

Perspectives of Substituted Ferrites in Current Scenario

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ABSTRACT

During last few decades, there has been a growing degree of interest in ferrites. The magnetic, electrical, optical and other properties of ferrites gain attention due to their use in various applications such as medical diagnostics, rechargeable lithium batteries, high frequency media, solar energy devices and magnetic fluids. The high resistivity and low eddy currents makes ferrites the better choice over metals. The aim of this review paper consists of an overview on the ferrites, classification of ferrites, synthesis methods and its potential applications in different fields of technology as well as summarize the major researches in the field of Mn-Zn ferrites on one platform.

Keywords : Ferrites, Magnetic Fluids, Spinel, Retentivity, Nanoparticles

I. INTRODUCTION

Ferrites are chemical compounds obtained as powder or ceramic bodies with ferrimagnetic ordering due to the superexchange or interaction between the magnetic moments properties formed by iron oxides as their main component. Ferrites have the molecular formula of MFe_2O_4 , where M stands for the divalent metals such as Fe^{2+} , Mn^{2+} , Co^{2+} , Ni^{2+} , Cu^{2+} , Mg^{2+} , Zn^{2+} or Cd^{2+} . Like most other ceramics, ferrites are hard and brittle. They are also insulating or semiconducting metal oxides that exhibit high coercivity, high electrical resistance, low eddy current and dielectric loss with moderate permittivity. No other material has such a wide range of properties and therefore these materials are exploited for vast applications in various fields like transducers, activators, recording media, permanent magnets, phase shifters, electrode material for Lithium ion batteries, solid oxygen fuel cells and computer

technology. The ferrite materials also exhibit dielectric properties and do not conduct electricity easily; therefore ferrites became an alternative for the metal magnets like iron and nickel. Ferrites are also magnetic dielectric materials that allow an electromagnetic wave to penetrate via it, thereby permitting an interaction between the wave and magnetization within the medium. This makes them suitable for high frequencies application because an ac field does not induce undesirable eddy currents in an insulating material; even in microwave frequencies and they find very important applications in the field of microwave and optical communications. Therefore, due to the technological importance of ferrites increasing continuously as many discoveries required the processing of these materials is important to modify its properties as per the desired applications. Ferrites are structure sensitive materials and their properties critically depend on preparation method,

sintering condition, and amount of constituent metal oxides, grain size, porosity and the dopants or substituted elements. A. B. Gadkari, et al. reported that; ferrites are highly sensitive to preparation method, sintering condition, amount of constituent metal oxides, various additives include in dopants and impurities. These ferrite materials can be prepared by conventional synthesis methods such as; high-temperature solid-state reaction method, sol-gel method, co-precipitation, pulsed laser deposition, high-energy ball milling and hydrothermal synthesis methods.

II. METHODS AND MATERIAL

Types of ferrite materials

Ferrite materials can be classified depending on both crystal structure and magnetic properties. Depending on their crystal structural ferrites are classified in to three types: spinel ferrites, garnets, and hexagonal ferrites.

2.1 Spinel ferrites

Spinel ferrites are represented by the formula unit AB_2O_4 . Most of the spinel ferrites form cubic spinel structure with oxygen anions in face centered cubic (fcc) positions and cations in the tetrahedral and octahedral coordinated interstitial lattice sites, forming the A and B sublattices. The spinel ferrites possess the general mineral spinel structure of $MgAl_2O_4$ which was first determined by Bragg and Nishikawa in 1915. They are also called cubic ferrites with chemical formula of MFe_2O_4 where M is a divalent transitional metal ions such as Co^{2+} , Mn^{2+} , Zn^{2+} , Fe^{2+} , Mg^{2+} , Ni^{2+} , Cd^{2+} , Cu^{2+} and etc. Depending upon the magnetic or non-magnetic nature and distribution of cations among A and B sublattices, spinel ferrites can exhibit properties of different type magnets, like: ferrimagnet, antiferromagnet and paramagnet. Among the broad classification of magnetic oxides, the spinel ferrites, due to their high magnetic permeability and low

conduction losses find wide use in high frequency devices. S. Sugimoto et al. reported that spinel ferrites are the most widely used family of ferrites because of their high values of electrical resistivity and low eddy current losses make them ideal for their use at microwave frequencies. They are used in magnetic recording media, a microwave device, and magnetic resonance imaging. The spinel ferrites have been classified in to three types due to the distribution of cations on tetrahedral (A) and octahedral (B) sites. They are normal, inverse and partial inverse spinel ferrite.

