

## Enhanced Photocatalytic Degradation of Maxillon Blue Dye (GRL) by Using ZnO NPs in Aqueous Solutions

Esraa Ahmed Said\*, Mohammed Hadi<sup>1</sup>, Hasan Mohammed Abdullah<sup>2</sup>,  
Fadhil A. Rasen<sup>3</sup> and Montather F. Ramadan<sup>4</sup>

Department of Dentistry, Al-Noor University College, Nineveh, Iraq

<sup>1</sup>College of Medical Technology/ Medical Lab Techniques, Al-Farahidi University, Baghdad, Iraq

<sup>2</sup>Department of Optical Techniques, Al-Zahrawi University College, Karbala, Iraq

<sup>3</sup>Department of Medical Engineering, Al-Esraa University College, Baghdad, Iraq

<sup>4</sup>College of Dentistry, Al-Ayen University, Thi-Qar, Iraq

✉ esraa.ahmed@alnoor.edu.iq

*Received June 17, 2023; revised and accepted June 30, 2023*

**Abstract:** The ZnO nanoparticles (ZnO NPs) catalyst was prepared using the hydrothermal method. Properties of nanoparticles were investigated by utilising some techniques (FE-SEM, TEM, and EDX). The photocatalytic degradation of Maxillon Blue (GRL) under UV irradiation has been studied. The use of zinc oxide as a catalyst in the photo catalysis process under different experimental conditions. The influence of several factors includes the effect of mass catalyst and the concentration of Maxillon blue (GRL) dye. Through the results, the photocatalytic efficiency increases by increasing the weight of the ZnO NPs catalyst starting from 0.1 to 0.4 g per 200 mL, also when the amount of ZnO NPs increased, the rate of degradation goes up first and then keeps unalterable on a certain scale, but when the quantity reaches to 0.4 g per 200 mL, the rate of degradation shows slight change. The highest photodegradation efficiency reaches 87% indicating a good surface activity.

**Key words:** Photocatalytic, ZnO nanoparticles, Maxillon Blue (GRL), advanced oxidation processes.

### Introduction

Water pollution is one of the utmost important and serious problems faced by humanity at this time; in addition to the pollution of drinking water with industrial dye, due to its widespread use in industry, such as dyeing fabrics and papers, as well as its presence in very high concentrations in the water that prevents the metabolism of marine organisms, so it was classified as one of the most dangerous pollutants. There are many modern, easy and inexpensive ways to get rid of pollutants, one of utmost importance is photocatalytic degradation (Abdulrazzak, 2016; Aljeboree et al., 2019; Aseel et al., 2019; Abbas et al., 2020; Alkaim

and Ajobree, 2020; Chkirida et al., 2021; Wared and Radia, 2021). Therefore, in the present time, water pollution is one of the most dangerous and unwanted problems that most countries of the world suffer from, as a result of waste emitted from factory and hospital waste (Alqaragully et al., 2015; Aljeboree et al., 2023). With regard to living organisms and their infection with many diseases, there are many methods of treatment to conserve water from pollutants, the most important methods of wastewater treatment, include ozone adsorption and photo-oxidation, advanced oxidation processes (AOPs), use of catalysts with ultraviolet or near ultraviolet rays, to get rid of dyes, proven photocatalytic oxidation, especially ZnO NPs photocatalysis,

\*Corresponding Author

is a very effective technique for the disposal of resistant dyes (Abdulrazzak, 2016). Zinc oxide is an *n*-type semiconductor with a broad direct band gap of 3.37 eV, and it is a promising photo-catalytic material due to its low- cost, high photo catalytic activity and environmental friendliness (Aneesh et al., 2007; Amin et al., 2011). Chemically and photo punctures are highly oxidised. Therefore, zinc oxide is considered to have a high ability to oxidise and convert dangerous organic compounds into non-hazardous environmentally friendly compounds.

