MUZ KABUĞU EKSTRAKTI KULLANARAK YEŞİL SENTEZLE ÜRETİLEN ZNO NP'LERİNİN FOTOKATALİTİK BOYA BOZUNMASINA ETKİSİ VE ANTİBAKTERİYEL POTANSİYELİ

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ÖZET

Fiziksel, kimyasal ve/veya biyolojik yöntemlerle sentezlenebilen nanoparçacıkların karakteristik özellikleri (şekil, boyut, toksisite vb.) sentez yöntemine göre değişebilmektedir. Nanopartikülleri biyolojik olarak sentezlemek için bitkiler ve bitki özütleri, mikroorganizmalar, algler, enzimler ve biyomoleküller ile endüstriyel ve tarımsal atıklar kullanılabilmektedir. Yeşil sentez olarak da bilinen biyolojik sentez, çevre dostu ve uygun maliyetli bir yöntem olarak diğer yöntemlerden daha üstündür. Bu yöntemde, tehlikeli kimyasallara ve yüksek sıcaklık ve basınç gibi maliyetli ve zor işlemlere gerek yoktur. Bu çalışmada, muz kabuğu ekstraktları kullanılarak çinko oksit nanoparçacıklarının (ZnO NPs) yeşil sentezle üretilmesi ve elde edilen malzemenin kristal viyole boyasının fotokatalitik degradasyonunda etkinliğinin ve antibakteriyel etkisinin belirlenmesi amaçlanmıştır. Yeşil sentez metodu ile üretilen ZnO NP'ler, fotokatalitik boya degradasyon işleminde farklı konsantrasyonlarda (0,05-0,1-0,25-0,5 g/100 mL) özel lambalar (gün ışığı -18 W, 1700 lumen) kullanılarak gerçekleştirilmiştir. Fotodegradasyon verimi, muz kabuğu ekstraktı ile üretilen 0,25 g ZnO NPs konsantrasyonunda %79,58 olarak elde edilmiştir. Bu arada, iki farklı bakteri üzerinde gerçekleştirilen antibakteriyel potansiyel çalışmaları DMSO içinde hazırlanan ZnO NPs süspansiyonları kullanılarak agar kuyu difüzyon metoduna göre yapılmıştır. Buna göre maksimum ortalama zon inhibisyonu, 0,04 mg ZnO NPs/mL konsantrasyonunda gram-negatif (*Escherichia coli*) bakteri için 15,43 mm ve gram pozitif (*Staphylococcus aureus*) bakterileri için 14,78 mm olarak ölçülmüştür. Sonuçlar, yeşil sentezlenmiş ZnO NP'lerin düşük konsantrasyonda bile kristal viyole boyasının fotodegradasyonu için fotokatalizör olarak kullanılabileceğini göstermiştir. Ayrıca bakterilerin büyümeleri üzerinde pozitif kontrole göre *S. aureus* için %42,43 ve *E. coli* için %61,3 oranında bir inhibisyon etkisinin olduğu belirlenmiştir.

Anahtar Kelimeler: Antibakteriyel Özellik, Fotodegredasyon, Muz Kabuğu, Nanopartikül, Yeşil Sentez

EFFECTS OF GREEN SYNTHESIZED ZnO NPs USING BANANA PEEL EXTRACT ON PHOTOCATALYTIC DYE DEGRADATION AND ANTIBACTERIAL POTENTIAL

ABSTRACT

The characteristic properties (shape, size, toxicity, etc.) of nanoparticles that can be synthesized by physical, chemical and/or biological methods may vary according to the synthesis method. In order to biosynthesize nanoparticles, plants and their extracts, microorganisms, algae, enzymes and biomolecules, as well as industrial and agricultural wastes can be used. Although biological synthesis is also known as green synthesis, it is superior to other methods as an environmentally friendly and cost-effective method. In this method, there is no need for hazardous chemicals and costly and difficult processes such as high temperature and pressure. In this study, it was aimed to green synthesize of zinc oxide nanoparticles (ZnO NPs) using banana peel extracts and to determine the effectiveness and antimicrobial effect of the obtained material in photocatalytic crystal violet dye degradation. Photocatalytic dye degradation process was realized with green synthesized ZnO NPs at different concentration (0.05-0.1-0.25-0.5 g / 100 mL) using special lamps (daylight - 18 W, 1700 lumen). The photodegradation yield was obtained as 79.58% at the concentration of 0.25g ZnO NPs with banana peel extract. By the way, antibacterial potential studies on two different bacteria were performed according to the agar well diffusion method using ZNO NPs suspensions prepared in DMSO. The maximum average zone inhibition was measured for gram-negative (*Escherichia coli*) as 15.43 mm and for gram-positive (*Staphylococcus aureus*) bacteria as 14.78 mm at the concentration of 0.04 mg ZnO NPs/mL in DMSO. The results showed that green synthesized ZnO NPs can be used as photocatalysts for photodegradation of crystal violet dye even at low concentration. In addition, compared to the positive control on the growth of bacteria, it was determined that it had an inhibition effect of 42.43 % for *S. aureus* and 61.3 % for *E. coli*.

Keywords: Antibacterial Potential, Banana Peel, Green Synthesis, Nanoparticle, Photodegradation

1. INTRODUCTION

It has been determined that dyes are colored-aromatic chemical compounds with a strong affinity for the substrate, which is widely employed in the textile sector. These non-biodegradable materials need to be removed from the environment because they pose a serious threat to the it. To decrease the dyes, a variety of techniques are applied, including flocculation, UV-light degradation, electro-coagulation, and redox potential treatments (El-Kemary et al., 2011). The science of nanotechnology is what makes it possible to create materials at the nanoscale that have various properties depending on their size and structure. Nanoscale materials including nanoparticles, nano sensors, and nanotubes are manufactured for a variety of uses, and the application fields of nanotechnology are quite diverse. Nanomaterials include advantages such as easy functionality, high surface area, and compact size.

ZnO nanoparticles (ZnO-NPs) are well-suited for a variety of tasks, including photocatalytic degradation of organic contaminants in water (Wannapop et al., 2022; Zheng et al., 2022; Wang et al., 2022), solar cell technology (Platzer-Björkman et al., 2006), photocatalytic antibiotic degradation (Shawky et al., 2022), chromium (VI) reduction (Poliukhova et al., 2022), antibacterial research (Kumar et al., 2014), cosmetics, electronics, and optics. ZnO is a chemical that has wide band-range dielectric characteristics and is an n-type semiconductor with a significant exciton binding energy and a band gap of 3.37 eV (60 meV) (Andrade et al., 2017). ZnO has a rather wide bandgap, however solar light illumination can enable surface redox processes like the hydrogen evolution reaction (HER) (Abdullah et al., 2022). Comparatively to other metals, it also has a high electron mobility. It effectively transfers electrons and produces photocurrent (Nie et al., 2021). Because of their unique physical and chemical characteristics, including as high electron mobility, photosensitivity, and stability, ZnO nanoparticles have attracted the attention of researchers. ZnO can replace TiO_2 due to TiO_2's scarcity, robustness, low cost, and harmless nature. It is suitable for catalysis because it has a wide surface area and many active sites (Umar et al., 2011). For the reduction of certain pigments, ZnO has more photocatalytic activity than TiO_2 (Kansal et al., 2009).

ZnO-NPs have been created using both chemical and physical methods. However, these techniques have a number of disadvantages, including a high energy requirement, high pressure and temperature, the need for expensive equipment, an unfriendly environmental impact, and the production of waste that is harmful to the environment (Agarwal et al., 2017). Utilizing of several dangerous, toxic, and damaging chemical substances leads to problems with the environment and biological systems. ZnO-NPs can be produced using plant extracts to prevent these effects. This is a promising choice since it is easy to use, takes less time, inexpensive, non-toxic, and good for the environment, and is effective because plant sources are more widely available and less expensive (Saratale et al., 2018).

Phytochemicals and organic compounds included in plant extracts serve as reducing and capping agents to help stabilize the production of nanoparticles and control crystal development during synthesis (Singh et al., 2018).

The solution undergoes a redox reaction, and the extracts help the metal ions transfer electrons to form nanoparticles. For the manufacture of ZnO-NPs, a number of plant extracts have been studied, including *Scutellaria baicalensis* (Chen et al., 2019), *Berberis vulgaris* (Anzabi, 2018), *Suaeda japonica* (Shim et al., 2019), and *Solanum nigrum* (Ramesh et al., 2015). In this study, it was aimed to green synthesize of zinc oxide nanoparticles (ZnO-NPs) using banana peel (BP) extracts and to determine the effectiveness and antimicrobial effect of the obtained material in photocatalytic crystal violet (CV) dye degradation when exposed to visible light.

2. RESEARCH AND RESULTS

2.1. Experimental Design

2.1.1. Materials

The following ingredients were obtained from Sigma-Aldrich: Ethanol (C_2H_6O), sodium hydroxide (NaOH), and zinc acetate dihydrate ($Zn(CH_3COO)_2.2H_2O$, >%98). All other chemicals used in this study were of analytical grade. The two-stage Millipore Direct-Q_3 was used to create distilled water (DI).

2.1.2. Preparation of the BP extract

We purchased Mersin Anamur bananas from a store in Mersin, Turkey. The fresh BPs were initially removed and repeatedly washed with tap water. The BPs were then given a second deionized water wash to remove any contaminants that could still be on them. The peels were then cut into little pieces after drying on paper towels overnight at room temperature. To 100 mL of deionized water, up to 100 g of peels were added. The peels were then cooked for 20 minutes at 80 °C. The aqueous extract was then allowed to cool to room temperature before being filtered twice through Whatman No. 1 filter paper.

2.1.3. Green synthesis of ZnO-NPs

A typical biosynthesis of the ZnO-NPs was as follows: 33.33 g of the zinc precursor salt ($Zn(CH_3COO)_2 \cdot 2H_2O$) was dissolved in 1000 mL distilled water that prepared as stock solution. Then 150 mL of this stock solution was added to 90 mL of the BP extract and the mixture was vigorously stirred for 10 min. Subsequently, NaOH was added accordingly up to pH 12.0. The solution was continuously stirred at room temperature for 2 h to produce ZnO-NPs. The obtained precipitate that revealed the presence of BP-ZnO was filtered and washed with deionized water repeatedly. Finally, the precipitate was dried in an oven overnight at 60 °C. The light-yellow powder of BP-ZnO NPs was ground into a fine powder, and then stored for the characterization.

2.1.4. Characterization techniques

The morphology of BP-ZnO NPs was studied using scanning electron microscopy (SEM, FEI, Quanta FEG 250) with Cu-K radiation. To ascertain the chemical composition of the BP-ZnO NPs and the purity of the phase, X-ray Diffraction Spectroscopy (XRD, Bruker, D8 Advance) with a 2 scan was utilized.

2.1.5. Photocatalytic degradation of CV

A recognized strong oxidant in photocatalytic systems is the hydroxyl radical ($OH^{\bullet}$), which is produced by zinc oxide (ZnO), one of the metallic nanoparticles with demonstrated efficacy. The BP-ZnO nanoparticles used in this study as a catalyst helped the visible-light photocatalytic activity against CV dye.100 mL of CV (10 ppm) were added to a beaker containing 0.05-0.5 g/100 mL of BP-ZnO NPs. The solution was exposed to a daylight (Philips TL18 W*3 lamp, 1700 lumen) after being left in the dark for 30 min. The mixture was kept under stirring and samples were withdrawn at every 15 min to observe the variation in the dye's absorbance in the 595 nm by using a UV–vis spectrophotometer (Hach Lange 3900). In order to achieve homogeneous mixing of the catalysts, ambient air (150 mL/min) was supplied to the system via an air-pump from the bottom of the reactor. Before taking color measurements on the samples, all of the samples were centrifuged at 6000 rpm for 10 minutes. A UV-VIS spectrophotometer (HACH-DR3900) was used to detect maximum absorbance in order to determine color removal efficiency. Every experiment was carried out triplicate, and the degradation efficiency percentage was determined by the following equation (Eq.1):

$$Color\ Removal\ Efficiency\ (\%) = \frac{(C_{0} - C_{t})}{C_0} x100$$

2.1.6. Antibacterial Activity

The agar well diffusion method was used to determine the antibacterial effects of green synthesized ZnO NPs from banana peel extract on a gram-positive (*Staphylococcus aureus*) and a gram-negative (*Escherichia coli*) bacterial colonies. In the first step, the bacterial colonies were inoculated to a Tryptic Soy Broth (TSB) and incubated at 37 ^{o}C for 24 h. After the incubation, *S. aureus* and *E. coli* active culture was transferred to new tubes include TSB and incubated again at 37 ^{o}C for 5 h. Then the bacterial cultures were transferred to TSB again and the absorbance was determined at 625 nm (~ 106-7 cfu/mL). For the spread plate technique, the Triptic Soy Agar (TSA) was poured into petri dishes, and after the solidification, 0.1 mL of activated bacterial cultures were spread on the surface of TSA homogenously. The 10 mm diameter wells were formed, and the wells were filled with 100 μL of three concentrations of green synthesized BP-ZnO NPs (0.01, 0.02, 0.04 g/mL) solved in DMSO (dimethylsulfoxide) (10%) (Lal et al., 2022). The DMSO and antibiotic (Amoxicillin, 50 μg) were employed as a negative control (NC) and a positive control (PC), respectively. The petri dishes were left for the incubation at 37 ^{o}C for a night and the zone diameters (the diameters around the wells) were determined via Vernier caliper (Aldeen et al., 2022). All the antibacterial assays were performed in triplicate.

2.2. Experimental Results

2.2.1. Characterization of synthesized BP-ZnO NPs

In order to verify the formation of BP-ZnO nanoparticles, XRD analysis was conducted. The diffractograms were presented in Fig. 2.1. The BP-ZnO nanoparticles that were prepared at pH 12.0 exhibited diffraction peaks at 31.76°, 34.41°, 36.26°, 47.56°, 56.68°, 62.84°, 66.48°, 67.88°, 69.16°, 72.43°, and 76.9°, corresponding to the (100), (002), (101), (102), (110), (103), (200), (112), (201), (004), and (202) lattice planes, respectively. The hexagonal wurtzite crystalline structure of ZnO nanoparticles is characterized by these peaks (JCPDS card number: 36-1451). (Kaliraj et al., 2019). No peaks connected to impurities were discovered. These results support earlier findings (Abdullah et al., 2020; Rana et al., 2019). It is interesting to note that ZnO made by green synthesis is completely pure and incredibly crystalline in its unaltered native condition. We conducted the experiment at ambient temperature rather than using high temperatures, in contrast to earlier investigations (Anbuvannan et al., 2015; Karnan et al., 2016). This implies that a quick, easy, and environmentally friendly method of synthesis might be able to create highly crystalline nanomaterial.

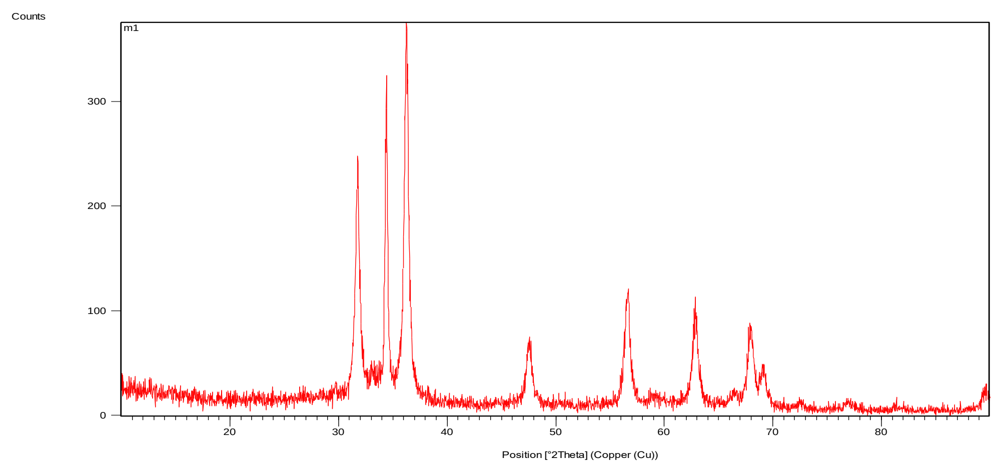

Figure 2.1. XRD patterns of ZnO

SEM was employed to observe the surface morphology of the prepared ZnO. Fig. 2.2 illustrates the morphological structure of ZnO prepared at pH 12.0. According to Fig. 2.2, it was seen that BP-ZnO NPs had a granulated and broken structure. It was also discovered that the synthesized BP-ZnO NPs had a circular-like form. As the pH value rises, the amount of H^+ and OH ions varies, which has an impact on the structure, morphology, and formation of ZnO. Because there are more OH ions present, they are more strongly attracted to the positively charged Zn^{2+}, which encourages the development of strong Zn-O bonds in the structure (Wahab et al., 2009). At high pH, smaller nanoparticles form. According to Wang et al. (2008), altering the basicity of the solution allows for the customization of various ZnO morphologies, including rods and flowers.

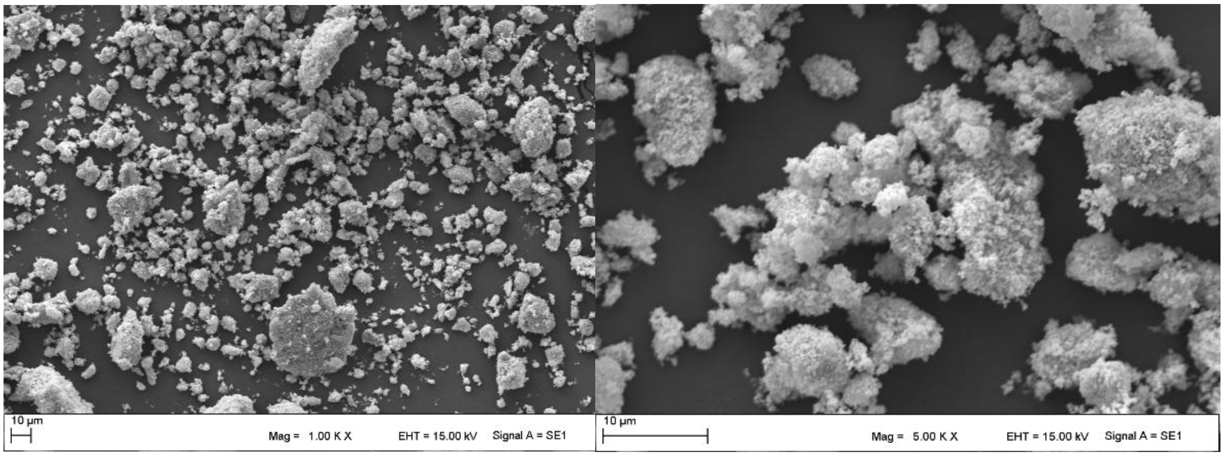

Figure 2.2. SEM images of BP-ZnO NPs prepared with magnifications of 1.000× and 5.000×

2.2.2. Photodegradation of CV

The photodegradation results of crystal violet (CV) using green synthesized ZnO nanoparticles with banana peel extract was shown in Fig. 2.3. It was observed that the lowest concentration of BP-ZnO NPs (0.05 g/100 mL CV) has not effect on the dye degradation under daylight lamps (18 W, 1700 lumen) after 120 min. Besides that, the increasing BP-ZnO NPs concentration increased the dye degradation after 30 min at almost all other concentrations (0.1-0.25-0.5 g/100 mL BP-ZnO NPs). The maximum dye degradation was determined at the 0.25 g/100 mL BP-ZnO NPs as 79.58 % after 120 min as like Ameur et al. (2019). The authors showed that the photodegradation efficiency of ZnO NPs on CV under sunlight irradiation was 78% after 210 min. Besides that, Abdullah et al (2020) synthesized ZnO nanoparticles using banana peel extract to determine the effectiveness on methylene blue dye degradation. The authors showed that the 0.02 g synthesized materials exhibited a good visible-light photocatalytic activity as 98 %. Aldeen et al. (2022) determined that the same results on methylene blue degradation using green synthesized ZnO NPs by palm leaves after 105 min under UV illumination.

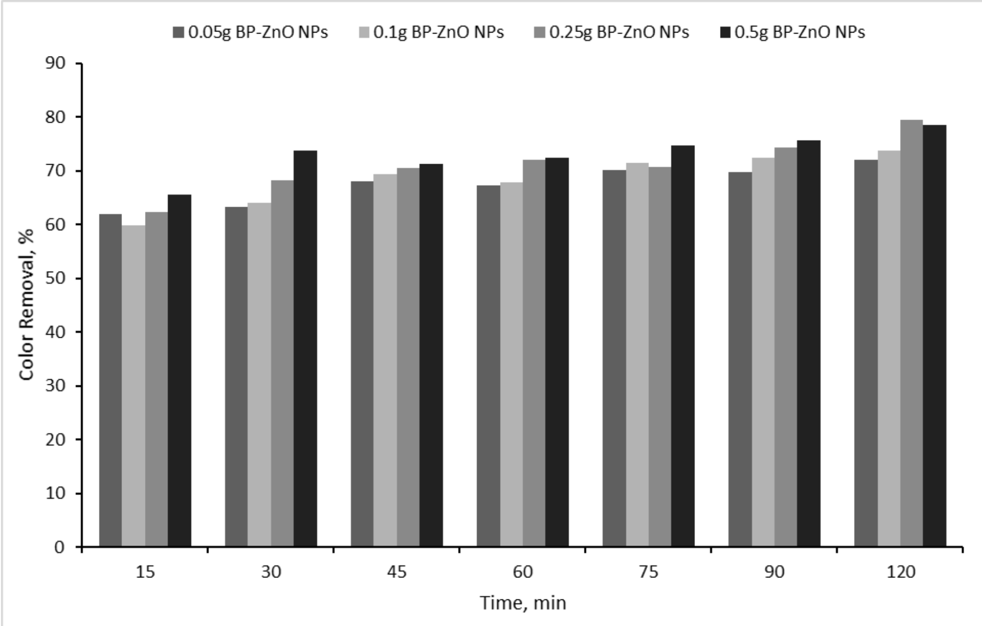

Figure 2.3. Photocatalytic crystal violet dye degradation at different concentration of synthesized ZnO NPs using banana peel extract under daylight lamps

2.2.3. Antibacterial Activity

To evaluate the antibacterial potential studies on two different bacteria were performed according to the agar well diffusion method using BP-ZnO NPs suspensions prepared in DMSO. The Fig.2.4a showed the effectiveness of BP-ZnO NPs on bacterial strains and Fig. 2.4b showed the average zone inhibition of BP-ZnO NPs on *Staphylococcus aureus* and *Escherichia coli*. It was determined that the maximum average zone inhibition was measured for gram-negative (*E. coli*) as 15.43 mm and for gram-positive (*S. aureus*) bacteria as 14.78 mm at the concentration of 0.04 mg ZnO NPs/mL in DMSO. Besides that, the DMSO solution used as a negative control had no effect on antibacterial activity. Results showed that the gram-negative bacteria were more sensitive to BP-ZnO NPs than gram-positive bacteria. These results are in accordance with obtained by Ruangtong et al. (2020) and Marfuah et al. (2020). This is caused by their thin cell wall and the interaction between Zn^{+} ions and gram-negative bacteria surface charge (Aldeen et al. 2022).

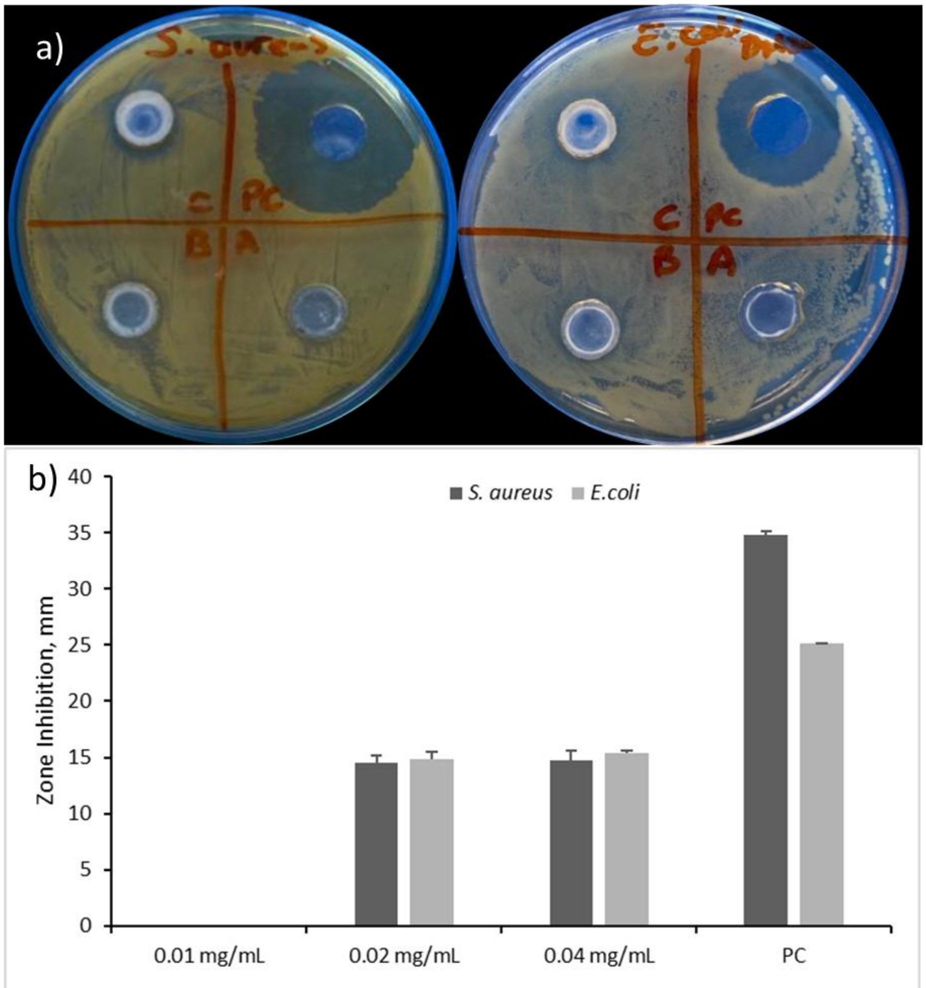

Figure 2.4. Antibacterial activity of green synthesized ZnO NPs using banana peel extract on *S. aureus* and *E. coli*

3. CONCLUSION

- In this study, the potential antibacterial activity of ZnO NPs synthesized by using banana peel extract and their effects on decolorization of crystal violet in water namely photocatalytic activity were evaluated.
- The results showed that green synthesized BP-ZnO NPs can be used as photocatalysts for the degradation of crystal violet dye even at low concentration (0.25 g/ 100 mL).
- Green synthesized ZnO NPs by using banana peel extract had inhibition effects on *S. aureus* and *E. coli* as 42.43 % and 61.3 %, respectively compared to the positive control.

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GÜNEYCE (GÜDÜL-ANKARA) VE ÇEVRESİNDEKİ YERALTI SULARININ KİMYASI VE BAZI KİRLİLİK PARAMETRELERİNİN İNCELENMESİ

Ziya ŞAHİN