2.1.1 Normal spinel ferrites

In normal spinel ferrites, all the divalent cations occupy tetrahedral site and trivalent cations occupy the octahedral sites. The general formula is: $[M^{2+}]_T [M^{3+}_2]_O O_4$ Where letter 'O' indicates octahedral site occupancy and the 'T' indicates tetrahedral site occupancy. Here, octahedral sites are occupied by only one kind of cations. In other words, in this structure, the non-magnetic ions occupy the A sites and consequently there is no AB interaction. The negative BB interaction now becomes dominant and the trivalent cations align themselves in an anti-parallel fashion; thereby producing zero net magnetization. Examples of normal spinel ferrites are $ZnFe_2O_4$ and $CuFe_2O_4$.

2.1.2 Inverse spinel ferrites

In inverse spinel ferrites, the trivalent cations occupy both the octahedral and tetrahedral sites and divalent cations occupy only octahedral sites. The general formula is; $[M^{3+}]_T [M^{2+}M^{3+}]_O O_4$ In other words, in this case there are 8 M^{2+} ions that occupy 8 octahedral sites and the 16 Fe^{3+} ions are divided into 8 octahedral sites and 8 tetrahedral sites. $NiFe_2O_4$ and $CoFe_2O_4$ has an inverse spinel crystal structure.

2.1.3 Mixed spinel ferrites

In mixed spinel ferrites, the divalent and trivalent cations are randomly distributed among the tetrahedral and octahedral sites. It is intermediate

cation distribution between the normal spinel and inverse spinel. The cation distribution is given by the general formula: $(M_{3+}^{\delta} M_{2+}^{1-\delta})[M_{3+}^{2-\delta} M_{2+}^{\delta}]O_4$

where δ is the degree of inversion which depends on the synthesis techniques, calcination and sintering temperature with a value of zero for the normal and one for the inverse distribution. $MnFe_2O_4$ is an example of mixed spinel structure.

2.2 Garnets

Garnets have the general formula $M_3Fe_5O_{12}$ where $M = La, Y, Sm, Eu, Gd, Tb$ etc. and have applications in microwave systems. They have complex crystal structure with the cell shape is cubic and the edge length is about 12.5\AA . The coordination of the cations is considerably more complex than spinels, with 24 M_{3+} in dodecahedral sites, 24 Fe_{3+} ions in tetrahedral sites and 16 remaining Fe_{3+} in octahedral sites. Yoder and Keith reported that substitutions of cations can be made in ideal mineral garnet of $Mn_3Al_2Si_3O_{12}$. They produced and reported the first silicon free garnet $Y_3Al_5O_{12}$ by substituting Y_{3+} Al_{3+} for Mn_{2+} Si_4 . Bertaut and Forret prepared $Y_3Fe_5O_{12}$ (YIG) in (1956) which is well-known garnet and measured their magnetic properties. In (1957) Geller and Gilleo also prepared and investigated silicon free garnet $Gd_3Fe_5O_{12}$ which is also a ferromagnetic compound. Similar to spinels and hexagonal ferrites, a wide range of transition metal cations can substitute M_{3+} or Fe_{3+} ; especially rare earth ions may replace the ions on octahedral and dodecahedral sites. Each type of lattice site will accept other metal ions at dodecahedral sites, octahedral sites and at tetrahedral sites. Thus, in garnets, pentavalent ions such as V^{5+} and As^{5+} can occupy tetrahedral sites, while Ca^{2+} substitute ions on dodecahedral sites. They are important due to their applications in memory structure.