### Experimental Part

The textile dye (Maxillon Blue GRL) was obtained from the textile fabric factory (Al-Hilla - Iraq). Where a standard solution of dye was prepared at a concentration of 1000 mg/L, a weight of ZnO NPs of about (1.0 g) was taken and dissolved in 1000 ml of DW. As a model basic dye GRL, the chemical structure appears in Figure 1a, chemical formula:  $C_{20}H_{26}N_4O_6S_2$ ,  $\lambda_{max} = 599$  nm.

To calculate the  $\Lambda_{max}$  of (Maxillon Blue GRL) dye the UV-Visible spectroscopy of the GRL solution was recorded within a range of 200-1000 nm.  $\Lambda_{max}$  GRL= 599 appears in Figure 1.

### Preparation of Zinc Oxide NPs by Hydrothermal Process

The Hydro-thermal method was used to prepare Zinc Oxide NPs by means of three steps: Step one: zinc acetate ( $Zn(CH_3COO)_2 \cdot 2H_2O$ ) about (5 g) used as a source of zinc dissolved in 25 ml DW with stirred in magnetic stirrer 10 minutes. Step two: oxalic acid  $(COOH)_2 \cdot 2H_2O$  about 4 g was added in 50 ml of DW and stirred for about 15 minute. Step three: Under optimum conditions, with continuation of experiment of step one and two gradually mixture by stirring for 15

minutes to give a white solution. After that transferred into a Teflon-lined sealed stainless steel auto-clave and maintained in an oven at 165 °C for 24 hr. These steps yielded a white product that was washed several times by DW and dried at 75 °C overnight. After this step, the powder was calcined at 500 °C for 2h to obtain nano powder that was used in the experiment (Aneesh et al., 2007; Amin et al., 2011) .

### Experiments of the Photocatalytic

The photocatalytic activity of the zinc oxide nanoparticles catalyst was determined via the degradation of GRL. Wholly, experiments were carried out in a beaker of 250 ml. The reaction was placed under ultra-violet light, taking into account the distance between the surface of the solution and the light source. Before each test, to obtain accurate results, the lamp is heated for 10 minutes. Thus, a weight of ZnO NPs of about 0.2 g was added to solution GRL with a 200 ml beaker reaction, and the experiment was initially conducted for 10 mins. Usually called the adsorption (or dark process). Influence of different parameters such as weight of ZnO NPs (0.1–0.4 g L<sup>-1</sup>), the concentration of GRL (10–50 mg·L<sup>-1</sup>). The PDE% of GRL and apparent rate first order constant were calculated using Equation 1

$$PDE (\%) = (C_0 - C_t) / C_0 \times 100 \quad (1)$$

where  $C_0$ : Concentration of GRL dye and  $C_t$  concentration of photolysis (mgL<sup>-1</sup>).

## Results and Discussion

### Characterisation

The FE-SEM image was used to study the morphology and properties of ZnO NPs as appears in Figure 2a. Micro-graphs of ZnO NPs show that the surface

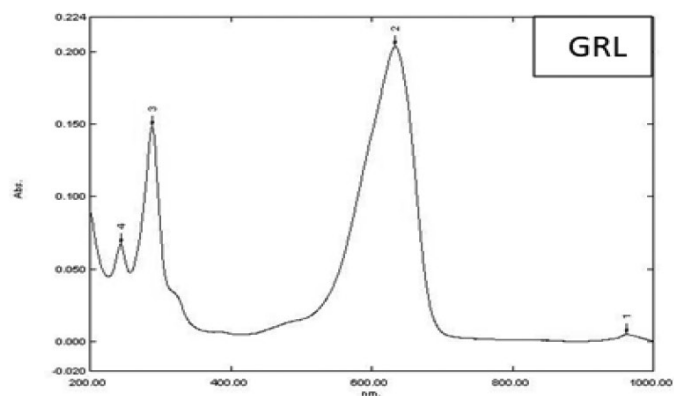
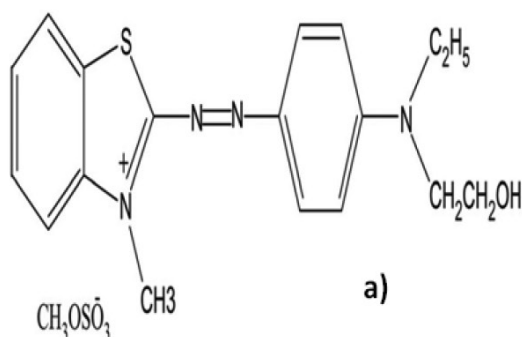


