SYNTHESIS, CHARACTERISATION, SOLID-STATE STUDIES AND APPLICATIONS OF COBALT-NICKEL FERRITES

M.Sc. Dissertation

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DISSERTATION

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DECLARATION

I hereby declare that the matter presented in this dissertation entitled ***'SYNTHESIS, CHARACTERISATION, SOLID-STATE STUDIES AND APPLICATIONS OF COBALT-NICKEL FERRITES '*** was carried out by me during the year 2021-2022 under the guidance of Dr. Kedar Umakant Narvekar. In keeping with the general practice of reporting scientific observations, due acknowledgements have been made wherever the work described is based on the findings of other investigation.

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INTRODUCTION

The term ferrite is generally used to describe a family of magnetic oxide compounds containing iron oxide as the main component [1,2]. Ferrites, in general can adopt three different crystal lattices namely spinel ferrites (cubic), hexaferrites (hexagonal) and garnets (cubic). Each of these structures give rise to magnetic properties which in turn may change with the chemical composition and ordering of the ions in the crystal lattice. The spinel ferrites have a general formula of AB_2O_4 [2]. Spinel ferrites generally contain two cation sites for metal cation occupancy. There are eight A sites where the metal cations are tetrahedrally coordinated by oxygen and sixteen B sites with octahedral coordination. It is important to know that when A sites are occupied by M^{+2} cations and B sites are occupied by Fe^{+3} cations, the ferrite structure is that of normal spinel [3].On the other hand when A sites are occupied by trivalent metal ions i.e., Fe^{+3} and B site possesses both the divalent (M^{2+}) and trivalent ions (Fe^{+3}), the ferrite structure is that of inverse spinel [4]. However, in most spinels the cation distribution possesses an intermediate degree of inversion wherein both sites contain a fraction of the M^{+2} and Fe^{+3} cations [3].

Among all the spinel ferrites, $CoFe_2O_4$ has a partially inverse spinel structure and $NiFe_2O_4$ is a completely inverse spinel [2]. Also, $CoFe_2O_4$ is a well-known hard magnetic material with extraordinary magnetic properties as well as high thermal and chemical stability, whereas $NiFe_2O_4$ is a soft magnetic material with good permeability and high electrochemical stability known. By combining $CoFe_2O_4$ and $NiFe_2O_4$, superparamagnetic nanoparticles of mixed spinel ferrite such as $Ni_{1-x}Co_xFe_2O_4$ or $Co_{1-x}Ni_xFe_2O_4$ can be obtained [5]. Significant advancements have been made by many researchers and scientists in the production of magnetic cobalt-nickel nanoparticles and it was found that the physical properties of cobaltnickel ferrite nanoparticles are influenced by the synthesis process [6].Thus, in general magnetic properties of spinel ferrites are strongly affected by factors such as synthesis method, particle size, composition, microstructure, temperature etc. [7]. Several production methods such as micro-emulsion, coprecipitation, sol-gel and hydrothermal are available to obtain cobalt-nickel ferrite nanoparticles. It is significant to note that magnetic materials in the scale of nanometre show different properties and thus have different applications than that of their bulk-form which stem from effects like finite-size effects and increase in surface area[1]. For applications especially in electronic and electrical devices such as multi-layer chip inductor, a nano-particle powder of the material would be inappropriate and high-density plus fully consolidated bulk material would be required or needed and hence this can be achieved by using a technique such as the Spark Plasma Sintering or SPS technique which is a advanced sintering technique used to consolidate nanoparticle powders into high-density samples [6].

Mixed ferrites such as cobalt-nickel ferrites are a common approach to optimize magnetic properties of materials for particular and specific applications. Since the magneto-crystalline anisotropy of nickel ferrite and cobalt ferrite have opposite signs, one can adjust the magnetic anisotropy of nickel-cobalt ferrites over a wide range by changing the relative amount of cobalt and nickel [8].

Cobalt-Nickel ferrites are important electronic materials which are used in electronic devices suited for high-frequency applications in the field of telecommunication. The excellent electromagnetic properties of these materials make them suitable for size reduction of highfrequency application devices [9]. Cobalt-Nickel ferrite-based supercapacitors are presented as a new idea to overcome the energy-deficit. Cobalt-Nickel ferrite nanoparticles also have numerous applications as gas sensors, recording devices, energy-storage materials, ferrofluids and catalysts [10].

LITERATURE REVIEW

I. EXPERIMENTAL METHODS OF PREPARATION

Several synthesis methods can be used to obtain cobalt-nickel ferrites as reported in the literature including sol-gel, co-precipitation, hydrothermal, mechano-chemical, refluxing, precursor, microwave processing, sol-gel auto-combustion, reverse-microemulsion, glycolthermal method etc. [5,11]. Some of the most commonly used experimental methods of preparation mentioned in the literature are discussed below:

i. Co-precipitation method: Cobalt-Nickel ferrites ($Co_{1-x}Ni_xFe_2O_4$; where the range of x values can be chosen as per the requirement of the analysis) can be synthesized or prepared using the chemical co-precipitation method [3,10]. For synthesis, chlorides of iron, nickel and cobalt can be chosen as the starting materials or precursors [3], or nitrates of cobalt, nickel and iron along with citric acid can be chosen as the starting materials or precursors [10]. Alkaline solution of NaOH can be used as the precipitating agent. This method basically involves weighing the required stoichiometric amounts of the starting materials (chlorides, nitrates etc.), preparing a metal ion solution by dissolving the starting materials in deionized water, dropwise addition of the precipitating agent along with constant stirring (so as to obtain the desired precipitate), filtration of the solution containing the precipitate followed by washing the precipitate several times with water (to remove impurities) and then acetone. The precipitate obtained should then be dried in an electric oven and the dried powder obtained must then be homogenously grounded using mortar and pestle and treated thermally in a muffle furnace so as to obtain the desired cobalt-nickel ferrite nanoparticles [3,10].

ii. Sol-Gel Method: Cobalt-Nickel ferrites can be synthesized or prepared using the solgel method. Cobalt nitrate, nickel nitrate and ferric nitrate can be used as starting materials [5,12,13]. The simplest way of using this method involves using cobalt nitrate, nickel nitrate, ferric nitrate, citric acid and ammonia wherein cobalt nitrate, nickel nitrate, ferric nitrate and citric acid are dissolved in distilled water so as to form a clear solution followed by gradual dropwise addition of ammonia solution so as to balance the pH value and form the gel. After the gel is formed, the solution must be stirred on a hot plate with continuous stirring wherein the temperature should be maintained ~100^{o}C. The gel should then be burned in the form of flame and the desired particles of cobalt-nickel ferrites are finally obtained [12]. Also, in another way cobalt-nickel ferrite particles can be obtained using a polyol-based sol-gel method [5] or a PVA based sol-gel method [13].

iii. Sol-Gel Auto Combustion Method: Cobalt-Nickel ferrites can be synthesized or prepared using the sol-gel auto combustion method. The raw materials used for this synthesis include cobalt nitrate, nickel nitrate, ferric nitrate, citric acid and NH_4OH, all of analytical purity [4,8,14,15]. In this particular method, nitrates act as oxidants and citric acid acts not only as a fuel but also a chelating agent [4]. In this method, appropriate amounts of metal nitrates must be dissolved in minimum amount of double distilled water. The solution must then be placed on a hot plate at a constant heating rate and with continuous stirring till a clear solution is obtained. When the temperature reaches to about 65°C or so, a desired amount of citric acid must be added to the nitrate solution in the molar ratio of citric acid: metal nitrates of 3:1 to provide complete combustion and not to leave any residues of NO_3^- ions remaining. The mixture must then be allowed to cool to room temperature while continuing the stirring process and also the mixed solution should be neutralised to a pH of 6.5 through drop-by-drop addition of liquor ammonia. Such a pH provides full combustion and absence of ammonia or nitrate in the final product. Stirring must be vigorously continued till 100°C where the solution boils and froths due to dehydration and under continuous elimination of water, the solution then transforms into a gel. The dried gel then burns in a self-propagating combustion manner until all of it is completely burnt out. The as prepared sample should firstly be preheated at 500°C for 1 hour and then calcined at 1200°C for 2 hours in order to obtain the final product [4,8,14,15].

