### Controllable Synthesis and Characterization of Cu-CoZnFe2O4 Nano Ferrites by Auto-combustion Route

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**ABSTRACT**

The methods employed to prepare nanocrystalline mixed ferrite materials during experimentation significantly affect the resulting particle size. The quality of ferrite materials greatly influences the performance of devices. The chemical composition and microstructure of ferrites, which determine their properties, are both determined by the preparation process. The CoCuZnFe2O4 ferrite nanoferites were prepared by sol gel method. The prepared samples were calcinated at 750 oC shows sharper X-ray diffraction peaks, indicating increased crystallinity of the materials. Average crystallite sizes were obtained in the ranges 30.4 - 42.1 nm for the samples calcinated at 450 ℃ and 750 ℃, respectively. Images using scanning electron microscopy (SEM) showed clumps of particles. The overall acidity was reduced for the samples that were calcinated at higher temperatures. The findings highlight how the preparation circumstances affect nanoferrites’ crystallite size, shape, and magnetic characteristics.

**Keywords:** XRD, Nanoferrites, DSC, TGA, SEM

1. **INTRODUCTION**

Nanotechnology-often abbreviated as “nanotech”-is the study of manipulating matter at the atomic and molecular levels. Typically, nanotechnology entails creating materials or electronics that fit inside 100 nanometers or smaller structures in at least one dimension. Nanotechnology encompasses a wide range of disciplines, from inventing novel materials with nanoscale dimensions to examining if we can directly alter matter on the atomic scale. It ranges from extensions of standard device physics to whole new methodologies based upon molecule self-assembly.

Nanotechnology holds the promise of producing a wide variety of novel materials and gadgets with a diverse range of use, including in medicine, energy production, and electronics. On the other hand, nanotechnology brings many of the same problems as when new technology is introduced, such as worries about the economy [1]. Materials scaled down to the nanoscale display different properties from those they do at the macroscale, opening up new applications. For instance, solids that are opaque become translucent (copper), solids that are stable become flammable (aluminium), and solids that are insoluble become soluble (gold). At nanoscales, a substance like gold, chemically inert at larger dimensions, can act as a powerful chemical catalyst. These quantum and surface phenomena that matter demonstrates at the nanoscale are a large part of what attracts people to nanotechnology.

Ferrites, such as spinel ferrite, have a crystal structure with two interstitial sites - tetrahedral and octahedral - that can support a variety of cations. This allows for a wide range of characteristics in ferrites, as other divalent metal ions can be substituted for ferrites. Previous literature has indicated that substituting Al3+ in cobalt ferrite nanoparticles can alter their electromagnetic characteristics, making them a potential candidate for microwave and power device applications. However, replacing Zn in MgFe2O4 can increase resistivity while decreasing magnetic properties. Semiconducting behaviour becomes a trend after Cr3+ substitution. Substituting Cr3+ ions in the nickel-zinc ferrite system can also increase resistivity, making the prepared samples suitable for higher frequency ranging systems. Ferrites can also improve the activation energy and thermal decomposition of propellants and can be easily separated using an external magnet [24]. Various methods such as sol-gel, hydrothermal, and coprecipitation are utilised to synthesise ferrite nanoparticles. This article reports on the effect of cobalt copper zinc ferrite (CoCuZnFe2O4) using a low-cost sol-gel method.

1. **MATERIALS AND METHODS**

All the chemicals used were of analytical grade without further purification and available from commercial sources: cobalt(II)nitrate hexahydrate [Co-(NO3)2.6H2O, 99%], copper(II)nitrate trihydrate [Cu- (NO3)2.3H2O, 95%], zinc(II)nitrate hexahydrate [Zn- (NO3)2.6H2O, 99%], ferric(III)nitrate nonahydrate [Fe- (NO3)3.9H2O, 98%], and sodium hydroxide (NaOH), Glycine. Laboratory-setup triple distilled water was used as a solvent.

Cobalt copper zinc ferrite powder was synthesized by the auto-combustion method. The stoichiometric amounts of nitrates to the fuel are taken as 1:3. Initially, all the nitrates were mixed into the distilled water and fuel is added to the solution. These solutions are mixed using a magnetic stirrer and heated for about 45 min. The solution is continuously stirred till the temperature reaches 80 0C and pH 7 is maintained by using ammonia of suitable quantity, then, the mixed solution is transferred into a heating mantel which is maintained at a temperature of about 100 oC. Upon heating, the solution is turned into dried gel. The dried gel undergoes auto-combustion and forms a foam-like structure. After cooling, the obtained foam is converted into ash, poured into the mortar, and will be grinded to form a fine powder. The obtained fine powder is kept under sintered at 600 oC temperature for about 6 hours using a muffle furnace. Finally, the Cobalt Copper Zinc Ferrites are obtained.

