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A Current Review on the Synthesis and Magnetic Properties of M-Type Hexaferrites Material

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Abstract

After the discovery of hexagonal ferrites or hexaferrites, it has been become important materials commercially and technically to study which is still growing on. In this article, we have reviewed about the M-type hexaferrites including their structural, synthesis techniques and important magnetic properties. The role of experimental synthesizing techniques adopted for preparation of M-type hexaferrites on the various parameters studied in this review paper. The substitution of holonium in BaM ferrite reduces the value of coercivity but not saturation magnetization and ramanence and the cobalt-titanium substituted ferrites were the most important M-type ferrites in the field of application in microwave properties and magnetic field industry.

Keywords

Hexagonal Structure, Sol-Gel Synthesis Technique, Co-Precipitation Technique, TEM, VSM

1. Introduction

In the recent years, hexagonal ferrites are of great importance to study because of its usage in technological applications such as permanent magnets and also of their excellent magnetic properties and potential applications in various fields

[1] [2]. Richard Feynman [3] (2005) stated that ferrites are one of the most difficult topics theoretically to study but for the practical applications and studies, it is one of the best topics to come into practice. This will be especially true only when dealing with hexaferrites or hexagonal ferrites (ferrites which have hexagonal crystallographic structure). The world's first permanent magnet came into existence in 1951 which was based on ferroxdure-$BaFe_{12}O_{19}$ (also called BaM) [4].

Since after the discovery of hexagonal ferrites in 1950s, the scientist and the technologists showing a lot of interest in studying hexagonal ferrites and it is still going on and in 1955, applications of gyromagnetic properties of hexaferrites started in systematic way [5] [6] [7]. But, nowadays, a lot of theoretical and experimental research work on the various properties and synthesis of hexagonal ferrites have been going on in the laboratory [8]. Recently, there has been much work done in nanotechnology magnetoelectric/multiferroic applications and also in the development of orientation and alignment effects in ferrite fibres [9]. Commercially and technologically, hexagonal ferrites have number of applications in different areas. Also these materials account for the bulk of total magnetic materials which are massively important and are manufactured globally. The different types of hexagonal ferrites are: M-type ferrites such as $BaFe_{12}O_{19}$ (BaM or barium ferrite), $SrFe_{12}O_{19}$ (SrM or strontium ferrite), Z-type ferrites ($Ba_3Me_2Fe_{24}O_{41}$) such as $Ba_3Co_2Fe_{24}O_{41}$ or Co_2Z, Y-type ferrites ($Ba_2Me_2Fe_{12}O_{22}$) such as $Ba_2Co_2Fe_{12}O_{22}$ or Co_2Y, W-type ferrites ($BaMe_2Fe_{16}O_{27}$) such as $BaCo_2Fe_{16}O_{27}$ or Co_2W, X-type ferrites ($Ba_2Me_2Fe_{28}O_{46}$) such as Ba_2Co_2 $Fe_{28}O_{46}$ or Co_2X and U-type ferrites ($Ba_4Me_2Fe_{36}O_{60}$) such as $Ba_4Co_2Fe_{36}O_{60}$ or Co_2.

The main purpose of this review is to be focused on the hexagonal ferrites specially, having barium and strontium as divalent cation (BaM or SrM) and their important substituent. The magnetic properties of hexagonal ferrites are linked to their crystal structures intrinsically and they are all ferrimagnetic in nature. However, the magnetic properties of hexagonal ferrites shall be studied practically here and the reasons and the theory behind these magnetic characteristics can be seen in the review by Özgür *et al.* [9]. This review will totally focus on the synthesis and magnetic properties of hexagonal ferrites especially M-type hexaferrites.

2. Structure of Hexaferrites

All the six types of hexagonal ferrites such as M, Y, W, Z, X, U are summarized in **Table 1** and the structure of hexagonal can be described by the two approaches such as spinel-based model and block-based model.

2.1. Spinel-Based Model

The structure of hexagonal ferrites was same to that of spinel structure which is composed of plates of close packed cubic oxygens with smaller metal ions and

Table 1. Stiochiometry and basic properties of six types of hexagonal ferrites where * represents 180° rotation of block around the c-axis [10].

Ferrite	Formula	Spinel plates and hexagonal layers	S/R/T structure
M	$BaFe_{12}O_{19}$	$2M_5 = B_1S_4B_1S_4$	SRS*R*
W	$Ba_2Me_2Fe_{16}O_{27}$	$2W_7 = B_1S_6B_1S_6$	SSRS*S*R*
X	$BaMe_2Fe_{16}O_{27}$	$3X_{12} = B_1S_4B_1S_6B_1S_4B_1S_6B_1S_4B_1S_6$	SSRS*S*R*
Y	$Ba_3Me_2Fe_{14}O_{41}$	$2Y_6 = B_2S_4B_2S_4B_2S_4$	3 (ST)
Z	$Ba_2Me_2Fe_{28}O_{46}$	$2Z_{11} = B_2S_4B_1S_4B_2S_4B_1S_4$	STSRS*T*S*R*
U	$Ba_4Me_2Fe_{36}O_{60}$	$U_{16} = B_1S_4B_2S_4B_1S_4$	SRS*R*S*T*

these metal ions are present at octahedral and tetrahedral interstices. The interstices between these layers are filled with three octahedral or one octahedral and two tetrahedral sites and the spinel structure is obtained by the layers of four oxygen atoms repeat three vertical layers to form an ABCABC lattice. In the hexagonal ferrites, two types of plates (S_4 and S_6) are present which are joined together by two barium containing layers ($B_1$and B_2) in which the M, Y and Z consist of only one plate (S_4), W structure consist of only one plate (S_6) and the X structure consist of two pates (S_4 and S_6). B_1 layer appears in case of M, W, Z ferrites, is a single hexagonal layer which contains the barium atom in position of four oxygen atoms and the B_2 layer appears in case of Z ferrites is two hexagonal layers which contain the two barium atoms in position of eight oxygen atoms. The oxygen atoms remains in B_1 layer are arranged in two sets of triangles and the structure of hexagonal ferrite can be studied more deeply by combining the S and B layers which are S_2-two spinel layers, M_5 = aB_1-sandwiched between the four spinel layer, Y_6-two B_2 layers sandwiched between the fourspinel layers as show in **Figure 1** [11].

2.2. S, R and T Blocks

The S block has double spinel layer and has the unit formula $Me_2Fe_40_8$ where Me is a divalent ion. The S block composed of two layers of oxygen atoms with three metal atoms. The three metal atoms are present between each layer of four oxygen atoms at four octahedral sites and two tetrahedral sites. At the octahedral sites, the cation is surrounded by six oxygen anions and at the tetrahedral sites, the four oxygen anions surrounded the cation (**Figure 2**). The R block has the unit formula $BaFe_6O_{11}$ and composed of three layers of oxygen atoms where the barium atom takes the position of one of oxygen atom in the centre layer. If the larger barium atom takes the position of the central oxygen atom, then, it creates asymmetry in the cation sites resulted in five octahedral sites, no tetrahedral sites and five coordinate trigonal bipyramidal sites and the S block is seen in the M-type ferrites and is the M_5 unit in the top and bottom layers removed. The T block has the unit formula $Ba_2Fe_8O_{14}$ and composed of three layers of oxygen atoms where the barium atom takes the position of one of the oxygen atoms in