iv. Reverse Microemulsion Method: Cobalt-Nickel ferrites can be synthesized or prepared using the reverse microemulsion method. In general, the microemulsion route is defined as the co-precipitation of water in oil system and it has the ability to control the size and morphology of nanoparticles during the synthesis process. In the reverse microemulsion method, numerous water droplets are surrounded by continuous oil phase and a surfactant is placed at the water and oil interface which significantly reduces the tension between these two phases. The chemical reactions between the materials are carried out in the droplets to in turn form ultra-fine particles or rather it can be said that the droplets are used as templates to form the nanoparticles. Reverse microemulsion is a very important method. The size and morphology of the synthesized particles depend directly on the size and shape of the droplets and hence these two factors can be controlled through change in micro emulsion composition such as aqueous phase, organic phase and surfactant ratio. The most important parameter is the surfactant to water proportion. With respect to formation of particles in small droplets, growth of particles and agglomeration is limited. Accordingly, the final particles produced are mono-disperse and nano-scale [1].

v. Glycol-Thermal Method: Cobalt-Nickel ferrites can be synthesized or prepared using the glycol-thermal method. Generally, in this method high purity metal chlorides and ammonium nitrate are used as starting materials. Stoichiometric amounts of the metal chlorides must first be mixed in deionized water followed by careful mixing of the metal solution for about 20 minutes via the use of a magnetic stirrer. Ammonium nitrate solution must then be added dropwise to the chloride mixture under rapid stirring until a pH of ~ 9 is obtained. The precipitate obtained must then be washed a number of times with deionized water until all the chloride ions are removed (which can be confirmed by performing the chloride test using $AgNO_3$). During the washing stage, the precipitate must be generally washed over a Whatman glass microfiber filter. The clean wet precipitate obtained must then be dispersed in 250 mL of ethylene glycol by rapid stirring. The clean precursor obtained should then be transferred into a glass lining in a stainless-steel pressure vessel. The pressure vessel must be heated to 200^{o}C and pressure must gradually be increased to 400 psi. These conditions have to be maintained for six hours. Finally, after cooling to room temperature the product obtained must be filtered and washed using deionized water and finally using ethanol. The final product obtained must be dried under an infrared lamp and finally homogenized using a mortar and pestle [16].

