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A Study the Application of Nano Ferrites & Structural, Electrical, Magnetic & Optical Properties of Various Substituted Zinc Ferrite

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ABSTRACT

Nano ferrites find wide range of applications in magnetic memories, noise filters, isolators, ferro fluids, transformer cores, gas sensors, smart sensors etc. Nano zinc ferrite is an important member of the nano ferrite family due to its important properties like chemical stability, high electromagnetic performance, mechanical hardness, low coercivity& moderate saturation. Due to these unique properties, zinc ferrite finds a wide range of applications in gas sensors, photo catalyst, MRI contrast agent, catalyst and so on. This study deals with the variations observed in the structural, electrical, magnetic & nonlinear optical properties of various divalent and trivalent ion substituted zinc ferrite. Furthermore, Chlorella pyrenoidosa, a type of fresh water microalga, has been studied in relation to the effects of particle size, concentration, & divalent ion concentration (Co2+ ion) of zinc ferrite nanoparticles.

KEYWORDS- Nanoparticles, Ferrites, Ferrimagnetism, Magnetic, Electrical

INTRODUCTION

Ferrites are well known ferrimagnetic materials which are in commercial use for long time. In 1948, L. Néel provided a theoretical understanding of ferrites and word ferrimagnetism is invented by him [Chikazumi S., 1964]. Ferrites are combination of remarkable properties like magnetic, electrical and optical [Smit J. 1959]. They are electrically insulating with low dielectric losses and eddy currents and having high electrical resistivity while magnetically they acquire moderate to high saturation magnetization, coercivity and permittivity. No material other than ferrites having such extensive properties exists and as a result ferrites have applications in almost all fields. Iron and metal oxides are the main elements of the ferrites. In early 12th century Chinese were used lodestones (Fe3O4) in navigation compasses [Smit J. 1959]. The practical uses of ferrites have started in the year 1930 and since then they are widely studied by researchers.

Ferrites are categorised in two categories based on their magnetic behaviours:

a) Soft ferrites: Soft ferrites can reverse their magnetization by using small amount of energy. They have low to moderate values of coercivity and remnant magnetization. These ferrites have high electrical resistance and low eddy current losses so they are used in inductors and transformers. They are usually made by transition metal oxides.
b) Hard ferrites: Hard ferrites are usually called permanent magnets. They contain moderate to high values of coercivity and remnant magnetization. Classically hard ferrites are made of barium, strontium or iron oxides. They are inexpensive and used in variety of applications.

CLASSIFICATION OF FERRITES

On the basis of atomic and molecular structure ferrites are classified into three types: Spinel ferrites (Cubic ferrites), Hexagonal ferrites and Garnets [Standley K. J., 1972]. Since the present work is centred on spinel ferrites, hence we discuss spinel ferrites in details in the following section and rest of two in brief.

- **SPINEL FERRITES**

Spinel ferrites are the most widely used magnetic oxides having cubic structure and their interesting properties make them scientifically the most studied materials [Joshi S., 2014]. Their excellent properties make them suitable for many applications like colour imaging, catalytic activity, magnetically guided drug delivery, ferro-fluids, magnetic refrigeration systems, gas sensors and biomedical applications [Joshi S., 2014]. The spinel structure of ferrites was first determined by scientists Bragg and Nishikawa in 1915 [Standley K. J., 1972].The spinel ferrite is expressed by general formula MFe_2O_4 where M is divalent metal ion like Zn^{2+}, Co^{2+}, Mg^{2+}, Fe^{2+}, Cd^{2+}, Ni^{2+} and Cu^{2+} or their mixture etc.

- **Crystal structure of spinel ferrite**

Spinel ferrites possess fcc structure having eight formula units per unit cell. The oxygen ions form 96 interstitial sites in which 24 cations are placed (8 cations in 64 tetrahedral (A) sites while 16 cations in 32 octahedral (B) sites). In the tetrahedral geometry, three oxygen ions (anions) in a plane touch each and forth anion sits above at the centre and metal ion (cations) resides at the void. Figure 1(a) shows tetrahedral configuration where O is the anion and M is the metal ion (cation).

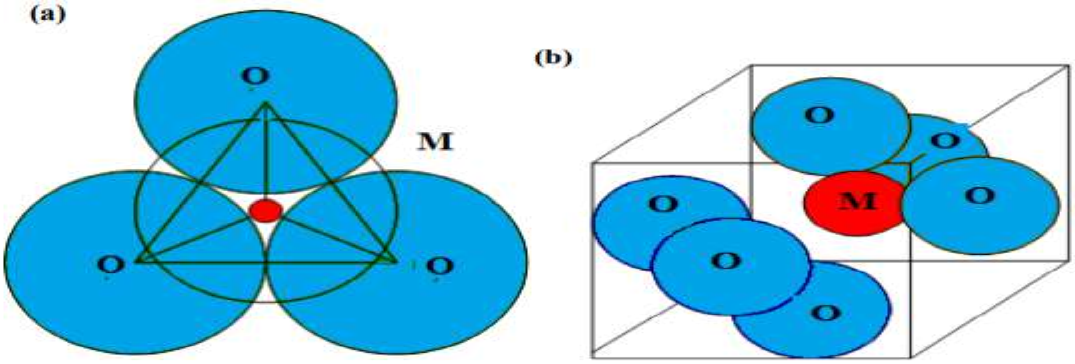

Figure 1: (a) tetrahedral configuration (b) octahedral configuration

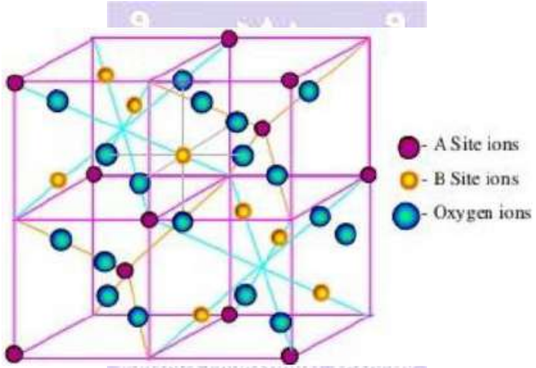

Figure 2: FCC structure of ferrite

In an octahedral geometry, four anions in a plane touch each other while the other two anions sit at above and below the centre of the plane and cation occupy the void created by these anions as shown in Figure 1(b). There are 8 cations to occupy 64 tetrahedral sites while 16 cations to occupy 32 octahedral sites in fcc lattice as shown in Figure 2. Spinel ferrites are further classified in three classes according to the allocation of cations at A and B sites as (a) normal spinel ferrites, (b) inverse spinel ferrites and (c) mixed spinel ferrites.

(a) Normal Spinel Ferrites

In this type of spinel ferrites, A sites are occupied by divalent cations (M^{2+}) while B sites are occupied by trivalent cations (M^{3+}). Normal spinel ferrites are expressed as $(M^{2+})A[M^{3+}]BO_4$. Where M^{2+} and M^{3+} are notations used for divalent and trivalent ions, respectively. Parentheses '()' represents A site while square bracket '[]' represents the B site.

(b) Inverse Spinel Ferrites

In inverse spinel ferrite, M^{3+} ions occupy both A and B sites while M^{2+} ions occupy B sites. Inverse spinel ferrites are expressed as $(M^{3+})A[M^{2+}M^{3+}]BO_4$.

(c) Mixed Spinel Ferrites

In these type of spinel ferrites cations are distributed like the combination of normal and inverse spinel types and hence they are represented as $(M_{1-\delta}^{2+} M_{\delta}^{3+})A[M_{\delta}^{2+} M_{2-\delta}^{3+}]BO_4$ where δ is termed as inversion parameter. The value of δ depends on many factors like synthesis technique, conditions and character of the components of ferrites. For normal spinel and inverse spinel ferrites, the values of δ are 0 and 1respectively, however for mixed spinel ferrites the value of δ varies between 0 to 1.

