Synthesis and characterization of lead-doped copper ferrite nanoparticles

Síntese e caracterização de nanopartículas de ferritas de cobre dopadas com chumbo

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ABSTRACT
Magnetic nanoparticles of lead-doped copper ferrite, Pb$_x$Cu$_{1-x}$Fe$_2$O$_4$, where $x = 0.0, 0.1$, and $0.2$, were synthesized by the combustion method. The samples were characterized by X-ray diffraction (XRD) to determine crystallographic phases and crystallite sizes. The results showed the presence of trigonal and tetragonal phases for $x = 0$, with crystallite sizes of $37$ nm and $72$ nm, respectively, and for $x = 0.2$, with crystallite sizes of $23$ nm and $21$ nm, respectively. For $x = 0.1$, only the trigonal phase was observed, with a crystallite size of $27$ nm. Scanning electron microscopy (SEM) and Transmission electron microscopy (TEM) showed cubic, hexagonal, and rod-shaped structures for all values of $x$, indicating a polycrystalline material with various spatial orientations. Energy-dispersive X-ray spectroscopy (EDS) identified the chemical elements present in the samples and Mössbauer spectroscopy revealed the formation of spinel-type ferrite. Vibrating sample magnetometry (VSM) displayed typical ferrimagnetic behavior and the Jahn-Teller effect was observed in all samples.

Keywords: Ferrites, Mössbauer Spectrometry, Magnetic Nanoparticles, Jahn-Teller Effect.

RESUMO
Nanopartículas magnéticas de ferrita de cobre dopadas com chumbo, Pb$_x$Cu$_{1-x}$Fe$_2$O$_4$, onde $x = 0.0, 0.1$ e $0.2$, foram sintetizadas pelo método de combustão. As amostras foram caracterizadas por difração de raios X (XRD) para determinar as fases cristalográficas e os tamanhos dos cristalitos. Os resultados mostraram a presença de fases trigonais e tetragonais para $x = 0$, com tamanhos de cristalitos de $37$ nm e $72$ nm, respectivamente, e para $x = 0.2$, com tamanhos de cristalitos de $23$ nm e $21$ nm, respectivamente. Para $x = 0.1$, apenas a fase trigonal foi observada, com um tamanho de cristalito de $27$ nm. A microscopia eletrônica de varredura (SEM) e a microscopia eletrônica de transmissão (TEM) mostraram estruturas cúbicas, hexagonais e em forma de bastão para todos os valores de $x$, indicando um material policristalino com várias orientações espaciais. A espectroscopia de dispersão de energia de raios X (EDS) identificou os elementos químicos presentes nas amostras e a espectroscopia Mössbauer revelou a formação de ferrita do tipo espinélio. A magnetometria de amostra vibrante (VSM) exibiu
comportamiento ferrimagnético típico e o efeito Jahn-Teller foi observado em todas as amostras.

**Palavras-chave:** Ferritas, Espectroscopia Mössbauer, Nanopartículas Magnéticas, Efeito Jahn-Teller.

**RESUMEN**
Nanopartículas magnéticas de ferrita de cobre dopadas con plomo, Pb$_x$Cu$_{1-x}$Fe$_2$O$_4$, donde $x = 0, 0, 1$ y 0,2, fueron sintetizadas por el método de combustión. Las muestras fueron caracterizadas por difracción de rayos X (XRD) para determinar las fases cristalográficas y los tamaños de los cristalitos. Los resultados mostraron la presencia de fases trigonal y tetragonal para $x = 0$, con tamaños de cristalitos de 37 nm y 72 nm, respectivamente, y para $x = 0,2$, con tamaños de cristalitos de 23 nm y 21 nm, respectivamente. Para $x = 0,1$, solo se observó la fase trigonal, con un tamaño de cristalito de 27 nm. La microscopía electrónica de barrido (SEM) y la microscopía electrónica de transmisión (TEM) mostraron estructuras cúbicas, hexagonales y en forma de varilla para todos los valores de $x$, lo que indica un material policristalino con varias orientaciones espaciales. La espectroscopía de rayos X de dispersión de energía (EDS) identificó los elementos químicos presentes en las muestras y la espectroscopía Mössbauer reveló la formación de ferrita tipo espinela. La magnetometría de muestra vibrante (VSM) mostró un comportamiento ferrimagnético típico y se observó el efecto Jahn-Teller en todas las muestras.