2.3 Hexagonal ferrites

Hexagonal ferrites are well established magnetic materials represented by the general Formula: $M_2Fe_{12}O_{19}$ where $M = Ba, Sr, Ca, Pb$, etc are

important in permanent applications. They can be found in cost-effective hard magnets as well as in components for high frequency applications and they are widely used as permanent magnets and characterized by possessing a high coercivity. The hexagonal ferrite lattice is similar to the spinel structure, with the oxygen ions closely packed, but some layers include metal ions, which have practically the same ionic radii as the oxygen ions. This lattice has three different sites occupied by metals: tetrahedral, octahedral, and trigonal bipyramid surrounded by five oxygen ions. Hexagonal ferrites have a variety of magnetic structures and properties that are determined by structure and particular composition which is identified by Went et al. (1952) & Jonker et al. (1956). Out of iron oxides, hexagonal ferrites are a broad subset which is of great practical importance as well as scientific interest. These systems are ferrimagnets as dominant interaction between magnetic ions and oxygen-mediated anti-ferromagnetic superexchange. They are widely used as permanent magnets and have high coercivity. Hexagonal ferrites have larger ions than that of garnet ferrites and are formed by the replacement of oxygen ions. Most of these larger ions are barium, strontium or lead.

2.4 Classification of ferrites on the basis of magnetic property

Depending on the magnetic properties, ferrites are often classified as hard and soft ferrites. This classification is based on their ability to be magnetized or demagnetized not their ability to withstand penetration or abrasion.

2.4.1 Hard ferrites

Hard ferrites are characterized by a large value of retentivity and coercivity after magnetization that means; hard ferrites are difficult to magnetize or demagnetize as soft ferrites easily which is an essential characteristic of a permanent magnets, so they find applications as permanent magnets in radios; e.g., strontium and barium ferrite. Their maximum

magnetic field is about 0.35 T and magnetic field strength is about 30 to 160 kA/m. Hard ferrites are ferromagnetic materials that are typically oxides of mixed transition metals that containing the iron.

For example, $MnFe_2O_4$ and Mg-Zn ferrite are described as $Mn_{1-x}Zn_xFe_2O_4$. They are usually insulating in nature and like most other ceramics, they are hard and brittle. These hard ferrites are used in applications such as magnetic components in microelectronics. The most commonly used hard ferrite is Cobalt ferrite, $CoFe_2O_4$ ($CoO \cdot Fe_2O_3$), it is used in magnetic recording applications such as audio-/video-tape and high-density digital recording disks. Most hard ferrite particles used in synthesizing magnetic fluids exhibit a spinel structure. This structure consists of a cubic closed packed case of oxygen ions with the metallic ions occupying the tetrahedral A and octahedral B sites. Magnetically hard ferrites have these advantages and particular properties: economical raw materials, very good resistance against corrosion and chemicals and easy to magnetize.

2.4.2 Soft ferrites

Ferrites which are magnetically soft are often described as soft ferrites. They are characterized by a small value of coercivity so they cause low hysteresis loss at high frequency owing to which they are widely used in electromagnetic cores of transformers, switching circuits in computers and radio field inductors. Examples of soft ferrites are manganese-zinc ferrite, nickel ferrite, and lithium ferrite. Soft ferrites are those that can be easily magnetized or demagnetized. This shows that soft magnetic materials have low coercive field and high magnetization that is required in many applications. Due to the low coercivity of soft ferrites, their magnetization can switch direction without much energy requirement or heat generation. For this reason; soft ferrites find wide applications in electronic industries or in cores of modern electronic components such as recording

heads, filters, switching power supply transformers, amplifiers, etc.

2.5 Ferrites synthesis methods

There are varieties of available synthesis methods, which can be successfully used to synthesize ferrite materials. The most commonly used synthesis methods are discussed below.