- **Magnetic Properties of Spinel Ferrites**

As stated earlier ferrites are ferrimagnetic oxides and their magnetic properties are similar to ferromagnetic materials. The magnetic properties of ferrites depend on their domain structure.

Magnetic domain and hysteresis

Magnetic materials consists some magnetically saturated regions called magnetic domains. Weiss has given the concept of domain. Each domain is wholly magnetized itself but the net magnetization of material is zero as there is random arrangement of domains in different directions of material. In presence of external field domains are aligned themselves in the field's direction and hence net magnetization is observed.

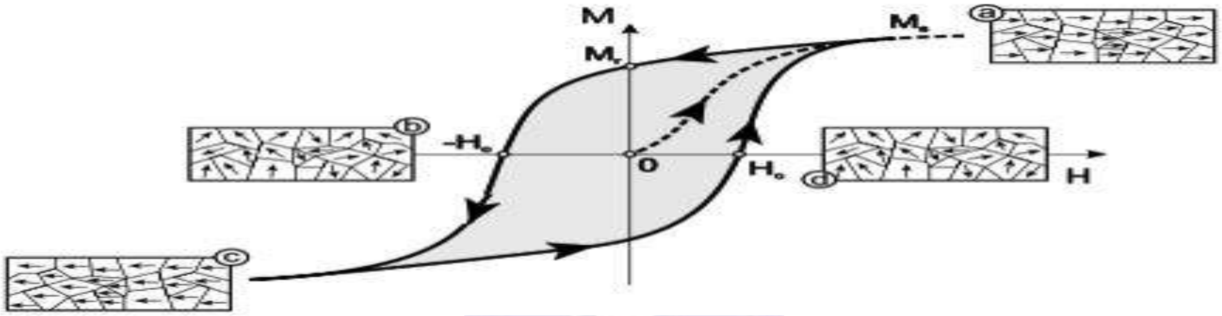

Figure 3: The magnetization 'M' vs magnetic field 'H' curve

Hysteresis in M-H curve is present due to the existence of imperfections in material. These imperfections act as barrier in domain wall motion. When the applied field enhances, then due to the energy of the applied field these obstacles are conquered and the intensity of the magnetization increases gradually. The hysteresis curve with concept of domain is shown in Figure 3. As the saturation magnetization approaches domain rotation needs more energy than applied field and hence, increase in magnetization becomes slower i.e., the slope of the M-H curve decrease. Now when applied field is removed the defects prevent the walls to come again to their original positions. When applied field is reduced to zero, the material retains some magnetization because of the presence of favourably oriented domains. This retained magnetization is called remanent magnetization (Mr). The demagnetization curve does not follow the reverse magnetization curve. Thus, to revisit the domain structure, with zero net moment, an applied field in the reverse direction is needed and is called as coercive field (Hc) [Morish A. H., 1965]. The appearance of hysteresis loop depends on the movement of the domain walls and hence physical and chemical formation of the materials.

Magnetic interactions in ferrites

The interactions among various atoms play important role in governing the magnetic properties of the materials. Two types of interactions (i) magnetic dipolar interactions and (ii) exchange interactions are possible between magnetic ions of magnetic materials. Among these two interactions dipolar interactions are very weak and exchange interactions are dominant interactions [Standley K. J., 1972]. The exchange interactions are again of two types (i) direct and (ii) indirect or superexchange interactions (important one in ferrites). These super exchange interactions are possible via oxygen ions through superexchange process. Three types of super exchange interactions A-A, B-B and A-B are possible in ferrites [Valenzuela R., 1994]. The potency of these interactions depends on the distance and angle between cations (metal ions) and anions (oxygen ions). The interaction is greatest when the interatomic distances are shortest and angle is of 180o . The distances are large and the angles are also very small for A-A and B-B super exchange interactions. In a perfect (undistorted) spinel lattice, the A-O-A angle is about 80o , the B-O-B angles are nearly 90° and 125° while A-O-B angles are about 125° and 154°.

Hence A-B super exchange interaction is strongest and dominant than that of B-B super exchange interaction while the A-A superexchange interaction is weakest and most unfavourable among all three. The pictorial representation of superexchange interaction is shown in Figure 4.

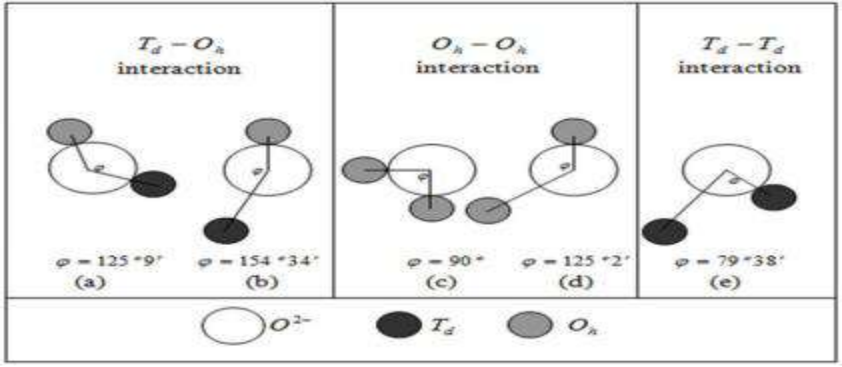

Figure 4: Superexchange interactions of anions, O^{2-} , with tetrahedral (Td) and octahedral (Oh) cations

The spins of A and B sublattices are oppositely aligned and confer the total magnetic moment equivalent to the difference of moments of ions at A and B sites. According to Neel's two sublattice model, the saturation magnetization of spinel ferrite can be written as: $Ms = MB-MA$. However, in actual there is deviation in calculated and observed net magnetization due to several factors such as: (i) the cation distribution on A and B sites is not as ideal as it is predicted, (ii) the direction of the spins at both A and B sites are not completely antiparallel in interactions, rather they are canted. Ferrites also exhibit hysteresis phenomenon and the shape of the hysteresis curves is different for different ferrite. In case of ferrite nanoparticles, they illustrate different coercive field and low saturation magnetization as compared to their bulk equivalent. These changes arise due to surface effects and finite size scaling effects which are effective in the nano region. In nano regime cation distribution is different from its ideal condition and hence existence of different types of interactions leads to different magnetic properties.

- **Electrical Properties of Spinel Ferrites**

In general electrically spinel ferrites fall somewhere between insulators and semiconductors. They possess low dielectric losses and eddy currents and have high electrical resistivity as compared to other ferromagnetic materials. This is a major advantage of spinel ferrites over other magnetic materials.

Conduction mechanism in ferrite

Ferrites are semiconductors and their conductivity is due to the presence of Fe^{2+} and Fe^{3+} ions at octahedral (B) sites. The conduction takes place due to the movement of electrons from Fe^{2+} to Fe^{3+} ions. When divalent metal ions occupy B-sites the conductivity of ferrites is reduced. The conduction mechanism of ferrites and semiconductors are very different from each other. The band type conduction found in ferrites (semiconductors), but each cation according to the site is enclosed by four or six anions (oxygen) and each cation is isolated from other cations. It means the electron associated with a particular cation is mostly isolated and therefore a model for localized electrons is much more suitable than a band model. This behaviour discriminate the ferrites from the semiconductors and directs to the hopping electron model for ferrites. Many models are proposed to explain the electrical behaviour of ferrites and some of the models are discussed below.

