Investigations on Optical and Electrical Conductivity of \( \text{Ba/Ni/Zn/Fe}_{16}\text{O}_{27} \) Ferrite Nanoparticles

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Abstract: The Barium w-type ferrite powder with the composition formula \( \text{BaNi}_{2-x}\text{Zn}_{x}\text{Fe}_{16}\text{O}_{27} \), with \( x=0.0, 0.4, 0.8, 1.2, 1.6, \) and 2) were synthesized by using the ceramic method. The X-ray analysis confirms the formation of a single-phase w-type hexagonal structure. The lattice constant increases slightly with increasing the zinc concentration. UV-VIS analysis of the sample was carried out. The band gaps can be obtained at different concentrations from the zinc and have band gaps in the semiconducting region. The optical constants of absorption coefficient, extinction coefficient, refractive index, and reflection showed systematic variation with Zn concentration. The optical and electrical conductivity were be observed increases with photon energy, and the conductivity peak of the samples increases with a decrease in Zn content. The electric susceptibility decreases with an increase in the Zn concentration.

Keywords: nano barium ferrite; band gaps; optical conductivity; ceramic method.

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1. Introduction

Investigation of ferrite nanoparticles recently became much more interesting due to improving their electrical and magnetic properties and their excellent physical properties. The w-type hexa-ferrites have been widely used in various fields of science, industrial and technological applications, including humidity sensors, recording disks, microwave devices, catalysts, Information storage, magnetic resonance imaging, ceramic coatings for solar cells, optoelectronic devices, and electronic devices. [1,2]. The optical properties of the dielectric materials generally became of interest because of their sensible transmission within the optical part of the spectrum as compared with different categories of materials. So that in order to study the optical properties of the ferrite materials, the optical constants such as absorption coefficient, transmission, reflection, and index of refraction are vital parameters to explain the optical properties of the ferrite materials. [3,4]. The reportable by Ram et al. about the \( \text{Zn}_2\text{W} \) ferrites was different by substituting \( \text{Zn}^{2+} \) ions, which prefer tetrahedral positions or inhibit grain growth and the magnetic properties tailored with careful addition [5]. Nakamura et al. reported that the \( \text{Co}_2\text{W} \) had very low permeability, with a rise in permeability and a reduction in FMR frequency, within the zinc-substituted Ba \( (\text{Co}_{1-x}\text{Zn}_x)\text{Fe}_{16}\text{O}_{27} \) [6]. Rani et al. reported a decrease in the saturation magnetization when increasing the zinc concentration for the cobalt zinc ferrite composition synthesis by the ceramic method [7]. The general behavior for the nickel ferrites prefers to
occupy the octahedral sites while the zinc prefers to occupy the tetrahedral site. The interactions between the tetrahedral ions and octahedral sites will alter the electrical and magnetic properties of ferrites. The octahedral sites are occupied by Ni$^{2+}$ and Fe$^{3+}$, whereas the tetrahedral sites are occupied by Zn$^{2+}$ and Fe$^{3+}$. The properties of the ferrite are also strongly dependent on the synthesis method, sintering temperature, chemical composition, etc. [8–10]. The knowledge of types of bandgap energy, direct or indirect structure, is very important for deciding what material can be used in a device requiring light output. Thus, in the present study, we synthesize the BaNi$_{2-x}$Zn$_x$Fe$_{16}$O$_{27}$ ($x = 0, 0.4, 0.8, 1.2, 1.6,$ and 2) composition ferrite via the ceramic method and investigate the influence of Zn addition on their structure and optical behavior and bandgap energy for the synthesis ferrite.

2. Materials and Methods

The synthesis of the zinc substituted barium-nickel ferrite (BaNi$_{2-x}$Zn$_x$Fe$_{16}$O$_{27}$ with $x=0.0, 0.4, 0.8, 1.2, 1.6,$ and 2) was done using the Ceramic method according to their molecular formula. The initial ingredients BaCo, ZnO, NiO, Fe$_2$O$_3$, with very high purity, were weighed and mixed in the correct Stoichiometric ratio and grounded for six hours using an agate mortar. The resulting mixture was air-dried and pre-sintered at 1270K in the air for 10 hours. Temperature is controlled by a sensor thermocouple within the furnace. Then the ferrites were grounded again for three hours. After that, the granulated powder was then pressed into pellets with the help of a hydraulic press. The pellets were finally sintered at 1470K for six hours and then slowly cooled to room temperature. Finally, the samples were grounded to fine powder. After that, the characterization of samples was performed using different analytical methods. The X-ray diffraction (XRD) patterns of the samples were recorded using Cu Kα ($\lambda = 1.54$ Å) in the 2θ range of 5° to 75° at a step size of 0.02°/s using an X-ray diffraction (XRD) instrument model Shimadzu EDX-720. The optical properties of this sample were investigated by using a UV-Visible absorption spectrophotometer (Shimadzu PC 1650 model) and fluorescence spectra taken from a Spectro fluorophotometer (Shimadzu RF 5301 model).

3. Results and Discussion

3.1. XRD analysis.

Figure 1 shows the XRD pattern of BaNi$_{2-x}$Zn$_x$Fe$_{16}$O$_{27}$ hexaferrite. The phase compositions of the sample's powders are made up entirely of W-type hexaferrite without any anther impurity phases detected. In other words, this figure only shows a single phase of W-type hexagonal ferrite. The samples were also found to be in good agreement, confirmed with the BaNi2 W-type hexaferrite structure of p63/mmc. The standard powder diffraction data of JCPDS,54-0097 is compatible with Zn doped BaNi2-xZnxFe16O27. The lattice parameters of hexagonal structures are computed using the relation (1) [11].

$$\frac{1}{d_{hk\ell}^2} = \frac{4}{3} \frac{h^2 + hk + k^2}{a^2} + \frac{l^2}{c^2}$$

(1)

where $d_{hk\ell}$ is determined by using relation (1).
Figure 1. X-ray diffraction patterns for BaNiZnFe₁₆O₂₇.

Figure 2. The relation between lattice parameters (a, c) and Zn²⁺ concentration for BaNiZnFe₁₆O₂₇.

Figure 2 shows the fluctuation of the lattice parameters a and c, as well as their ratio c/a, versus Zn concentration. Both lattice constants rise with increasing Zinck concentration.
which can be explained by the fact that Zn$^{2+}$ has a larger ionic radius (0.074 nm) than Ni$^{2+}$ (0.069 nm). As a result, when the ionic radius of substituted Zn$^{2+}$ is greater than (0.074 nm), which is larger than of Ni$^{2+}$ ion (0.068 nm).

3.2. Optical measurements.

All samples were subjected to optical tests at room temperature. The spectral optical transmission of BaNi$_{2-x}$Zn$_x$Fe$_{16}$O$_{27}$ (x = 0.0, 0.4, 0.8, 1.2, 1.6, and 2) was measured using UV-visible absorption spectra as shown in Figure 3a at longer wavelengths ($\lambda > 400$ nm), all samples were transparent, and no light was scattered or absorbed in the non-absorbing zone (R+T=1). Absorption causes the inequality (R+T<1) at short wavelengths ($\lambda < 400$ nm), which is known as the absorbing zone.

![Optical measurements graph](image)

Figure 3. Plot of (a) Transmittances & Reflectance; (b) Absorbance; (c) Extinction coefficient; (d) Absorption Coefficient, versus wavelength spectra of BaNi$_{2-x}$Zn$_x$Fe$_{16}$O$_{27}$ ferrites.

The optical transmittance edge for the samples likewise shifts slightly to the lower wavelength, indicating that the energy gap will be slightly expanded with Zn concentration. It's also worth noting that, with the exception of around 350 nm, transmittance rises with increasing wavelength in all cases between 200 and 400 nm. The transmittance of all samples increases as the wavelength increases in the 300-400 nm wavelength range, then saturates at higher wavelengths. This material has a high absorbance at 350 nm wavelengths, making it suitable for use in reagents or solar cells. The visible and infrared parts of the spectrum have high transmittance, while the ultraviolet area has poor transmittance. As shown in Figure 3b, as wavelength increases, absorbance falls in all situations, so the absorbance was highest at 334 nm in the visible range and 234 nm in the ultraviolet zone.
Furthermore, when the Zn content increases, the optical density decreases. With increased Zn content, the absorption edge shifts to the short wavelength side. And it gets smaller as the wavelength gets longer. This explains why a photon incident cannot irritate an electron and move it from the valence band to the conduction band. Its energy is insufficient to bridge the energy gap. The visible and infrared parts of the spectrum have high transmittance, while the ultraviolet area has poor transmittance. On the other hand, in Figure 3, the reflectance of the spectrum as a function of wavelength is shown, and it is evident that the absorbance spectrum behaves oppositely as the transmittance spectrum.

The nanocomposite's reflectance versus wavelength in the range of 200 nm–800 nm is shown in Figure 3a. It can be seen that when the amount of Zn in the composite increases, the reflectance decreases, but the reflectance vs. wavelength profile remains unchanged. This indicates that the absorption in the visible and near-IR ranges is low.

3.2.1. Absorption coefficient (α).

By using the flowing equation, the absorption coefficient (α) has been computed [12],

\[ \alpha = 2.303A/t \]  

where (t) is the sample thickness and A is the absorbance. The largest absorption coefficient emerges in two places in Figure 1d, the first in the violet range at (\(\lambda=334\)nm), which has a 30 percent absorption ratio. Direct electronic transitions are expected to occur at those wavelengths (high photon energy). The second was made in the ultraviolet range, which has a 70 percent absorption ratio at (\(\lambda=270\) nm). The figure demonstrates that the value of the absorption coefficient decreases as the incident wavelength increases for all ferrite samples up to a particular wavelength that varies depending on the ferrite sample, at that wavelength (320-300 nm) in the visible area \(\alpha\) almost becomes invariant. The absorption coefficient values decline in the UV region as the wavelength (\(\lambda<220\)nm) decreases and the values decrease. This is owing to the Zn element's absorption properties.

3.2.2. Extinction coefficient (K).

The extinction coefficient is a unique property of a substance's composition that indicates how well it absorbs light of a specific wavelength. The fraction of light lost due to scattering and absorption per unit average distance is used to calculate it. Also, the extinction coefficient (k), which can be calculated using equation (3) [13], is a measurement of the fraction of light lost through scattering and absorption per unit distance.

\[ K = \alpha\lambda/4\pi \]  

In Figure 3c, the sample's extinction coefficient has been demonstrated to behave similarly to the absorption coefficient's behavior as a function of wavelength. It gets smaller as the wavelength gets longer. The extinction coefficient value falls as the zinc level rises. The extinction coefficient indicates the percentage of light lost in this location owing to scattering and absorption. In addition, the nature of the curve (k) and the curved absorption coefficient were shown to be identical.

3.2.3. Optical bandgap.

For the designing of optoelectronic devices, the optical band gap is another important parameter required. The optical band gap was calculated from the optical absorption coefficient \(\alpha\) by using the following relation [14]:

\[ E_g = \frac{\hbar c}{\lambda} \alpha \]
\[
\alpha = \frac{A(h \nu - Eg)^n}{h \nu}
\]  
(4)

where A is the constant depending on the probability transition. \( Eg \) refers to the optical band gap, \( h \nu \) the photon energy, and \( n \) is the constant equal to 1/2 or 2 for allowed direct or indirect transition and 3 or 3/2, for forbidden indirect or direct transition probability. From the Tauc plots \((\alpha h \nu)^2 \) vs. \((h \nu)\) graph, the optical energy band gaps \( (E_g) \) were calculated. \((\alpha h \nu)^{1/2}\) against \((h \nu)\) curves are plotted and shown in Figure 4. to evaluate the value of the indirect bandgap of produced samples. The S-shape of the \((\alpha h \nu)^{1/2}\) against \((h \nu)\) curves is owing to the presence of two separate slopes and indicates that phonons play a role in the absorption process [15,16]. The plots of the prepared samples are shown in Figure 4. The value of indirect bandgap energy \( (E_g) \). Obtained from the extrapolation of a straight line at \((\alpha h \nu)^{1/2}=0\) for all the synthesized samples.

\[\text{Figure 4. The plot of bandgap versus photon energy spectra of BaNi}_{2-x}\text{Zn}_x\text{Fe}_{16}\text{O}_{27} \text{ferrites.}\]

Figure 4 depicts the plots of the prepared samples. The indirect bandgap energy value \( (E_g) \). Extrapolation of a straight line at \((\alpha h \nu)^{1/2}=0\) for all the variables samples that have been synthesized the predicted indirect band energy values for \(\text{BaNi}_{2-x}\text{Zn}_x\text{Fe}_{16}\text{O}_{27} \) (\(x=0.0, 0.4, 0.8, 1.2, 1.6,\) and 2) are listed in Table 1. The results demonstrate that as the concentration of \( \text{Zn}^{2+} \) increases, the band gap widens (Figure 4), which could be due to quantum confinement phenomena [17-19]. It has been discovered that the size of the crystallites can influence \( E_g \), with smaller crystallites resulting in more discrete energy levels and hence higher \( E_g \) [20].

\[\text{Table 1. The optical band gap of BaNi}_{2-x}\text{Zn}_x\text{Fe}_{16}\text{O}_{27} \text{ferrites nanocomposites.}\]

| Sample         | Zn Concentration | Bandgap (eV) |
|----------------|------------------|--------------|
| \(\text{BaNi}_2\text{Fe}_{16}\text{O}_{27}\) | 0                | 2.89 eV      |
| \(\text{BaNi}_{1.6}\text{Zn}_{0.4}\text{Fe}_{16}\text{O}_{27}\) | 0.4             | 2.9 eV       |
| \(\text{BaNi}_{1.2}\text{Zn}_{0.8}\text{Fe}_{16}\text{O}_{27}\) | 0.8             | 2.89 eV      |
| \(\text{BaNi}_{0.8}\text{Zn}_{1.2}\text{Fe}_{16}\text{O}_{27}\) | 1.2             | 2.94 eV      |
| \(\text{BaNi}_{0.4}\text{Zn}_{1.6}\text{Fe}_{16}\text{O}_{27}\) | 1.6             | 2.95 eV      |
| \(\text{BaZn}_2\text{Fe}_{16}\text{O}_{27}\) | 2                | 2.958 eV     |

All \( \text{Zn} \)-doped samples \((x = 0.0, 0.4, 0.8, 1.2, 1.6, 2)\) have a blue shift, indicating that they may have visible–light photoactivity. In contrast to the general properties of light, the optical energy of the bandgap grows with particle size. All \( \text{Zn} \)-doped samples \((x = 0.0, 0.4, 0.8, 1.2, 1.6, 2)\) have a blue shift, indicating that they may have visible–light photoactivity. In contrast to the usual features of quantum bonding, the optical energy of the bandgap grows...
with particle size. ONOCHIE, Uche Paul, *et al.* [21] discovered that doping Zn\(^{2+}\) causes an increase in bandgap as the Zn concentration rises.

3.2.4. Refractive index (n).

The refractive index of optical materials must be evaluated for applications in integrated optical devices, and the refractive index of the material is also the fundamental parameter for device design. On the other hand, due to its relationship with the electronic polarization of ions and the local field inside the material, the refractive index (n) is considered one of the most important features of optical materials. The refractive index is calculated using the formula (5) [22]:

\[
n = \left( \frac{1+R}{1-R} \right) + \left( \frac{4 \times R}{(1-R)^2 - k^2} \right)^{1/2}
\]  

(5)

where R is the reflectance and k the extinction coefficients.

![Figure 5. The plot of refractive index versus photon energy spectra of BaNi\(_2\)\(_x\)Zn\(_x\)Fe\(_{16}\)O\(_{27}\).](image)

For samples with varying Zn: Graph 5. It's worth noting that the refractive index grows as the wavelength increases but begins to fall in the visible area. In general, it can be demonstrated that as the number of Zn nanoparticles increases, the refractive index decreases.

For a more detailed description, the refractive index increases with increasing wavelength up to 334 nm and then drops with increasing wavelength. As the amount of Zn in the solution grows, the curve's form remains constant. With the increase in photonic energy and the increase in Zn content for the sample, the extinction coefficient values and refractive index generally show an increasing trend. The use of photonic energy This spectrum and dopant dependence of optical constants (n, k) will be important in establishing the materials' suitability for usage in optical data storage devices.

3.2.5. Optical conductivity.

Optical conductivity is an excellent technique for investigating matter's atomic structure. It connects the current density to the material's exposure to different wavelengths of light in the electric field and is highly dependent on the material's absorption coefficient and refractive index. Eq. (6) [23]. is used to explore the optical conductivity of BaNi\(_2\)-xZn\(_x\)Fe\(_{16}\)O\(_{27}\) (x = 0.0, 0.4, 0.8, 1.2, 1.6, and 2) ferrite at various light photon energies with varying amounts of Zn.

\[
\sigma_{opt} = \frac{\alpha nc}{4\pi}
\]  

(6)
where $\alpha$ is the absorption coefficient, $n$ is the refractive index, and $c$ is the light velocity. In Figure 6, the optical conductivity of BaNi$_{2-x}$Zn$_x$Fe$_{16}$O$_{27}$ ($x = 0.0, 0.4, 0.8, 1.2, 1.6,$ and 2) ferrite varies with input photon energy. It can be seen that the optical conductivity increases with light energy irradiation and that at 5 eV, the optical conductivity grows fast until it reaches a maximum of around 5.5 eV. Its value exceeds the previous value by more than 20 times.

The rise in absorption coefficient in the photon energy range between 3.2 and 5.58 eV can be attributed to an increase in optical conductivity in this range. The discrete wavelengths at the two regions are responsible for the existence of two photoconductivity peaks.

Two peaks of photoconductivity are attributed to the discreet wavelengths in the two regions. This material appears to be more absorbed at a critical wavelength in the visible region and UV region. The conductivity peak of the samples increases with a decrease in Zn content. Also, it is observed that the optical conductivity directly depends on the absorption coefficient.