**Palabras clave:** Ferritas, Espectometría Mössbauer, Nanopartículas Magnéticas, Efecto Jahn-Teller.

**1 INTRODUCTION**

Currently, nanotechnology emerges as one of the most promising areas within the realm of scientific and technological knowledge. In the context of basic science supporting technological innovations, magnetic nanoparticles, particularly ferrites, stand out as materials with vast potential [1].

Nanostructured materials foment great interest in the scientific community due to their unique physical properties. These materials, characterized by small dimensions, exhibit properties that strongly depend on size and shape, offering materials for various applications [2]. In the class of magnetic nanomaterials, ferrites are a large family of compounds of trivalent iron, oxygen and metal ions. Depending on the crystal
arrangement, they can be categorized into three main forms: spinel ferrites, garnet ferrites, and hexagonal ferrites. Among them, spinel ferrites are the most promising due to their simple and stable lattice structure [3-7]. In some spinel ferrites there is a distortion of the crystal lattice, meaning that the unit cell of the spinel undergoes a slight elongation in one of its edges. This elongation changes the symmetry from cubic to tetragonal in what is called the Jahn-Teller effect [8,9].

The copper spinel ferrite, CuFe2O4, stands out for some notable characteristics. In particular, it exhibits excellent properties for catalysts and chemical sensors, anodes of lithium-ion batteries and microwave absorbers [10,11]. Another important magnetic ferrite is the lead spinel ferrite (PbFe2O4). Though less studied than copper ferrite, it has also been investigated for some applications [5,6]. Nanosized spinel ferrites can be employed in high-density magnetic recording media, ferrofluids, microwave absorbers, and magnetically guided drug carriers. Recently, they have found applications as transducers due to their minimal eddy current losses resulting from high permeability and high resistivity [12-15].

Thus, evidences show that the magnetic properties of ferrites are profoundly influenced by their microstructure. Any effort to control the properties of a product must begin with a good understanding of the fundamental properties of the material, as well as of the manufacturing process that shapes its final microstructure [16,17]. Hence, this study aims to synthesize and characterize some lead-doped copper spinel ferrites contemplating potential applications of these mixed ferrites.

2 MATERIALS AND METHODS

2.1 SYNTHESIS

Lead-doped copper ferrite nanoparticles were synthesized using the solution combustion method, employing iron nitrate, Fe(NO₃)₃·9H₂O, copper nitrate, Cu(NO₃)₂·3H₂O, and lead nitrate, Pb(NO₃)₂, as reactants, and glycine (C₂H₅NO₂) as fuel. The molar ratio between glycine and total nitrates was 1.0. This study focused on
obtaining samples with composition PbxCu1-xFe2O4, where x = 0.0, 0.10, and 0.20. For the synthesis of PbxCu1-xFe2O4, the reactants were dissolved in distilled water at room temperature. Water was used as a homogenization medium.

The solution was heated to 100 °C on a heating plate, leading to evaporation of the water and formation of a highly viscous mass, which, upon ignition, resulted in a nanometric powder. The powder was easily deagglomerated through mechanical maceration in a mortar.

The exposure to high synthesis temperatures promotes the formation of small crystallite sizes and the technique does not require subsequent heat treatment [18]. Figure 1 illustrate the material before, during, and after the combustion process.

Figure 1. Images of the combustion method (a) before, (b) during and (c) after the process

Source: Elaborated by the authors.

2.2 CHARACTERIZATION TECHNIQUES

The XRD technique was used to confirm the presence of the ferrite phase and to determine the average crystallite sizes of the nanoparticles. The diffraction patterns were obtained using a PANalytical X Pert Pro diffractometer with a copper target (λ = 0.154 nm). The parameters employed were as follows: source voltage 40 kV, source current 40 mA, scanning angle from 10° to 90°, angular increment of 0.05°, and counting time of 150 s.

