Samples Preparation of Ferrite for Absorbing Spectrum of X-Band Waves

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Abstract

Compound of spinel ferrite was prepared in this research in order to investigate and evaluate the microwaves absorption at X-band with the range of frequencies (8-12GHz). Conventional Ceramic techniques (the solid state reaction method) were used to prepare Cu-Zn ferrite micro particles (38 µm), with formula (Cu$_{1-x}$Zn$_x$Fe$_2$O$_4$), at x=0.6 where x is the proportion of zinc in the ferrite, as bulk samples (3 disks) of (3) cm in diameter. The resonance peaks of Cu-Zn ferrite appeared at the frequencies (9, 10, 11.5) GHz. For all samples it was found that the effect of increase the sintering temperature increased the values of attenuation and absorbance, the best values of attenuation coefficient and the absorbance are at 1150°C. X-ray diffraction results showed that the structure was polycrystalline. The density of samples greatly depends on the temperature of sintering, witch increases proportionally with the temperature of the sintering.
تحضير نماذج من الفيرايتيت لامتصاص طيف من موجات النطاق السيني

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الخلاصة

مركبة من فيرايتيت السبينل حضر في هذا البحث لتحقيق وتقسيم الامتصاص للموجات الدقيقة ضمن النطاق X للمدى التردد (12-8 GHz). استخدمت التقنيات السيراميكية (طريقة تفاعل الحالة الصلبة) لتحضير فيرايتيت النحاس- زنك ذات حجم حبيبي (بالصيغة Cu₁ₓZnₓFe₂O₄ (38 µm) (x = 0.6) عند (38 µm) حسب الشكل الثلاثي في الفيرايتيت، بحيث تكون النماذج بشكل أقراس وعدها (3) ذات قطر (3 cm) زنكي عند الترددات (9, 10, 11 GHz). ظهرت القمم الرنينية لفيرايتيت النحاس- زنك عند الترددات (9, 11.5 GHz) بالنسبة لجميع العينات وجد بأن تأثير زيادة درجة حرارة التبلد يؤدي إلى زيادة قيم التوهين والامتصاصية، وتكون أفضل قيم لمعامل التوهين والامتصاصية عند درجة حرارة تبلد 1150ºC. ولقد أظهر نموذج الحيوية السينية أن تركيب العينات متعدد البلورات. أن كثافة العينات تعتمد بشكل كبير على درجة حرارة التبلد، حيث تزداد الكثافة مع زيادة درجة حرارة التبلد.
1. Introduction

The general composition of such ferrite is $\text{MeFe}_2\text{O}_4$, where Me represents the divalent transition metals such as Mn, Zn, Ni, Cu, Fe, Mg, the most popular being (Ni-Zn) ferrite $[^1]$. The spinel crystal structure made up of the closest possible packing of layers of oxygen ions, with the metallic ions fit in at the interstices. The unit cell of the spinel crystal structure is which contains 32 oxygen ions, these have been designated by the large sphere because the anion have a larger atomic radius, there are 16 trivalent ions in B sites and 8 divalent ions in A sites, thus, each unit cell consist of eight ‘sub-dice’ ($8\text{MeFe}_2\text{O}_4$) $[^2]$.

There are three types of the spinel ferrite according to the metallic ion position $[^3]$:

a. Normal spinel ferrite: - for Zn and Cd ferrites, where the eight metallic ions are at the A site, while the 16 trivalent metal ions occupy the B site.

b. Inverse spinel ferrite: - most of the simple ferrites, like $\text{NiFe}_2\text{O}_4$, are of this type, in which trivalent ferric ion Fe$^{+++}$ is at the site A while the remaining trivalent ferric ion and the divalent metallic ion are at the B site.

c. Random spinel ferrite: - which is intermediate case between both type a and b, such as Ni-Zn ferrite and Mn-Zn ferrite.
Electromagnetic waves propagation through a material medium is governed by the intrinsic physical parameters of the medium, i.e. its permittivity, permeability and conductivity. From Maxwell’s equation and the constitutive relations, the travelling electromagnetic waves in the materials can be describe from the following eq’s.\(^4\) -

\[
\nabla \times E = -\frac{\partial B}{\partial t} \quad \text{.......................... (1)}
\]

\[
\nabla \times H = \frac{\partial D}{\partial t} + J \quad \text{.......................... (2)}
\]

Where \(B\) is magnetic flux density, \(E\) is the electric field intensity and \(H\) is the magnetic field intensity.