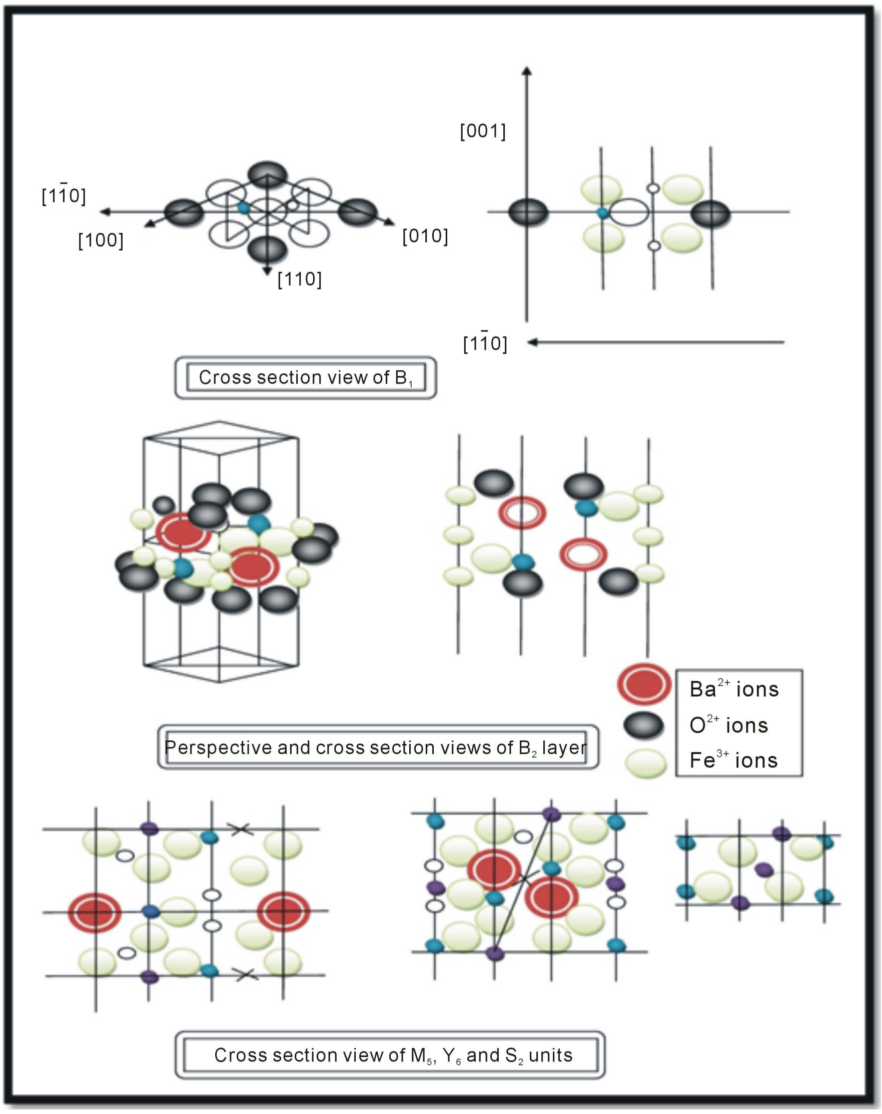

Figure 1. Cross sectional view of B_1 layer, B_2 layer and M_5, Y_6 and S_2 units [11].

the centre two layers. This is the Y_6 unit with the top and bottom layers removed [12] [13].

2.3. Structure of M-Type Ferrites

The M-ferrite is type of hexagonal ferrite which is composed of one S block and one R block. These blocks are overlapped with hexagonally and cubic packed layers and also, the plane which contains the barium atom is called mirror plane where the S and R blocks are 180˚ rotation around the c-axis, so, the mirror block R, R* providing the unit cell formula SRS*R* where * shows the 180˚ rotation of the given block around the c-axis. Also, it was demonstrated that the BaM ferrite composed of stacking of spinel like blocks and substituted M-type ferrite has been investigated on large scale such as $BaFe_{12-X}A_XO_{19}$. This has been investigated for improving the magnetic properties of BaM ferrite

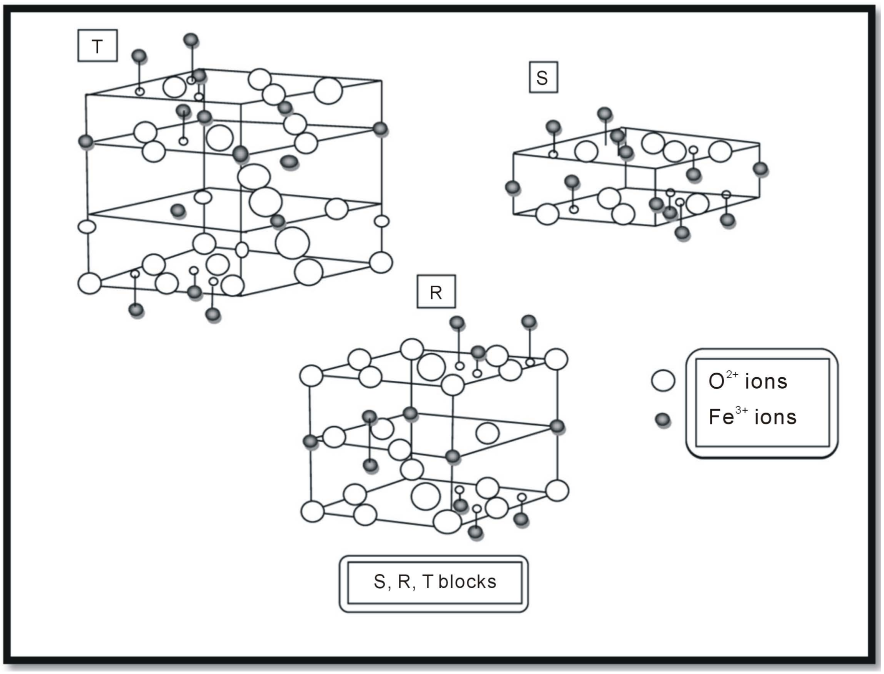

Figure 2. Perspectiveand ball and sticky view of S, R and T blocks [14].

[14] [15] It was also reported that in case of BaM ferrite, the lattice parameters are c = 23.17 Å and a = 5.89 Å but in case of SrM ferrite, the lattice parameters are c = 23.03 Å and a = 5.86 Å [16] and also, for PbM ferrite, the lattice parameters are c = 23.02 Å and a = 5.88 Å [17].Cross section view of the M-type ferrite structure are shown in **Figure 3** [18] [19] [20].

3. Synthesis Techniques for the M-Type Hexaferrites

A simple method through which the hexagonal ferrites can be produced commercially is standard ceramic method and is ineffective in controlling the grain size and morphology of ferrite powder product. However, there are some other chemical methods through which ferrite precursors powders shall be produced. During synthesizing ferrite powders, we have to take into practice some important factors:

- Heat treatment;
- Chemicals used in starting powders;
- Stoichiometry of powder.

The various methods with the help of which the ferrite precursor powders can be prepared are explained as:

3.1. Conventional Ceramic Method

The conventional ceramic method is a simple method which includes mixing and sintering of metal oxides and carbonates precursor powders which should be present in appropriate molar ratios [21] [22] but if the starting sample is mixed at the atomic or ionic level before the reaction occurrence then, the

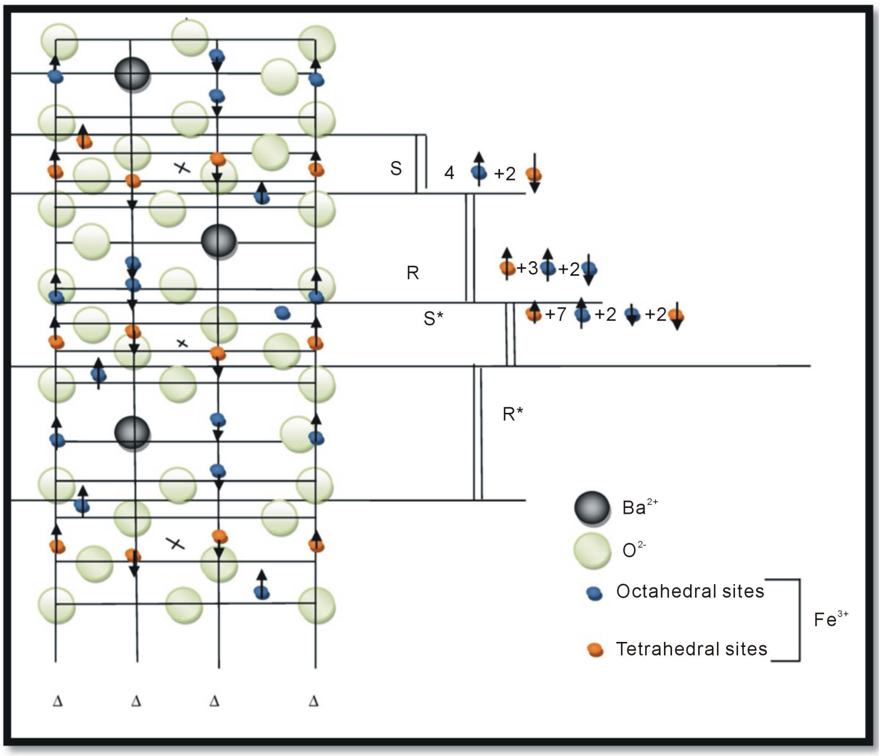

Figure 3. Cross section view of the M-type ferritestructure ($BaFe_{12}O_{19}$) [18] [19] [20].