The SEM images were acquired using a FEI Quanta FEG 250 instrument in high vacuum with an acceleration voltage of 10 kV. The TEM images were acquired using a JEOL JEM-2100F instrument. Selected area electron diffraction (SAED) images were
correlated with crystal planes using the DigitalMicrograph software. The mapping of elements by TEM/EDS for the Pb$_{0.1}$Cu$_{0.9}$Fe$_2$O$_4$ and Pb$_{0.2}$Cu$_{0.9}$Fe$_2$O$_4$ ferrites aimed to identify and visualize the spatial distribution of the chemical constituents in the samples, revealing material heterogeneity. The combination of TEM with EDS not only facilitated the identification of elements but also enabled the exploration of interfaces between different phases and the analysis of microstructures.

The $^{57}$Fe Mössbauer spectra were obtained using a $^{57}$Co/Rh source with an activity of 40 mCu. The velocities were calibrated using an $\alpha$-Fe foil which exhibits a single hyperfine field sextet of 33 T at room temperature. The spectrum fitting was performed using the Normos program. Spectra were collected at 300 K and 4.2 K.

The VSM measurements were conducted using a Quantum Design VSM-SQUID MPMS 3 instrument. The experiments were conducted at 300 K with a maximum applied field of 3T. After each measurement, an oscillating field was applied to demagnetize the superconducting coils responsible for generating the external magnetic field.

### 3 RESULTS AND DISCUSSION

#### 3.1 X-RAY DIFFRACTION

The diffractograms shown below were refined using TOPAS-Academic software version 4.1, employing the Rietveld method to compare theoretical data with experimental results. Experimental data are represented by the blue line, while fitted data are represented by the red line and the gray line represents the difference between the two. The theoretical model is based on crystallographic data, using the crystallographic card JCPDS 77-0010 for copper ferrite.

It was observed that the lead-free samples and those with 0.2 lead content contain ferrites with trigonal and tetragonal structures, while the sample with 0.1 lead content contains only ferrite with a trigonal structure (see Table 1). This transformation from cubic to tetragonal and trigonal structures is evidence that the samples are undergoing the Jahn-Teller effect.
Table 1 Parameters obtained from the Rietveld refinement of the ferrite samples.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Ferrite</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>CuFe₂O₄</td>
</tr>
<tr>
<td>GOF</td>
<td>2.82</td>
</tr>
<tr>
<td>Phases</td>
<td>CuFe₂O₄</td>
</tr>
<tr>
<td>Crystal Structure</td>
<td>Trigonal</td>
</tr>
<tr>
<td>Crystallite size (nm)</td>
<td>37.0</td>
</tr>
<tr>
<td>Lattice parameter (nm)</td>
<td>0.303</td>
</tr>
</tbody>
</table>

Source: Elaborated by the authors.

Figure 2. Parameters obtained from the Rietveld refinement of the ferrite samples.

Source: Elaborated by the authors.

Figure 3. X-ray Rietveld refinement of the Pb₀.₁Cu₀.₉Fe₂O₄ sample.

Source: Elaborated by the authors.

Figure 4 X-ray Rietveld refinement of the Pb₀.₂Cu₀.₈Fe₂O₄ sample.

Source: Elaborated by the authors.
3.2 SCANNING ELECTRON MICROSCOPY

The SEM micrographs of the samples are shown in Figure 5. They display particle clusters in a porous framework, a complex and heterogeneous structure. Additionally, they exhibit intercluster pores that suggest the occurrence of gas release during combustion. The observation of intercluster pores is relevant, since they may change the physical and chemical properties of the material, including the surface area. It is also possible to observe that the samples are composed of particles with various shapes. These morphological variations can influence several properties of the material, especially those of a magnetic nature. The shape and orientation of the particles, for instance, can affect the magnetic anisotropy of the material. Additionally, clusters or specific shapes can impact the organization of the particles, affecting magnetic properties such as coercivity [10]. The material response to external magnetic fields can also be influenced by the morphology of particles, resulting in nonlinear responses or anomalous magnetic behavior [19].
Figure 5: (a) SEM micrograph of (a) the CuFe$_2$O$_4$ sample (b) the Pb$_{0.1}$Cu$_{0.9}$Fe$_2$O$_4$ sample (c) the Pb$_{0.2}$Cu$_{0.8}$Fe$_2$O$_4$ sample.

Source: Elaborated by the authors.

3.3 TRANSMISSION ELECTRON MICROSCOPY

Figure 6 shows TEM micrographs of the lead-free sample, CuFe$_2$O$_4$. Figures 6 (a) and (b) reveal a non-uniform distribution of particle sizes, with a predominance of spherical and cubic shapes. The SAED image presented in Figure 6 (c) corroborates the results obtained by X-ray diffraction, displaying the characteristic spots of a crystalline material. These spots were indexed with the crystallographic card JCPDS 77-0010 for copper ferrite.

The agreement between the data obtained by SAED and XRD reinforces the crystalline nature of the material under analysis.

In all samples, XRD data corroborated those of SAED for the following crystallographic planes: (111), (220), (222), (311), (422), (440), and (551).
Figure 6 TEM images of CuFe$_2$O$_4$ at different magnifications (a), (b), with the corresponding SAED image (c);

Source: Elaborated by the authors.
Figure 7 TEM images of Pb$_{0.1}$Cu$_{0.9}$Fe$_2$O$_4$ for different magnifications (a), (b), with the corresponding SAED image (c).

Figure 7 shows TEM micrographs of the Pb$_{0.1}$Cu$_{0.9}$Fe$_2$O$_4$ sample. Figures 7 (a) and (b) depict a non-uniform distribution of particle sizes, displaying spherical, cubic, and hexagonal morphologies, suggesting an influence of lead doping. Figure 7 (c), corresponding to the SAED pattern, complements the results obtained by XRD diffraction, showing characteristic spots of crystalline materials. These spots were identified and associated with the crystallographic card JCPDS 77-0010 for copper ferrite, the same card used to index the peaks in the XRD pattern. These results suggest a well-defined crystalline structure in the sample, highlighting the possible modification of particle morphology due to lead doping. The presence of porous particles may indicate distinct structural characteristics in the sample.
Figure 8 shows TEM micrographs of the Pb$_{0.2}$Cu$_{0.8}$Fe$_2$O$_4$ sample.

![TEM micrographs of Pb$_{0.2}$Cu$_{0.8}$Fe$_2$O$_4$ sample](image)

Source: Elaborated by the authors.

Figures 8 (a) and (b) depict a non-uniform distribution of particle sizes, highlighting a distinct morphology from the lead-free sample and the Pb$_{0.1}$Cu$_{0.9}$Fe$_2$O$_4$ sample. The presence of rod-shaped structures is noted, along with the occurrence of cubic and spherical structures. The SAED image, shown in Figure 8 (c), reinforces the results obtained by XRD, displaying the characteristic spots of a crystalline material.

3.3.1 Mapping of Elements by TEM/EDS of Pb$_{0.1}$Cu$_{0.9}$Fe$_2$O$_4$ and Pb$_{0.2}$Cu$_{0.9}$Fe$_2$O$_4$.

Figures 9 and 10 illustrate the element mapping conducted by TEM/EDS for the Pb-doped ferrites.
The elemental mappings confirm that the ferrite is composed of the elements Pb, Cu, Fe, and O. The concentration of iron, identified by the green color, is the highest, confirming that the ferrite is predominantly composed of iron atoms, which are well distributed. The amounts of lead, represented by the pink color, and copper, represented by the orange color, are lower.

### 3.4 Mössbauer Spectroscopy

Figures 11, 12 and 13, show, respectively, the Mössbauer spectra of CuFe$_2$O$_4$, Cu$_{0.9}$Pb$_{0.1}$Fe$_2$O$_4$, and Cu$_{0.8}$Pb$_{0.2}$Fe$_2$O$_4$. 
Ferrite nanoparticles exhibit Mössbauer spectra consisting of combinations of patterns known as sextets, associated with ferrimagnetism, and doublets associated with superparamagnetism [22].

Figure 11 Mössbauer spectrum of the CuFe$_2$O$_4$ sample at 300 K.

![Mössbauer spectrum](image)

Source: Elaborated by the authors.

Figure 11 presents the Mössbauer spectrum of the CuFe$_2$O$_4$ sample at room temperature. The dots represent experimental data, while the solid lines depict the data fitting performed by the Normos program.