Figure 1: The chemical structure (a) Maxillon Blue GRL dye and (b) maximum wavelength of the textile dye (Maxillon Blue GRL dye).

has large, irregular, coarse, particles, and also data from spherical clusters resulting from ZnO NPs (Al-Mashhadani et al., 2021). Figure 2b showed the TEM image of ZnO NPs, where results from spherical clusters resulting inside surface. EDX of ZnO NPs is shown in Figure 2c. The presence of O, C, and Zn indicates the existence of ZnO. The lowest and highest values of the elements that existed in the modified zinc oxide weighed 76.5 wt.% and 8.7 wt.%, respectively (Ali et al., 2019).

### Effect of Different Parameters on Photocatalytic Efficiency

#### *Effect of Weight on ZnO NPs*

The effect of the weight of zinc oxide (ZnO NPs) catalyst was studied by taking weights of (0.1-0.4 g) under experimental conditions for each of the

temperatures 25°C, dye concentration 40 mg/L, and pH 6 as appeared in Figures 3 and 4.

When the quantity of ZnO NPs rises, the rate of degradation goes up first and then keeps getting unalterable on a certain scale, but when the quantity goes to 0.4 g/L as compared with 0.3 g/L, the rate of degradation showed little change; therefore, 0.3 g/L was chosen the best amount of ZnO NPs giving PDE% of about 85.3% . It is possibly due to the aggregation and the effect of screening (Sauer et al., 2002).

When the weight of the zinc oxide NPs is lower than the ideal value, the effectiveness of catalyst ZnO NPs and the absorption of the light are the main parameters that limit the rate of photocatalytic degradation (Akyol et al., 2004; Wang et al., 2007). The primary rise in the rate of GRL degradation with a rise in the quantity of

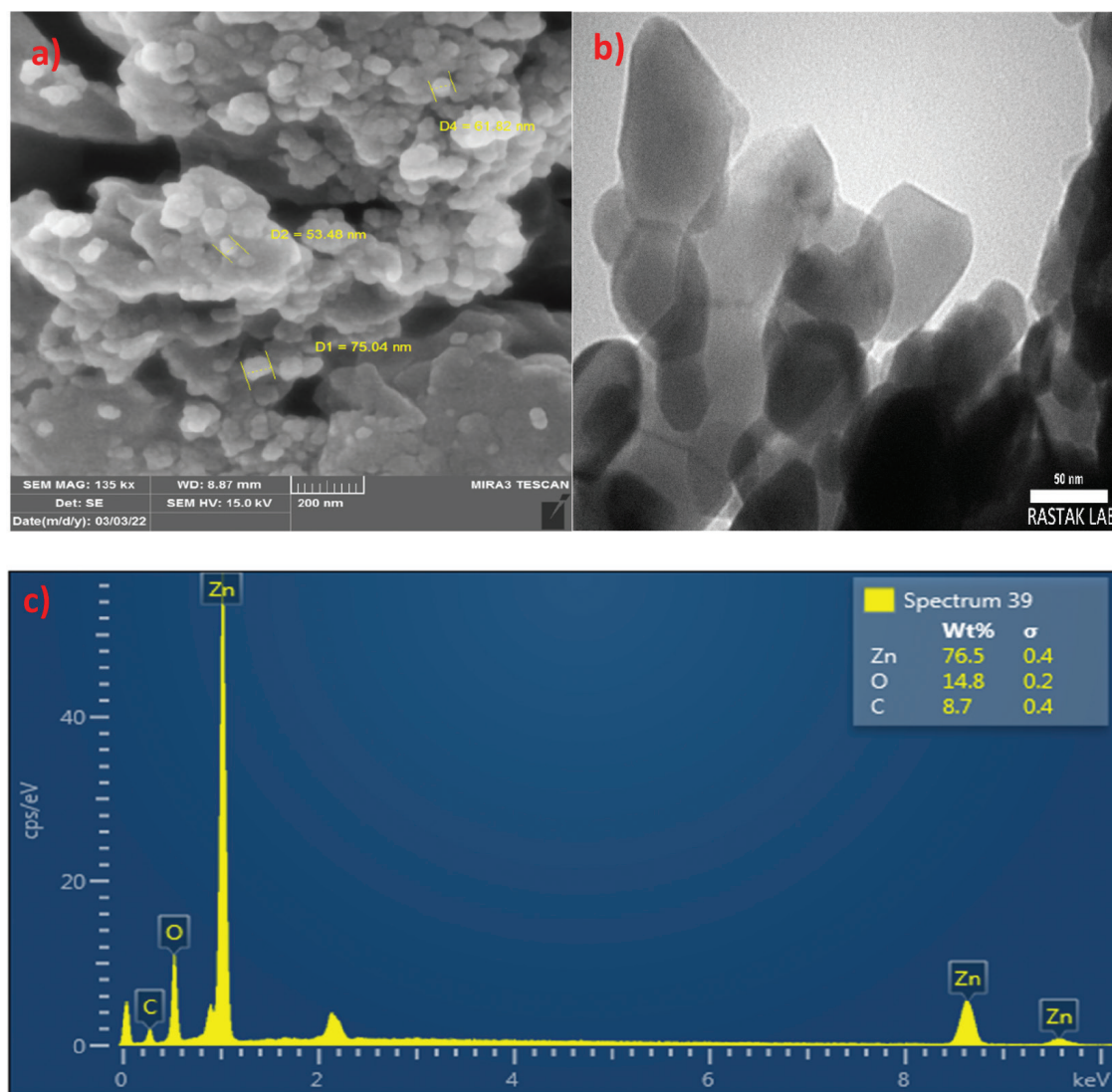


Figure 2: (a) FESEM of ZnO NPs, (b) TEM of ZnO NPs and (c) EDX of ZnO NPs.

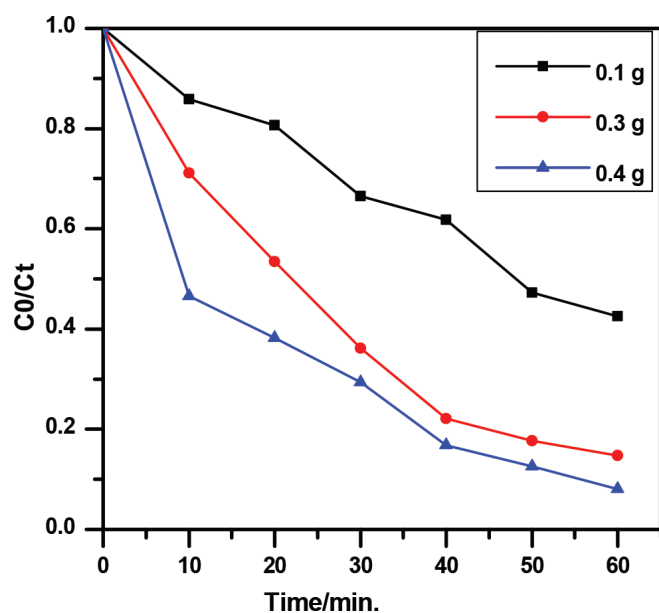


Figure 3: Effect weight of ZnO NPs on photo catalytic de-gradation of GRL dye.

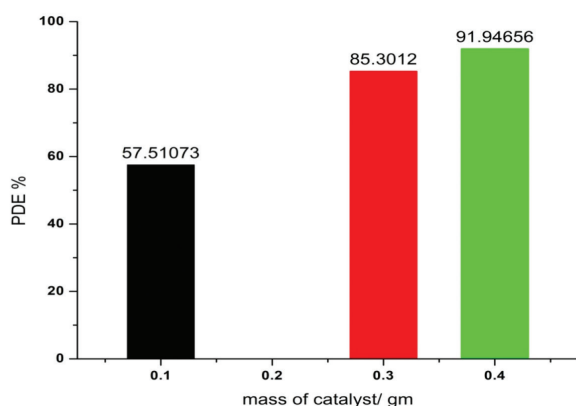


Figure 4: Effect of the weight of catalyst on photo catalytic degradation efficiency PDE%.

ZnO NPs is because of the rise in the number of active sites on the ZnO NPs of the photocatalyst. Due to this, there is a rise in the number of photo-generated pair electronholes. Thus increasing the number of ( $\text{OH}^\cdot$ ) responsible for the degradation of GRL dye (Kaur et al., 2014; Mashkour et al., 2011; Wang et al., 2007).

#### Effect of GRL Dye Concentration

The primary GRL dye concentration essential plays a pivotal role in deciding the rate of GRL degradation (Daneshvar et al., 2004; Kaur et al., 2014; Pare et al., 2008). In this study, GRL dye concentration was about  $10\text{--}50 \text{ mg}\cdot\text{L}^{-1}$  under optimum conduction temperature  $25^\circ\text{C}$  and mass of ZnO NPs  $0.3 \text{ g/L}$ , as shown in Figures 5 and 6. As the primary GRL concentration

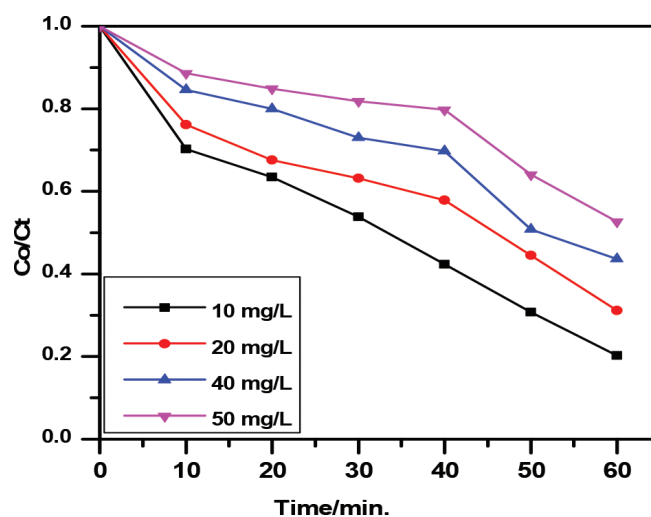


Figure 5: Effect of GRL concentration on to time.

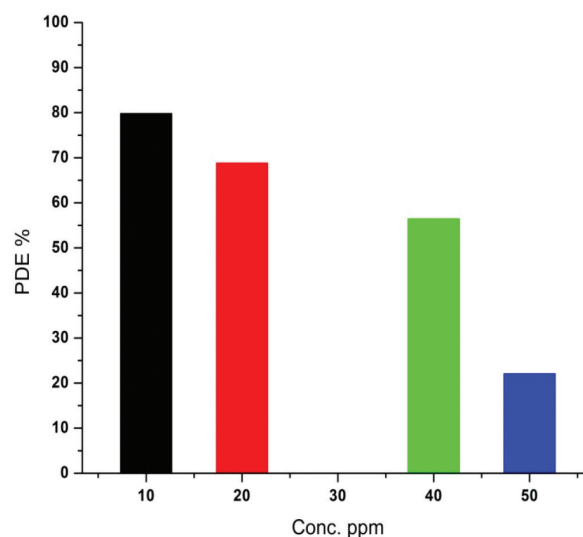


Figure 6: Effect of concentration GRL dye on PDE%.

risks, the rate of photocatalytic degradation decreased. The data reveal that the concentration of GRL affects the degradation capacity severely. With the rise of the concentration of GRL dye, the de-gradation capacity decreases amazingly, especially when the concentration of GRL dye is about  $10\text{--}50 \text{ mg/L}$ . The negative influences of the concentration of GRL dye are ascribed to the competence among GRL and ion  $\text{OH}^-$  adsorption on the ZnO NPs. The adsorption of GRL depresses the ion  $\text{OH}^-$  adsorption, which data in the reduction on the formation of ( $\text{OH}^\cdot$ ). Together, the initial concentration of dye increases and the path length of photons entering the solution decreases. Hence, in the solution with constant catalyst concentration, the formation of ( $\text{OH}^\cdot$ ) that can attack the contaminant decreases, therefore, leading to a



lower de-colonisation capacity (Behnajady et al., 2006; Yang et al., 2010).

### Conclusions

It was found that GRL dye can be easily get degraded by using the photo catalytic process in the presence of zinc oxide NPs as a catalyst under UV irradiation. Several factors affect the photocatalytic process of a dye, these factors increased the photodegradation efficiency. It was found that increasing the weight of the surface (ZnO NPs ) increased the dye's cracking; on the contrary, while increasing concentration of GRL dye, the cracking efficiency would get decreased as the photocatalytic decomposition of GRL was the most effective in the solution with lower primary concentration.