2.5.1 Solid state reaction synthesis

The physical, chemical and electrochemical properties of materials depend to a great extent on the synthesis methods. Several synthesis methods have been developed for the preparation of materials. Some of them are solid state synthesis, co-precipitation, sol-gel process, hydrothermal method and etc. Solid state reaction synthesis method is one of the common methods employed for preparing powder materials from oxides, carbonates, hydroxides, nitrates, sulfates, acetates, oxalates, and other metal salts. This technique involves heating mixtures of two or more solids to form a solid phase product. In this synthesis method, solvents are not used. Since, solids do not react with each other at room temperature, solid state synthesis needs much higher temperatures and longer heating time than other techniques. Solid state reaction differs from solution reaction, since in solution all ions and molecules are available for reaction. However, this is not the case in a solid state reaction. Here, the reaction takes place only at that point where the reactions are in intimate contact with each other. Once the product layer is formed at the interface between the reactants, further progress of the reaction depends upon the diffusion of one or both reactants through this product layer. This depends on various factors such as the size of the diffusion ions, the reaction temperature, and also the presence of defects and the history of the sample.

2.5.2 Sol-gel synthesis

Sol-gel processing is a wet chemistry technique, which can be used to synthesize solid electrolyte ferrite materials and powders as well as nanoparticles by a process preparation of a sol, gelation and aging of

it and removal of the liquid . A sol is a stable colloidal suspension of solid particles or molecular precursors in a liquid solvent. The colloidal particles are agglomerates and form polymer chains creating a polymer gel, a three dimensional continuous network including a liquid phase. The most widely used precursors for the sol-gel preparation are metal alkoxides, metal chlorides, nitrates and acetates which undergo hydrolysis and poly-condensation reactions. The sol-gel process works by following mechanism. Firstly, formation of stable solutions of the alkoxide or solvated metal precursor. Then process of gelation resulting from the formation of an oxide or alcohol bridged network by a poly-condensation or polyesterification reaction resulting in a dramatic increase in the viscosity of the solution. Within the next step is the process of gel aging during which the gel transforms into a solid mass. The next stage includes drying of the gel, when water and other volatile liquids are removed from the gel network. Dehydration is the fifth phase, where the gel is stabilized against rehydration. Calcining the monolith at temperature up to 800°C is normally required in order to achieve the final product. The sol-gel method has many advantages. The mixing of precursors taking place in a very short period of time and homogeneous gel can be obtained. During the sol-gel method, the chemical reactions occur more readily, with a much lower reaction temperature, which is advantageous in comparison to for instance solid-state reaction synthesis. Along with the advantages, the disadvantages also exist. For example, the precursors used for the synthesis could be expensive and in the case of organic precursors these could be toxic. One of the major disadvantages that the sol-gel process itself takes long processing times or several steps are involved.

2.6 Applications of ferrites

In the past decade MnZn have attracted a large amount of attention in academia due to its advantageous features that make MnZn ferrites

suitable to be used in many applications of daily life. . Due to useful magnetic, electrical and optical properties of ferrite nanoparticles, researchers are taking interest in the synthesis of ferrite nanoparticles and making their use in a lot of applications that include medical field, information technology, antenna, microwave absorbing materials, biosensors and many electronic applications.

MnZn ferrites have a broad area of applications due to high saturation magnetization , high initial permeability , low power loss. The application area of MnZn ferrites include power applications, microwave devices, magnetic fluid , radar absorbing system, high frequency applications , bio-medical , water purification etc. Use of MnZn ferrites in the field of power application attracted great attention in the research areas. From last many years the MnZn ferrites are synthesized to be used in power applications for making current convertors, power inductors with magnetic cores , electronic transformer cores , high frequency applications , electronics and communication.

