate, as the grain size and coercivity can be reduced significantly without much loss of magnetisation in the ferrite [79]. Therefore, it was found that cobalt–titanium doped M-type hexagonal ferrites fulfilled the criteria of materials with switching fields, low magnetic losses, and good signal-to-noise ratios [80]. Recent breakthroughs in M-type hexagonal ferrites with ferroelectric-like behaviour at ambient temperature and polarisation switching by modest magnetic fields might lead to innovative magnetically-rewritable ferroelectric memories and electrically rewritable magnetic memories in the future.

5.4 RAM and microwave/EM wave absorption application

The wide usage of microwave-frequency equipment in our immediate surroundings, like radar and wireless and mobile communications, has led to a significant rise in electromagnetic interference, and the intensity of this ever-present non-ionising radiation. As a result, there is an increasing demand for microwave absorbing materials to decrease interference, as well as shielding devices, shield rooms, and chambers for electromagnetic compatibility testing and to prevent the damaging effects of electromagnetic waves on biological tissues [81]. Ferrites are employed as microwave absorbers because they can absorb microwave energy at the same frequency as ferromagnetic resonance. They're already being employed as electromagnetic absorbers in rooms and chambers where new items and equipment are being tested for electromagnetic compatibility at microwave frequencies. Absorbers are commonly made of plasto-ferrite tiles for electromagnetic applications, although they can also be made of ceramics as composites. By altering the volume fractions of the component phases, the electromagnetic characteristics of composites may be successfully modified. In some composites, a synergistic impact between the characteristics of the component phases has also been found. The X and K bands (8–40 GHz), which are typically pulsating over a range of frequencies, are used by current military radar because the short wavelength allow for better resolution [82]. Radar-absorbing paints or coatings composed of ferrites have been employed to coat military aircraft for stealth operations, as well as ships and other naval and ground-based assets. They absorb a few of the radar signal that is directed

towards the target, decreasing or hiding the radar cross section signature that is received at the other end; however, the object is not completely radar “invisible”. This technique is referred to be “stealth” or “low observable”. Hexaferrites as radar absorbing materials used in electronic warfare are gaining a lot of attention, especially for their tuneable properties or wide absorption range to covering a wide spectrum of frequencies, as well as increased directional capabilities in oriented or fibrous hexaferrites. Because cost is less of a factor in these applications, most of the research into more unusual forms of hexaferrites is now being driven by aerospace and defence demands. The majority of this material, however, is confidential or classified, and most of it is not available in the public domain.

5.5 Magnetolectric (ME) and multiferroic (MF) applications

The utilization of the connection between electric and magnetic characteristics is a major importance in magnetoelectric and multiferroic materials. This could contribute to plenty of new devices, including dual electrical and magnetic field tuneable devices at microwave and millimetre wave frequencies, extremely sensitive magnetic field sensors, switches and activators, high-frequency signal processing devices and gyrators [83,84]. The composite materials comprised of phases with good electrical and magnetic characteristics can have significantly stronger magnetoelectric interactions, and they have already shown tremendous potential. Although there has been less research on hexagonal ferrites in this sector, it is suitable for investigation; devices including yttrium iron garnet and a piezoelectric phase such as PZT, BT, or BST have showed strong magnetoelectric coupling constants in the hundreds of $\text{V cm}^{-1} \text{Oe}^{-1}$ range. Enhancing magnetoelectric effects by resonance coupling offers a lot of promise, particularly at high GHz frequencies, and the low magnetic fields required substantially expands the variety of real-world applications. They might potentially be employed as current sensors or transformers, particularly during resonance, when the voltage gain effect is enhanced. Most significantly, all of these phenomena, especially in composites, can occur at room temperature.

5.6 Plastroferrites

Plastroferrites, often known as bonded magnets, are magnetic composite materials composed of a powdered ceramic ferrite integrated into a resin or thermoplastic matrix. Because of their composite nature, they have diluted magnetic properties, yet the products are not brittle and fragile, and are difficult to shatter or shatter. The plastroferrites can be cut in any form of shape in a very mass-productive and hence, it is a low-cost process. One of the examples of plastroferrites are flexible sheets either long or

in thin form. Plastroferrites are well known as refrigerator door magnets, both decorative on the exterior and magnetic sealing strips within the door's rim [85].

5.7 Other important applications

Several designs for ferrites-based sensors have been suggested, including a humidity sensor based on resistance changes in a copper–zinc ferrite [86]. Hexaferrites can be used in biomedical purposes such as nuclear magnetic resonance imaging and magnetomotive biomedical implants. However, the toxicity of a few of the constituent elements notably barium, limits their usage in applications where particles or fluids must be injected into the human body. Like SrM grains were surface modified with oleic acid to increase their dispersibility in water with nearly negligible loss of magnetism, and they were then processed with oleic acid to form large polymeric microspheres with a magnetic core that should be biocompatible [87]. Similarly, Co₂Z coated magnetic force microscopy cantilevers were employed to provide better high-frequency measurements than normal CoCr coated tips, with imaging up to 2 GHz and 20 nm resolution. The imaging, characterization, and quality control of high-density magnetic disc drives and recording heads at high frequencies might be of special importance to industry [88].

Conclusions

In the current chapter, we have tried to summarize the basics of hexa-ferrites including the classification of all ferrites. We have categorically focussed on M-type of hexa-ferrites especially the magnetic properties and their linkage with the crystalline structure of hexaferrites. Various potential applications of hexaferrites constitute a part of this chapter. Owing to the higher crystalline field of hexagonal ferrites, their suitability of applications in GHz range is appreciated and proposed.

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