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## Effectiveness of non-magnetic ions Doping on optical energy, surface properties and antimicrobial activity of Ba-Based nanoferrites

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### Abstract:

The main mechanism of barium hexaferrite nanoparticles (BFNPs) as antibacterial activity was investigated in the present study. A ceramic method was used to synthesized barium nickel ferrite doped with zinc ions. The mean crystallite size and surface area were determined by X-ray diffraction, which showed that the sample Zinc ions equal to 0.4 had a smaller crystallite size and a high specific surface area than other samples. This is useful in establishing the relationship between band gaps and antimicrobial activity, as electrons move from the valence band to the conduction band. BFNP powder concentrations were tested against two types of gram-positive and gram-negative bacteria. All nanoparticles inhibited both gram-positive and gram-negative bacteria at the higher concentration range of 3 ml. In terms of antimicrobial activity, *Bacillus subtilis* growth was inhibited to a lesser extent (16 mm) and *Pseudomonas aeruginosa* growth was inhibited to a greater extent (30 mm). The difference in antimicrobial activity against gram-positive and gram-negative bacteria may be due to the composition of the cell walls and differences in the interaction of the membrane with the cell at the molecular and cellular level. Nanoparticle size, microstructure and specific surface area also significantly influence antibacterial activity. Components of the underlying antimicrobial mechanism have been identified as cell membrane damage, protein leakage and intracellular reactive oxygen species production. These studies indicate the potential of PFN as a material for biomedical applications and as an effective antimicrobial agent.

**Keywords:** Nanoferrites; Antibacterial activities; Inhibition zone; Optical energy; Surface area.

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## Introduction

Recent research has focused on the applications of nanomaterials in the medical field, in particular anticancer, antifungal, antiviral and antibacterial. Using them indiscriminately reduces their effectiveness against bacteria. Nanoparticle ferrites have been studied as agents with antibacterial activity<sup>1,2</sup>. Due to their favorable biomedical applications and ferromagnetic properties, some metal oxides such as zinc ferrite, barium ferrite and nickel ferrite have been extensively investigated<sup>3,4</sup>.

Changing the size of the nanoparticles of ferrite NPSs results in changes in the chemical, structural, optical and electrical properties; some authors believe that these changes result from the amount of confined surface area, and therefore lead to new applications<sup>5,6</sup>. For example, when the size of nanoparticles is reduced to nanometers, there is an increase in bacterial activity<sup>7</sup>. Expressly point of view antibacterial applications, in terms of the preferred organic material, have a lower toxicity, improved stability, lower resistance and a good choice<sup>8,9</sup>. The efficacy of ferrite nanoparticles in terms of antibacterial activity has been reported<sup>10,11</sup>.

The literature also refers to this<sup>12-14</sup>, that the cations Cu<sup>2+</sup>, Zn<sup>2+</sup> and Ba<sup>2+</sup> are essential for the health of human beings as they are good disinfectants with antibacterial activity. These cations have a very important antibacterial activity by attaching to the cell walls of bacteria and destroying the DNA. Previous<sup>11-15,16</sup> have demonstrated antibacterial activity have shown that ultra-fine magnetic nanoparticles can easily enter the bacterial cell, interact with the cell membrane, induce oxidative stress and lead to the destruction of DNA<sup>17-19</sup>.

The surface modulation is responsible for the automatic toxicity of ferrite NPs on bacteria. Dispersal of the NPs is controlled by the intrinsic structural and chemical properties of the NPs and by doping.

The NPS ferrites are dispersed using the intrinsic structural, physical and chemical properties of NPs and type doping. The antibacterial activity is known to be related to the size of the nanoparticle<sup>20</sup>. The confluence of magnetic materials with antibacterial properties can make this material important for biomedical applications. To the best of our knowledge, there are no reports on the improvement of zinc-doped barium-nickel ferrites using a ceramic method for biomedical applications. Considering its availability, and suitability, an effectively designed hybrid system of zinc-doped barium-based nickel ferrite would be valuable for possible application as an antibacterial agent to targeted organs in the human body. Thus, this may help to clarify the most desirable collection of Ni and Zn ions to achieve the best antimicrobial effect. Therefore, in the present study, attempts were made to synthesize zinc-metal doped  $\text{BaNi}_{x-2}\text{Zn}_x\text{Fe}_{16}\text{O}_{27}$  ( $x=0.0, 0.4, 0.8, 1.2, 1.6, \text{ and } 2$ ) to explore the suitability of powder nanoparticles for antibacterial application.

## Materials and Methods

### Study design

#### Study Design

The samples used in this study with the starting materials BaCO<sub>3</sub>, ZnO, NiO and Fe<sub>2</sub>O<sub>3</sub> were mixed according to their molecular weight ratio to obtain different compositions of BNZFeO-NPs. The well-known ceramic method was used to prepare the zinc-doped W-type hexagonal barium-nickel ferrite nanoparticles. The processes used to prepare the nanoparticles are detailed elsewhere<sup>21,22</sup>.

### Antibacterial activity Test

The antibacterial activity against gram-positive *B. subtilis* and gram-negative *P. aeruginosa* was evaluated using dimethyl sulfoxide (DMSO) and complexes. Activities were carried out using an agar-well diffusion method, where a pure isolate is grown to confluence on a Müller-Hinton plate

using a sterile swab. The plates were desiccated and an 8 mm diameter antiseptic cork punch was used to puncture the plates. The plates were desiccated and a well was made in each agar plate using an 8 mm diameter antiseptic cork punch. A 20  $\mu$ L volume of each compound at 200  $\mu$ g/ml concentration was pipetted into the Muller-Hinton agar plate wells using a micropipette. Each compound's antibacterial activity was determined by measuring inhibition zones in well <sup>23,24</sup>. The antibacterial activity of each compound was determined by measurement of the inhibition zone diameter. The mean diameter of the inhibition zones was calculated and the experiment was performed in triplicate.

## Results

### Surface Area and optical energy:

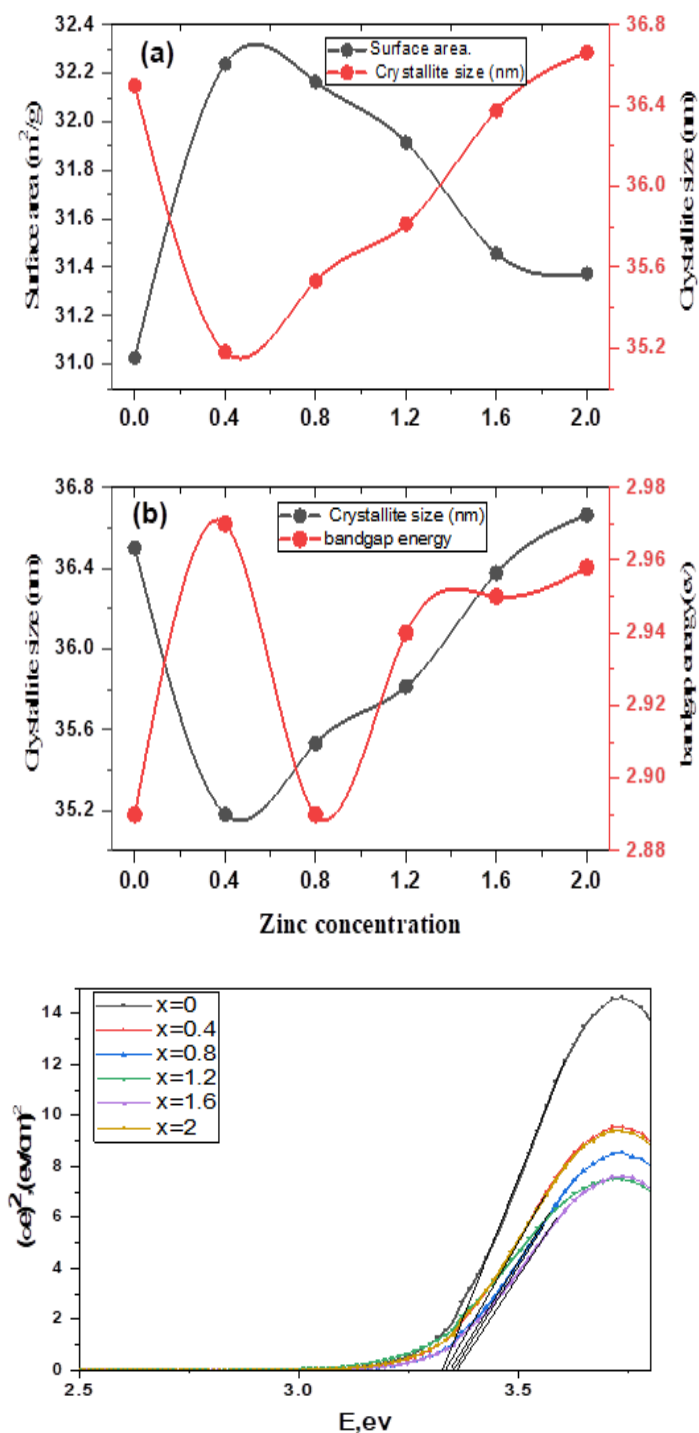
The structural composition of the nanoparticle ferrites was studied and determined by X-ray diffraction analysis. The mean crystal size and surface area were calculated from the X-ray data and the results are shown in Fig1. The results show that the average crystallite size and surface area of  $\text{BaNi}_{2-x}\text{Zn}_x\text{Fe}_{16}\text{O}_{27}$  nanoparticles were modified by doping with non-magnetic ions, as shown in the average crystallite size and surface area of  $\text{BaNiFe}_{16}\text{O}_{27}$  nanoferrite. Therefore, they had a high specific surface area (Figure 1.a), allowing more contact with the microbial surface. In terms of antimicrobial properties, these results could also be applied. This is due to its small size and high specific surface area, allowing more contact with the microbial surface. The hexagonal structure is another reason. The (001,008) face can adsorb oxygen molecules and  $\text{OH}^-$  ions due to its unsaturated oxygen coordination and positive charge. This was explained by the fact that as the size decreases, the number of NPs per volume increases, as does the surface area, resulting in greater  $\text{H}_2\text{O}_2$  generation. This results in a higher rate of production of  $\text{H}_2\text{O}_2$  and  $\text{OH}^-$

radicals and hence increased antimicrobial activity. The increase in antibacterial activity is described by the collective response of the reduced size of the crystallite particles, and the increased surface area also generates a higher capacity for ROS generation.

The bandgap energy of the nanoferrites is another factor that confirms the relationship with antibacterial activity, as shown in Figure 1. b and c. The inter-valence charge transition cations in the synthesised materials can be physically explained by the presence of ions ( $\text{Fe}^{2+}/\text{Zn}^{2+}/\text{Ni}^{2+}/\text{Ba}^{2+}$ ). Usually, the smaller magnetic nanoparticles. In general, the smaller magnetic nanoparticles will have a higher absorption value than the larger ones. For  $\text{BaNi}_{1.6}\text{Zn}_{0.4}\text{Fe}_{16}\text{O}_{27}$  a higher energy band gap was obtained than for the other samples. The decrease in energy gap is due to the insertion of extra energy levels in the valence band of  $\text{BaNi}_{2-x}\text{Zn}_x\text{Fe}_{16}\text{O}_{27}$ . This idea of moving electrons from the valence band to the conduction band is useful to find a relationship between band gap and antibacterial activity.

### Antibacterial activity:

The antibacterial activities of barium-nickel nanoferrites powders have been investigated by means of *Bacillus subtilis* Gram-positive and *Pseudomonas aeruginosa* Gram-negative. The method used was the agar well diffusion technique. Bacteria were selected based on common infections affecting the community by ingesting them when present in contaminated water. For example, *Pseudomonas aeruginosa* causes whooping cough in adults and children. Bacterial infections, endocarditis, pneumonia and septicaemia have been attributed to *B. subtilis*. However, patients who were free of infection have been found to have these infections. Due to the presence of these dangerous bacteria in water, there is a need for the efficient development of nano-based antibacterial inhibitors to reduce their growth and functionality in the water system.

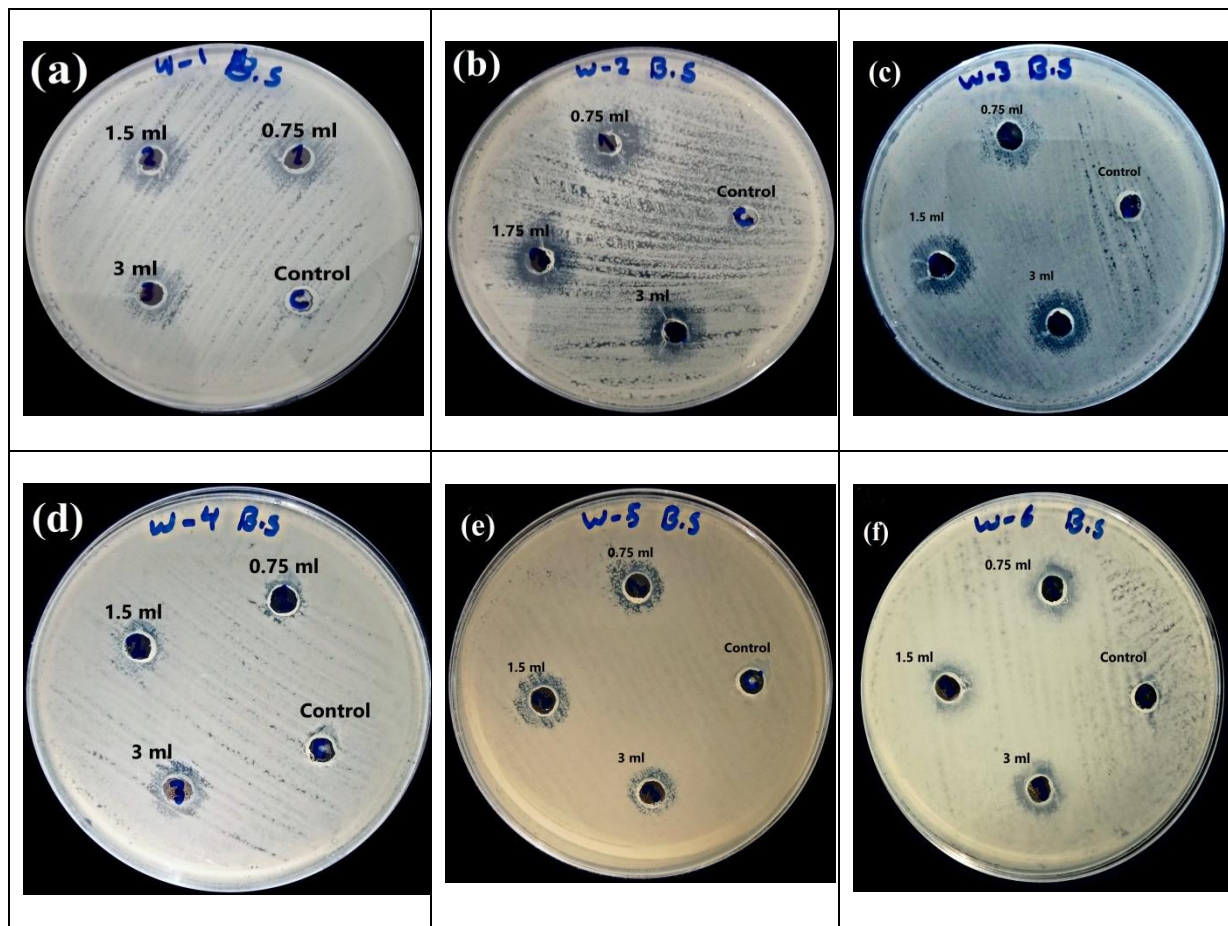


**Figure 1.**(a) relationship between crystallite size and surface area; (b): relationship between

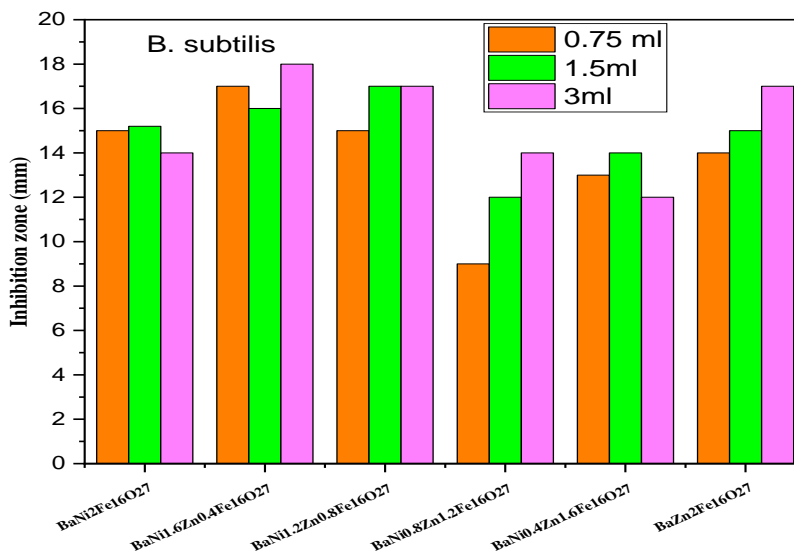
crystallite size and band gap energy; (c): band gap energy.

The ratio of the concentration mass of the barium nickel ferrite nanopowders to the solvent (DMSO) was varied to determine the efficacy and the best concentration charge for the best antibacterial activity. The results of the antibacterial activities of Ba-Ni nanoferrite doped with zinc ions against *Bacillus subtilis* gram-positive type are presented in the form of the area of the inhibition zone. The minimum inhibitory concentrations of 3 mL, 1.5 mL and 0.75 mL, respectively, are shown in Fig. 2. Histogram of inhibition zone diameter of prepared samples on bacterial growth. As can be seen in Fig. 2, b :c, the sample BaNi1.6Zn0.4Fe16O27 at a concentration (3 mg/mL) was the most effective against gram-positive bacteria *Bacillus subtilis* with an inhibition zone of 18.2 mm. These results correlated the effects of nanoparticle size and antibacterial activity, as the large inhibition zone due to the small crystal size, as found in the structural results and published<sup>25</sup>. led to a high interaction between the sample and the cell membrane. In the second order, the sample BaNi1.2Zn0.8Fe16O27, at a concentration (1.5 and 3 mg/ml), showed a significantly higher antibacterial activity against the gram-positive bacterium *Bacillus subtilis*, with a zone of inhibition of 16.8 mm, due to the presence of Ni<sup>2+</sup> and Zn<sup>2+</sup> ions in a large ratio delayed at the antibacterial lifetime<sup>24</sup>. In addition, the accumulation of Zn ions caused the pitting of the cell wall structure and the continuous release of membrane proteins and lipopolysaccharides, leading to the death of the bacteria at a concentration (3 mg/ml) of BaZn2Fe16O27 with an inhibition zone of 16 mm<sup>25,26</sup>. This is because of their large crystal sizes, resulting in a low level of interaction with cell membranes.

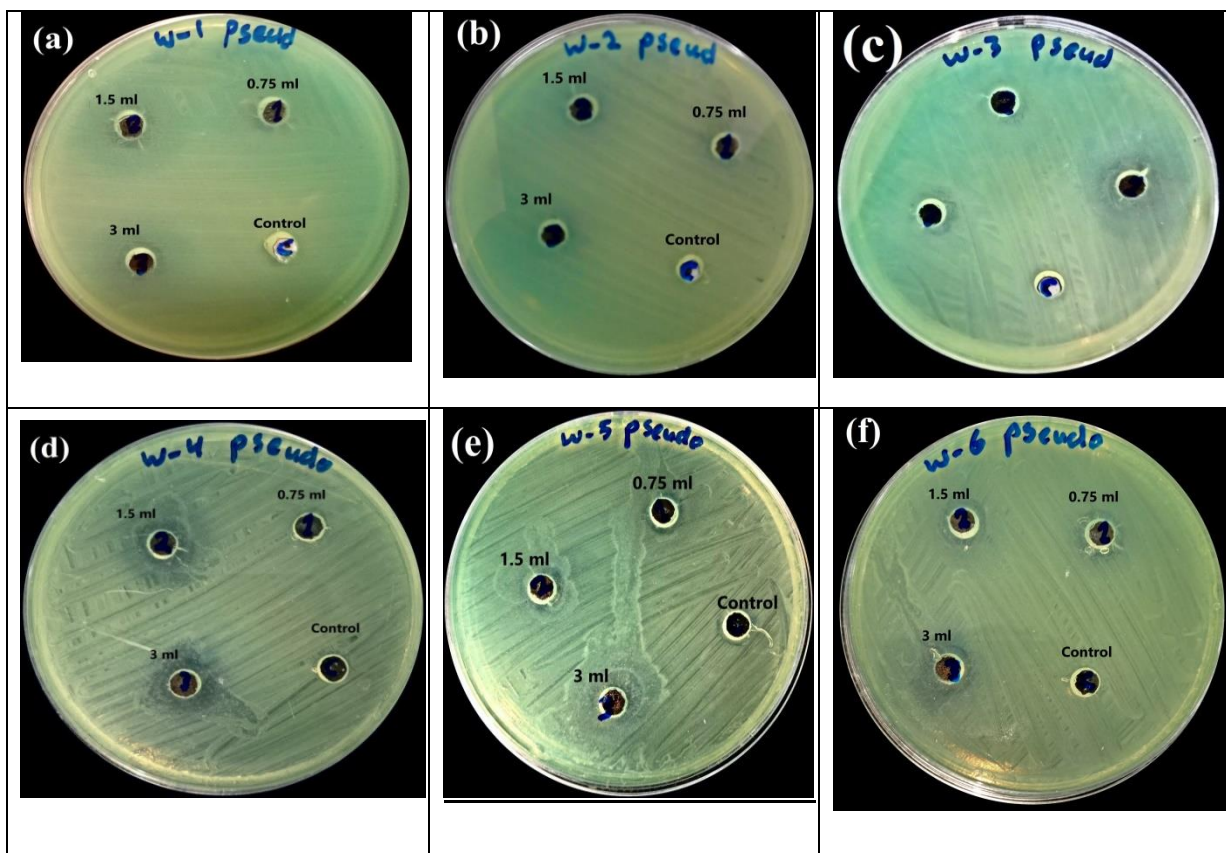




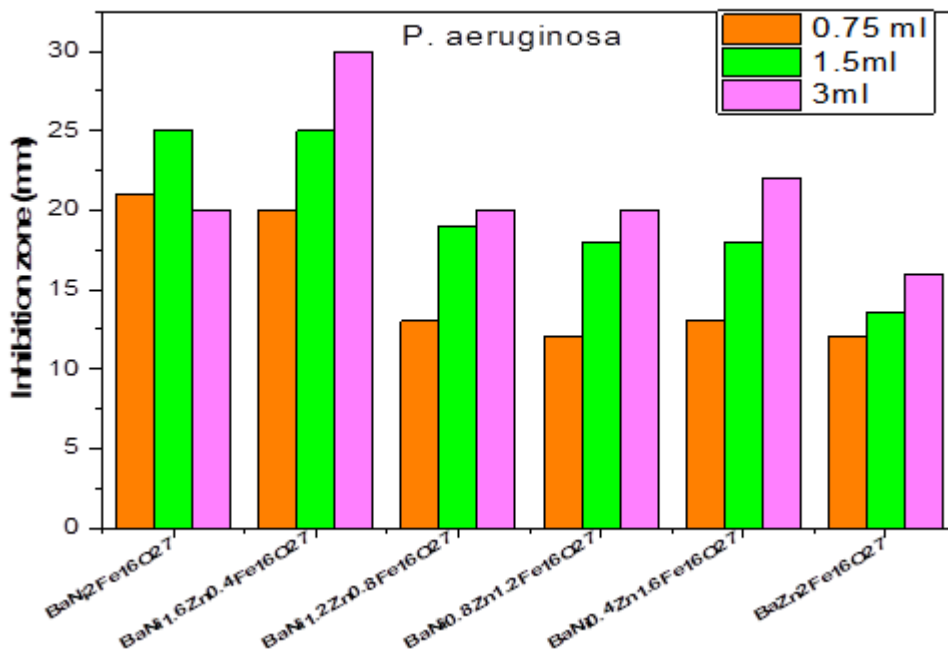
**Plate 1.** Antibacterial activity of (a)  $BaNi_2Fe_{16}O_{27}$ ; (b)  $BaNi_{1.6}Zn_{0.4}Fe_{16}O_{27}$ ; (c)  $BaNi_{1.2}Zn_{0.8}Fe_{16}O_{27}$ ; (d)  $BaN_{0.8}Zn_{1.2}Fe_{16}O_{27}$ ; (e)  $BaN_{0.4}Zn_{1.6}Fe_{16}O_{27}$ ; (f)  $BaZn_2Fe_{16}O_{27}$ ; against bacteria *Bacillus subtilis*. Numbers refer to the concentration of BFNPs: 0.75, 1.5, and 3  $\mu$ g/disk of BfNPs.



**Figure.2.** Average diameter zone (mm) growth inhibition activity of nanoparticle’s ferrites at various concentrations against Gram-positive *Bacillus subtilis*.



**Plate 2.** Antibacterial activity of (a)  $BaNi_2Fe_{16}O_{27}$ ; (b)  $BaNi_{1.6}Zn_{0.4}Fe_{16}O_{27}$ ; (c)  $BaNi_{1.2}Zn_{0.8}Fe_{16}O_{27}$ ; (d)  $BaN_{0.8}Zn_{1.2}Fe_{16}O_{27}$ ; (e)  $BaN_{0.4}Zn_{1.6}Fe_{16}O_{27}$ ; (f)  $BaZn_2Fe_{16}O_{27}$ ; against bacteria *Pseudomonas aeruginosa*. Numbers refer to the concentration of BFNPs: 0.75, 1.5, and 3  $\mu$ g/disk of BfNPs.



**Figure 3.** Average diameter zone (mm) growth inhibition activity of nanoparticle's ferrites at various concentrations against Gram-negative *Pseudomonas aeruginos*

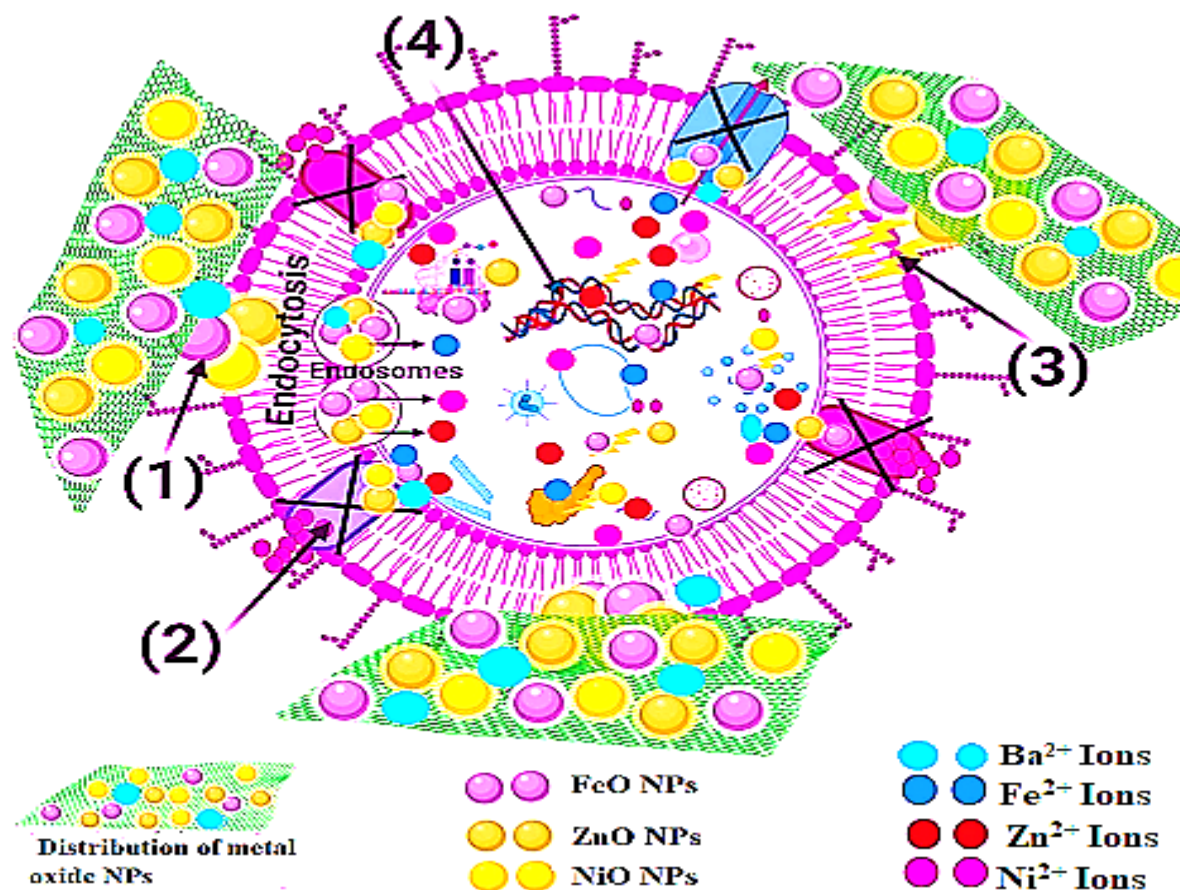
(a) *Bacillus subtilis*, (b) *P. aeruginosa*, (c) NPs either interact directly with bacterial cells or produce secondary products which damage bacterial cell walls. There are several mechanisms underlying antibacterial activity of BFNPs. The following factors were responsible for the antimicrobial activity of the preparation<sup>28</sup>. Photo generation of ROS on the surface of metal oxide nanocomposites such as superoxide anion radical ( $O_2^-$ ), hydroxyl radical (OH) and hydrogen peroxide ( $H_2O_2$ ). Subsequent to oxidation, a valence band hole is attracted by electrons of water or hydroxyl anion to produce most reactive radical (OH).

Peroxide ( $H_2O_2$ ) is produced by the reaction between the hole-electron pairs and the superoxide anion. Serious damage to DNA, nucleic acids, carbohydrates and lipids is caused by the hydroxyl radical (OH) and superoxide anion radical ( $O_2^-$ ). Among the ROS, ( $H_2O_2$ ) and (OH) are the most powerful oxidizing agents and can directly penetrate the bacterial cell membrane to cause injury and prevent cell growth, resulting in bacterial death. Increased ROS generally depends on increased surface area, corresponding crystallite size reduction, increased oxygen vacancies, and facilitated diffusion and mass transport of reactant molecules. Antibacterial activity also involves other mechanisms. Parasitism of the nanoparticles in the cell membrane of the bacteria and restriction of the mesosome. The surface area of the bacterial cell membrane is increased by disrupting the mesosomal functions of cellular respiration, DNA replication and cell division. Oxidative stress, caused by the generation of ROS due to cell death, is induced by these intracellular functional changes. When the metals ( $Ni^{2+}$ ,  $Ba^{2+}$ ,  $Fe^{2+}$  and  $Zn^{2+}$ ) are liberated from the surface of the nanocomposite, they come into contact with the cell membrane of the

microorganism. The cell membranes are negatively charged and ( $Fe^{2+}$ ,  $Ni^{2+}$ ,  $Ba^{2+}$  and  $Zn^{2+}$ ) positively charged. This creates an electromagnetic attraction between the nanoparticles. They attract each other and ( $Fe^{2+}$ ,  $Ni^{2+}$ ,  $Ba^{2+}$  and  $Zn^{2+}$ ) pass through cell membranes and react with thiol groups (-SH) of proteins present on bactericidal cell surfaces. This causes the microbes to oxidise and die instantly. Nanomaterials inactivate proteins and reduce the permeability of the membrane, leading to the death of microbes<sup>25</sup>. This process is illustrated in Fig. 3. 4. The main mechanisms of the antibacterial activity of BFNPs include the following First, BFNPs nanoparticles ferrite cleaved to the microbial cell surface and spread over it to stop the release of BFNPs metal oxide. As a result, the membranes deteriorate and the transport activities change. Second, the transfer of ions to and from the microbial cell is blocked by ferrite NPs ( $NiO$ ,  $BA$  and  $ZnO$ ). Thirdly, metallic NPs generate and enhance ROS, promoting cell damage. Fourth, metal oxide nanoparticles penetrate into microbial cells and interact with cellular organelles and biomolecules (such as plasmid DNA, ribosomes, chromosomal DNA and mesosomes), thereby affecting their respective cellular machinery<sup>29</sup>.

Metal oxide NPs ( $NiO$ ,  $FeO$  and  $ZnO$ ) can act as transporters to effectively deliver  $Ni^{2+}$ ,  $Fe^{3+}$  and  $Zn^{2+}$  ions to the microbial cytoplasm and membrane, where proton propulsion would lower the pH below 3, enhancing the release of  $Ni^{2+}$ ,  $Fe^{3+}$  and  $Zn^{2+}$  ions<sup>30</sup>.





**Figure 4.** Characteristic diagram of antibacterial activity mechanism.

## Conclusion

Nanoparticle ferrite BFNPs were successfully synthesized by the ceramic method, the X-ray results are evidence that the average crystallite size and surface area showed that the sample Zn=0.4 had a smaller crystallite size and a high specific surface area than other samples. The decrease in energy band gap for this sample was further confirmed by the UV results, which are useful to establish a relationship between band gap and antimicrobial activity by moving electrons from the valence band to the conduction band. The antibacterial activities of zinc doped with barium-nickel ferrites have been reported for gram-negative bacteria such as

*Pseudomonas aeruginosa* and the gram-positive bacterium *Bacillus subtilis*. The antimicrobial effects of zinc nanoparticles doped with barium-nickel ferrite on microorganisms showed that the nanoparticles had a low effect on the growth of 16 mm *Bacillus subtilis*. A further increase in ferrite concentration to 3 mL and Zn of 0.4 proved the inhibition characteristics of the base *Pseudomonas aeruginosa* and *Bacillus subtilis*. The maximum inhibition zones against the gram-negative bacteria *Pseudomonas aeruginosa* and the gram-positive bacteria *Bacillus subtilis* were found for the BaNi<sub>1.6</sub>Zn<sub>0.4</sub>Fe<sub>16</sub>O<sub>27</sub> sample consisting of Zn =0.4 BFNPs. Against the bacteria tested, the samples showed relatively



good antibacterial activity. The rate of bacterial growth seems to be another factor influencing the sensitivity of bacteria to antibiotics and nanoparticles. There is evidence that nanoparticles with small crystallite sizes are more active against bacteria than those with large crystallite sizes. They also interact with membrane proteins, which can damage bacterial membranes. We also found that the metal content of the core was related to the results we obtained. To support the results, we also proposed a plausible mechanism for the formation of Zn<sup>2+</sup> NPs. The good antimicrobial properties of barium-nickel-ferrite nanoparticles inhibit the progression of *Bacillus subtilis* and *Pseudomonas aeruginosa*, which makes it a good source of disinfecting agents with effective properties to enhance bacterial inhibition. *Pseudomonas aeruginosa* and *Bacillus subtilis*, a potential biomedical material.

Therefore, the development of zinc nanoparticles doped with barium-nickel ferrite has shown a high antibacterial effect on *Pseudomonas aeruginosa* and *Bacillus subtilis*, making it a potential material for biomedical applications.

### Conflict of interest

The authors declare no conflict of interest.

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