2.6.1 Microwave devices

Ferrite nanoparticles have low electrical conductivity and low dielectric losses , so they can be used in microwave devices. MnZn ferrites are most suitable for their use in the microwave devices because of high permittivity, high resistivity, high stability, high value of saturation magnetization, high curie temperature with low eddy current and low magnetic losses. Due to the use of ferrite nanomaterials, electronic devices can be mechanically hard, chemically stable and permit the materials to operate properly at a wide frequency range. There are a lot of advantages of the use of MnZn ferrites in the microwave devices. There is a decrease in the emission of unwanted EM waves from the device and also it absorbs the incoming EM waves that may harm the microwave device. MnZn ferrites are used in microwave systems because of their low loss and high saturation magnetization. Wang et al. synthesized MnZn ferrite nanoparticles

and the result showed that because of high reflection loss and broad absorbing band in low frequency (10 MHz to 1 GHz) these ferrites can be used in electromagnetic microwave absorbing field.

2.6.2 Radar absorbing devices

The radiations emitting from radar results in the increase in electromagnetic radiation pollution in the environment. These radiations reduce the efficiency and performance of electronic instruments and thus decrease their lifetime and safety. As MnZn ferrite belongs to the class of soft ferrites having high electrochemical stability, high permeability, high saturation magnetization and low power losses, it is used in many electronic applications. Ferrite nanoparticles can be used in the radar absorbing devices due to their high value of Curie temperature and temperature stability. Also the ferrite nanoparticles are environmentally safe that make their use easier in the radar absorbing devices. The application of MnZn ferrites in radar absorbing system is also attracting the researchers. Praveena et al. synthesized $\text{Ni}_{0.4}\text{Zn}_{0.2}\text{Mn}_{0.4}\text{Fe}_2\text{O}_4$ nano ferrites for radar absorbing. The high value of Curie temperature indicated homogeneity and temperature stability. The EPR spectra showed reduction in the peak width and increase in relaxation with increase in sintering temperature. These all results showed that the ferrite nanoparticles can be used for radar absorbing from few MHz to 2 GHz and also these materials are environmentally safe.

2.6.3 Image based diagnostics

A one-pot thermal decomposition method was used to synthesize a series of Zn^{2+} doped nanoparticles of $(\text{Zn}_x\text{Mn}_{1-x})\text{Fe}_2\text{O}_4$ and $(\text{Zn}_x\text{Fe}_{1-x})\text{Fe}_2\text{O}_4$ ($x = 0, 0.1, 0.2, 0.3, 0.4, \text{ and } 0.8$). By carefully controlling Zn^{2+} doping level, nanoparticles of size 15 nm with single crystallinity and size monodispersity ($s < 5\%$) and having high magnetization value (175 emu/g) were obtained. The nanoparticles provided the large MRI contrast effects ($r_2 = 860 \text{ mm}^{-1}\text{s}^{-1}$) with an eight to fourteen fold increase in MRI contrast and a fourfold

enhancement in hyperthermic effects compared to conventional iron oxide nanoparticles. This enhancement was significant for clinical purposes as the nanoparticle probe dosage level can be progressively lowered when using probes that have improved contrast enhancement effects. For $(\text{Zn}_x\text{Mn}_{1-x})\text{Fe}_2\text{O}_4$ nanoparticles, Zn^{2+} ions mainly occupy tetrahedral sites of the spinel matrix which was confirmed by using extended X-ray absorption fine structure (EXAFS) analysis to examine the Zn and Fe K-edges. To detect small sized pathogenic targets precisely at an early stage, MRI contrast agents are often used to highlight those specific areas of interest. Due to high imaging contrast effects, magnetic nanoparticles can increase the difference between pathogenic targets and normal tissues via MRI. One of the most appropriate ways to increase the MR contrast effects is the optimization of saturation magnetization (M_s) that is directly related to the relaxivity coefficient (r_2). The relaxivity coefficient (r_2) is determined by a slope of R_2 against nanoparticle concentration and often used as an indicator for contrast effects. The relaxivity coefficient (r_2) of contrast agents can be tuned and further enhanced by engineering magnetic parameters.