1. **CHARACTERIZATION**

The X-ray diffraction patterns of the synthesized samples were recorded using the Panalytical X-Pert Pro MPD instrument. The samples were analyzed in the 2θ range of 10 - 80º, with a scanning speed of 5º/min and step size of 0.02º. The morphological analysis of the synthesized samples were performed using the FESEM CARL ZEISS instrument. This microscope is equipped with a field emission gun, operating with accelerating voltage variable from 0.5 to 30 kV, with 2 nm resolution. Thermal properties of the prepared samples were carried out using TGA/DTA.

**A. X-Ray Diffraction Studies on as prepared CuZnFe2O4 and CoZnFe2O4 Nano Ferrites**

Structural analysis of the CoZnFe2O4 and CuZnFe2O4 were obtained from powder XRD which is depicted in Figures 1 and 2. A single-phase spinel cubic structure can be seen in the prepared sample according to the XRD peaks (1 1 1), (0 2 2), (1 1 3), (2 2 2), (2 2 4), (3 3 3), and (0 4 4). The secondary hematite phase with a cubic spinel-phase structure is partially forming in the other prepared samples. According to the reported literature, single-phase Co-Zn ferrite nanoparticles that were prepared by the sol-gel process and annealed below 600 oC have been discovered [17, 18]. The standard JCPDS card nos. 52-0277 and 89-0599, which correspond to spinel Co-Zn ferrite and secondary hematite phase, respectively, and the diffraction peaks are in good accord. The peak strength of the secondary hematite phase declined at high cobalt doping concentrations. Using Scherer’s equation [19], the average crystallite size of all the synthesized samples has been determined from the full-width half maximum (FWHM) of the most prominent peak (1 1 3) of the XRD patterns.

where D is the average crystallite size, is the intensity FWHM in radians and is the wavelength of X-ray and is the Bragg’s angle.

It is discovered that cobalt doping results in an increase in crystallite size (D) from 25 to 31 nm. Nearly the same crystallite size (D) was achieved at x=0.5% (28 nm) and x=0.5% (30 nm). Due to this slight dopant fluctuation, other estimated structural parameters at x = 0.03 and 0.09 have values that are nearly identical. The spacing (d) and lattice constant (a) [21] are two structural parameters that are impacted by Co doping and have been determined by using the relation:



**Figure 1: XRD pattern of CoZnFe2O4 Nanoferrite**



**Figure 2: XRD pattern of CuZnFe2O4 Nanoferrite**

**B. FESEM Analysis**

CuZnFe2O4 and CoZnFe2O4 Nano Ferrites were synthesized and dried at 90 0C in a vacuum and calcined at 500 0C for 45 min, respectively, in Figure 3. It is evident that crystal agglomeration occurs as can be seen from Figure 3 (a). It typically results from the formation of smaller crystals. There are several spherical crystals that are substantially smaller or have nanometer dimensions that are below 100 nm. This is consistent with the XRD pattern, which showed the peak broadening for these powder specimens. The grain size was assessed from a SEM micrograph, the crystal size increased (Fig. 3b). This value is consistent with the XRD data results. The extremely narrow XRD peaks show that larger grains are larger than the nanoscale area. Low-resolution micrographs clearly show the impact of heat treatment on the fine-grain size of the specimens (Fig. 3b).

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| **stub 743**  **(a)** |
| **stub 744**  **(b)** |

**Figure 3: FESEM micrograph of (a) CuZnFe2O4 and (b) CoZnFe2O4 sintered at 500 0C.**

**C. DSC and DTG Analysis**

The DSC/TGA curve of CoCuZnFe2O4 nanoferrites is shown in Figure 4. The initial weight loss in the CuZnFe2O4and CoZnFe2O4 nanoferrites was around 10% when the temperature was less than 100 oC. The sample was thermally stable and the weight change was insignificant. Due to the degradation of the polymer chains, there is a weight loss of about 50% in the second phase in the temperature range of 100 oC to 450 oC and a greater weight loss (about 68%) in the temperature range of 500 oC to 800 oC. It was observed that the quantity of CuCoZnFe2O4 nanoferrites contained in the composite affected the breakdown temperature of the nanoferrites [13, 14].



**Figure 4. DSC/TGA Spectra of CoCuZnFe2O4 Nanoferrite**

1. **CONCLUSIONS**

The current work illustrated the structural, thermal and chemical characteristics of prepared CuZnFe2O4 and CoZnFe2O4 nanoferrites. XRD, FE-SEM, and DSC analysis techniques were used to investigate the prepared samples. Samples exhibit a single-phase spinel phase cubic structure according to XRD analysis. CuZnFe2O4 and CoZnFe2O4 nanoferrites create a spinel phase cubic structure with a partial secondary hematite phase (α-Fe2O3) development. Because Co+2 ions have higher ionic radii than Cu+2 ions, the crystallite size (D) increases with Co and Cu doping. FESEM gave information about the crystallinity of the prepared samples. From TGA, the thermal stability was studied.

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