Which in the phasor form can be written as: -

\[
\nabla \times E = -j \omega B \quad \text{.......................... (3)}
\]

\[
\nabla \times H = j \omega \sigma E + \sigma E \quad \text{...........(4)}
\]

Where, \(D = \varepsilon E\), \(B = \mu H\), \(J = \sigma E\), \(\omega = 2 \pi f\)

Where, \(D\) is the electric displacement, \(J\) is the current density, \(\omega\) is the angular velocity.
The complex permittivity and complex permeability can be defined as:

\[
\varepsilon = \varepsilon' - j\varepsilon'' \quad \text{(5)}
\]

\[
\mu = \mu' - j\mu'' \quad \text{(6)}
\]

Also, equation (3), (4) can be written as:

\[
\nabla^2 E = -\omega^2 \mu \varepsilon E + j\omega D \varepsilon E \quad \text{(7)}
\]

where, \( \varepsilon = \varepsilon_0 \varepsilon_r , \mu = \mu_0 \mu_r \), and \( \varepsilon_r , \mu_r \) are the relative permittivity and the relative permeability, respectively.

Maxwell equation (7) can be written in the form:

\[
\nabla^2 E = \frac{\varepsilon_r \mu_r}{c^2} \cdot \frac{\partial^2 E}{\partial t^2} + \frac{\sigma \mu_r}{c^2 \varepsilon_0} \cdot \frac{\partial E}{\partial t} \quad \text{(8)}
\]

The solution of this equation in x-direction is:

\[
E = E_0 e^{io(\frac{t}{v} - \frac{x}{v})} \quad \text{(9)}
\]

where, \( x \) is the distance, \( v \) is the electromagnetic wave velocity.

By derivation eq. (9) and substituted it in eq.(8), one get:

\[
\frac{c^2}{v^2} = \varepsilon_r, \mu_r - j \frac{\sigma \mu_r}{\omega \varepsilon_0} \quad \text{(10)}
\]
but

\[ N = \frac{c}{v} = n - jk \]  

where, \( n \): the refractive index.

\( k \): the absorption coefficient.

Thus,

\[ 2n k = \frac{\sigma \mu_r}{\omega \varepsilon_o} \]  

\[ n^2 - k^2 = \varepsilon_r \mu_r \]

Where \( \sigma \) is the conductivity.

Equations (12) and (13) represent the relation between the optical parameters and the electromagnetic parameters of the absorber materials.

This research used the electromagnetic parameters \( \varepsilon_r, \mu_r \) to describe the material properties and the interaction nature between microwaves and the magnetic absorber materials.
2. Experimental section

a) Materials

The weight of the used raw materials are accurately calculated from its atomic weights. The raw materials made in England and Germany. For instance, to prepare one mole of \((\text{Cu}_{0.4}\text{Zn}_{0.6}\text{Fe}_2\text{O}_4)\) compound at \(x=0.6\) we calculated the amount of its raw materials as shown \(^{[5]}\):

\[
\begin{align*}
\text{Fe}_2\text{O}_3 &= (2*55.8)+(3*16)=159.6 \text{ gm} \\
\text{ZnCo}_3 &= 65.4+12+(3*16) =125.4 \text{ gm} \\
\text{CuO} &= 63.5+16=79.5 \text{ gm} \\
\text{Total} &= 0.4*79.5+0.6*125.4+159.6=266.6 \text{ gm}
\end{align*}
\]

b) Microwave Absorber measurements: This paper used the waveguide method for measuring the attenuation. In waveguide, one can measure the reflection coefficient by measuring the Voltage Standing Wave Ratio \((V_{\text{SWR}})\) in transmission line.

Where it is received by the detector associated with a scale of \(V_{\text{SWR}}\), then to calculate the attenuation coefficient in \((\text{dB})\) unit, which is equal to :-
Atten.Coeff. = 20 log|Γ| \hspace{1cm} (14)

Where, Λ  The reflection coefficient which is equal to :-

\[ |Γ| = \frac{V_{SWR} - 1}{V_{SWR} + 1} \] \hspace{1cm} (15)

After calculating the reflection coefficient of eq. (15) can be adjusted to obtain absorbance and reflectivity of the following equation \[^6\] :-

\[ Γ^2 + A^2 = 1 \] \hspace{1cm} (16)

Where, A The absorption coefficient.