### Reference

- Akyol, A., Yatmaz, H.C. and M. Bayramoglu (2004). Photocatalytic decolorization of Remazol Red RR in aqueous ZnO suspensions. *Appl Catal B: Environ.*, **54(1)**: 19-24.
- Abbas, A.M., Abdulrazzak, F.H., Radhi, I.M., Abdullatif, A.I., Himdan, T.A. and F.H. Hussein (2020). Purification techniques for cheap multi -walled carbon nanotubes. *Journal of Physics: Conference Series*, **11(2)**: 1115-1123.
- Abdulrazzak, F.H., Hussein, F.H., Alkaim, A.F.A., Ivanova, I.E. and D.W. Bahnemann (2016). "Sonochemical/ hydration-dehydration synthesis of Pt-TiO<sub>2</sub> NPs/decorated carbon nanotubes with enhanced photocatalytic hydrogen production activity. *Photochemical and Photobiological Sciences*, **15(11)**: 1347-1357.
- Abdulrazzak, F.H. (2016). Enhance photocatalytic activity of TiO<sub>2</sub> by carbon nanotubes. *International Journal of ChemTech Research*, **9(3)**: 431-443.
- Al-Mashhadani, Z.I., Aljeboree, A.M., Radia, N.D. and O.K.A. Alkadir (2021). Antibiotics removal by adsorption onto eco-friendly surface: Characterization and kinetic study. *International Journal of Pharmaceutical Quality Assurance*, **12(4)**: 252-255.
- Ali, R., Radhi, I.M., Ismail, A.A. and F.H. Abdulrazzak (2019). Modified ZnO for efficient photo-catalysis by silver/graphite oxide nanoparticles. *Journal of Global Pharma Technology*, **11(7)**: 143-150.
- Aljeboree, A.M., Alshirifi, A.N. and A.F. Alkai (2019). Activated carbon (as a waste plant sources)-clay micro/nanocomposite as effective adsorbent: Process optimization for ultrasound-assisted adsorption removal of amoxicillin drug. *Plant Archives*, **19(2)**: 915-919.
- Aljeboree, A.M., Essa, S.M., Kadam, Z., Dawood, F.A., Falah, D.A. and F.A. Ayad (2023). Environmentally friendly activated carbon derived from palm leaf for the removal of toxic reactive green dye. *International Journal of Pharmaceutical Quality Assurance*, **14(1)**: 12-15.
- Alkaim, A.F. and A.M. Ajobree (2020). White marble as an alternative surface for removal of toxic dyes (methylene blue) from aqueous solutions. *International Journal of Advanced Science and Technology*, **29(5)**: 5470-5479.
- Alqaragully, M.B., Al-Gubury, H.Y. Aljeboree, A.M., Karam, F.F. and A.F. Alkaim (2015). Monoethanolamine: Production plant. *Research Journal of Pharmaceutical, Biological and Chemical Sciences*, **6(5)**: 1287-1296.
- Aneesh, P.M., Vanaja, K.A. and M.K. Jayara (2007). Synthesis of ZnO nanoparticles by hydrothermal method. *Nanophotonic Materials*, 66390J.
- Aseel, M. and A.F.A. Aljeboree (2019 ). Role of plant wastes as an ecofriendly for pollutants (crystal violet dye) removal from aqueous solutions. *Plant Archives*, **19(2)**: 902-905.
- Amin, G., Asif, M.H., Zainelabdin, A., Zaman, S., Nur, O. and M. Willander (2011). Influence of pH, precursor concentration, growth time, and temperature on the morphology of ZnO nanostructures grown by the hydrothermal method. *Journal of Nanomaterials*, **2011**: 269692.
- Behnajady, M.A., Modirshahla, N. and R. Hamzavi (2006). Kinetic study on photocatalytic degradation of C.I. Acid Yellow 23 by ZnO photocatalyst. *J. Hazard Mater.*, **133(1-3)**: 226-232.
- Cai, S., Yang, Z. and Y. Gui (2007). Comparison of dye degradation efficiency using ZnO powders with various size scales. *J. Hazard Mater.*, **141(3)**: 645-652.
- Chkirida, S., Zari, N., Achour, R., Hassoune, H., Lachehab, A., Qaiss, A.e.k. and R. Bouhfid (2021). Highly synergic adsorption/photocatalytic efficiency of Alginate/Bentonite impregnated TiO<sub>2</sub> beads for wastewater treatment. *Journal of Photochemistry & Photobiology, A: Chemistry*, **412**: 113215.
- Daneshvar, N., Salari, D. and A.R. Khataee (2004). Photocatalytic degradation of azo dye acid red 14 in water on ZnO as an alternative catalyst to TiO<sub>2</sub>. *J. Photochem. Photobiol. A*, **162(2-3)**: 317-322.
- Kaur, J. and S. Singhal (2014). Heterogeneous photocatalytic degradation of rose bengal: Effect of operational parameters. *Physica B*, **450**: 49-53.
- Mashkour, M.S., Alkaim, A.F., Ahmed, L.M. and F.H. Hussein (2011). Zinc oxide assisted photocatalytic decolorization of reactive red 2 dye. *Int. J. Chem. Sc.*, **9(3)**: 969-979.
- Pare, B., Jonnalagadda, S.B., Tomar, H., Singh, P. and V.W. Bhagwat (2008). ZnO assisted photocatalytic degradation of acridine orange in aqueous solution using visible irradiation. *Desalination*, **232(1-3)**: 80-90.
- Sauer, T., Neto, G.C., José, H.J. and R.F. Moreira (2002). Kinetics of photocatalytic degradation of reactive dyes in

- a TiO<sub>2</sub> slurry reactor. *J. Photochem. Photobiol. A*, **149**: 147-154.
- Wang, H., Xie, C., Zhang, W., Cai, S., Yang, Z. and Y. Gui (2007). Comparison of dye degradation efficiency using ZnO powders with various size scales. *Journal of Hazardous Materials*, **141(3)**: 645-652.
- Wared, S.H.H. and N.D. Radia (2021). Synthesis and characterization of sodium alginate-g-polyacrylic acid hydrogel and its application for crystal violet dye adsorption. *International Journal of Drug Delivery Technology*, **11(2)**: 556-565.
- Yang, L., Dong, S., Sun, J., Feng, J., Wu, Q. and S. Sun (2010). Microwave-assisted preparation, characterization and photocatalytic properties of a dumbbell-shaped ZnO photocatalyst. *J. Hazard Mater.*, **179(1-3)**: 438-443.