On another hand, the samples exhibited the value of $\sigma_{opt}$ in the range from $8.8 \times 10^9$ to $1 \times 10^{11}$ s$^{-1}$ over the ultraviolet region and in the range of $9.1 \times 10^8$ to $8.8 \times 10^9$ s$^{-1}$ above the visible light range, confirming that these specimens maintain good photosensitivity. The results of the optical conductivity for the samples were in agreement with the literature [23-25]. The electrical conductivity has been calculated by using the relation [26].
\[ \sigma_e = \frac{2\lambda \sigma_{\text{opt}}}{\alpha} \]  

(7)

Figure 7 presents the electric conductivity with the wavelength with an increase in the Zn concentration, the electrical conductivity decreased. However, with increasing, the wavelength decreased to reach 400nm after that started increasing.

Electrical susceptibility indicates the degree of polarization of dielectric material in response to an applied electric field. The greater the electric susceptibility, the greater the ability of a material to polarize in response to the field, thereby reducing the total electric field inside the material. Electrical susceptibility (\(\chi\)) has been calculated from the optical constants by using the following relation \[27\]:

\[ \varepsilon_r = \varepsilon_0 + 4\pi\chi_c = n^2 + K^2 \]

\[ \chi_c = \frac{n^2 - k^2 - \varepsilon_0}{4\pi} \]

where \(\varepsilon_0\) is the dielectric constant in the absence of any contribution from free carriers.

Figure 8. The plot of Electrical conductivity versus wavelength of BaNi$_{2-x}$Zn$_x$Fe$_{16}$O$_{27}$ ferrites.

The value of electrical susceptibility is 0.0961 at \(\lambda=800\) nm. Figure 8 presents the electric susceptibility (\(\chi\)) with the wavelength with an increase in the Zn concentration; the value of electrical susceptibility decreased from 2.8 to 0.86.

4. Conclusions

The ceramic method has been used to synthesize the (BaNi$_2$-xZn$_x$Fe$_{16}$O$_{27}$ with x value 0.0, 0.4, 0.8, 1.2, 1.6, and 2) ferrite. The XRD analysis results confirmed the formation of a single-phase hexagonal structure. Parameters of structural lattice constant increase with increasing zinc concentration due to larger ionic radii of zinc than nickel ion. Shows variations in crystallite size with an increase in Zinc contents. The crystallite size of barium-based ferrites (BaNi$_{2-x}$Zn$_x$Fe$_{16}$O$_{27}$) for x = 0.0 was 36.5nm, and with Zinc substitution (x=0.4), crystallite size decreased to 35.18nm. Then gradually increased to reach 36.7nm as Zinc concentration increased to x= 2. This increase in crystallite size with Zinc concentration shows that nickel acts as a crystallite inhibitor.

The bandgap energy for the samples was obtained from the Tauc plot and showed that with increasing the zinc concentration, the energy increased from 2.258 eV to 2.8306 eV. The transmittance for all samples increases with increases in the wavelength in the range of about
(300-400nm) and then saturates at higher wavelengths. This material is good in absorbance at wavelengths of (350 nm). The absorption edge shows a shift to the short wavelength side with increasing Zn content. The value of the absorption coefficient decreases with the increase in the wavelength for all the samples of ferrite up to a certain wavelength depending on the ferrite sample; at that wavelength (320-300 nm) in the visible region, αt almost becomes invariant. In UV region, the absorption coefficient values of α decrease by decreasing the wavelength (λ<220nm) and the values of α decreased. This is due to the absorbing nature of the Zn element. The absorption edge shows a shift to the short wavelength side with increasing Zn content. The value of the absorption coefficient decreases with the increase in the wavelength for all the samples of ferrite up to a certain wavelength depending on the ferrite sample; at that wavelength (320-300 nm) in the visible region, αt almost becomes invariant. In UV region, the absorption coefficient values of α decrease by decreasing the wavelength (λ<220nm) and the values of α decreased. This is due to the absorbing nature of the Zn element. The absorption edge shows a shift to the short wavelength side with increasing Zn content.

The refractive index and extinction coefficient value shows an overall increasing trend with the increase in photonic energy and the increase of Zn content for the sample. This spectral and dopant dependence of optical constants (n, k) with the photonic energy will be helpful in deciding on the suitability of this system for application in optical data storage devices. The conductivity peak of the samples increases with a decrease in Zn content, confirming that these specimens maintain good photosensitivity.

The results show an increasing value of the bandgap as the concentration of Zn\(^{2+}\) is increased.

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**Conflicts of Interest**

The authors declare no conflict of interest.

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