II. CHARACTERIZATION OF THE SAMPLE

The prepared Cobalt-Nickel ferrite samples can be characterized by using several techniques as mentioned in the literature. The X-Ray Diffraction technique (XRD) is used to characterize the structure of the prepared sample. In order to confirm the formation of a phase and check crystallinity of the sample, X-Ray Diffraction technique (XRD) is used [2,3,4,10,12,15,17] The average crystallite size of particles of the prepared sample can be calculated using the Scherrer's formula which is given as d=0.9 l/bcosq where d is the crystallite size, b is the full width of the diffraction line at half maxima (FWHM) measured in radians, l is the X-Ray wavelength and q is the angle (Bragg's angle) [3]. The analysis of the shape and size distribution of the particles of the prepared sample powder is done using a Scanning Electron Microscope (SEM) [3]. The microstructure and elemental composition of the sample can also be examined using a Scanning Electron Microscope (SEM) [2]. Morphological characterization of the prepared sample can be performed by using a Transmission Electron Microscope (TEM) [17]. The Vibrating Sample Magnetometer (VSM) is used to evaluate the room temperature magnetic properties of the prepared ferrite [15]. Electrochemical properties of the prepared sample are revealed using the Cyclic Voltammetry (CV) technique [10]. Morphological studies of the prepared ferrites can also be carried out using Atomic Force Microscopy (AFM) technique [15]. The optical thickness and absorbing properties of the cobalt-nickel ferrites can be analysed using the Vector Network Analyzer (VNA) technique [17]. X-Ray Photoelectron Spectroscopy can be used to determine whether the synthesized Cobalt-Nickel ferrite nanoparticles exist in multiple oxidation states or not [10]. Thermal decompositions of the prepared Cobalt-Nickel ferrite nanoparticles can be studied or analysed in the desired temperature range using Thermogravimetric Analysis (TGA) [2,15]. The chemical bonding structure in the prepared Cobalt-Nickel ferrite samples can be analysed using Fourier Transform Infra-Red (FTIR) technique [7]. Mossbauer Spectroscopy can also be used for characterization of the prepared sample [2]. Elemental ratios of the ions in the prepared sample can be studied quantitatively using Atomic Absorption Spectroscopy (AAS) technique[1].

III. SOLID STATE STUDIES OF COBALT-NICKEL FERRITES

i. Magnetic Studies:

Synthesized Cobalt-Nickel ferrite nanoparticles ($Co_xNi_{1-x}Fe_2O_4$; where $0 \leq x \leq 1$) using the PVA sol-gel method. It was found increase in cobalt concentration yields the monotonic increase of maximum magnetization M_s . The residual magnetization ratio M_I/M_s and coercivities H_c were found to increase in the range of low cobalt concentration and then decrease in the range of high concentration, which could have originated from the variation of anisotropy induced by Co^{+2} ions in the octahedral sites [13].

Synthesized Cobalt-Nickel ferrite nanoparticles ($Ni_{1-x}Co_xFe_2O_4$; where x= 0.6, 0.8, 0.9) using the sol-gel combustion method. The various magnetic properties like saturation magnetization and coercivity were estimated from the hysteresis curve. It was found that the saturation magnetization M_s of the nano-sized cobalt-nickel ferrites increased with increase in grain or particle size. The coercivity values were found to be smaller for the particles calcined at 300°C (smaller crystallite site) and at 900°C (higher crystallite size) when compared to these for particles calcined at 500°C (intermediate crystallite size). Thus, it was found that magnetic behaviour depends strongly on the particle size. Further, it was observed that the M_s and H_c values decreased with increase of cobalt content which could be due to migration of Co^{+2} ions in the B-sub-lattice [15].

Synthesized Cobalt-Nickel ferrite nanoparticles ($Ni_{0.5}Co_{0.5}Fe_2O_4$) using oxide-precursors by mechanical alloying and studied the effect of annealing temperatures on the magnetic properties of the synthesized Cobalt-Nickel ferrites. It was found that magnetic properties of powders were extensively changed by variations of annealing temperatures. Coercivity of annealed powders was reduced due to particle growth and strain recovery of milled particles. Magnetization saturation was found to increase with annealing temperature as a result of relaxation from the non-equilibrium cation distribution and canted spin arrangement formed by mechanical alloying synthesis route [18].

Synthesized Cobalt-Nickel ferrite nanoparticles ($Ni_{0.5}Co_{0.5}Fe_2O_4$) using the microemulsion based method. It was found that the magnetic properties of the synthesized nanorods were largely influenced by the calcination temperatures. It was seen that as the calcination temperature was increased from say 300°C to 1000°C, the saturation magnetization increased from 8.2 to 51.1 emu/g while the coercivity initially showed an increase reaching a maximum value 973.37 Oe at 800°C and then showed a decrease [19].

