



## Sonochemical and photocatalytic degradation of brilliant green in aqueous medium as an effective technique for water treatment

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### Abstract

This study investigates the sonolysis of Brilliant Green dye in aqueous solution, focusing on the effects of varying concentrations of hydrogen peroxide ( $H_2O_2$ ) and the use of  $TiO_2$  and  $ZnO$  as photocatalysts. The degradation efficiency of the dye was analyzed through advanced oxidation processes (AOPs), including sonolysis alone, as well as Fenton and photo-Fenton reactions under ultrasonic radiation. Kinetic studies revealed that the degradation process follows a first-order mechanism, with correlation coefficients ( $R^2$ ) of 0.9852 and 0.9918 for two different  $H_2O_2$  concentrations. The findings indicate that the AOP methods employed are effective for dye removal, with the photo-Fenton process under ultrasound demonstrating the highest degradation efficiency. Additionally, this research highlights the potential for AOPs to convert azo dyes into non-toxic end products, providing a promising avenue for wastewater treatment applications. Further exploration of the degradation mechanisms and byproducts generated will enhance the understanding of these processes and their practical implications.

**Keywords:** Brilliant Green, sonolysis, photo-Fenton, sonocatalytic, sono-photo-Fenton.

### 1. Introduction

Brilliant Green, a triphenylmethane dye from the malachite-green series, is widely utilized as a topical antiseptic due

to its effectiveness against gram-positive microorganisms. Additionally, it serves in dyeing wool and silk. However, the introduction of textile dyes like Brilliant Green into aquatic ecosystems poses

serious environmental risks, adversely affecting living organisms. Annually, global dye production exceeds 1,000,000 tons, originating from various industries, including textiles, food, cosmetics, and pharmaceuticals. The nature of industrial waste varies based on the processes employed, and water pollution has detrimental effects on plant and animal life, as well as human health.

Industrial activities are significant sources of diverse waste products, necessitating effective wastewater treatment strategies. These methods are categorized into three main types: physical, biological, and chemical. Physical methods involve unit operations such as filtration, adsorption, air flotation, flocculation, and sedimentation. Biological methods, considered secondary treatments, include aerobic and anaerobic processes, along with activated sludge systems. Chemical methods, referred to as advanced processing, encompass techniques like chemical precipitation, oxidation, ion exchange, and incineration.

To tackle water contamination by organic pollutants, numerous advanced oxidation processes (AOPs) have been developed and implemented. These include photocatalysis [1–6], piezocatalysis [7,8], pyrocatalysis [9,10], electrochemical catalytic oxidation [11–

13], and Fenton catalytic processes [14–16]. AOPs are recognized as effective technologies for treating wastewater containing pollutants that exhibit low biodegradability or high chemical stability [17,18]. While these processes are advantageous for treating persistent pollutants, the costs associated with complete mineralization via chemical oxidation can be significant. To alleviate these costs, AOPs are often combined with biological treatments.

The fundamental principle of AOPs is the generation of hydroxyl free radicals ( $\bullet\text{OH}$ ), which are powerful, non-selective oxidants capable of degrading organic compounds resistant to conventional oxidants like oxygen and chlorine [19]. Hydroxyl radicals are effective in reacting with a wide range of organic chemicals, making them highly valuable in wastewater treatment. In Fenton-like processes, transition metals such as  $\text{Fe}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{Cu}^{2+}$  are critical due to their low cost, high efficiency, and environmental friendliness. However, traditional heterogeneous Fenton reactions often face limitations, including the necessity for acidic conditions ( $\text{pH} < 4$ ) and reaction rate constraints due to the  $\text{Fe}^{2+}/\text{Fe}^{3+}$  transition, which restricts their industrial applicability [22,23].

AOPs can be classified into homogeneous and heterogeneous methods, with the latter further divided based on the energy input required. Homogeneous AOPs typically utilize UV radiation to degrade compounds that absorb within specific spectral ranges. In contrast, heterogeneous methods focus on catalytic materials to generate active species [25]. Common heterogeneous AOPs include photocatalytic degradation systems [4], photoelectrochemical processes, and catalytic ozonation.

The use of ultrasound in wastewater treatment offers both opportunities and challenges, particularly concerning the potential effects of additional inorganic ions. Sonochemical processes convert electrical energy into acoustic power, generating  $\bullet\text{OH}$ ,  $\text{HO}\bullet$ , and  $\text{O}\bullet$  radicals. The cavitation effect produced by ultrasonic waves enhances degradation efficiency for emerging contaminants [26]. To improve degradation capacity, strong oxidants such as persulfate or hydrogen peroxide are often added [27].

Research indicates that  $\bullet\text{OH}$  radicals effectively mineralize hydrophilic and non-volatile organic pollutants, while thermal degradation can eliminate volatile and hydrophobic species [28]. The sono-Fenton process is particularly effective in enhancing treatment efficiency through

increased  $\bullet\text{OH}$  radical production. The combination of sonication with Fenton reagents significantly improves contaminant mineralization, making the integration of these processes more effective than their individual applications [29].

This study aims to investigate the removal of Brilliant Green dye from aqueous solution using sonochemical methods, both with and without hydrogen peroxide as an oxidizing agent. It will also explore the sonocatalytic activity of  $\text{TiO}_2$  and  $\text{ZnO}$  in the degradation of Brilliant Green, evaluating the effects of UV radiation on the efficiency of these processes.

## 2. Materials and methods

### 2.1. Materials

Brilliant Green dye was obtained from Merk Co., USA. The chemical formula for Brilliant Green is  $\text{C}_{27}\text{H}_{34}\text{N}_2\text{O}_4\text{S}$ , Molecular weight is 482.6 g/mol (Fig. 1). Its solubility in water is 100 g/L at 20 °C. It shows absorption spectra at 625 nm.

Nitric acid and sodium hydroxide used for adjusting pH values were of reagent grade chemicals. Hydrogen peroxide ( $\text{H}_2\text{O}_2$ , 30%) was obtained from Piochem Company, Egypt. Titanium dioxide and zinc oxide were percussed

from Piochem Co., Egypt. Ferrous sulfate heptahydrate ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ) was obtained from Alpha Chemika, India, Titanium dioxide ( $\text{TiO}_2$ ) and zinc oxide ( $\text{ZnO}$ ) were percussed Piochem Co., Egypt and ADWIC Co., Egypt, respectively. All solutions were prepared with distilled water. All chemicals were of AR grade and were used as received. Distilled water was used for preparing the working solution.

## 2.2. Apparatus

The main apparatuses used in this study are: Dual-beam UV-VIS spectrophotometer (JASCO V-770), Digital pH meter (AD11& AD12, Adwa Instrument Company Romania), Digital ultrasonic cleaner, CD-4830, Zenith Lab Inc., 36 KHz), Centrifuge (LC-04R, MCS Instrument Co.), UV-Lamp (UV-C G13 T8 20W, Philips Co.).

## 2.3. Experimental

A stock solution of the dye was prepared by dissolving 500 mg of the dye in one liter of distilled water. The desired

dye concentrations were prepared by proper dilution of the stock. 250 mL of dye solution were added to a glass beaker (500-mL capacity) centered and fixed to the bottom of the ultrasonic device. Definite volumes of  $\text{H}_2\text{O}_2$  (30%) and doses of solid  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  were added to the dye solution at the beginning of the experiments. Photolysis and photocatalysis experiments were performed by exposing the reaction medium to a UV-Lamp fixed at 20 cm upper the dye solution surface.

Small volumes of the dye solution were withdrawn from the working solution and their absorptions were measured by the spectrophotometer. The removal efficiency ( $R\%$ ) was calculated by using Eq.1:

$$R\% = \frac{C_o - C_e}{C_o} \times 100$$

(1)

Where,  $C_o$  is the initial concentration of the dye (mg/L),  $C_e$  indicates the concentration of the dye solution (mg/L) at time  $t$ .

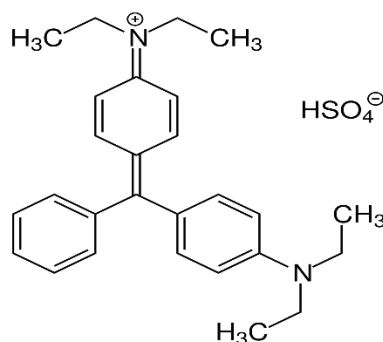
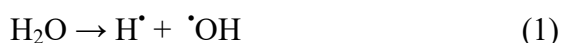


Fig. 1. Chemical structure of Brilliant Green dye.