Equation (16) shows that there are two parameters only are the absorption coefficient and the reflection coefficient, where the transmission coefficient is equal to zero because there is a short-circuit.

c) **Measuring the density:** The density measurement of the prepared samples has been measured for \((\text{Cu}_{0.4}\text{Zn}_{0.6}\text{Fe}_2\text{O}_4)\) after a process of sintering in which the completely dry samples have been weighted, and the size of the sample which is a disk with diameter \((D=3\text{cm})\) and thickness \((t=3\text{mm})\) is measured by using micrometer.
The radius of the sample is R (R=D/2), and its size is V (V=πR²t); then the density of the sample ρ is (ρ=m/V), where the mass (m) is measured by the unit of g and V is measured by the unit of cm³ [7].

**d) X-ray Diffraction Tests:** To investigate the crystal structure of the prepared samples after calcination (sintering), phase analysis was done by X-ray diffraction (XRD) using Cu-Kα radiation, and wavelength λ = 1.54060 Å; the range of the Braggs angles are taken (2θ=20° - 60°) for the samples. By using Braggs law [8]:

\[
2d \sin \theta = n \lambda \ldots \ldots (17)
\]

the interplaner distance (d) can be measured, and then comparing the resultant X-ray patterens with international standard (ICDD) International Centre for Diffraction Data which is the American Standard for Testing Materials (ASTM).

**3. Results and Discussions:** The absorbance tests of the Cu-Zn ferrite samples have been carried out for x =6 at the X-band range (8-12 GHz). The samples were sintered at 1050°C, 1100°C, and 1150°C. This paper calculated the V_{SWR}, Γ, Atten. Coeff., Γ², and A² and recorded all the values in the tables, as for the graphs which include curves of attenuation coefficient and
absorbance curves as a function of frequency at three sintering temperatures and as follows:

It is noticed from the following tables (1), (2) and (3) that the values of $V_{SWR}$ vary with frequency, and then all the parameters have been changed such as reflection coefficient, attenuation coefficient, reflectivity and absorbance, due to the absorption of ferrite of the waves depends on the frequency.

Also it is noticed that the decrease in the value of $V_{SWR}$, reduces the reflection coefficient, increases the attenuation coefficient (negative value), reduces the values of reflectivity and increases in the values of absorbance, this means that the best desired results when the values of $V_{SWR}$ are at the minimum value, and all the values of parameters in tables depended upon the values of sintering temperature of the samples.

The figures (1) and (2) and the tables show the appearance of peaks at frequencies (9, 10, 11.5) GHz at 1050°C, while at 1100°C and 1150°C the peaks overlap to become one peak due to the matching between the values of the relative permeability and the relative permittivity values at frequency (11.5) GHz. The highest values of the attenuation coefficient at 1150°C are (-15.92, -12.40) dB at frequencies (9, 10) GHz respectively and at 1100°C is (-14.43) dB at frequency (11.5) GHz. While the corresponding absorbance values at 1150°C are (97.4%, 94.2%)
respectively and at 1100°C is (96.4%), the good bandwidth of the absorption at all frequencies except the frequency (10.5) GHz at 1150°C, any limits (3.5) GHz of X-band, as at 1100°C the good bandwidth is (1.5) GHz of the frequencies (8 - 9.5) GHz and (0.5) GHz of the frequencies (11.5-12), any limits (2) GHz of X-band.

Table (1): Listed the Parameters of $\text{Cu}_{0.4} \text{Zn}_{0.6} \text{Fe}_2\text{O}_4$ samples with thickness (3 mm) at 1050 °C