Hopping electron model:

The electrons hopping among Fe^{2+} and Fe^{3+} ions of octahedral (B) site are mainly responsible for conduction in ferrites. Due to the sintering condition or electron exchange between divalent metal ion and Fe^{3+} ions ($M^{2+} + Fe^{3+} \rightarrow M^{3+} + Fe^{2+}$), a small amount of Fe^{2+} is present in most of the ferrites [Valenzuela R., 1994]. Both M^{3+} and Fe^{2+} ions prefer to occupy B-sites and hence both participate in conduction mechanism. In equilibrium state the electrons or holes (charge carriers) can not move freely in the crystal. However, the probability of motion of charge carriers is increased due to the lattice vibrations. This type of conductivity is not arisen because of the number of charge carriers in the sample but due to the mobility of charge carriers.

Small polaron model:

In semiconductors, especially most of oxides like ferrites have strong electron-phonon coupling in which Coulomb interaction between lattice ions and conduction electron is found. Though, there is no real phonon present in the lattice but still the electron is constantly enclosed by the cloud of virtual phonons [18]. This cloud of virtual phonons and the electron are engaged together as a composite particle known as polaron [Byrnes S.J.F., 2008]. When the radii of these polarons are greater than the lattice constant of material, polarons are named as large polarons else are called small polarons. Hence, a small polaron is a defect like structure which migrates by a hopping mechanism. Conduction by small polaron formation takes place in materials which have incomplete inner (d or f) shells [Tuller H.L., 1977]. The hopping of small polaron involves both the electron and the virtual phonons cloud. The low value of mobility and temperature independence of Seebeck coefficient of ferrites are explained by small polaron .

Phonon induced tunnelling:

In this process electrons involved in Fe2+→ Fe3++ eexchange process at octahedral sites are firmly tied to the lattice and transfer from one site to the other site of lattice via phonon-induced transfer process known as tunnelling [Srinivasan G., 1981].

Polarization permittivity in ferrite

Ferrite materials demonstrate high dielectric constant with dispersion in the frequency range from 10 Hz to 1 GHz [Rabinkin L.T. 1960]. The decrease in dielectric constant is frequency dependent which is swift at lower frequencies and sluggish at higher frequencies. This dispersion is explained by Maxwell-Wagner (M-W) model based on Koop's theory [KoopsC.G 1951]. According to M-W model in terms of the dielectric, ferrites are made of two layers. The first one is larger layer of ferrite grains and is well-conducting while second shorter layer of grain boundaries is comparatively poor conductor and is found in between the grains. At higher frequencies first layer is effective while at lower frequencies second layer dominates [Koops C.G., 1951]. Ferrites act like dipoles due to the existence of Fe3+ and Fe2+ ions. At low frequency the electrons hopping among Fe3+ ↔ Fe2+ ions occurs in ferrite lattice and charge carrier (electrons) reaches at the grain boundaries. The piling up of electrons at grain boundaries takes place and generates interfacial polarization which in turn ends in high permittivity. At high frequency, the direction of motion of electrons reverse and chances of getting electrons at the grain boundaries reduce which reduces the permittivity. At very high (>MHz) frequency alternating field make no effect on electron exchange and therefore almost no change in permittivity is noticed [Rabinkin L.T1960].

HEXAGONAL FERRITES

Hexagonal ferrites were first invented in 1952 by Went, Rathenau, Gorter& Van Oostershout and then in 1956 by Jonker, Wijn& Braun [Standley K. J., 1972]. They are ferrimagnetic oxides having hexagonal or rhombohedral structure of MFe12O19 formula where M is an element which is mostly Ba (Barium), Pb (Lead) or Sr (Strontium).

Importance of Nickel-Cobalt Ferrites

Cobalt ferrite has been extensively investigated because of its interesting magnetic behaviour, chemical stability and mechanical hardness [Szotek Z., 2006]. Cobalt ferrite, CoFe2O4, crystallizes in a partially inverse spinel structure [Franco A. 2010]. Nano size ferrites have been prepared by various techniques such as sol–gel combustion, modified oxidation process, forced hydrolysis, hydrothermal process, ball milling and the micro-emulsion method [Verde E.L., 2012; Singh R.K., 2012] On the other hand cobalt ferrite is a promising candidate material for biomedical drug delivery [Kambale R.C., 2011], magnetic resonance imaging, biocompatible magnetic nano particles for cancer treatment and magnetooptical devices. Many of these (soft and hard magnetic) properties make them very promising candidates for different applications in electronic (recording technology) and in biomedicines [Shinde T.J., 2012; Bharathi K.K., 2012]. CoFe2O4 and NiFe2O4 both exhibit ferrimagnetism, originating from the magnetic moment of anti-parallel spins between Fe3+ ions at tetrahedral sites and Co2+ or Ni2+ ions at octahedral sites [Bharathi K.K 2009]. The doping of cobalt in nickel ferrites replaces nickel ions in octahedral sites as it has the same inverse spinel structure and affects its magnetic properties [Pant R.P., 2010; Naik S.R. 2012]. The effect of doping of a highly magneto-crystalline anisotropic and magnetostrictive material (cobalt ferrites) in a material with very high resistivity (nickel ferrites) causes the pinning of domain walls [Naik S.R. 2014; Kamble V.B 2014], due to an increase in anisotropy and has an impact on the magnetic permeability as well as the coercivity and saturation magnetization. Ni–Co ferrites are one of the important materials for high-density magnetic recording media because of their moderate magnetic nature, mechanical hardness, and chemical stability.

Magnetic nano particles for Biomedical Applications

Due to the lack of well-known research, using magnetic nanoparticles for biological & medicinal applications remains difficult. Using magnetic, core-shelled Fe3O4 nanoparticles, Buteic et al. 2009 improved colloidal dispersion & controlled particle sizes. The inhibition zone widths for cephalosporins are found to be larger than those for the cephalosporin-nano fluid in an experiment to ascertain the antibacterial capabilities of Fe3O4 coated with cephalosporins. The fact that the nano fluid served exclusively as an antibiotic carrier was a benefit of this approach. Additionally, tiny magnetic nanoparticles made it possible to transport an antibiotic directly to organs like the liver & brain that were being treated. Sun et al. [1972] devised a thermal decomposition procedure to produce Fe3O4 nanoparticles from a mixture of an iron salt, 1,2-hexadecanediol, oleic acid, oleylamine, & biphenyl ether. Silver is then applied to the Fe3O4 nanoparticles to enhance bacterial activity. Precision control of particle size, shape, dispersion, or any external influences that may affect these features is necessary to achieve the specific properties of magnetic nanoparticles needed for biomedical applications. In principle, it is necessary to stabilize the magnetic nano particle dispersion in the aqueous environment. Thus, coating the magnetic nano particles with a polymer shell, including organic (e.g., polyethylene glycol, dextran, chitosan, polyethyleneimine and phospholipids or inorganic (e.g., silica) materials, leads to highly dispersed and high quality nano particles with good biocompatibility.

APPLICATIONS OF FERRITES

Both soft and hard ferrites are extensively used in many applications like transformer core, telecommunication, medical, computer, industrial electronic system and as permanent magnets in loud speakers. Saturation magnetization (MS), coercivity (HC), dielectric constant (ε \ ) and dielectric loss (ε \\ ) are the important parameters to decide quality and applications of ferrites. Some of the novel applications of ferrites are shortly described below

- **INDUCTORS**

Ferrites are widely applicable as many inductive components in electronic circuits like amplifiers, filters, oscillators and impedance matching networks. Ferrite multi layers are used as key technology for integrated devices. The critical elements to fabricate the inductance are a metallic coil with soft ferrite. Nowadays magnetic field sensors use layered samples of ferrites with piezoelectric oxides within a two-step method. The first step is to induce mechanical strain in magnetic material by applying magnetic field and next step is generation of a voltage by transfer of strain in piezoelectric layer. High sensitivity and very less power consumption are few advantages of these sensors.

