



# Green Chemistry Synthesis of Ag/ZnO (ashwater) Using Bark Ash Water and Mango Extract for Enhanced Photocatalytic Degradation of Rhodamine B

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## Abstract

Ag/ZnO (BAW) was synthesized via a green chemical precipitation method and evaluated as a visible-light photocatalyst for Rhodamine B (RhB) degradation. X-ray diffraction (XRD) analysis confirmed the coexistence of hexagonal wurtzite ZnO and metallic Ag phases, while energy-dispersive X-ray spectroscopy (EDX) verified the presence of Zn, O, and Ag. Scanning electron microscopy (SEM) revealed mixed rod-like and short tubular morphologies. UV-Vis diffuse reflectance spectroscopy demonstrated enhanced visible-light absorption of Ag/ZnO (BAW) compared with ZnO (BAW), and Tauc plot analysis showed a reduction in the optical band gap from 3.16 to 3.10 eV after Ag incorporation. Under visible-light irradiation, Ag/ZnO (BAW) achieved 99.69% RhB degradation within 120 min, compared with 89.51% for ZnO (BAW). The degradation followed a pseudo-first-order kinetic model, with Ag/ZnO (BAW) exhibiting an apparent rate constant of  $0.0428 \text{ min}^{-1}$ , approximately 2.6 times higher than that of ZnO (BAW) ( $0.0165 \text{ min}^{-1}$ ). The addition of 1,4-benzoquinone (BQ) markedly suppressed RhB degradation, suggesting the involvement of superoxide radicals ( $\text{O}_2^{\bullet-}$ ) in the photocatalytic mechanism. However, the possible contributions of other reactive species cannot be excluded. The enhanced photocatalytic activity of Ag/ZnO (BAW) is associated with improved light absorption and enhanced photoinduced charge-carrier separation induced by Ag incorporation. These findings demonstrate the potential of Ag/ZnO (BAW) as an efficient and environmentally friendly photocatalyst for dye degradation under visible-light irradiation.

**Keywords :** Ag/ZnO; Green synthesis; Photocatalysis; Rhodamine B; Pseudo-first-order

## Introduction

Rhodamine B (RhB) is widely utilized as a synthetic dye, particularly applied in the textile industry and the manufacturing of various commercial products, including ballpoint pens, paints, leather goods, dye lasers, carbon paper, stamp pad inks, fireworks, and explosive materials [1]. In the case of Rhodamine utilization, it has raised serious environmental concerns that RhB has been reported to induce mutagenic and oncogenic effects in living organisms [2, 3]. Consequently, effective RhB removal is essential for environmental

remediation and human health protection. Metal oxide photocatalysts have attracted considerable attention for dye degradation because they can utilize UV and visible light to generate reactive species that decompose pollutants. Among them, ZnO has been widely studied due to its excellent electrical, optical, and catalytic properties, showing significant potential for the removal of persistent synthetic dyes from wastewater. There are various synthesis methods of ZnO, such as thermal decomposition, hydrothermal treatment, sol-gel processing, and chemical precipitation, have been used. Among these methods, the chemical precipitation

method is widely employed because of its simplicity, cost-effectiveness, and controllability of material particle size and morphology. To handle posing environmental problems, a green synthesis method has been chosen instead of previous approaches avoiding hazardous chemicals and by products generated during the process. The green method has started by using plant extracts or waste-derived precursors for reducing and stabilizing agents. This method decreases dangerous chemicals created in the process, reduces production costs, and aligns with principles of sustain-able chemistry [4].

Recent studies of the potential capability of ZnO nanoparticles in the dye degradation process. ZnO has been synthesized using Hibiscus rosa-sinensis leaf extract and has shown effective RhB degradation under sunlight [5]. In addition, polysaccharide is applied for ZnO nanoparticles synthesis using *Sechium edule* showing high photocatalytic performance under visible light irradiation [6]. Moreover, ZnO nanoparticles synthesized from *Cyanometra remiflora* leaf extract illustrated comparable activity, confirming the capability of green synthesis methods. Despite this capability, the wide band gap of ZnO (about 3.2 eV) restricts its excitation mainly to UV light, limiting its efficiency under natural sunlight, which is dominated by visible light. To overcome these challenges, surface modification and metal nanoparticle doping have been extensively studied to enhance visible-light-driven photocatalysis. Ag particle added into the process has shown remarkable effectiveness in reducing the band gap, extending the adsorption, and improving separation efficiency. The silver on nanoparticles are helpful to transfer and suppress electron hole recombination, leading to a higher stage of photocatalytic activity [7]. Hybrid photocatalysts, such as Ag-modified Al-doped ZnO (AZO), have demonstrated excellent performance in both hydrogen production and dye degradation under UV and visible light irradiation [8]. The improvement of photocatalytic activity is primarily controlled by the efficiency of charge transfer (CT) at the interface between metals, such as Ag, and semiconductor materials, such as ZnO. These interfacial interactions play a major role in supporting the rapid migration of photogenerated electron-hole pairs leading to restrain their

recombination and significantly improving light adsorption capability.

In term of these considerations, the current study focuses on the green synthesis of Ag/ZnO (BAW) photo catalysts. ZnO (BAW) is prepared by using bark ash water as a natural precursor, while Ag is extracted through mango extract [9] through a simple precipitation method. The photocatalytic efficiency of the synthesized Ag/ZnO (BAW) is subsequently studied and compared with the pure bark ash water. As a result, their performance in the RhB dye degradation is particularly emphasized under light irradiation.

## Objectives

To study the synthesis of Ag/ZnO (BAW) through a green chemical method using bark ash water as a main precursor as well as mango extract as a reducing and stabilizing agent, and to evaluate its photocatalytic capability efficiency in the RhB degradation compared with the bark ash water.

## Methods

### 1. Synthesis of Zinc Oxide from Bark Ash Water using the Co-precipitation method

Zinc oxide (ZnO) was synthesized from Bark Ash Water using a co-precipitation method adapted from Phongarthit et al. (2019). Briefly, 2.19 g. of zinc acetate ( $\text{Zn}(\text{CH}_3\text{COO})_2$ ) was dissolved in 20 ml. of deionized water under stable stirring. Consequently, 50 ml. of Bark Ash Water with pH value 12 was added into the solution over 3 minutes with continuous stirring. The mixture was heated to 80 degrees Celsius and stirred for an hour before finishing and allowing precipitation to occur. The precipitated material was repeatedly washed with deionized water and ethanol to remove residual impurities, followed by filtration and drying at 100 degrees Celsius for an hour. The dried material was ground into a fine powder and subsequently calcined in a high temperature furnace at 900 degrees Celsius for an hour to obtain ZnO derived from Bark Ash Water (ZnO (BAW)). The final obtained product was ground again after calcination to achieve equal particlesize

## 2. Preparation of Ag/ZnO

To prepare the Ag/ZnO (BAW) composite material, 0.1 g of calcined ZnO (BAW) powder was dispersed in 50 ml of a silver nanoparticle solution synthesized from mango extract. Fresh mango fruits were washed thoroughly, peeled, rinsed with deionized water, and cut into small pieces. A 2.5 g portion of the mango flesh was mixed with 100 mL of deionized water and heated at 50 °C for 1 h. After cooling to room temperature, the extract was filtered through Whatman No. 1 filter paper. The preparation of the mango extract and the synthesis of silver nanoparticles (AgNPs) were performed according to the method reported by Ninaksorn et al. [2023]. The extract was added into the ZnO suspension under continuous stirring for an hour to ensure homogeneous deposition of Ag on the ZnO surface. The result of the precipitation was collected by filtration, thoroughly rinsed with deionized water to remove residual impurities, and subsequently dried in a hot-air oven for an hour. The dried material was finely ground into powder to be the Ag/ZnO (BAW) composite, which was used for photocatalytic efficiency testing in the degradation of Rhodamine B (RhB).

## 3. Characterization of Ag/ZnO (BAW)

SEM-EDS analysis confirmed the formation and elemental distribution of the Ag/ZnO (BAW) composite. The optical properties were evaluated by UV-Vis diffuse reflectance spectroscopy, and XRD analysis verified the hexagonal wurtzite ZnO structure with additional peaks attributed to metallic Ag, confirming the successful formation of Ag/ZnO (BAW).

## 4. Photocatalytic Study

The photocatalytic activity of Ag/ZnO (BAW), synthesized using ZnO (BAW) prepared from Bark Ash Water, was examined based on its performance to degrade Rhodamine B (RhB) dye under visible light irradiation. RhB solution with concentration of  $1.0 \times 10^{-5}$  M was prepared as the prototype pollutant. For each experiment, 0.1 g of the Ag/ZnO (BAW) catalyst was added to 250 ml beaker containing 150 ml. of the RhB solution. The suspension was first stirred in the dark for 30 minutes to generate adsorption-desorption equilibrium, after which it was allowed to settle for 2 minutes.

Then, a 3 ml. aliquot of the supernatant was subsequently withdrawn and centrifuged at 3,000 rpm for 3 minutes to remove any residual catalyst particles, and the result of clear solution was retained for further analysis. Following this step, The suspension was irradiated under visible light using a 25 W xenon lamp fitted with a 420 nm long-pass optical filter to exclude ultraviolet irradiation. The distance between the light source and the reaction vessel was fixed at 16 cm, and the suspension was continuously stirred throughout the photocatalytic reaction. At predetermined irradiation times (10, 20, 30, 45, 60, 90, and 120 min), aliquots were withdrawn, allowed to settle, and subsequently centrifuged before UV-Vis spectrophotometric analysis. The absorbance of each sample was then measured using a UV-Vis spectrophotometer. The photocatalytic degradation efficiency of RhB was calculated from the absorbance values at different time intervals using Equation (1):

$$\%D.E. = (C_0 - C_t) / C_0 \times 100 \quad (1)$$

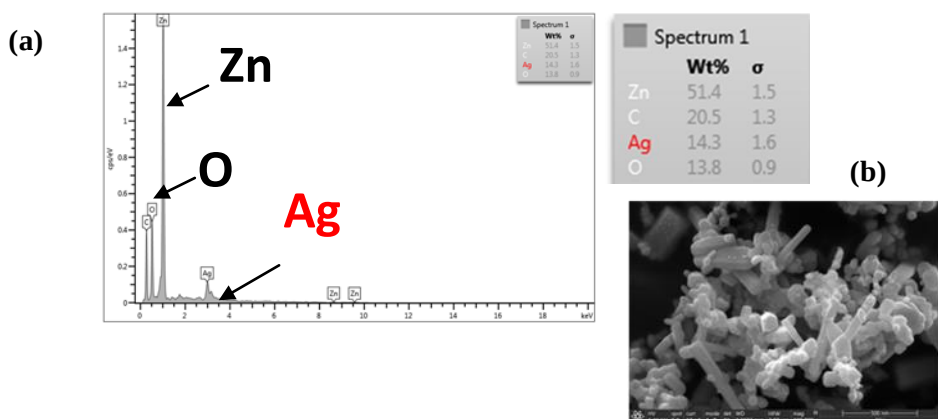
where  $C_0$  and  $C_t$  represent the initial concentration and the concentration of Rhodamine B at irradiation time  $t$ , respectively. According to the Beer-Lambert law, the absorbance of Rhodamine B is directly proportional to its concentration under the experimental conditions employed. Therefore, the concentration ratio ( $C_t/C_0$ ) was determined from the corresponding absorbance ratio ( $A_t/A_0$ ).

## Results and Discussion

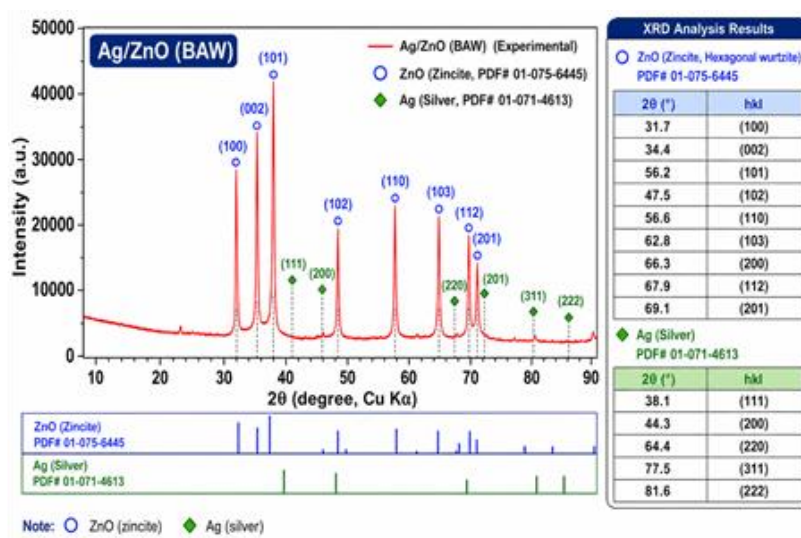
The synthesized Ag/ZnO (BAW) was characterized using Energy-dispersive X-ray spectroscopy (EDX) to determine its elemental composition and to confirm the successful incorporation of Ag on the ZnO (BAW) surface. As illustrated in **Figure 1 (a)**, The EDX analysis confirmed the presence of Zn, O, and Ag, indicating the successful formation of the Ag/ZnO (BAW) composite. The Ag loading was estimated to be 14.3 wt%, demonstrating the effective deposition of Ag onto the ZnO (BAW) surface. The deposited Ag nanoparticles are expected to form a metal-semiconductor (Schottky) junction with ZnO (BAW), which promotes interfacial electron transfer, suppresses the recombination of photogenerated electron-hole pairs, and consequently enhances the

photocatalytic activity under visible-light irradiation. The morphological characteristics of the composite were further investigated by

Scanning Electron Microscopy (SEM), as shown in **Figure 1(b)**, revealing a mixture of rod-like and short tubular structures.



**Figure 1** EDX spectrum of Ag/ZnO (BAW) (a) and morphology of Ag/ZnO (BAW) synthesized via the coprecipitation method (b)



**Figure 2** XRD pattern of Ag/ZnO (BAW), showing the characteristic diffraction peaks of hexagonal wurtzite ZnO and metallic Ag. The observed peaks agree well with the standard reference patterns PDF# 01-075-6445 for ZnO and PDF# 01-071-4613 for Ag, confirming the formation of the Ag/ZnO (BAW) composite

From **Figure 2** The XRD pattern confirmed that the synthesized Ag/ZnO (BAW) composite predominantly exhibited the hexagonal wurtzite crystal structure of ZnO (BAW). In addition, characteristic diffraction peaks corresponding to metallic Ag were observed, confirming the successful formation of the Ag/ZnO (BAW)

composite. The absence of any noticeable structural changes in the ZnO (BAW) diffraction pattern indicates that the incorporation of Ag did not alter the crystal structure of ZnO

**Figure 3 (c)** presents the UV-Vis diffuse reflectance (UV-Vis DR) spectra of the samples. The strong absorption observed at wavelengths

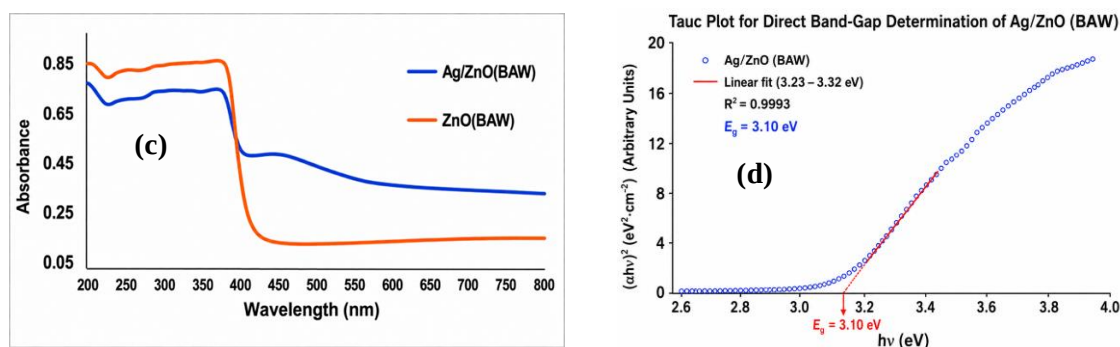
below 400 nm is attributed to the electron transition from the valence band to the conduction band of ZnO (BAW). Significantly, an additional absorption band emerges at wavelengths above 400 nm in the visible region. Notably, the spectrum of Ag/ZnO (BAW) displays a importantly broader absorption band in the visible range compared to that the pure ZnO (BAW). The Tauc plot analysis in **Figure 3 (d)** revealed that the estimated band-gap energy decreased from approximately 3.18 eV for ZnO (BAW) to 3.10 eV for Ag/ZnO (BAW). This slight reduction in the apparent band-gap energy is associated with the modification of the ZnO (BAW) surface by Ag nanoparticles. In addition, Ag nanoparticles enhance visible-light absorption through their surface plasmon resonance (SPR) effect, enabling the photocatalyst to harvest a broader portion of the visible-light spectrum. These effects facilitate the separation of photogenerated electron-hole pairs and contribute to the enhanced photocatalytic degradation of Rhodamine B under visible-light irradiation.

**Figure 4 (e)** shows the time-dependent degradation spectra of Rhodamine B (RhB), with the characteristic absorption peak at 554 nm gradually decreasing during visible-light irradiation, confirming the photocatalytic activity of Ag/ZnO (BAW). **Figure 4 (f)** compares the photocatalytic degradation of RhB over ZnO (BAW), Ag/ZnO (BAW), and Ag/ZnO (BAW) in the presence of 1,4-benzoquinone (BQ). After 120 min, Ag/ZnO (BAW) achieved

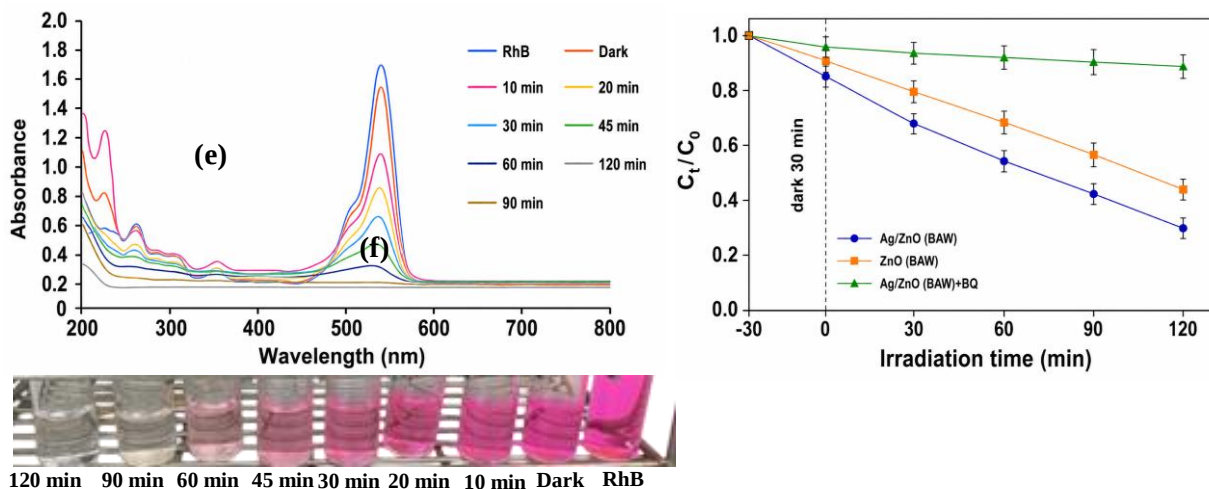
99.69% degradation, compared with 89.51% for ZnO (BAW), whereas the addition of BQ reduced the degradation efficiency to 25.38%. These results demonstrate the enhanced photocatalytic performance of Ag/ZnO (BAW) and suggest the involvement of superoxide radicals ( $O_2^{\bullet-}$ ) in the RhB degradation mechanism, although the possible contributions of other reactive species cannot be excluded.

The photocatalytic degradation kinetics were evaluated using the pseudo-first-order model. As shown in **Figure 5**, the approximately linear relationship between  $\ln(C_0/C_t)$  and irradiation time indicates pseudo-first-order behavior. Ag/ZnO (BAW) exhibited an apparent rate constant ( $k$ ) of  $0.0428 \text{ min}^{-1}$ , approximately 2.6 times higher than that of ZnO (BAW) ( $0.0165 \text{ min}^{-1}$ ), demonstrating its enhanced photocatalytic activity. In the presence of 1,4-benzoquinone (BQ), the rate constant decreased markedly to  $0.0021 \text{ min}^{-1}$ , suggesting the involvement of superoxide radicals ( $O_2^{\bullet-}$ ) in RhB degradation.

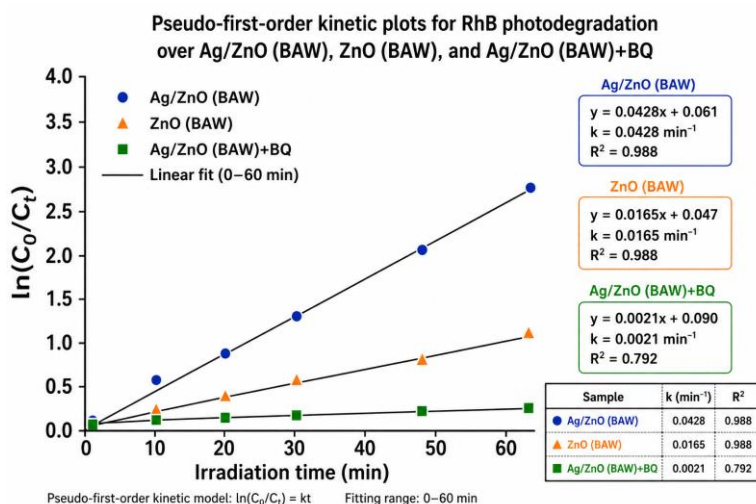
This was demonstrated by the addition of 1,4-benzoquinone a well-established scavenger for superoxide radicals, to the Rhodamine B solution containing the Ag/ZnO (BAW) photocatalyst. Under these conditions, the degradation efficiency decreased significantly, indicating that superoxide radicals play an important role in the photocatalytic degradation process. The experimental results, presented in **Figure 6**.



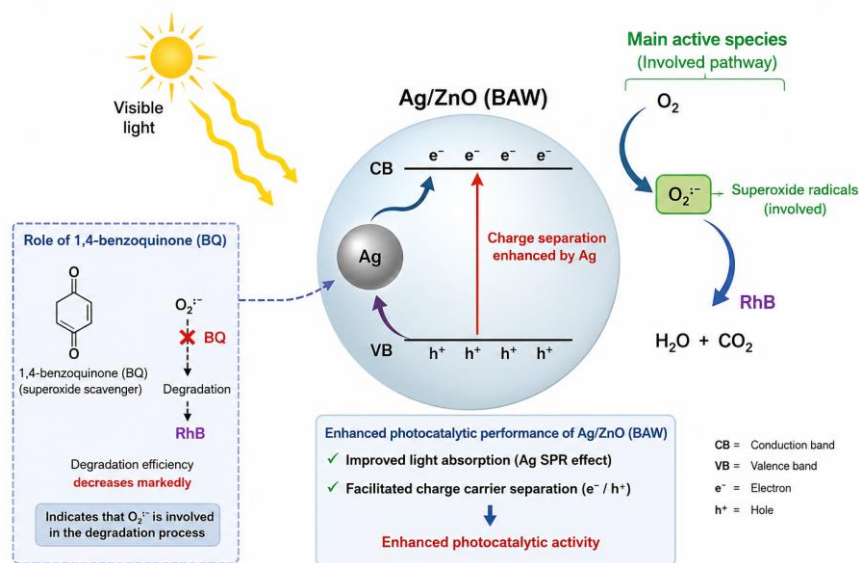
**Figure 3** (c) UV-vis DR spectra of Ag/ZnO (BAW) compared with ZnO (BAW)  
(d) Tauc Plot for Direct Band-Gap Determination of Ag/ZnO (BAW)



**Figure 4** (e) Time-dependent UV-Vis absorption spectra of RhB over Ag/ZnO (BAW) (f) Photocatalytic degradation of Rhodamine B over ZnO (BAW), Ag/ZnO (BAW), and Ag/ZnO (BAW) with 1,4-benzoquinone (BQ) under visible-light irradiation. Prior to irradiation, the suspension was equilibrated in the dark for 30 min. Error bars represent mean  $\pm$  SD ( $n = 3$ ).



**Figure 5** Pseudo-first-order kinetic plots for the photocatalytic degradation of Rhodamine B over ZnO (BAW), Ag/ZnO (BAW), and Ag/ZnO (BAW) in the presence of (BQ) benzoquinone under visible-light irradiation.



**Figure 6** Possible scheme for charge-carrier transfer process in a visible light-irradiated Ag/ZnO (BAW) system

**Table 1** Comparison of the photocatalytic performance of Ag/ZnO (BAW) with previously reported ZnO-based photocatalysts

| Reference                        | Photocatalyst                                                    | Pollutant                                                  | Light Source                         | Time (min) | Degradation (%)          | Band Gap (eV)   | Rate Constant, $k$ ( $\text{min}^{-1}$ ) |
|----------------------------------|------------------------------------------------------------------|------------------------------------------------------------|--------------------------------------|------------|--------------------------|-----------------|------------------------------------------|
| <b>Chai et al. (2019)</b>        | Green synthesized ZnO NPs (using <i>Hibiscus rosa-sinensis</i> ) | Palm Oil Mill Effluent (POME)                              | UV Pen-Ray Lamp (254 nm)             | 240        | 82% (COD removal)        | 3.34            | Not Specified                            |
| <b>Hadi &amp; Jasim (2025)</b>   | Ag/ZnO nanocomposites                                            | Brilliant Blue (BB) dye                                    | UV light (UVA LED)                   | 60         | 99.87% (at 10 mg/L)      | ~3.37 (for ZnO) | Not Specified                            |
| <b>Phongarthit et al. (2020)</b> | AgCl/ZnO (Green approach)                                        | Rhodamine B (RhB), Reactive Orange (RO), Bisphenol A (BPA) | Visible Light (25W Xe lamp, >420 nm) | 60         | >90% (RhB and BPA)       | ~3.2            | 0.07318 (RhB)                            |
| <b>Bharathi et al. (2022)</b>    | Green synthesized ZnO NPs (using <i>Sechium edule</i> )          | Rhodamine B (RhB)                                          | Visible Light (Tungsten halogen)     | 75         | 95%                      | 3.08            | Not Specified                            |
| <b>Trang et al. (2020)</b>       | AZO-20 (Ag@ZnO hybrid nanostructure)                             | Rhodamine B (RhB)                                          | UV and Visible Light                 | 120        | 97% (UV) / 82% (Visible) | 3.07            | 0.025 (UV) / 0.014 (Visible)             |
| <b>This work</b>                 | Ag/ZnO (BAW)                                                     | Rhodamine B (RhB)                                          | RhB Visible                          | 120        | 99.69%                   | 3.10            | 0.0428 (RhB)                             |

As shown in Table 1, the photocatalytic performance of Ag/ZnO (BAW) was compared with previously reported ZnO and Ag/ZnO-based photocatalysts. The present Ag/ZnO (BAW) exhibited a high RhB degradation efficiency of 99.69% within 120 min under visible-light irradiation, with an optical band gap of 3.10 eV and an apparent rate constant of  $0.0428 \text{ min}^{-1}$ . Although direct comparison among studies should be made with caution because of differences in photocatalyst composition, pollutant type and concentration, catalyst dosage, light source, and experimental conditions, these results demonstrate the competitive photocatalytic performance of Ag/ZnO (BAW) relative to previously reported ZnO-based systems.

## Conclusions

This study successfully synthesized Ag/ZnO (BAW) via a coprecipitation method. XRD and EDX analyses confirmed the coexistence of hexagonal wurtzite ZnO (BAW) and metallic Ag, while SEM revealed rod-like and short tubular morphologies. UV-Vis DR and Tauc plot analyses showed enhanced visible-light absorption and a reduced band gap from 3.16 eV for ZnO (BAW) to 3.10 eV for Ag/ZnO (BAW). Ag/ZnO (BAW) achieved 99.69% Rhodamine B degradation within 120 min, compared with 89.51% for ZnO (BAW). The degradation followed pseudo-first-order kinetics, with Ag/ZnO (BAW) exhibiting a rate constant of  $0.0428 \text{ min}^{-1}$ , approximately 2.6 times higher than ZnO (BAW) ( $0.0165 \text{ min}^{-1}$ ). The scavenger experiment using 1,4-benzoquinone (BQ), a well-established scavenger for superoxide radicals, indicated that superoxide radicals ( $\text{O}_2^{\cdot-}$ ) are involved in the photocatalytic degradation of Rhodamine B, as the degradation efficiency decreased markedly after the addition of BQ. The enhanced performance of the Ag/ZnO (BAW) composite is attributed to the presence of silver, which improves light absorption and facilitates charge carrier separation, thereby enhancing photocatalytic activity. Overall, Ag incorporation enhanced light absorption and photocatalytic activity, demonstrating the potential of Ag/ZnO (BAW) for efficient dye degradation.

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