<table>
<thead>
<tr>
<th>Freq. (GHz)</th>
<th>$V_{\text{swr}}$</th>
<th>$\Gamma$</th>
<th>Atten. Coeff. (dB)</th>
<th>$\Gamma^2$</th>
<th>$\Lambda^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>2.40</td>
<td>0.41</td>
<td>-7.74</td>
<td>0.168</td>
<td>0.832</td>
</tr>
<tr>
<td>8.5</td>
<td>1.66</td>
<td>0.25</td>
<td>-12.04</td>
<td>0.063</td>
<td>0.937</td>
</tr>
<tr>
<td>9</td>
<td>1.53</td>
<td>0.21</td>
<td>-13.56</td>
<td>0.044</td>
<td>0.956</td>
</tr>
<tr>
<td>9.5</td>
<td>3.33</td>
<td>0.54</td>
<td>-5.35</td>
<td>0.292</td>
<td>0.708</td>
</tr>
<tr>
<td>10</td>
<td>2.42</td>
<td>0.42</td>
<td>-7.54</td>
<td>0.176</td>
<td>0.824</td>
</tr>
<tr>
<td>10.5</td>
<td>4.96</td>
<td>0.66</td>
<td>-3.61</td>
<td>0.436</td>
<td>0.564</td>
</tr>
<tr>
<td>11</td>
<td>1.88</td>
<td>0.31</td>
<td>-10.17</td>
<td>0.096</td>
<td>0.904</td>
</tr>
<tr>
<td>11.5</td>
<td>1.70</td>
<td>0.26</td>
<td>-11.70</td>
<td>0.068</td>
<td>0.932</td>
</tr>
<tr>
<td>12</td>
<td>1.85</td>
<td>0.30</td>
<td>-10.46</td>
<td>0.090</td>
<td>0.910</td>
</tr>
</tbody>
</table>
Table (2): Listed the Parameters of Cu$_{0.4}$ Zn$_{0.6}$ Fe$_2$O$_4$ samples with thickness (3 mm) at 1100 °C

<table>
<thead>
<tr>
<th>Freq. (GHz)</th>
<th>V$_{swr}$</th>
<th>Γ</th>
<th>Atten. Coeff. (dB)</th>
<th>Γ$^2$</th>
<th>A$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>1.92</td>
<td>0.32</td>
<td>-9.90</td>
<td>0.102</td>
<td>0.898</td>
</tr>
<tr>
<td>8.5</td>
<td>1.58</td>
<td>0.23</td>
<td>-12.77</td>
<td>0.053</td>
<td>0.947</td>
</tr>
<tr>
<td>9</td>
<td>1.41</td>
<td>0.17</td>
<td>-15.39</td>
<td>0.029</td>
<td>0.971</td>
</tr>
<tr>
<td>9.5</td>
<td>1.50</td>
<td>0.20</td>
<td>-13.98</td>
<td>0.040</td>
<td>0.960</td>
</tr>
<tr>
<td>10</td>
<td>2.23</td>
<td>0.38</td>
<td>-8.40</td>
<td>0.144</td>
<td>0.856</td>
</tr>
<tr>
<td>10.5</td>
<td>3.17</td>
<td>0.52</td>
<td>-5.68</td>
<td>0.270</td>
<td>0.730</td>
</tr>
<tr>
<td>11</td>
<td>2.15</td>
<td>0.37</td>
<td>-8.64</td>
<td>0.137</td>
<td>0.863</td>
</tr>
<tr>
<td>11.5</td>
<td>1.47</td>
<td>0.19</td>
<td>-14.43</td>
<td>0.036</td>
<td>0.964</td>
</tr>
<tr>
<td>12</td>
<td>1.55</td>
<td>0.22</td>
<td>-13.15</td>
<td>0.048</td>
<td>0.952</td>
</tr>
</tbody>
</table>

Table (3): Listed the Parameters of Cu$_{0.4}$ Zn$_{0.6}$ Fe$_2$O$_4$ samples with thickness (3 mm) at 1150 °C

