DIELECTRIC BEHAVIOR OF Ni$_{0.1}$Zn$_{0.9}$Fe$_2$O$_4$-POLYPROPYLENE COMPOSITES AT LOW MICROWAVE FREQUENCIES

Tan Foo Khoon$^{1,*}$, Jumiah Hassan$^1$, Nurhidayaty Mokhtar$^1$, Mansor Hashim$^1$ and Nor Azowa Ibrahim$^2$

$^1$Department of Physics, Faculty of Science, University Putra Malaysia, 43400 Serdang, Selangor.

$^2$Department of Chemistry, Faculty of Science, University Putra Malaysia, 43400 Serdang, Selangor.

*Corresponding author: fookhoon1202@hotmail.com

ABSTRACT

In the last decade, studies and research toward polymer-clay composites draw significant attention for a suitable filler that can improve mechanical, thermal, electrical, optical and pharmaceutical properties as compared with pure polymer. Ni$_{0.1}$Zn$_{0.9}$Fe$_2$O$_4$ (NZF) was prepared using conventional solid-state method. A two-phase composite was fabricated with blend filled Ni$_{0.1}$Zn$_{0.9}$Fe$_2$O$_4$ added to isotactic polypropylene matrix. The samples were characterized by XRD and dielectric measurements were done using Agilent 4291B Impedance/Material Analyzer. It was observed that the composition of 30 wt% NZF gave the highest dielectric constant in the frequency range of 1 MHz to 1.8 GHz at room temperature.

Keywords: Nickel-zinc ferrite; solid-state; polypropylene; dielectric properties;

INTRODUCTION

Ferrites which have many applications at microwave frequencies are very good dielectric material. The useful characteristics of ferrites such as electrical resistivity, low dielectric loss, chemical stability and others played a vital roles in both domestic and industry sector [1, 2]. Dielectric properties of ferrites are dependent on several factors including the method of preparation, sintering temperature, sintering time and chemical composition [3].

Polypropylene (PP) has very attractive solid-state properties such as high modulus and tensile strength, rigidity, excellent heat resistance [4]. Besides, PP can be easily tailored for variation of performance characterization [5]. Therefore, the development of composites combining mineral and polymer is definitely one of the most interesting perspectives for isotactic PP [6].
EXPERIMENTAL DETAILS

NZF was prepared using conventional solid-state reaction method. The starting materials, NiO (99.7% purity), ZnO (99.9% purity) and Fe₂O₃ (99.7% purity) were weighed according to the stoichiometric equation (1) below:

\[ 0.1(\text{NiO}) + 0.9(\text{ZnO}) + \text{Fe}_2\text{O}_3 \rightarrow \text{Ni}_{0.1}\text{Zn}_{0.9}\text{Fe}_2\text{O}_4 \] (1)

The powders were dry milled and pre-calcined in air at 1100 °C for 10 hours. They were grinded and sieved using a 45 micron siever to ensure homogeneity particle sizes of the powders. These powders were finally calcined in air at 1250 °C for 10 hours and the grinding and sieving process repeated to ensure homogeneous particle sizes of the powders when mixing with PP.

PP and NZF were weighed according to the percentage (5% - 30%) of NZF added. PP was heated and melted to a molten state at 160 °C. The desired amount of NZF was added to PP and blend for 10 minutes. The composite was removed from the Brabender and hot pressed to approximately 1 mm thickness.

All the samples were examined by XRD to determine the microstructure and purity of the samples. The dielectric properties of the samples were determined using the Agilent 4291B Impedance/Material Analyzer.

RESULT AND DISCUSSION

Figure 1 showed the XRD patterns for NZF–PP composites at different compositions. The composite matched with 00-050-2397 and 00-008-0234 of the ICDD database that showed polypropylene and nickel zinc iron oxide respectively. NZF (filler) is in crystalline structure while PP is a semicrystalline structure [7]. Thus, there are lots of small peaks obtained throughout the angle. The samples were pure because there were no unwanted peaks shown in the XRD pattern.

Figure 2 showed the dielectric constant of NZF-PP composites from 1 MHz to 1.8 GHz. It can be easily seen that with increasing of NZF content, the dielectric constant increases. According to effective medium theory, a higher dielectric constant of the polymer-based composite can be obtained by adding high dielectric constant filler into the low dielectric constant polymer matrices as shown in Figure 2 [8]. Hence, it can be concluded that NZF with higher dielectric constant can be added to improve the dielectric constant of PP. However, the addition of NZF compensates the dipole moment of PP. As a result, the amount of NZF added in 5 wt% NZF composite is not significant and the dielectric constant is lower than pure PP. The composition of 30 wt% NZF added gave the highest value of the dielectric constant which is 2.66 at 100 MHz. This may be due to the Fe²⁺ and Fe³⁺ of the n-type and the hole exchange between Ni³⁺ and Ni²⁺ of the p-type ferrites resulting in local displacements of electrons or hole in the direction of electric field which may cause polarization [1].
Figure 1: XRD patterns for Ni$_{0.1}$Zn$_{0.9}$Fe$_2$O$_4$ – Polypropylene composites at different compositions of NZF