crystallization will exists there at lower temperatures and lower sintering temperatures and this can be achieved by ball milling [23]. Ball milling can be done in two ways-dry milling or wet milling. In most of the cases, firstly the powders are mixed which is then milled by balls made of tungsten carbide, stainless steel [24] [25] and then, finally, it is dried and compressed by applying pressure to the responded shapes for full densification and then, at high temperatures, the compacts are sintered. It was found that about approximately 90% of the theoretical density of 5.28 g/cm^3 for BaM and 5.11 g/cm^3 for SrM. It was demonstrated in a study that pellets of wet milled BaM powder compacted at pressure of approximately 250 MPa and sintered at 1220˚C for 3 hours were reported to have 90% of the theoretical density [26], at a pressure approximately 500 MPa and sintered at 1100˚C for 2 hours were reported to have approximately 80% - 93% of the theoretical density [27] and at pressure approximately 30 MPa and sintered at pressure 1150˚C have less than 70% of the theoretical density [28].

3.2. Coprecipitation Method

The coprecipitation method is used for the synthesis of magnetic nanoparticles. In this method, metal chlorides or nitrates are dissolved in appropriate molar ratios to obtain homogenous solution. Add a base dropwise in the solution for the precipitation of metal powders and then, the co-precipitated powder is washed and dried. This dried powder is, then, sintered at temperature lower than the conventional solid state route and the required M-type ferrite were obtained [29] [30] [31]. Several workers are working on the properties and quality of M-type ferrite powders prepared by co-precipitation method and they are doing such investigation on the basis of experimental conditions [32] [33]. In this method, an appropriate amount of Fe $(NO_3)_3.9H_2O$ was added to NACLO and NACL solutions which are produced through a concentrated solution of NAOH when chlorine gas was bubbled through a solution of NAOH and at Fe:Ba = 10 ratio, $BaCl_2.2H_20$ was added to the BaM solution. After this, the solution was stand over for one day which is heated for one hour at 80˚C which was further filtered and rinsed in order to remove the chlorine and alcohol residue and by thisprocess, crystalline barium hydroxide and amphorous ferrihydrite are formed which was structurally related to the α-Fe_2O_3 indicates the formation of a pure BaM phase at a low temperature of 800˚C [34]. The increase in PH will result in increase in coercivity which is probably due to the small grain size or particle size and low coercivity is due to the large grain size. With Fe:Ba = 10, the grain size of BaM hexaferrites decreased with increasing PH from 11 to 12.5 but the grain size of BaM was reported to increase with the increasing PH with Fe:Ba = 10.5 or 11. For Fe:Ba = 10, it was reported that the saturation magnetization is 66.1 emu/g at PH = 11 which was sintered at 920˚C is decreased to 43.6 emu/g at PH = 12.5 and the coercivity increases from 3400 Oe to 4334 Oe. For Fe:Ba = 11, it was reported that the saturation magnetization decreases from 60.1 emu/g at PH = 11 to 46.2 emu/g at PH = 12.5 and the coercivity decreases with the increase in PH from 4585 Oe to 4435 Oe. Also, with the increase in sintering temperature from 640˚C to 920˚C, it was observed that the saturation magnetization of co-precipitated powder increases from 25 emu/g to 65 emu/g and also, the coercivity increases from 5264 Oe to 5791 Oe which represents single domain particles with typical high coercivity [35] [36]. The steps involved in the preparation of Al_2O_3-Fe_2O_3 compounds by coprecipitation technique are given in flow chart shown in **Figure 4** [37].

3.3. Sol-Gel Method

The sol-gel method is basically used for the manufactured of magnetic powders. In this method, under constant stirring, metal nitrates or metal chlorides water solutions are mixed and if we are working with citrate sol gel method, then, we add to the solution, an appropriate amount of citric acid under constant stirring. After this, a basic solution drop by drop is added to the solution under constant stirring. Due to this, the PH of the solution is maintained between the range 7 - 9 and the solution is evaporated at 80˚C. After drying and sintering the resulted gel, hexaferrite phase is produced and this is one of the best things that obtained throughout the process. The scientists are working on improving the quality of the products by doing a large numbers of modifications on the method and this is carried out by doing a number of experiments. The SrM ferrites were prepared by sol gel technique after making some modifications in this technique

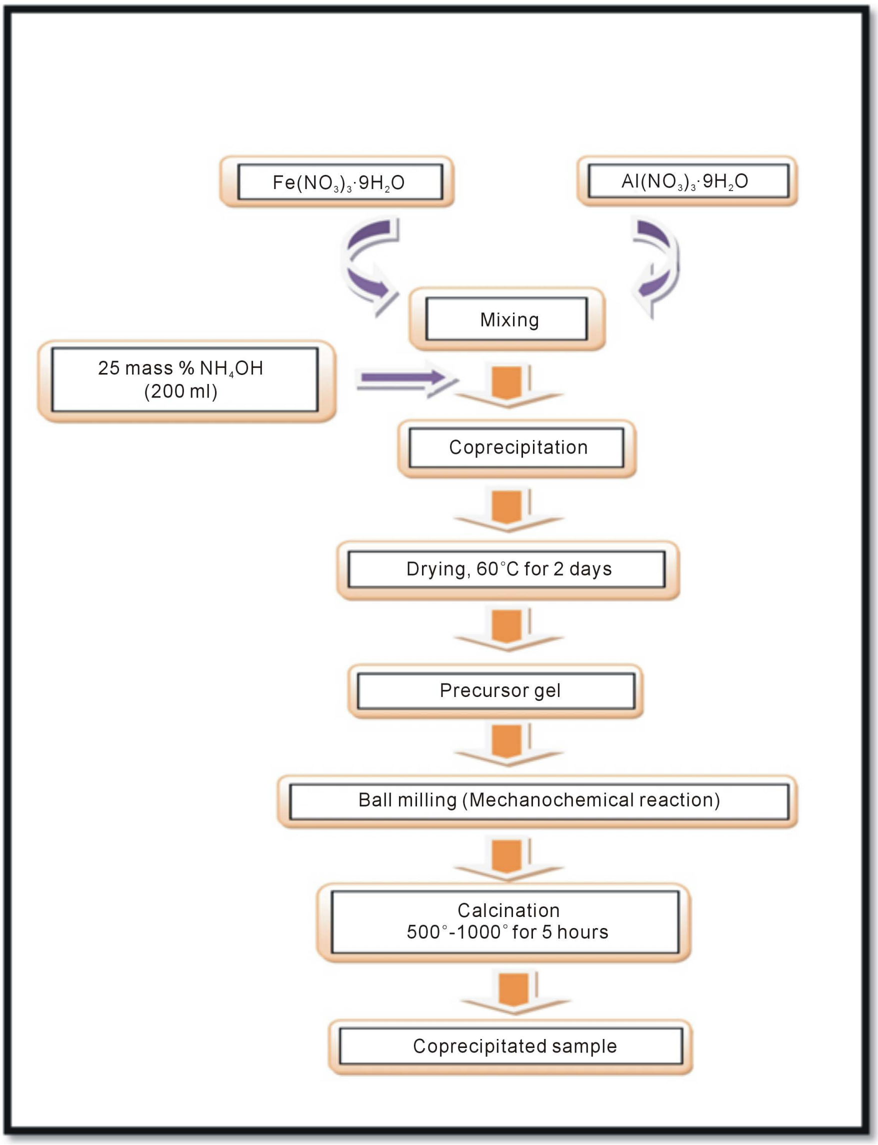

Figure 4. Flow chart of co-precipitation method [37].

which illustrates coercivity of 2213 Oe and a saturation magnetization of 30.61 emu/g but previously we studied [38], with the help of ammonia, $Fe(NO_3)_3$water solutions was precipitated and these precipitates were dissolved in citric acid with $BaCO_3$. After that, benzoic acid and ethylene glycol were added to the solution and the given solution was evaporated at 60˚C and then, the viscous gel produced is dried with the help of heat treatment given at 170˚C in two ways described below-Route (a) In this route, a heat treatment of 1050˚C was a heating rate of 4.5˚C/min was given to the gel samples placed in the furnace and these samples are heated at different temperatures for different periods of time. Route (b) In this route, the process is slight different. Firstly, the samples are heated at 450˚C for 5 h and after this, they have again given a heat treatment in the range 500˚C to 1250˚C for 5 h with a same heating rate of 4.5˚C/min. The various phases were obtained at different temperatures by the samples of route "a" and route "b".

- At temperatures 300˚C - 500˚C, γ-Fe_2O_3 and $BaCO_3$ phases was formed by the samples of route a having saturation magnetization of 25 - 34 emu/g.
- At temperatures ≥ 550˚C, BaM phase was formed.
- At temperatures 550˚C - 900˚C, α-Fe_2O_3 intermediate phase was formed.
- At temperature 650˚C - 750˚C, $BaFe_2O_4$ phase was formed and saturation magnetization increased which was reported 44emu/g prepared at 650˚C and the coercivity increased sharply above 650˚C.
- At temperatures 800˚C to 900˚C, the coercivity comes close to 4200 Oe and the saturation magnetization increase which was reported 65 emu/g but at temperatures above 900˚C, the coercivity decreases which comes closes to about 3400 Oe. Also, for this temperature, the saturation magnetization reaching about 69 emu/g by the samples of route a.

On the other hand, a improvement in the magnetic properties were observed by samples of route b in the temperature ranges specified above but at the temperature range 900˚C - 950˚C, the samples prepared by route b was reported to have saturation magnetization and coercivity of 70 emu/g and 5950 Oe [39]. The steps involved in the sample preparation buy sol-gel technique are given in flow chart shown by **Figure 5** [40].

3.4. Auto-Combustion Method

The auto-combustion method is known as modified sol-gel citrate method [41] [42] in which the solutions are made from the metal nitrates and citric acid with the appropriate molar solutions present and this solution prepared above is the basic solution made for this technique. After this, a base is drop by drop added to the solution due to which the PH of the solution reaches in the range 7 - 8 [43]. Then, a brown or yellow gel is obtained after heating the solution at 80˚C which results in the formation of auto-combustion foamy powder by further heating at 220˚C - 240˚C [44] [45]. This foamy powder is grinded and sintered at temperatures above 60˚C which results in the formation of the ferrite powder that is the thing we are looking for and the properties of the these ferrite powder is affected by the Fe:Ba ratio calcinated at 900˚C [45]. The magnetic properties of BaM was reported to be improved with the increase in Fe:Ba ratio from 1:1 to 1:2 reaching about saturation magnetization, coercivity, ramenance and energy product of 55 emu/g, 5000 Oe, 28 emu/g, 1.013 MGOe and also, it was reported that the saturation magnetization of the sample increases with the increase in the calcinated temperature from 700˚C to 1000˚C [46]. With Fe:Ba = 11, it was reported that the samples reaching about the saturation magnetization and coercivity of 51 emu/g and 4700 Oe but with Fe:Ba = 9, it was reported that the samples reaching about the saturation magnetization and coercivity of 67 emu/g and 5650 Oe [46]. The steps involved in the sample preparation by auto-combustion technique are given in flow chart shown in **Figure 6** [47].

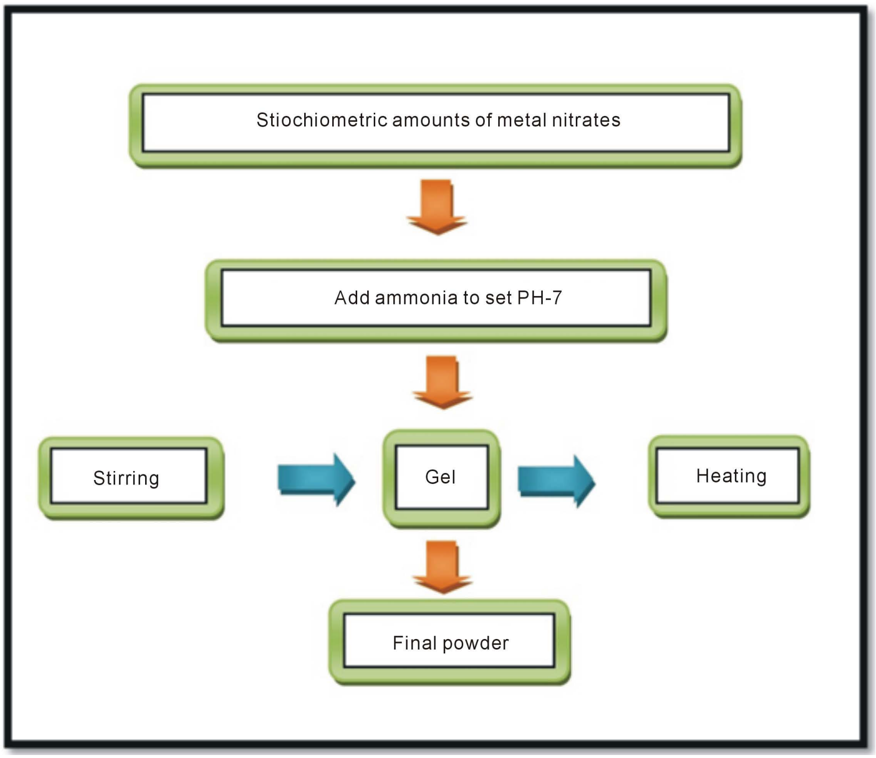

Figure 5. Flow chart of sol-gel technique [40].

3.5. Hydrothermal Method

From the last 15 years, the researchers and the scientists showing a great interest in the hydrothermal method and this method is useful in the processing of monostructural, nano-hybrid and nanocomposite materials [48]. In this hydrothermal method, base like NAOH or KOH plays an important role with the aid of hydroxide, nitrates and Fe: Ba ratios in the coprecipitation of aqueous solution of metal nitrates and then, the solution is given a heat treatment in the temperature range of 150˚C - 290˚C in autoclave. This solution contains the metal precipitates and then, these precipitates were filtered, washed and dried the particles that obtained in an oven after which the dried powder is sintered in the temperature range of 110˚C - 120˚C. The sintering of dried powders helps us to improve the magnetic characteristics of the resulted product which depends upon the parameters like hydroxide: nitrate ratio, Fe:Ba ratio and the sintering temperature [49]. In the study it was reported that the BaM phase was obtained when there was hydroxide:nitrate ratio > 2 but the intermediate iron oxide phase was obtained when there was have low hydroxide: nitrate values [49] [50]and the supermagnetic particles obtained when the sample prepared at 150˚C having hydroxide:nitrate ratios = 16 [51] [52]. For example with Fe:Ba = 8 and hydroxide:nitrate = 2, as reported that the samples prepared at temperature range of 230˚C contains the two phases under specific heat treatments. The first phase that was obtained was α-Fe_2O_3 under heat treatments of 10 h and the second phase obtained was BaM phase under heat treatment of 25 h and also, under the

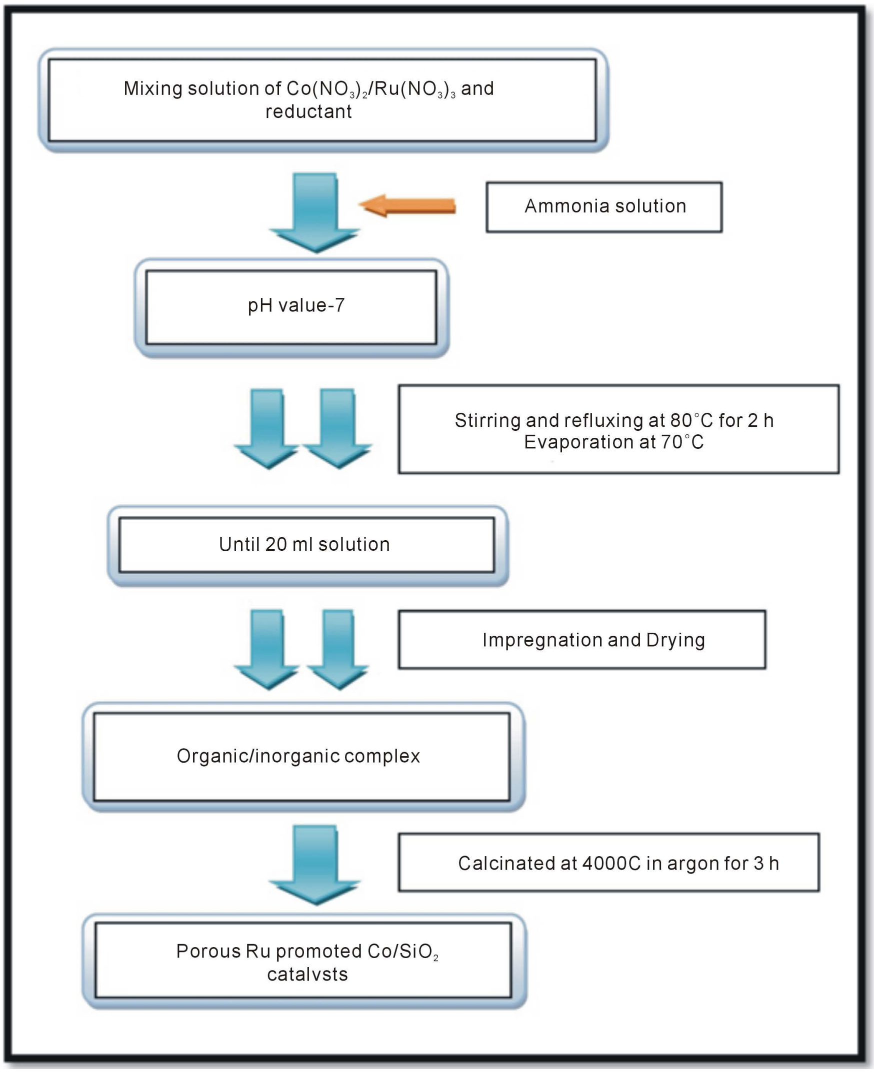

Figure 6. Flow chart for the preparation of catalysts by auto-combustion technique [47].

hydrothermal synthesis, the Fe:Ba ratio is most important in the production of M-type ferrites. The steps involved in the preparation of sample materials are given in flow chart shown in **Figure 7** with the help of hydrothermal method [53].

3.6. Citrate Precursor Method

The citrate precursor method is a simple synthesis method also known as pecchini method with the aid of which high yield ferrite powders are prepared at temperatures range reaching about 100°C [54] [55] and also, it is used for the production of ultrafine particles from the decomposed citrates at low temperatures range [55] [56]. In this method, firstly the appropriate molar ratios of iron and barium nitrates are dissolved in distilled water and then, the solution is mixed with citric acid with a cation:citric acid ratio of 1:1. The ammonia is then, added drop by drop to increase the PH of the solution forming a homogenous

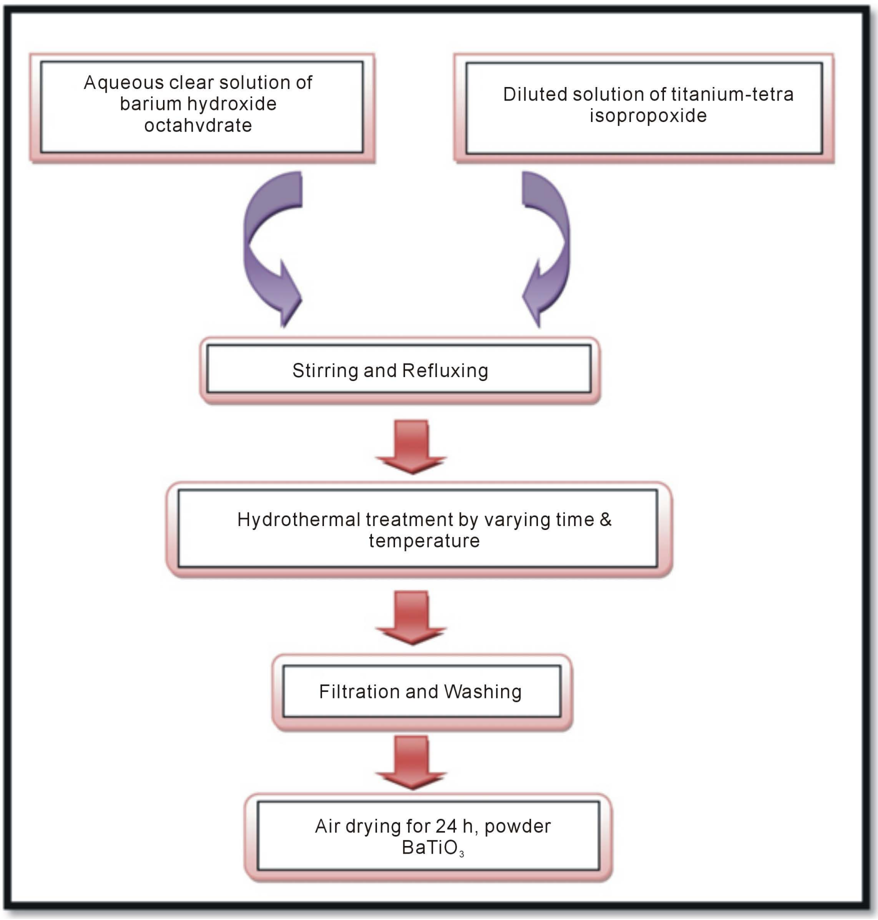

Figure 7. Flow chart of hydrothermal technique [53].

solution which was heated to remove the remaining ammonia in the solution at 80˚C. This solution is added into the ethanol drop by drop, after which the barium iron citrate complex precipitates out by alcohol dehydration and these precipitates are then, dried and heated at 425˚C - 470˚C for 48 h. This resulted into ~10 nm particle and these particles are sintered at specific temperatures to get the BaMpowder. It was reported that the powder sintered at temperature 600˚C and 700˚C resulted into 50 nm and 100 nm particles. It was found that the 50 nm particles shows the saturation magnetization of about 33 emu/g and coercivity about 580 Oe and the 70 nm particles shows a saturation magnetization of about 35 emu/g and coercivity of 4800 Oe but the BaM powder sintered at 7000 C resulted into 60 nm particles which shows a saturation magnetization of 61.5 emu/g prepared by this method [56] [57] [58]. The steps involved in the synthesis of materials by polymeric citrate precursor technique are given in flow chart shown in **Figure 8** [59].

3.7. Molten Salt Method

The molten salt method is also known as the salt melt method and is one of the best methods for the preparation of ceramic powders. The M-type ferrites can

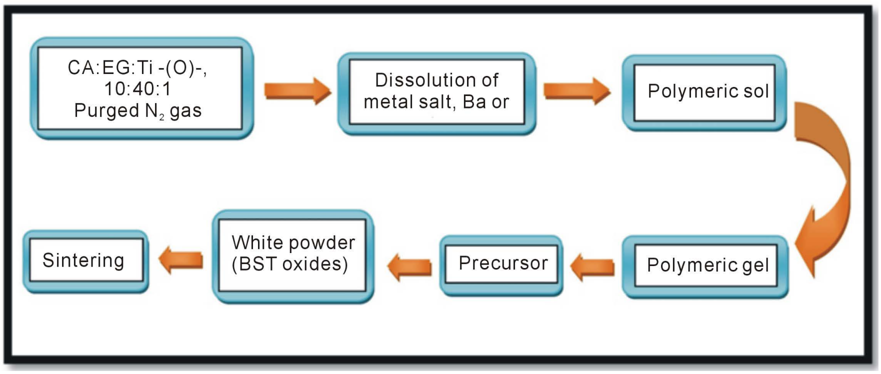

Figure 8. Flow chart of polymeric citrate precursor technique [59].

also be prepared by using this method. The starting solution is prepared by mixing barium carbonate and iron oxide precursor powders to which the potassium chloride and sodium chloride salt mixtures are added to it. Then, heated it at a temperature range of 800˚C - 1100˚C [60] and after that, the dried sample is crushed and washed with unionized water to get the salt out of the dry sample to get the magnetic powder. It has been demonstrated that the magnetic properties of BaM depends on the optimal experimental conditions such as saturation magnetization, coercivity etc. and the BaM with the good magnetic properties (saturation magnetization of 72 emu/g and coercivity of 4300 Oe) can be made with an appropriate molar ratio of Fe:Ba = 10 with the aid of molten salt method [60] [61]. The preparation procedure for the sample material is given in flow chart shown in **Figure 9** [62].

4. Further Developments in the Synthesis Techniques of M-Type Hexaferrites

It has been demonstrated that the BaM hexaferrites powders composed of three phases which are BaM, intermediate $BaFe_2O_4$ and Fe_2O_3 and these powders are prepared by conventional ceramic technique [63] which is the simple technique in controlling the grain size and morphology of ferrite powder product but addition of B_20_3 in small appropriate amounts to the BaM powder results in BaM phase only with the enhanced remanent magnetization. It was observed that the addition of B_20_3 in the BaM powder prepared helps in improving the magnetic characteristics of BaM hexaferrite. It has also been improved by doing etching with HCL solution along with B_20_3addition [64]. It was reported that addition of 1% of $B_20_3$to the BaM powder results in saturation magnetization of 54 emu/g, remanence magnetization of 28 emu/g and coercivity of 2000 to 3000 Oe but the addition of 0.1% of B_2O_3 to the BaM powder results in the saturation magnetization of 63.3 emu/g and ramenance magnetization of 34.9 emu/g [64].The BaM hexaferrite obtained powders with low coercivity prepared by oxalate precursor technique [65] but it has been reported that the coercivity and the saturation magnetization increases with the increase of temperature from 900˚C to 1100˚C when we prepared the powders by taking the starting solutions prepared by

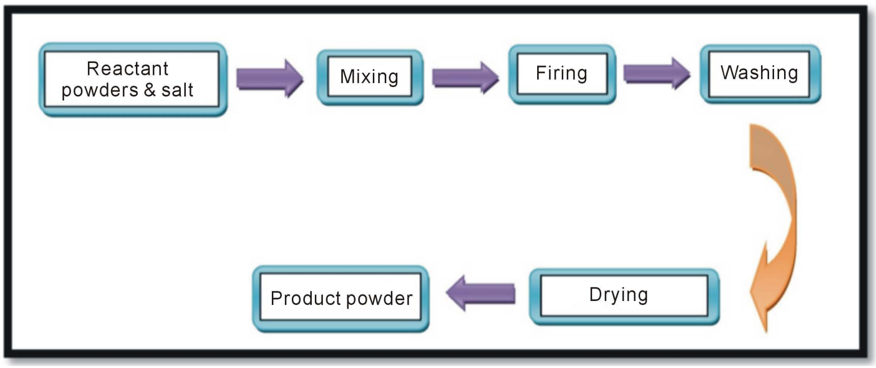

Figure 9. Flow chart of molten salt synthesis of ceramic powders [62].

dissolving the metal chlorides in equal amounts of oxalic acid with a ratio of Fe:Ba = 12 and clean the solution with constant stirring on stirrer about 15 minutes which was then, heated at 80˚C on magnetic stirrer with hot plate and dried it at 100˚C on stirrer with hot plate for 12 hours and then, finally calcinated it for 2 hours in the temperature range of 800˚C - 1200˚C. It was reported that the powder prepared exhibit saturation magnetization at a maximum value was about 66.36 emu/g and the coercivity at a maximum value was about 640 Oe. It was also demonstrated a new technique has been developed for preparing the high quality hexaferrites at low Fe:Ba ratios [66] called ammonium nitrate melt synthesis [67]. The starting solution is prepared by mixing the appropriate quantities of $BaCO_3$ and Fe_2O_3 in the ammonium nitrate melt. Stir it on magnetic stirrer with hot plate and then, heat it at 260˚C. This results in reddish precipitates which were further heated for 5 hours at 450˚C and the parts of the powder obtained was provided a heat treatment at 800˚C - 1200˚C for observing the optimal heat treatment. It was reported that the powder sintered at 1100˚C gives two phases $BaFe_2O_4$ and BaM but powder washed with HCL solution gives BaM phase only. This only one BaM phase observed with a saturation magnetization of about 66.7 emu/g, coercivity of 4228 Oe and remanent magnetization of about 38.5 emu/g. It has observed that hexaferrite powders prepared by the synthesis techniques described above directly affects the quality and magnetic characteristics of powders prepared and also, the hexaferrites having low values of coercivity and saturation magnetization are useful in the field of magnetic recording applications but the hexaferrites having high values of coercivity and saturation magnetization are useful in the field of permanent magnets (PM). In Table 2, some of the values are listed on the basis of experimental findings that deal with the magnetic properties of the hexagonal ferrites in the field of permanent magnets and the magnetic recording applications.

5. Magnetic Properties of M-Type Hexaferrites

M-type barium hexaferrite also called as the BaM hexaferrite is the most important M-type hexaferrite in the field of magnetic recording industry and microwave properties and it is ferrimagnetic in nature with a maximum density of $\rho =$

5.25 g/cm^3 and coercivity of 160 - 255 K·Am^{-1}. **Table 3** shows the magnetic properties or characteristics of M-type hexaferrite where $M_s$represents saturation magnetization, H_c represents coercivity and k_1 represents first order anisotropy constant [79].

5.1. Ba-M Ferrite

It has been demonstrated that the various observations have be made on the single domain size of BaM ferrite, initially its value was found to be as 1.3 um but after performing the experiments, its value found as being 1.8um [79]. If its value will be observed by using Kettle's theory, then, it is found as being 0.3 - 0.9 um [80] and also, the domain size increases with the increase in temperature. The BaM ferrite have high theoretical value of coercivity reported around 594 K·Am^{-1} and also have high value of saturation magnetization reported around 72 Am^2·Kg^{-1} but the BaM ferrite prepared by using conventional ceramic technique have low value of coercivity reported around 159 - 255 K·Am^{-1} and have high

Table 2. Magnetic properties of M-type hexaferrites obtained through the various experimental synthesis techniques [68].

Synthesis Technique	M_S (emu/g)	M_r (emu/g)	H_C (emu/g)	Applications	Reference
Conventional ceramic technique	61	32	2080	HPMR	[68]
	49	24	1050	HLMR	[69]
	71	37	4020	PM	[70]
Coprecipitation technique	64	31	4700	PM	[71]
	69	36	5440	PM	[75]
	60	15	860	HLMR	[72]
Sol-gel technique	61	37	4996	PM	[73]
	59	36	1920	HPMR	[74]
	61	36	5692	PM	[75]
Molten Salt technique	59	-	4820	PM	[76]
	72	-	4650	PM	[77]
	72	-	4300	PM	[77]
Hydrothermal technique	40	-	2500	PM, HPMR	[77]
	59	20	1350	HPMR	[78]
	60	-	2300	PM, HPMR	[78]

Table 3. Magnetic properties of BaM and SrM hexaferrites [79].

M-type Ferrite	Chemical formula	M_S	Coercivity (H_C)	Isotropic M_r/M_s	H_A (K·Am^{-1})	K_1 (10^5 jm^{-3})	T_C (°C)
SrM	$SrFe_{12}O_{19}$	92 - 74	High value	0.50	1592	3.5	460
BaM	$BaFe_{12}O_{19}$	72	High value	0.50	1353	3.3	450

value of saturation magnetization reported around 70 $Am^{2}Kg^{-1}$ [80]. However, it has been observed that with the increase in temperature, the coercivity also increases. Example from 135 $K\cdot Am^{-1}$ at −200˚C to 255 $K\cdot Am^{-1}$ at 25˚C and peak at 380 $K\cdot Am^{-1}$ at 250˚C. After this specified point, the coercivity decreases [81]. BaM ferrite prepared with the help of organic precursor synthesis gives high value of saturation magnetization reported around 70 $Am^{2}\cdot Kg^{-1}$ and low value of coercivity reported around 240 $K\cdot Am^{-1}$ at a temperature of 900˚C for one hour with the grain size of 200 nm but BaM ferrite prepared with the help of non-stoichiometric aqueous sol-gel synthesis gives high value of coercivity reported around 470 $K\cdot Am^{-1}$ and low value of saturation magnetization reported around 55.7 $Am^{2}\cdot Kg^{-1}$ with the same temperature and same grain size as in case of organic sol-precursor synthesis. **Table 4** shows the characteristics of BaM ferrite where ρ represents X-ray density, μ_B represents the number of Bohr magnetrons, molecular mass, measured M_s at room temperature, calculated M_s at zero Kelvin and T_c in case of BaM ferrite [81].

5.2. Substituted BaM Ferrrite

It has been observed the substitution was done in a ferrite for increasing its purity or we can say that for increasing the value of saturation magnetization. M-type barium hexaferrite also called as BaM is the most important M-type hexaferrite and in this case, substitution of non-magnetic ions such as zinc ions (Zn^{2+}) has been made for increasing the value of saturation magnetization and also, the zinc ions most probable occupy the tetrahedral sites which oppose the octahedral sites in the M-type ferrites to obtain the magnetic moment. Thus, the zinc ions increase the net magnetic moment by reducing the negative participation and zinc substituted BaM ferrite have high value of saturation magnetization reported around 65 $Am^{2}\cdot Kg^{-1}$ and coercivity around 199 $K\cdot Am^{-1}$. BaM doped with antimony for x goes to 1 by hydrothermal technique gives low value of coercivity reported around 36 KAm^{-1} and also, low value of saturation Magnetization. The low value of saturation magnetization is due to the presence of Sb_2O_3 produc. But, the BaM prepared by sol-gel technique doped with arsenic and antimony reduces the values of magnetic characteristics such as saturation magnetization, coercivity etc. [82]. The different values of saturation magnetization and coercivity are calculated by using the different synthesis techniques at the various specified temperatures for the BaM ferrite and substituted BaM ferrite are shown in **Table 5** [82].

5.3. Holonium Substituted BaM Ferrites

The effect of substitution of Holonium on the structural and magnetic properties of BaM ferrite was studied by using various techniques like XRD (X-ray diffraction), TEM (Transmission electron microscopy), Hysteresis loop etc.

5.3.1. Experimental Process

The starting powders were prepared by using combined method of sol-gel and

Table 4. Characteristics of BaM ferrite [81].

M-type ferrite	ρ (g/cm^3)	μ_B	Molecular mass (g)	Measured M_s at room temp. ($Am^2 \cdot Kg^{-1}$)	Calculated M_S at 0 K ($Am^2 \cdot Kg^{-1}$)	T_c(˚C)
BaM	5.28	20	1112	72	> 100	450

Table 5. RT saturation magnetization and coercivity values for BaM and substituted BaM ferrite [82].

M-type ferrite	Synthesis technique/composition	M_s ($Am^2 \cdot Kg^{-1}$)	H_c ($K \cdot Am^{-1}$)
BaM	Sol-gel unprefired, 1050˚C	70	286
	Sol-gel prefired, 900˚C	70	474
	Sol-gel derived fibers, 100˚C	58.4 - 63.8	401 - 428
Substituted BaM	Zn substituted BaM	65	199
	$BaFe_{12-x}B_xO_{19}$, 1400˚C	79	0.40

auto-combustion process for x goes from 0.01 to 0.04 (0.01, 0.02, 0.03, 0.04). The various techniques have been used for determine the crystal structure, particle sizeand morphology, magnetic behavior which are XRD, SEM and VSM. The magnetic measurements, magnetic behavior and magnetic properties of the prepared samples were examined by using VSM (vibrating sample magnetometer). The particle size and morphology of the prepared samples were examined by using SEM (scanning electron microscopy) but the structure, composition and properties of the prepared samples were examined by using TEM (transmission electron microscopy).

5.3.2. XRD Analysis

The XRD patterns of Holonium substituted BaM hexaferrites for variation of x= 0.1, 0.2, 0.3, 0.4 given in **Figure 10** which shows that with the increase in doping of Ho^{3+} ions, secondary phase of Fe_2O_3 was seen which was also observed by various other researchers [83] [84] [85]. With the help of Scherer's formula, the particle size "t" was obtained for (114) & (107) peaks and with the increase in the doping of Ho^{3+} ions, the particle size also decreases from 51 nm to 44 nm [86] [87] [88] and also, due to the difference in the ionic radii of Ho^{3+} (1.04 Å) [89] and Fe^{3+} (0.67 Å) [90], extra phase exists.

5.3.3. TEM Analysis

The particle size and microstructural morphology of prepared samples of Holonium substituted barium nanohexaferrites was confirmed by studying its TEM, SAED and HRTEM patterns given in **Figure 11** which shows that the average size increases from 45nm to 60 nm.

5.3.4. Magnetism

For ferrite based nanostructure materials all the magnetic parameters can be calculated from the M-H Study. **Figure 12** shows the hysteresis loop determined

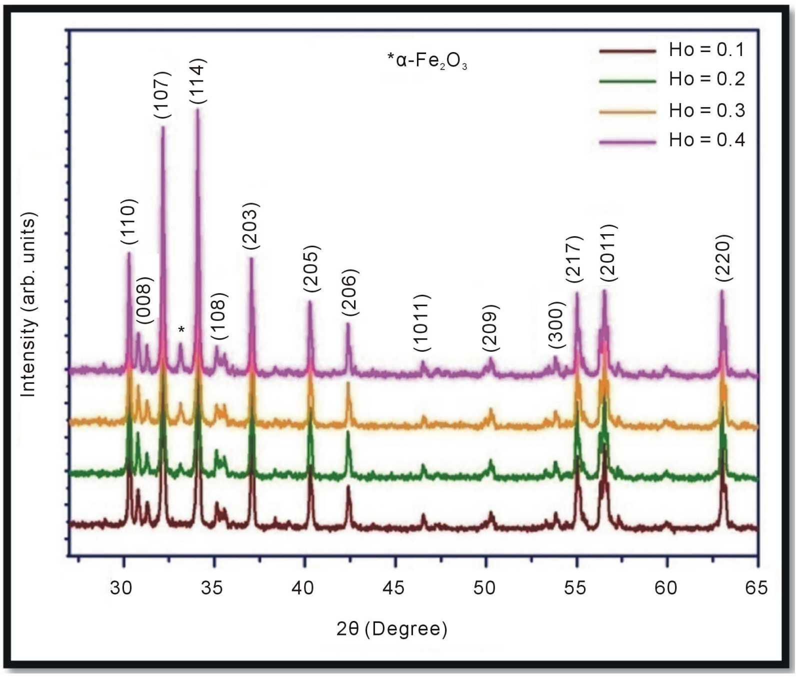

Figure 10. XRD patterns of BaHo$_x$Fe$_{12-x}$O$_{19}$ nanohexaferrites [91].