The fitting model includes two magnetic sites and a doublet. The sextet is related to the presence of Fe$^{3+}$ ions in two different sites, A (tetrahedral) and B (octahedral). The isomeric shift values for sites A and B are 0.300 and 0.336 mm/s, respectively.

The quadrupolar splitting values for sites A and B are 0.012 and 0.013 mm/s, respectively. Additionally, a third sextet, represented by the blue line, was observed, which is also present in other samples, but its origin could not be identified.

The spectrum also exhibited a doublet with an isomeric shift of 0.376 mm/s and a quadrupolar splitting of 0.630 mm/s, typical of superparamagnetic behavior, suggesting that the smaller particles in the sample are unblocked.
Figure 12 Mössbauer spectrum of the Pb$_{0.1}$Cu$_{0.9}$Fe$_2$O$_4$ sample at 300 K.

The spectrum of the Cu$_{0.9}$Pb$_{0.1}$Fe$_2$O$_4$ sample is shown in Figure 12. As in the spectrum of the lead-free sample, a doublet with an isomeric shift of 0.49 mm/s and a quadrupolar splitting of 0.61 mm/s was observed, attributed to unblocked particles, suggesting the occurrence of superparamagnetism.

In the case of blocked particles, the presence of two distinct sextets, represented by the pink and red lines, was observed. The intensity of the third sextet, represented by the blue line, is much higher than in the lead-free sample.

The Cu$_{0.8}$Pb$_{0.2}$Fe$_2$O$_4$ sample exhibits a spectrum similar to that of the Cu$_{0.9}$Pb$_{0.1}$Fe$_2$O$_4$ sample, as shown in Figure 13.
Figure 13 Mössbauer spectrum of the Pb$_{0.2}$Cu$_{0.8}$Fe$_2$O$_4$ sample at 300 K.

Source: Elaborated by the authors.

Figures 14, 15, and 16 show the Mössbauer spectra at 4 K for the CuFe$_2$O$_4$, Cu$_{0.9}$Pb$_{0.1}$Fe$_2$O$_4$, and Cu$_{0.8}$Pb$_{0.2}$Fe$_2$O$_4$ samples, respectively.

Figure 14 Mössbauer spectrum of the CuFe$_2$O$_4$ sample at 4 K

Source: Elaborated by the authors.
In the CuFe$_2$O$_4$ spectrum, sextets associated with the presence of Fe$^{3+}$ in sites A and B were identified, but the doublet observed in the 300 K spectrum was not present. This suggests that the blocking temperature of the smaller particles in the sample is above 4 K.

Figure 15 displays the Mössbauer spectrum of the Cu$_{0.9}$Pb$_{0.1}$Fe$_2$O$_4$ sample, revealing the presence of sextets associated with Fe$^{3+}$ ions in two distinct sites. The spectrum also shows the presence of a doublet with an isomeric shift of 0.36 mm/s and a low intensity quadrupolar splitting of 0.67 mm/s, similar to the much more intense doublet observed at 300 K. This observation indicates that, in the case of the ferrite doped with 1% lead, there are particles that remain unblocked at 4 K.

It is important to note that there was a change in the areas corresponding to sites A and B when the ferrite was doped with lead. This change suggests the influence of the Pb dopant on the cationic distribution of Fe ions.

Figure 15 Mössbauer spectrum of the Pb$_{0.1}$Cu$_{0.9}$Fe$_2$O$_4$ sample at 4 K

Source: Elaborated by the authors.

Figure 16 displays the spectrum of the Cu$_{0.8}$Pb$_{0.2}$Fe$_2$O$_4$ sample at 4 K, which is similar to that obtained at 300 K. In this spectrum there is a low intensity doublet with an
isomeric shift of 0.37 mm/s and a quadrupolar splitting of 0.63 mm/s, similar to the much more intense doublet observed at 300 K. This observation indicates that, as in the case of the ferrite doped with 1% lead, there are particles that remain unblocked at 4 K.

Figure 16 Mössbauer spectrum of the Pb_{0.2}Cu_{0.8}Fe_{2}O_{4} sample at 4 K.