Figure 2: Variations of the dielectric constant with respect to frequency for Ni$_{0.1}$Zn$_{0.9}$Fe$_2$O$_4$-Polypropylene composites
Figure 3 showed the dielectric loss factor of $\text{Ni}_{0.1}\text{Zn}_{0.9}\text{Fe}_2\text{O}_4$-Polypropylene composites at different compositions in the frequency range 1 MHz to 1.8 GHz. The dielectric loss factor decreases with increasing frequency. When the frequency approaches 1.8 GHz, the dielectric loss factor increases due to the higher frequency which may cause the electron to experience an unstable polarizing field and slowly move to its original position to be polarized.

![Graph showing dielectric loss factor vs frequency](image)

**Figure 3**: Variations of the dielectric loss factor with respect to frequency for $\text{Ni}_{0.1}\text{Zn}_{0.9}\text{Fe}_2\text{O}_4$-Polypropylene composites

Table 1 and Figure 4 showed the variations of the dielectric constant at different compositions of NZF-PP composites at 10 MHz, 100 MHz and 1 GHz. The dielectric constant increases with increasing NZF content. The dielectric constant at 100 MHz is stable and the highest compared to 10 MHz and 1 GHz. The dielectric constant at 10 MHz region is dispersed and not stable. This may be due to the changing priority of interfacial polarization to dipolar polarization at higher frequencies. Dielectric constant at 1 GHz has less ability to accumulate the mobile charges at the grain boundary resulting lower dielectric constant compared to 100 MHz.

Table 1 and Figure 5 showed the variation of the dielectric loss factor at different compositions of NZF-PP composites at 10 MHz, 100 MHz and 1 GHz. The dielectric loss factor at 10 MHz is higher compared to 100 MHz and 1 GHz. This phenomenon may due to the mobile charges at low frequency are not stable and the movement of mobile charges caused dielectric loss.
Table 1: Variations of the dielectric constant and loss factor with respect to composition at 10 MHz, 100 MHz and 1 GHz for Ni0.1Zn0.9Fe2O4-Polypropylene composites

<table>
<thead>
<tr>
<th>wt% NZF</th>
<th>10 MHz</th>
<th>100 MHz</th>
<th>1 GHz</th>
<th>10 MHz</th>
<th>100 MHz</th>
<th>1 GHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP</td>
<td>2.39</td>
<td>2.34</td>
<td>2.38</td>
<td>0.1042</td>
<td>0.0067</td>
<td>0.0014</td>
</tr>
<tr>
<td>5</td>
<td>2.23</td>
<td>2.32</td>
<td>2.30</td>
<td>0.0506</td>
<td>0.0052</td>
<td>0.0009</td>
</tr>
<tr>
<td>10</td>
<td>2.24</td>
<td>2.37</td>
<td>2.37</td>
<td>0.0407</td>
<td>0.0070</td>
<td>0.0023</td>
</tr>
<tr>
<td>15</td>
<td>2.37</td>
<td>2.42</td>
<td>2.40</td>
<td>0.0061</td>
<td>0.0045</td>
<td>0.0012</td>
</tr>
<tr>
<td>20</td>
<td>2.45</td>
<td>2.46</td>
<td>2.44</td>
<td>0.0155</td>
<td>0.0040</td>
<td>0.0021</td>
</tr>
<tr>
<td>25</td>
<td>2.58</td>
<td>2.57</td>
<td>2.55</td>
<td>0.0114</td>
<td>0.0099</td>
<td>0.0024</td>
</tr>
<tr>
<td>30</td>
<td>2.61</td>
<td>2.66</td>
<td>2.64</td>
<td>0.0069</td>
<td>0.0040</td>
<td>0.0050</td>
</tr>
</tbody>
</table>

Figure 4: Variations of dielectric constant with respect to composition for Ni0.1Zn0.9Fe2O4-Polypropylene composites
**CONCLUSION**

NZF-PP composites with high purity were successfully prepared. According to the results obtained, Ni$_{0.1}$Zn$_{0.9}$Fe$_2$O$_4$–Polypropylene composite with composition 30 wt% NZF showed the highest dielectric constant of 2.66 at 100 MHz. Addition of NZF more than 10 wt% resulted in a significant dielectric behavior improvement toward PP-based composites.

**ACKNOWLEDGEMENT**

The authors acknowledge the Graduate Research Fellowship given to the graduate student (Tan Foo Khoon) and the Physics and Chemistry department, Faculty of Science, UPM.

**REFERENCES**