Synthesized Cobalt-Nickel ferrite nanoparticles(($Ni_{0.5}Co_{0.5}Fe_2O_4$) using the glycol-thermal process. All the samples appeared to exhibit ferrimagnetic behaviour with remanent magnetization and coercive field at room temperature. It was found that magnetic properties of the nano ferrites are directly affected by measuring and annealing temperatures as well as by surface effects and finite sizes [7].

Synthesized Cobalt-Nickel ferrite nanoparticles ($Co_{1-x}Ni_xFe_2O_4$; where x=0, 0.25, 0.5, 0.75) using the auto-combustion method. It was observed that temperature and applied field have a significant effect on the magnetic structure of the particles. Nickel substitution was seen to increase the blocking temperature of the samples which made them suitable for ferro-fluid applications. The temperature dependant magnetic study M(T) curves and squareness value indicated a single domain magnetic ordering. The high coercivity, anisotropy and hysteresis loop strength (BH_{max}) values suggested that these particles show hard magnetic property and the hardness could be tuned with nickel substitution in the material [14].

Synthesized Cobalt-Nickel ferrite nanoparticles ($Co_{1-x}Ni_xFe_2O_4$; where x=0, 0.25, 0.5, 0.75, 1.0) using the reverse microemulsion method. It was observed that the magnetic properties of the nanoparticles were strongly dependant on the morphology, chemical composition and size of the synthesized nanoparticles. The spherical particles were found to show a perfect super magnetic behaviour with zero coercivity while the flaky particles obtained were seen to have a ferromagnetic property which was thought to be due to the presence of pinning factors. It was found that by increasing the size of nanoparticles, the saturation magnetization was found to increase. For spherical nanoparticles, it was seen that saturation magnetization went from 30 emu/gr to 17 emu/gr by decreasing the mean particle size from 10 nm to 3 nm respectively. In the case of flake-shaped nano ferrites, saturation magnetization and coercivity was measured as 7.5 emu/gr and 120 Oe as well as 12 emu/gr and 625 Oe and the variation between these values were thought to be due to different aspect ratio of the particles[1].

ii. Optical Studies

Synthesized nickel substituted cobalt ferrites nanoparticles using the gas phase nucleation and growth process. Diffused reflection spectra of the as prepared nickel substituted cobalt ferrite was recorded to do optical studies using UV-visible absorption spectroscopy. It was observed that band edge absorption values showed a decreasing trend with increase in the nickel substitution in cobalt ferrite. Also, it was seen that as the nickel substitution increased, the absorption spectra showed additional broad humps at around 489, 570, 640 and 757 nm. These humps showed a blue shift with increasing nickel content in cobalt ferrite whereas pure cobalt ferrite sample shows a sharp peak at around 850 nm, the intensity of which showed a decrease with increase in nickel substitution which was thus attributed to the appearance of a different intermediate state and the density of these states increased with increase in the nickel content. Further, the estimated band gap values were found to be around 1.15, 1.28, 1.36, 1.45, 1.50 and 1.62 eV for nickel content corresponding to 0, 0.2, 0.4, 0.6, 0.8 and 1.0 respectively which indicated that the blue shift was a function of nickel which could have been due to the presence of defects and sub-band gap energy level formation in the nanoparticles synthesized as a consequence of nickel substitution [11].

iii. Electrical studies

Synthesized nickel substituted cobalt ferrite nanoparticles using the glycol thermal technique. From the resistivity measurements performed on pellet samples it was observed that pellet samples annealed at 1100^{o}C showed evidence of a semiconducting behaviour. It was also found that there were differences in resistivity determined from each face of the pellet [20].

iv. Dielectric Studies

Synthesized Cobalt-Nickel ferrites using the co-precipitation method. The dielectric as well as the dielectric loss tangent measurements were carried out at room temperature as a function of frequency from 100 Hz to 3 MHz . It was thus found that both the dielectric constant and dielectric loss tangent showed an increase with increase in the cobalt concentration [21].

IV. APPLICATIONS OF COBALT-NICKEL FERRITES

a) Laser Propellant

Laser-matter interaction has opened several new avenues for scientists. Pulsed-layer ablation (PLA) of solids has been one subject of increasing interest due to its number of applications such as film deposition, material processing, cluster and nanostructure production etc. for last two decades. A novel use of laser has been recently reported for the refinement of crystal structure. Laser propulsion is an application of laser ablation and has potential advantages over chemical propulsion mainly due to the fact that the energy source and propellant are both decoupled. The impulse produced by laser ablation is expected to be useful for aerospace applications such as space-debris removal, orbit transfer and launching of small satellites from the ground. Reported $Co_{0.5}Ni_{0.5}Fe_2O_4$ as an efficient laser propellant. The level of thrust quantified via momentum coupling coefficient using single phase $Co_{0.5}Ni_{0.5}Fe_2O_4$ as planar and cavity targets with respect to design was found. Glass layer was used for the confinement of plasma expansion generated by Nd: YAG 532 nm laser. The transparent glass layer lead to plasma confinement and thus a high coupling coefficient was observed. The values without confinement were found to be in the range of $5.747*10^{-5}$ – $7.064*10^{-5}$ N-s/J while those calculated with cavity and confinement were in the range of $1.41*10^{-4}$ – $2.68*10^{-4}$ N-s/J. These values were obtained for laser fluencies of $4*10^{9}$ – $6*10^{9}$ J/m^2 [3].

b) Microwave Absorption Application

Various kinds of ferrites exhibit excellent microwave absorption properties and have been widely employed in military and civil fields as microwave absorbers due to their high absorption rates, broad absorption bands and thin matching thickness. The microwave absorption properties can be evaluated by the Reflection Loss (RL). Took $Ni_{0.8}Co_{0.2}Fe_2O_4$ as the experimental subject and noted that the minimum RL value that could be reached was -36.2 dB with a thickness of 2.5mm at 11.52 GHz which presented a good potential to absorb. It was also found that greater thickness would be better at lower frequencies. Another important phenomenon was the large bandwidth. To conclude, in the range of 9-12 GHz the RL values with the thickness of 1.5-2.5 mm all could reach -10 dB which indicated perfect microwave absorption properties [17].

c) Acetone-Sensing

Functional inorganic materials having specific architectures and acting as gas sensors have attracted significant attention for a highly efficient detection of all gases such as CO_2, SO_2, O_2, NH_3, ethanol, acetone etc which can be applied in many fields of research such as automotive, domestic detector, medical and security detector. Reported a facile strategy to fabricated hierarchical $Ni_xCo_{1-x}Fe_2O_4$ ($0.0 < x < 0.5$) composites via a combination of solvent-evaporation method and one-pot morphology-inherited annealing treatment of Ni^{2+} and Co^{+2} acetate salts and $Fe_4(Fe(CN)_6)_3$ MOF precursors. It was observed that the as-obtained $Ni_xCo_{1-x}Fe_2O_4$ ($0.0 \leq x \leq 0.5$) crystals assembled from numerous nanoparticles can demonstrate could demonstrate significant response signal towards acetone gas in comparison to other gases such as ethanol, methane, ammonia and triethylamine. The as-obtained composition-optimized hierarchical $Ni_{0.1}Co_{0.9}Fe_2O_4$ material featured selective response towards acetone in comparison to other gases along with an excellent reproducibility at working temperature of 240°C. The as-prepared hierarchical $Ni_{0.1}Co_{0.9}Fe_2O_4$ sensor device with a lose interior architecture exhibited a sensing reproducibility even after 14 cycles [22].

CONCLUSION

Amongst the different ferrites, Cobalt-Nickel ferrites having the reverse-spinel structure has attracted considerable attention due to a number of its potential applications in various fields. This mixed ferrite can be prepared using several methods and possesses excellent magnetic properties. Hence, more research in the field of cobalt-nickel ferrites should be done so as to optimize the magnetic properties for particular applications.

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