2.6.4 Electronic devices

MnZn ferrite nanoparticles are used in making many electronic devices due to their enhanced electrical properties such as high value of resistivity, low ac conductivity, low power losses etc. Dobak et al. studied miniaturization of components due to low loss MnZn ferrites. Also, Sun et al. studied effect of ZrO_2 addition on the microstructure and various properties of MnZn ferrites and found that the optimal values of initial permeability (2322), saturation magnetization (522 mT) and power loss (386 kW/m³) make it suitable for switch mode power supply applications. Due to suitable electrical and magnetic properties of the Sc^{3+} doped Mn-Zn ferrites, these were useful for modern technological application as well as for low and high frequency application. MnZn ferrites are also

used to construct power inductors , wireless power transfer applications and for making inductive components.

2.6.5 Telecommunication and others

One of the major use of MnZn ferrites is in telecommunication and high frequency applications. MnZn ferrites have applications in the field of bio-medical and hyperthermia. Hurtado et al. synthesized MnZn ferrite along with activated carbon composite for use in bio-medical applications. MnZn ferrites can be used to make ferrofluid due to high value of saturation magnetization. Arulmurugan et al. synthesized Co-Zn and Mn-Zn ferrite nanoparticles and found that because of low Curie temperature and high value of thermomagnetic coefficient, these ferrites can be used for preparing temperature sensitive ferrofluid. Praveena et al. synthesized Mn-Zn ferrite nanoparticles for high frequency applications. The ferrites had low power loss in frequency range 10Hz-1MHz. The constructed transformer with the ferrite material high efficiency and low surface temperature rise at frequency 1 MHz making it suitable for operating at high frequencies.

2.6.6 MnZn ferrites for ongoing COVID-19 pandemics

As nanomaterials are making a global impact on healthcare and socioeconomic development so are the viruses during pandemics. Nanoparticles of MnZn have unique physical and chemical properties that have associated benefits in development of potential therapeutic drugs, nanomaterial based environment friendly antiviral sprays, drug delivery and to develop anti-viral surface coatings in home appliances. This is attributed to the fact that the choice of synthesis method provides size and charge tunability properties to the MnZn ferrites. The size tunability ensures that large amount of drug can be delivered into anatomically privileged sites of the virus while charge tunability would facilitate entry of drug in to charged parts of the virus . In addition, biosensors for the early detection of viral strains such the COVID 19 can also

be developed with MnZn ferrites. For instance MnZn ferrites can readily be used to develop Giant magneto-resistance based sensors which have previously been used for virus detection.

III.CONCLUSION

The synthesis of MnZn particles has increased in the last ten years and most progress can be seen in the year 2016. Due to the fascinating properties of MnZn ferrites among the class of soft ferrites like high value of saturation magnetization, low value of coercivity, high initial permeability, narrow size distribution of the ferrite particles, low remenant magnetization, the researchers are taking interest in the synthesis of these ferrites. The co-precipitation and sol-gel method are the best for getting the fine crystallite size among all synthesis techniques. The XRD pattern of the MnZn ferrites has characteristic peaks showing the cubic spinel phase having Fd3m phase group. The shape of the prepared ferrite is nearly spherical but some distortion may be observed after doping. FTIR spectra confirmed the spinel phase of the ferrite nanoparticles having tetrahedral and octahedral sites. The value of saturation magnetization is highest when we synthesize the MnZn ferrites with proper amount of nickel doping by using sol-gel auto combustion method. Also, for getting the low value of coercivity sol-gel method is preferred. Generally, MnZn ferrites have a lot of applications including biomedical field, electronic devices, for making radar absorbing materials, for making ferrofluids etc. For enhancing the applications and advantageous properties of MnZn ferrite nanoparticles, further studies are required. The electrical and magnetic properties of MnZn ferrites can be enhanced by doping other metals such as cobalt, zinc, magnesium to make them suitable for use in agricultural and electrical applications. In the context of use of nanoparticles in the pandemic outbreak, such as in the recent COVID-19, MnZn soft ferrites can play a significant role in the development

of high contrast imaging dyes for viral strains in body fluids. Perhaps MnZn can also serve as a candidate nanomaterial for developing nanomaterial based medicines and therapeutics.

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