![Mössbauer spectrum of the Pb_{0.2}Cu_{0.8}Fe_{2}O_{4} sample at 4 K.](image)

Source: Elaborated by the authors.

3.5 VIBRATING SAMPLE MAGNETOMETRY

A study was conducted to investigate the magnetic behavior of ferrite nanoparticles using VSM. Figure 17 shows the magnetization curves at room temperature for the Pb_{x}Cu_{1-x}Fe_{2}O_{4} sample. It can be observed that these curves exhibit the characteristics of a magnetically soft material, a quality attributed to coercive field values less than 0.1 T. This indicates that the material can be easily magnetized and demagnetized.

In all curves, the presence of a gentle slope in saturation is observed. This slope is associated with the spin canting phenomenon, probably due to the presence of magnetic
disorder in the nanoparticles. In other words, it indicates the lack of complete alignment of all spins in the particles, typical of ferrimagnetic particles [17].

Figure 17 Magnetization curves of the three ferrites

![Magnetization curves](image)

Source: Elaborated by the authors.

Table 1 presents the values of saturation magnetization ($M_s$), coercive field ($H_C$), and remanent magnetization ($M_r$) for the three ferrites. An increase in the saturation magnetization was observed from the lead-free sample to the sample with 0.2% Pb. This increase in saturation magnetization is attributed to the mobility of domain walls, requiring an applied external field energy greater than the magnetic wall pinning energy [23]. Thus, the increase in grain size results in a smaller grain boundary extent, reducing the grain boundary area and favoring the interaction of atomic moments in the direction of the applied magnetic field, as discussed in Ref. [17].

The remanent magnetization decreased from 7.3 emu/g in the lead-free sample to 6.7 emu/g in the sample with 0.2% Pb. According to evidence in the literature, this reduction with lead doping is associated with an increase in porosity [13,16].
A reduction in the coercive field was also observed, from 145 Oe in the lead-free sample to 141 Oe in the sample with 0.2% Pb. This decrease in the coercive field possibly occurred due to the relationship between coercive field and grain size [17,18].

Table 2 Saturation magnetization ($M_s$), remanent magnetization ($M_r$) and coercitivity ($H_c$) of the ferrites

<table>
<thead>
<tr>
<th>Sample</th>
<th>$M_s$ (emu/g)</th>
<th>$M_r$ (emu/g)</th>
<th>$H_c$ (Oe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CuFe$_2$O$_4$</td>
<td>33</td>
<td>7.3</td>
<td>145</td>
</tr>
<tr>
<td>Pb$<em>{0.1}$Cu$</em>{0.9}$Fe$_2$O$_4$</td>
<td>37</td>
<td>6.8</td>
<td>134</td>
</tr>
<tr>
<td>Pb$<em>{0.2}$Cu$</em>{0.8}$Fe$_2$O$_4$</td>
<td>42</td>
<td>6.7</td>
<td>141</td>
</tr>
</tbody>
</table>

Source: Elaborated by the authors.

4 CONCLUSION

The combustion synthesis technique successfully yielded Pb$_x$Cu$_{1-x}$Fe$_2$O$_4$ ($x=0; 0.1; 0.2$) ferrite samples. The ferrites exhibit the inverted spinel configuration, since the Fe$^{3+}$ ions are evenly distributed between tetrahedral and octahedral sites, as shown by Mössbauer spectroscopy. In all samples, it was possible to observe the presence of characteristic sextets and doublets indicative of ferrites with the spinel structure in blocked and unblocked (superparamagnetic) states.

The XRD analysis allowed the verification of the crystalline structure of the ferrites and the occurrence of the Jahn-Teller effect in the samples, evidenced by the change from cubic to non-cubic structure observed in all samples.

Different textures were identified in SEM and TEM images for all samples, in the form of rods, hexagonal and cubic structures.

EDS mapping conducted on TEM images showed a high concentration of lead in rod-shaped structures.

The samples exhibited the magnetic behavior characteristic of soft magnetic materials, a quality attributed due to coercivity values less than 0.1 T. Magnetic characterizations indicated that all samples display ferrimagnetic behavior and that the smaller particles are superparamagnetic at room temperature.
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