<table>
<thead>
<tr>
<th>Freq. (GHz)</th>
<th>V$_{swr}$</th>
<th>Γ</th>
<th>Atten. Coeff. (dB)</th>
<th>Γ$^2$</th>
<th>A$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>2.17</td>
<td>0.37</td>
<td>-8.64</td>
<td>0.137</td>
<td>0.863</td>
</tr>
<tr>
<td>8.5</td>
<td>1.55</td>
<td>0.22</td>
<td>-13.15</td>
<td>0.048</td>
<td>0.952</td>
</tr>
<tr>
<td>9</td>
<td>1.38</td>
<td>0.16</td>
<td>-15.92</td>
<td>0.026</td>
<td>0.974</td>
</tr>
<tr>
<td>9.5</td>
<td>1.41</td>
<td>0.17</td>
<td>-15.39</td>
<td>0.029</td>
<td>0.971</td>
</tr>
<tr>
<td>10</td>
<td>1.63</td>
<td>0.24</td>
<td>-12.40</td>
<td>0.058</td>
<td>0.942</td>
</tr>
<tr>
<td>10.5</td>
<td>4.00</td>
<td>0.60</td>
<td>-4.44</td>
<td>0.360</td>
<td>0.640</td>
</tr>
<tr>
<td>11</td>
<td>1.57</td>
<td>0.22</td>
<td>-13.15</td>
<td>0.048</td>
<td>0.952</td>
</tr>
<tr>
<td>11.5</td>
<td>1.50</td>
<td>0.20</td>
<td>-13.98</td>
<td>0.040</td>
<td>0.960</td>
</tr>
<tr>
<td>12</td>
<td>1.65</td>
<td>0.25</td>
<td>-12.04</td>
<td>0.063</td>
<td>0.937</td>
</tr>
</tbody>
</table>
Fig. (1) Listed the attenuation coefficient curves as a function of frequency for Cu$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ samples.

Fig. (2) Listed the absorbance curves as a function of frequency for Cu$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ samples.
Table (4) shows the practical density which has been obtained for samples of the copper-zinc ferrite at x=0.6 and at different sintering temperatures.

As is stated in the international literature, the known real density of the copper-zinc ferrite is approximately 5.3 g/cm$^3$. In addition, the thermal treatment of the samples and the temperature of the sintering affect the measured density, which increases proportionally with the temperature of the sintering [9], in other words, the density of samples greatly depends on the temperature of sintering [10].

Table (4) listed the measured density of samples

<table>
<thead>
<tr>
<th>Type of Ferrite</th>
<th>Sintering Temp. °C</th>
<th>$\rho$ g/cm$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu$<em>{0.4}$ Zn$</em>{0.6}$ Fe$_2$O$_4$</td>
<td>1150</td>
<td>5.18</td>
</tr>
<tr>
<td>Cu$<em>{0.4}$ Zn$</em>{0.6}$ Fe$_2$O$_4$</td>
<td>1100</td>
<td>4.84</td>
</tr>
<tr>
<td>Cu$<em>{0.4}$ Zn$</em>{0.6}$ Fe$_2$O$_4$</td>
<td>1050</td>
<td>4.54</td>
</tr>
</tbody>
</table>
The XRD results shown in figure (3) demonstrate the completion of the $(\text{Cu}_{0.4} \text{Zn}_{0.6} \text{Fe}_2\text{O}_4)$ phase of the spinel structure at this temperature, obviously it is a polycrystalline. The pattern exhibits the Bragg reflection at the diffraction angles $(2\theta^\circ)$ which give interplaner distances $(d)$, which matched perfectly with the international standard (ASTM) as shown in Table (5). The peak analysis shows clear resemblance with the XRD pattern of $\text{Cu}_{0.4}\text{Zn}_{0.6}\text{Fe}_2\text{O}_4$ prepared by different ways\cite{11}. No external peak was detected in the prepared sample.

![Fig. (3) X-ray pattern of $\text{Cu}_{0.4}\text{Zn}_{0.6}\text{Fe}_2\text{O}_4$](image)

*Fig. (3) X-ray pattern of $\text{Cu}_{0.4}\text{Zn}_{0.6}\text{Fe}_2\text{O}_4$*

*Table (5) Listed the interplaner distances $(d)$ and $(2\theta^\circ)$ of X-ray pattern of $\text{Cu}_{0.4}\text{Zn}_{0.6}\text{Fe}_2\text{O}_4$ comparing with the ASTM card.*
4. Conclusions:

Throughout the absorption study of $\text{Cu}_{0.4} \text{Zn}_{0.6} \text{Fe}_2\text{O}_4$ samples the following conclusions are reached to:

1. The best value of sintering temperatures is at 1150°C this indicates that spinel ferrite needs to sintering temperature of more than 1100°C for the complete formation of ferrite and getting best absorbance because of high temperature sintering cancels all secondary phases that made up with ferrite.
2. The density of samples greatly depends on the temperature of sintering, witch increases proportionally with the temperature of the sintering.
3. From the results of the structure testing by XRD of the prepared samples one can find that the structure is polycrystalline.

5. References:


