Dielectric Properties of Mixed Ferrite (Mn$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ and Co$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$) Composites

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Abstract

Soft ferrite (Mn$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$) and hard ferrite (Co$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$) nanocomposites (x Mn$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ + (1-x) Co$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ where x= 0.25, 0.5, 0.75) were prepared with different weight percent. Crystal structures and microstructures of these composites have been investigated by using X-ray diffraction and scanning electron microscope for composite samples sintered at 900°C/30 min using microwave sintering technique. The effect of composite weight percent on dielectric properties of composites are examined over a wide frequency range (1MHz-1.8GHz) by studying the real and imaginary part of permittivity with available theories. Investigation show variation of mixed ferrites for resistivity and resonating frequency.

Keywords: Ferrites, Microwave sintering, Dielectric properties.

INTRODUCTION

Ferrimagnetic materials allows the fine tuning of the absorbing characteristics which are often limited by operating frequency, curie temperature, anisotropy of materials, operating parameters of ferrites as reported by several researchers [1-3]. Auzans et al. [4] have studied properties of Mn–Zn ferrite nanoparticles used for ionic and surfaced ferrofluids with different degrees of zinc substitution prepared by co-precipitation method. Chandana Rath et al. [5] have reported the dependence of cation distribution of particle size, lattice parameter and magnetic properties in nanosize Mn–Zn ferrite for different degrees of Zn substitution prepared by hydrothermal precipitation method. The ferrites such as Mn–Zn ferrites and Co–Zn ferrites contain a high quantity of nonmagnetic zinc ions which lowers the Curie temperature and moderates the thermo stability used for various applications such as transformers, spintronics etc [6-8]. Knowledge of electrical properties such as resistivity, dielectric constant and dielectric loss is important for processing of elements useful for electronic circuits since ferrites play useful role in many areas [6].

The microwave hydrothermal synthesis yields low particle size with higher densification [9]. Microwave sintering technique prevents low loss during sintering processing with improvement of properties [10]. Hence, for present work, nanopowders of Mn$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ [11] and Co$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ [12] are synthesized using microwave hydrothermal system. Mn-Zn ferrites is considered of magnetic nature with high permeability with low losses [13,15]. Zinc in cobalt ferrite promises to be important class for studies in elastic properties useful for transducers applications [14]. D.c. resistivity is measured under room temperature conditions using Kethley ammeter. The dielectric properties such as real part of permittivity ($\varepsilon'$) and imaginary part of permittivity ($\varepsilon''$) are measured for wide frequency range on the sintered composite samples and obtained results have been discussed. The present work is an attempt to understand the effect of mixing the ferrite-ferrite composite on real part of permittivity and imaginary part of permittivity up to frequency range of 1.8 GHz.

EXPERIMENTAL WORK

The nanopowders of Mn$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ [10] and Co$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ [11] are synthesized using microwave hydrothermal system as reported elsewhere. The synthesized nanopowders of Mn$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ and Co$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ are mixed at different weight percentage (%) to prepare the composites of x Mn$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ + (1-x) Co$_{0.4}$Zn$_{0.6}$Fe$_2$O$_4$ where 0.25, 0.5, 0.75 and then and named as sample A (x=0.75), sample B (x=0.50) and sample C (x=0.25). The samples are thoroughly mixed with agate mortar and then mixed with 2wt% polyvinyl alcohol as a binder. Then the powders are uniaxially pressed at a pressure of 1500 kg/cm$^2$ to form pellet (~1 mm in thickness, ~10 mm diameter) specimens. The samples are sintered by using microwave furnace at 900°C for 30 minutes. The d.c. resistivity ($\rho$) of the present samples, at room temperature is measured using a two probe method. The complex permittivity ($\varepsilon^*$) of the composite samples was measured over a wide frequency range of 1MHz to 1.8 GHz using Agilent 4291B Impedance analyzer.
In the present investigation, phase identification of the sintered samples are performed by the powder X-ray diffraction with a Philips (Panalytical) X-ray diffractometer with Cu Kα radiation (λ = 1.5406 Å). The ferrites are compared with the standard ICDD data for Mn_{0.4}Zn_{0.6}Fe_{2}O_{4} (74-2400) and CAS number for cobalt ferrite (22-1086) and zinc ferrite (89-1012). The XRD pattern suggests that the samples are having spinel phase of ferrite without any impurities or secondary phases even after mixing the two different ferrites compositions and sintering at high temperatures. The lattice parameter for the microwave sintered samples is calculated and tabulated in table 1. The results suggests that the lattice parameter increases (8.347 Å to 8.388 Å) with increase in the manganese content in the composite ferrite. Manganese has higher atomic radius when compared with cobalt or iron which are located in the octahedral sites and zinc radius which is present in the tetrahedral sites. It can be observed from the Table 1 that a low porosity value of 5% is obtained for the sample microwave sintered samples at 900°C/30 min. The grain size is estimated around 75 nm to 97 nm estimated from SEM (Fig.2) as shown in Table1. The increase in grain size may be due to increase of manganese ions which is having higher ionic radius.

![Figure 1](image1.png)

**Figure 1.** X ray diffraction pattern for composite ferrite samples.

![Composite Sample A](image2a.png) ![Composite Sample B](image2b.png) ![Composite Sample C](image2c.png)

**Figure 2.** Sintered SEM Pictures for the composite samples
D.c resistivity values of the microwave sintered sample are presented in the table 1. It can be observed that the Sample A has highest resistivity than the other two samples. This may be due to presence of less manganese ions and more number of cobalt ions. The low resistivity in the sample B and sample C is due to reduction of Mn$^{2+}$ content. The Fe$^{2+}$, Co$^{2+}$ ions show strong preference to octahedral sites i.e., B-sites. Since Co$^{2+}$ ions present with Fe$^{3+}$, it is possible that some of these ions can be oxidized to Co$^{3+}$ ions during the sintering [6,16]. The conduction mechanism in ferrites can be understood on the basis of hopping of electrons and understood using Verwey theory [17-18]. Microwave sintering due to rapid processing technique gives little time for material loss. D.c. resistivity depends on the micro structural depends on the properties such as grain size, porosity etc. Smaller grains shows greater surface to volume ratio by which higher oxidation and reconversion of ions takes place. The smaller grains effect in greater number of grain boundaries, which further acts as acts as scattering center for the flow of electrons leading to rise of resistivity [10, 19]. Many researchers have found the behavior of resistivity dependent on sintering conditions, temperature along with density of the samples [20-21].

<table>
<thead>
<tr>
<th>Sample</th>
<th>D.C. Resistivity (ρ&lt;sub&gt;d.c&lt;/sub&gt;) x10$^8$Ω.m</th>
<th>X ray density (ρ&lt;sub&gt;xray&lt;/sub&gt;) gm/cm$^3$</th>
<th>Bulk Density ρ&lt;sub&gt;bulk&lt;/sub&gt; gm/cm$^3$</th>
<th>Lattice parameter(a) A$^o$</th>
<th>Porosity (%)</th>
<th>Grain size from SEM (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>10.68</td>
<td>5.437</td>
<td>5.157</td>
<td>8.347</td>
<td>5.15</td>
<td>75</td>
</tr>
<tr>
<td>B</td>
<td>9.75</td>
<td>5.384</td>
<td>5.062</td>
<td>8.370</td>
<td>5.86</td>
<td>90</td>
</tr>
<tr>
<td>C</td>
<td>8.48</td>
<td>4.975</td>
<td>4.714</td>
<td>8.588</td>
<td>5.32</td>
<td>97</td>
</tr>
</tbody>
</table>

The frequency dependence of real part of permittivity (ε') and imaginary part of permittivity (ε") for microwave sintered composite ferrites is shown in Fig.3 and Fig.4. It can be seen the variation of dielectric constant decreases with increasing in frequency. It can be seen from Table 1 that the value of ε' for microwave sintered composite ferrites is 7 (sample A) at 1 MHz and remains constant up to 900 MHz. With further increase of frequency, the value of ε' also found to increase and shows a dispersion peak above 1.5 GHz. The lower value of dielectric constant [22] in sample A is due to reduction of interfacial polarization which occurs in smaller grains which acts high resistances grain boundaries regions. With increase in manganese content, resistance decreases which results in increases dielectric constant. With increase in grain size, due to interfacial polarization there is rise in dielectric constant as shown in table 1.
CONCLUSIONS

Mixed composite of Mn$_{0.6}$Zn$_{0.4}$Fe$_2$O$_4$ and Co$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ ferrite are prepared by adding different weight percentages and submitted for microwave sintering method. It is found that the value of real part of permittivity (ε’) and imaginary part of permittivity (ε”) remained low and constant for wide frequency range with good resistivity in the samples. The results indicate that ferrite-ferrite composite may be used for frequency operated devices.

REFERENCES: