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Research Article

Green Synthesis of Zinc Oxide Particles using Banana Peels and Tea Leaves Extracts for Rhodamine B Photodegradation

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Abstract
Rhodamine B is a widely used dye in the textile sector. However, the wastewater produced during the dyeing process presents a notable source of pollution, contaminating water and posing a threat to aquatic ecosystems due to its presence in liquid waste. Photocatalysis is a technique for breaking down toxic textile dye waste a semiconductor as a catalyst, valued for its high sensitivity and eco-friendly nature. In this research, zinc oxide particles were synthesized via a green synthesis approach using precipitation, employing natural capping agents from banana peel and tea leaf to degrade the synthetic dye of rhodamine B. The catalyst material was characterized using X-ray diffraction (XRD), scanning electron microscope (SEM), and ultraviolet-visible diffuse reflectance (UV-DRS). The photodegradation performance of rhodamine B was determined under UV light exposure for 3 hours. The XRD spectra of ZnO show the specific peaks of 2θ at 31.8°, 34.5°, and 36.3° with a crystallinity value of around 79.50%. The SEM result shows that the morphology of ZnO is in cotton-like form with a minimum band gap of 3.17 eV. The cotton-like ZnO particles demonstrated superior photodegradation efficiency for Rhodamine B, achieving 61.8%, compared to 47.9% with pure ZnO. It suggests that synthesizing ZnO particles with banana peels and tea leaf extracts boosts photodegradation efficiency by up to 20% compared to pure ZnO. This research highlights the potential of utilizing eco-friendly and sustainable methods as a greener approach for reducing waste in environmental applications.

Keywords: Banana peels, photodegradation, rhodamine B, tea leaves, zinc oxide

1. INTRODUCTION

The rapid worldwide industrial development has positive and negative impacts, including the decline in environmental quality due to water surface pollution [1]. Al-Buriahi, in 2022, reported that the textile industry in Indonesia produces approximately 749 tons of dyed textiles per year, and 10% of it is waste that contaminates the environment [2]. Among synthetic dyes, rhodamine B (RhB) is a widely used dye in textile industries due to its affordability. RhB is a red dye compound with high water solubility but highly toxic. The waste effluent containing RhB seriously threatens the environment and health, as it irritates the eyes, skin, respiratory tract, and digestive system [3–5]. Therefore, the wastewater treatment of RhB is crucial to reduce its hazardous nature.

A wide range of processes for treating and managing textile dye waste has been developed, including adsorption, biological treatment, membrane separation, solvent extraction, flocculation, photocatalysis, and other methods. Among these approaches, photocatalysis is advantageous due to its cost-effectiveness, efficiency, and lack of secondary pollutants. Photocatalytic techniques employ photons to decompose complex organic dyes into simpler eco-friendly compounds by employing semiconductors as catalysts, such as TiO_2 and ZnO [6–8].

Recent research shows ZnO outperforms TiO_2 because it has a wide bandgap (3.37 eV), a large exciton binding energy at room temperature (60 meV), affordability, high photosensitivity, high electron mobility, non-toxic, and environmentally friendly [9]. According to Suganya (2021), the size and shape of particles play a crucial role in photocatalytic activity as they can increase the surface area for energy absorption from the light source in photocatalytic reactions [4,10,11]. The nanostructures of ZnO show a larger surface area that results in a higher photocatalytic activity. Therefore, the use of ZnO particles for photodegradation purposes is widely proposed [4,12].

ZnO's controlling particle shape and size can be regulated during the synthesis step. In order to achieve nanoparticle size (10-100 nm), the utilization of a capping agent is employed to control the size, shape, and stability of ZnO particles [13]. Recently, natural capping agents from plant extracts have gained popularity due to their adherence to green chemistry principles –being environmentally friendly, low toxicity, generating minimal waste-—, cost-effective, and producing better morphology. Generally, natural capping agents are secondary metabolite compounds of plants, such as alkaloids, flavonoids, tannins, and polyphenols [14]. Based on research by Lyimo (2022), the waste of banana peels and tea leaves contains secondary metabolites that can be utilized as natural reducing and capping agents [15].

The Ambon banana (*Musa Paradisiaca S.*) is a widely cultivated fruit in tropical regions, particularly in Indonesia, where it is a staple in local diets. The peel of the Ambon banana is notable for its high content of natural antioxidants, including flavones and tannins [16]. In contrast, black tea leaves (*Camellia sinensis*) are recognized for their diverse phytochemical composition, containing significant amounts of alkaloids, flavonoids, and tannins [17]. These make both the banana peel and black tea leaves provide a synergistic effect due to their complementary properties and bioactive compounds, which enhance the synthesis process's efficiency and eco-friendliness and produce high-quality particles with desirable characteristics.

The Central Statistics Agency of Indonesia (BPS) reported that banana consumption in Indonesia in 2021 increased by 6.8% to 2020, with a nation's production of 8.7 Mtons. On the other hand, the tea consumption in 2021 reached 87 Mtons. The high production of bananas and tea in Indonesia corresponds to the waste generated. Considering the issue, this research purpose is to utilize waste from banana peels and tea leaves to synthesize and control the particle size of ZnO, which is one of the challenges in nanomaterial synthesis [18].

Based on the background, this research employed the green synthesis method of ZnO using waste from banana peels and black tea leaves as a capping agent. The synthesized material was characterized using X-ray diffraction (XRD), scanning electron microscope (SEM), and ultraviolet-visible diffuse reflectance (UV-DRS). Then, the photocatalytic activity in degrading rhodamine B was observed under ultraviolet light exposure. The expected outcome of this research is that the synthesized and studied material can be utilized as an alternative for applications in environmental science.

2. RESEARCH METHODS

Materials

All the chemicals used in this study were of analytical grade and were used without further purification. Sari Wangi black tea leaves and Ambon banana peels were the main materials collected from the local market in Indonesia. Zinc nitrate hexahydrate (ZNH, $Zn(NO_3)_2{\cdot}6H_2O$) and sodium hydroxide (NaOH) were purchased from Merck. Rhodamine B (RhB) was selected as a model organic pollutant. The water used in all experiments was double distilled.

Synthesis ZnO Particles using Banana Peels and Black Tea Extracts

Clean banana peels (100.00 g) were added to 300 mL of distilled water, then heated to 70 °C and stirred for 30 minutes. The solution was separated from solid residue by centrifugation at 6000 rpm for 15 minutes, then banana peel extract (supernatant) was stored at 4 °C [15].

Tea leaves (10.00 g) were added to 100 mL of distilled water, then heated and stirred for 2 hours at 80 °C. The solution was separated from solid residue by centrifugation at 6000 rpm for 15 minutes of tea leaf extract (supernatant), which was subsequently stored at 4 °C [15].

ZnO particles were synthesized over the green synthesis method of ZnO using waste from banana peels and black tea leaves as a capping agent[15]. 3.00 grams of $Zn(NO_3)_2{\cdot}6H_2O$ was dissolved into extracts of banana peels and tea leaves with volume ratio 1:1.15 pH of the obtained solution was adjusted to 7, 9, or 11 using 1M NaOH at 60-65 °C for 2 hours. Then, the solution was cooled and centrifuged at 10,000 rpm for 15 minutes. The resulting precipitate was washed with distilled water until neutral, dried in an oven at 50 °C for 24 hours, and calcined at 450 °C for 2 hours [19,20]. The ZnO was synthesized at pH 7, 9, and 11. A similar procedure was conducted by replacing the extracts with distilled water as a control.

Phytochemical Test

The tests conducted consist of flavonoid, alkaloid, and polyphenol tests [14].

a) Flavonoid test

The flavonoid test was conducted by adding 7-10 drops (about 0.5 mL) of 2M H_2SO_4 and Mg powder to the extract solution and allowing it to stand for 1 minute. The change of color to yellow-orange and the formation of bubbles indicate that a flavonoid is available in the extract.

b) Alkaloid Test

The alkaloid test was performed using two reagents: Mayer's reagent and Dragendorf reagent. The extract was added with 7-10 drops (about 0.5 mL) of 2M H_2SO_4, then dropped Mayer's or Dragendorf reagent, followed by homogenization. A positive result for alkaloids is indicated by the formation of a white precipitate for Mayer's reagent and an orange precipitate for Dragendorf reagent.

c) Polyphenol Test

The polyphenol test was carried out by adding a few drops of 10% (w/v) $FeCl_3$ to the extract, which was then homogenized. A positive polyphenol result is indicated by a change in color to greenish-brown.

Characterization

The obtained ZnO particles were characterized using X-ray diffraction (XRD, D8 Advance ECO Bruker, Germany) to observe the crystal structure with a range of 2θ of 10-90°. The scanning electron microscope (SEM, JEOL JSM-IT300LV, Japan) was used to examine the resulting morphology of the ZnO. Then, an ultraviolet diffuse reflectance spectrophotometer (SPECORD 210 PLUS, Germany) was used to determine the band gap of the ZnO.

Photodegradation of Rhodamine B using ZnO Particles

Photodegradation of rhodamine B using ZnO particles was conducted under dark conditions and UV light exposure. ZnO (50 mg) was added to rhodamine B solution (15 ppm) and stirred under dark conditions for 20 minutes. Then, the mixture was exposed to UV light (315 nm) for 3 hours. Further, the rhodamine B solution was separated from ZnO by centrifugation, and its absorbance was measured with a UV-Vis spectrophotometer at λmax 554 nm to determine the photocatalytic performance of ZnO. In addition, rhodamine B solution (15 ppm) was exposed to UV light without ZnO particles as a control. The standard calibration solutions of Rhodamine B were prepared with a range concentration of 0-50 ppm[3,21,22]

3. RESULTS AND DISCUSSION

Phytochemical Test for Banana Peels and Black Tea Extracts

Phytochemical testing aims to determine the content of secondary metabolite compounds in banana peels and tea leaf extracts. The results are shown in **Table 1** and **Figure 1**.

Table 1. Phytochemical test results of banana peel and tea leaves

Test	Extract	
	Banana peels	Tea leaves
Flavonoid	+	+
Alkaloid		
- Mayer	+	+
- Dragendorf	+	+
Polyphenol	+	+

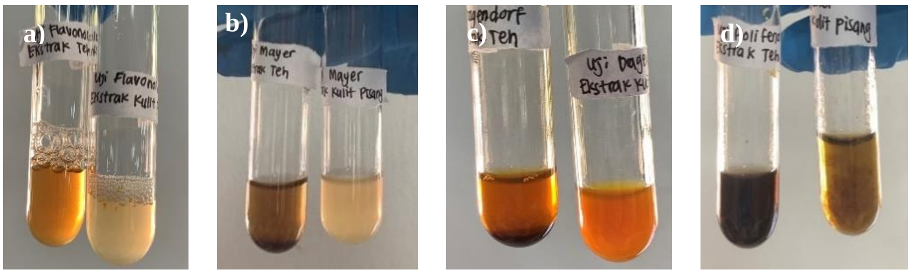

Figure 1. Phytochemical test (a) flavonoid, (b) Mayer test, (c) Dragendorf test, (d) polyphenol

Phytochemical analysis demonstrates that extracts from banana peels and tea leaves are rich in secondary metabolites, including flavonoids, alkaloids, and polyphenols. These bioactive molecules reflect the plants' inherent chemical characteristics and play a critical function in the production of zinc oxide particles. Acting as natural bio-capping agents during synthesis, these metabolites exert significant influence over the size and structural formation of the particles. In contrast to traditional chemical synthesis methods that typically rely on toxic reagents and high energy consumption, using banana peel and tea leaf extracts presents a more sustainable and environmentally friendly alternative. The bio-capping effect of secondary metabolites minimizes the environmental impact and enhances the particles' functionality, thereby improving their effectiveness in photocatalytic applications.

X-Ray Diffraction of ZnO Particles

The synthesized materials were thoroughly characterized using X-ray diffraction (XRD) to confirm the crystal structure and phases. The diffractogram displayed distinct peaks at 2θ values of 31.8°, 34.5°, and 36.3°, which closely match those of the standard powder sample (PDF Card-04-003-2106) [23]. These peaks correspond to the crystallographic planes (100), (002), and (101) of ZnO materials, as analyzed using DIFFRAC.EVA software (**Figure 2**). The XRD results validate the successful synthesis of ZnO particles using the green synthesis approach with banana peel and tea leaf extracts (ZnO-BPE). The presence of banana peel and tea leaf extracts as reducing and stabilizing agents during the synthesis process likely contributed significantly to the growth and crystallization of the ZnO particles, as reflected in the XRD patterns. The bioactive compounds in the banana peel and tea leaf extracts may have played a crucial role in influencing the nucleation and growth mechanisms, forming particles with distinct crystalline orientations.

The software assessed the synthesized material's crystallinity, calculating a 79.5% crystallinity value from the XRD spectra. The crystallite size of the ZnO particles was calculated using Debye-Scherrer's equation, as presented in the formula.

$$D = \frac{k\lambda}{\beta \cos \theta} \quad (1)$$

where D is the size of the crystals formed, k is the proportionality constant with a value of 0.9, λ denotes the wavelength of the CuKα X-ray radiation (1.54 Å or 0.154 nm), β refers to the full width at half maximum (FWHM) of the most intense diffraction peak, and θ corresponds to the Bragg angle.

The average crystallite size is estimated to be 10.62 nm. The crystallite size is notably smaller than that reported for ZnO synthesized using cherry extract[24] and *Myristica fragrans* [20] in green synthesis approaches. This relatively small crystal size indicates the success of the green chemistry approach employed in the synthesis method. However, the variation in peak intensities between the synthesized ZnO-BPE and the standard PDF card can be attributed to variations in the dominant exposed crystal facets of the ZnO particles. Such structural differences may influence the behavior and performance of the particles, particularly in photocatalysis, where factors such as surface area, crystallinity, and morphology are paramount to their efficacy.

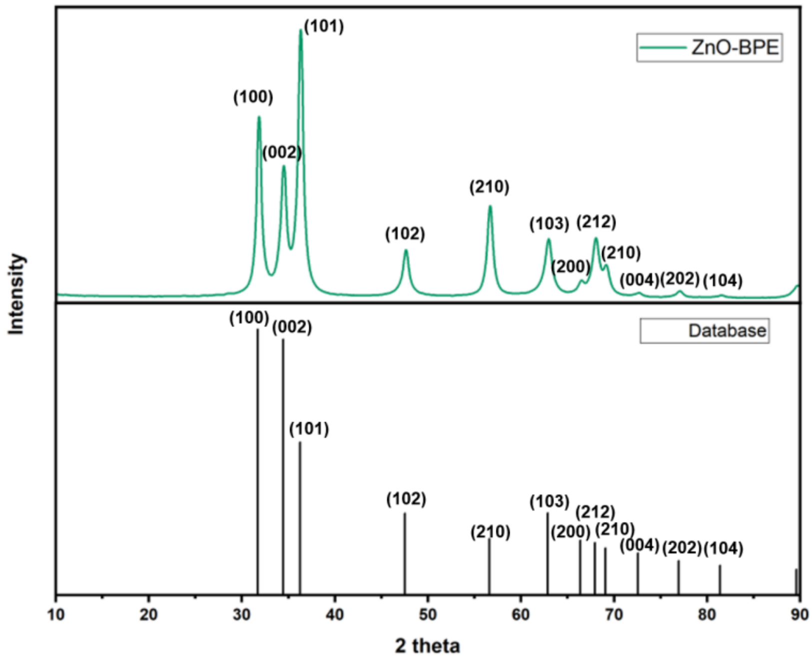

Figure 2. Diffractogram of ZnO-BPE that is synthesized using banana peels extract and tea leaves extract

The Morphology of ZnO

ZnO particles were characterized using a scanning electron microscope (SEM) to observe the surface morphology of materials. The characterization was performed by comparing the particles synthesized without using the extract (as a control) and those synthesized with the extract (**Figure 3**).

The SEM results show that synthesized ZnO particles without using the extracts had a range particle size of 200-300 nm. As shown in **Figure 3**, the large particle size is influenced by the increases in the pH solution during the synthesis process that causes agglomeration. When the pH solution exceeds 7, the OH^- ions bind with Zn^{2+} ions to form smaller particles. However, the particles are unstable and lead to agglomeration, forming aggregates of larger sizes [25].

On the other hand, the ZnO-BPE shows a cotton-like morphology, implying a particle size of less than 50 nm at a magnification of ×50,000 (Figure 3d to 3f). It indicates that adding the banana peels and tea leaf extracts has effectively decreased the particle size of ZnO particles. These extracts help stabilize the particles by preventing agglomeration, leading to improved dispersion and enhanced material characteristics. During the green synthesis process, the hydroxyl groups of secondary metabolite compounds (flavonoids, alkaloids, and polyphenols) in banana peels and tea leaf extracts interact with Zn^{2+} to form a stable structure. This interaction contributes to minimizing the interaction between Zn^{2+} and preventing the agglomeration, which allows for control of the particle size of materials [14].

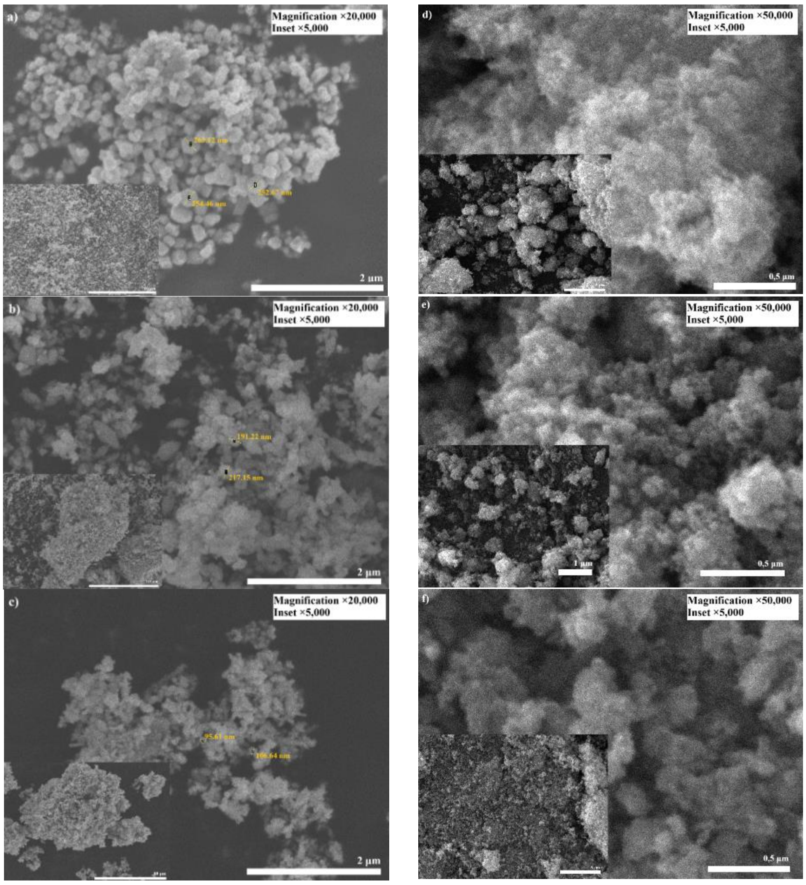

Figure 3. The morphology of pure ZnO (without extract): (a) pH 7; (b) pH 9; (c) pH 11 and ZnO-BPE: (d) pH 7; (e) pH 9; (f) pH 11 (note: magnitute of 5k, 20k and 50k)

Optical Properties

UV-Vis diffuse reflectance spectroscopy (DRS) analysis was employed to determine the band gap energy through Kubelka-Munk calculations. **Figure 4** shows that ZnO-BPE has a lower band gap energy than pure ZnO (without extract addition).

The optical band gap energies were calculated using the Tauc equation, and the plots are shown in **Figure 4**. The calculated band gap energies of ZnO-BPE synthesized at varying pH levels, were observed to range between 3.17 eV and 3.20 eV, slightly lower than the band gap energy of pure ZnO, which is typically 3.24 eV. Among the different pH conditions, the method involving the addition of plant extracts at pH 7 yielded the smallest band gap energy of 3.17 eV.

The observed reduction in band gap energy is of considerable importance for photocatalytic applications. A lower band gap allows the material to absorb lower-energy photons or light of longer wavelengths, thereby expanding its ability to utilize a broader portion of the light spectrum. This characteristic is essential for enhancing photocatalytic efficiency, as it promotes the more effective generation of electron-hole pairs under light irradiation. These charge carriers are crucial for initiating photocatalytic reactions, such as the degradation of organic contaminants, which rely on their reactive interactions to achieve efficient pollutant breakdown.

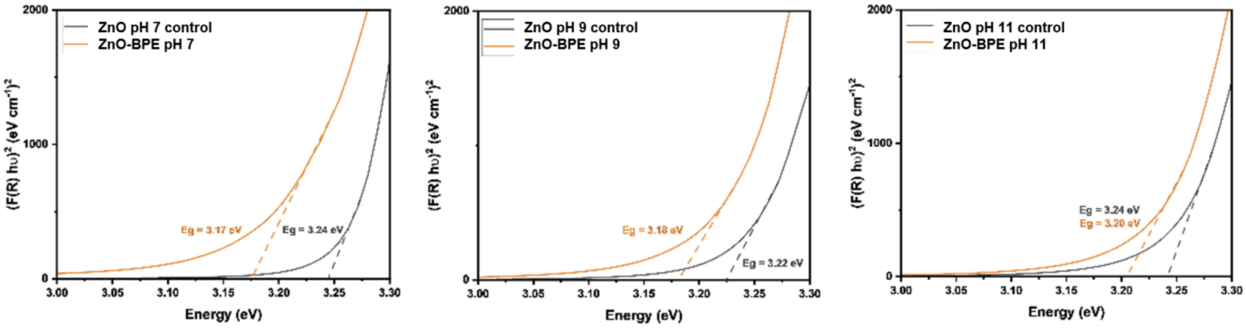

Figure 4. Zinc oxide band gap energy

The reduction in band gap energy observed in ZnO-BPE relative to pure ZnO is likely influenced by the bioactive compounds in banana peel and tea leaf extracts. These natural compounds may interact with the surface or influence the electronic configuration of ZnO particles during the synthesis process, inducing changes in their optical behavior. Furthermore, the differences in band gap energy on pH variations underscore the critical importance of optimizing synthesis parameters to achieve the desired photocatalytic properties of the material for enhanced performance.

Photodegradation of Rhodamine B

The synthesized ZnO-BPE was performed for photocatalytic degradation to determine their activity in degrading rhodamine B. The results show that the optimal degradation of Rhodamine B dye was achieved under synthesis conditions with the inclusion of banana peel and black tea leaf extracts. The photodegradation efficiencies were similar at pH 7 and 11, reaching around 61.7% and 62.6%, respectively (**Figure 5**). However, pure ZnO at pH 11 exhibited lower degradation efficiency than pH 7. This phenomenon can be attributed to the influence of pH on the distribution of particle sizes. In neutral conditions (pH 7), hydroxyl ions (OH^-) interact with positively charged Zn^{2+} ions, facilitating the synthesis of ZnO particles [24]. However, at elevated pH levels, the increase in crystallite size diminishes the available reaction surface area, thereby impacting the efficiency of the process [25].

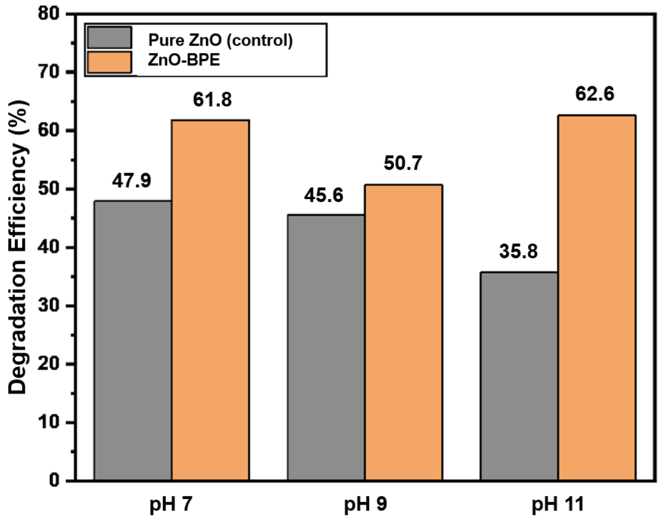

Figure 5. Photodegradation efficiency of ZnO particles towards rhodamine B based on synthesis conditions

4. CONCLUSIONS

Cotton-like ZnO particles were successfully synthesized using banana peels and tea leaf extracts that act as bio-capping agents. Adding banana peels and tea leaf extracts, as a representation of the green chemistry approach, effectively prevents ZnO particle agglomeration, enhancing photodegradation efficiency by up to 20% over pure ZnO. The surface area of ZnO particles is also impacted by the pH conditions maintained during their synthesis. The obtained ZnO particles using the green chemistry approach show a higher rhodamine B degradation efficiency.

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