- **HIGH FREQUENCY**

Ferrite materials are in very high demand for many applications at higher frequency (GHz) like telecommunications and radar. Ferrites can tolerate more penetration of EM fields in comparison to any metals which have very low limits of penetration at high-frequency due to skin effects. At high frequencies, absorption of microwave power takes place by spin dynamics as domain walls cannot follow the field at very high frequency. The spinel ferrites can handle EM frequencies up to 30 GHz while garnets and hexa ferrites work up to 10 GHz and 100 GHz, respectively. In spinel ferrites the losses involved in the absorption of microwaves are associated with defects, electrical conduction and anisotropy field. Ferrites transmit and absorb microwave field in different (opposite) directions of the field. Due to this the same device is used for broadcasting and reception of the signal. Some devices like circulators are used in radar systems and mobile phones and are typically made of garnets.

- **POWER SUPPLIES**

Ferrites are used in power supplies for appliances such as computers, TV, video systems, all types of small and medium electronic appliances. The ferrites are used in switched-mode power supplies in which ferrite transformers are used to increase efficiency by decreasing eddy currents. The ferrite core transformers which work up to 1 MHz are good for low-power applications. Soft spinel ferrites in power electronics show good results at low frequencies.

ELECTROMAGNETIC INTERFERENCE (EMI) SUPPRESSOR

The enhanced use of electronic equipment like computers, digital cameras, scanners, digital notebooks etc increases the chances of distressing each other by EM interference. Electromagnetic interference causes the degradation in performance of an electronic system. To avoid this we need low-pass filters to block signals of frequencies greater than a specified value (noise). Mostly soft ferrites, encapsulated magnetic particles, ferromagnetic metal, hexaferrite composites and carbon nanotube composites are used to construct EMI suppressor. Hexaferrites have high electrical resistivity and high permeability at microwave frequencies and therefore they are most commonly used as EMI suppressor components.

BIOSCIENCES

Ferrites mainly magnetite (Fe3O4), in the form of nanoparticles are used in a numerous of medical applications. Ferrite nanoparticles prepared for medical applications should have superior properties. They should have a specific size range, high chemical purity and should be crystallo-graphically perfect. Nowadays many biotechnological applications based on nanoparticles have been developed. In MRI, superparamagnetic nanoparticles of magnetite (Fe3O4) and cobalt ferrites nanoparticles are used in hyperthermia. Ferrite nanoparticles are also used in many applications like modification, detection and then isolation of biologically active compounds and cells to study and research in biosciences.

MAGNETIC SENSORS, POLLUTION CONTROL AND AS ELECTRODES

Magnetic sensors made of ferrites are used for temperature control, position and rotational angle sensors [5]. Japanese use few installations in which ferrite precursor’s precipitation is used for searching pollutant materials like mercury from wastes. The ferrites formed afterward can be separated magnetically along with the pollutant. Ferrites have high corrosion resistance so they can be used as electrode in applications mainly in chromium plating.

CONCLUSION

Nanoparticles of ferrites are attracting much responsiveness these days to their novel mechanical, electrical & magnetic properties which are much dissimilar from their bulk counterparts, due to the large surface to volume ratio that results from the nano scaling of these materials. Zinc ferrite (ZnFe2O4) is a prominent member of ferrite family with normal spinel structure showing paramagnetic property at room temperature but ferromagnetic behavior below 10K in the bulk form. Nano zinc ferrites have attracted much interest these days to their various properties like thermal and chemical stability, necessity of magnetic properties on particle size, mechanical hardness, high electromagnetic performance, low coercivity& moderate saturation. Apart from this, zinc ferrite also find numerous applications namely as catalyst, MRI contrast agent, gas sensors etc.

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