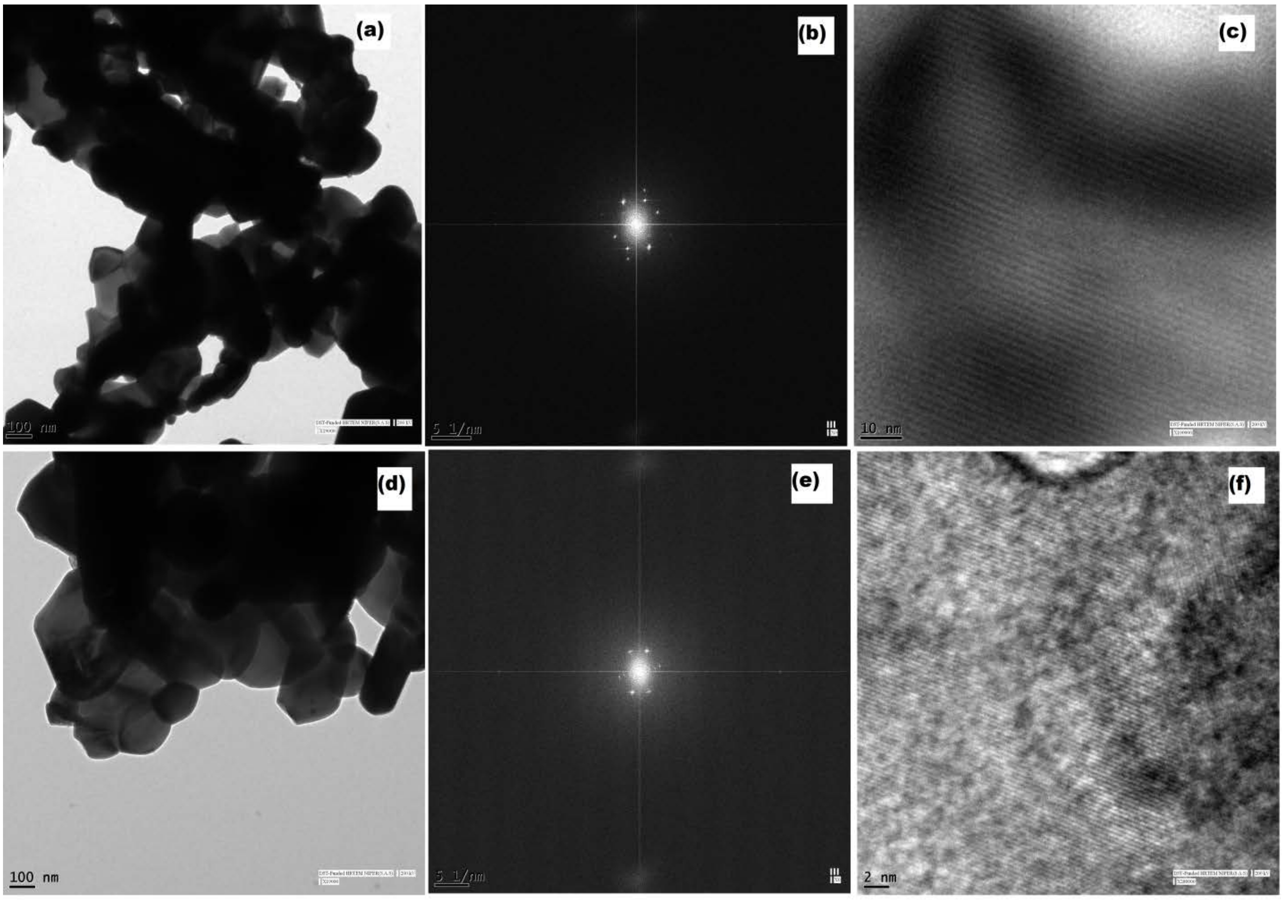

Figure 11. (a)TEM image (b) SAED pattern (c) HRTEM image of BaHo$_{0.3}$Fe$_{11.70}$O$_{19}$ nanohexaferrites sample and (d) TEM image (e) SAED pattern (f) HRTEM images of BaHo$_{0.4}$Fe$_{11.60}$O$_{19}$ nanohexaferrites sample [91].

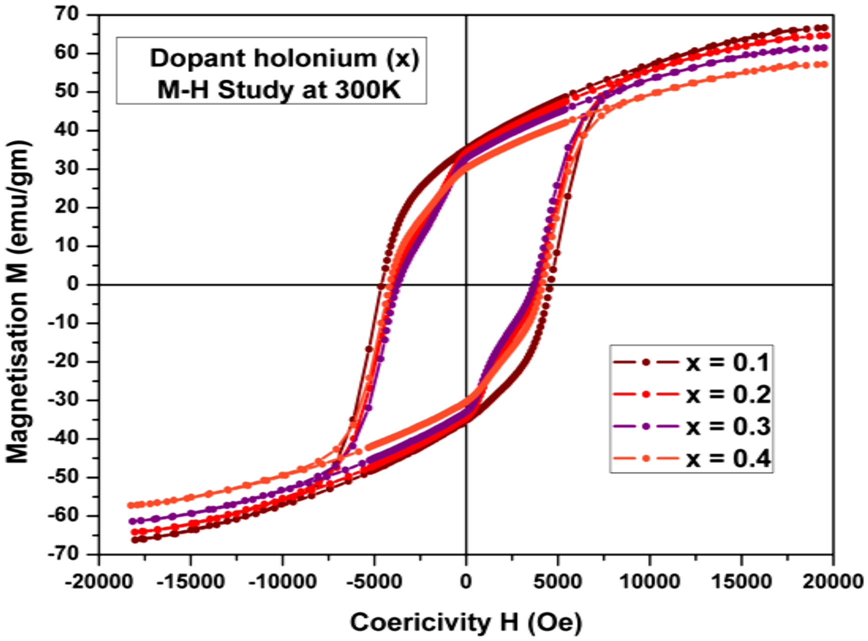

Figure 12. Room temperature variation of magnetization with applied coercive field for BaHox$Fe_{12-x}O_{19}$ (x = 0.1.0.2, 0.3, 0.4) [91].

as a function of applied magnetic field of Holonium substituted BaM nanohexaferrites for x = 0.1, 0.2, 0.3, 0.4 shows that the saturation magnetization decreases with the increase in doping of Ho^{3+} ions.

6. Conclusion

In this review paper, we have studied the effect of experimental techniques on the various magnetic properties of M-type hexaferrites. A number of techniques are used for synthesizing the M-type hexaferrites and also, for modifying their magnetic properties, at presently. It is depicted from various studies that chemical wet synthesis techniques are more effective as compared to conventional ceramic techniques. In addition to that, proper selection of dopants has direct effect on the structural and magnetic properties of M-type hexaferrites as shown in case of lanthanides substituted BaM hexaferrites. Magnetic parameters like saturation magnetization, magnetic retetentivity, coercivity, squareness ratio and magneton numbers may be explored on perfect substituent, intensively used in magnetic storage devices and recording industry. In the nut shell, we conclude that these types of hybrid magnetic materials have wide range of applications in the field of radar technology, wireless communication technology, satellite technology and miniaturization antenna technologies up to ultra high frequency range (1 - 50 GHz) where low frequency ferrite based magnetic materials have shown their limit.

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