### 3. Results and discussion

#### 3.1. US system

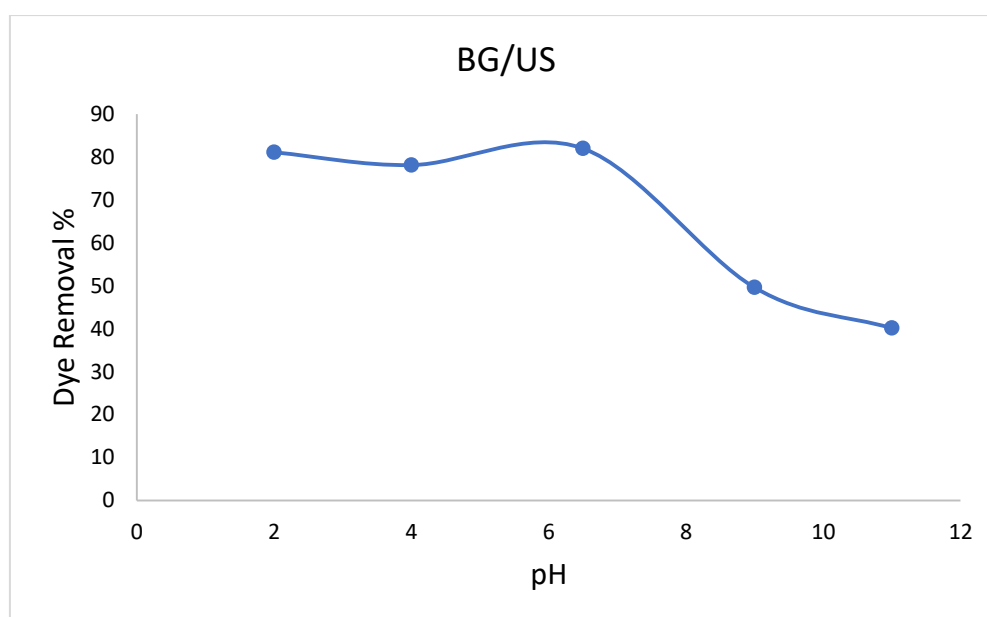
During sonolysis process in aqueous systems, reactive hydroxyl free radicals and hydrogen are formed as a result of the thermal dissociation of water vapor (Eq. 1) and reactions of oxygen gas (Eqs. 2– 5) [30].



When sonolysis is performed in the presence of an organic species, various chemical processes can occur, influenced by the solute's physical and chemical

properties. For instance, a volatile and hydrophobic pollutant may undergo pyrolysis within the cavitation bubbles, whereas a nonvolatile and hydrophilic compound is likely to be oxidized by radicals at the interface or within the bulk solution [31].

The effect of ultrasonic degradation on Brilliant Green (BG) dye at different pH values has been performed at pH values 2-11 using a 2 g/L dye solution. The sonolysis process was monitored for 180 minutes, with dye concentrations measured every 15 minutes. The relationship between pH and the percentage of dye removal after 180 minutes is illustrated in Fig.2. Maximum removal was observed at acidic media and decreased by increasing pH over neutral solution.



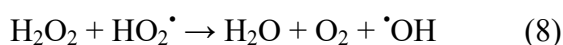
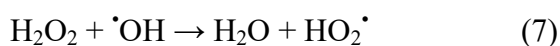
**Fig.2.** BG dye removal percent at different pH values after 180 min of US radiation.

### 3.2. US/H<sub>2</sub>O<sub>2</sub> and US/H<sub>2</sub>O<sub>2</sub>/UV systems

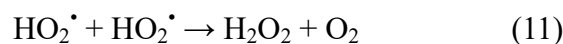
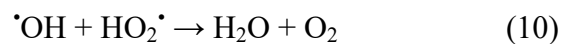
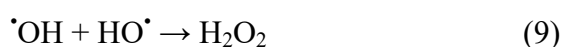
The combination of ultraviolet (UV) radiation and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) has gained significant attention as an advanced oxidation process (AOP) due to its capability to generate hydroxyl radicals ( $\cdot\text{OH}$ ), which are highly effective in degrading pollutants. The oxidative degradation facilitated by H<sub>2</sub>O<sub>2</sub> has garnered interest because of its low cost and non-toxic nature. Numerous researchers have investigated the mechanisms underlying this process [32, 33]. The primary reaction occurring in such systems can be described by the following equation [34]:



Under the effect of radiation, the production of OH radicals from the hydrogen peroxide molecules is accelerated. This decomposition is attributed to the cleavage of the O–O bond. Further OH production is initiated by the following reactions [35]:

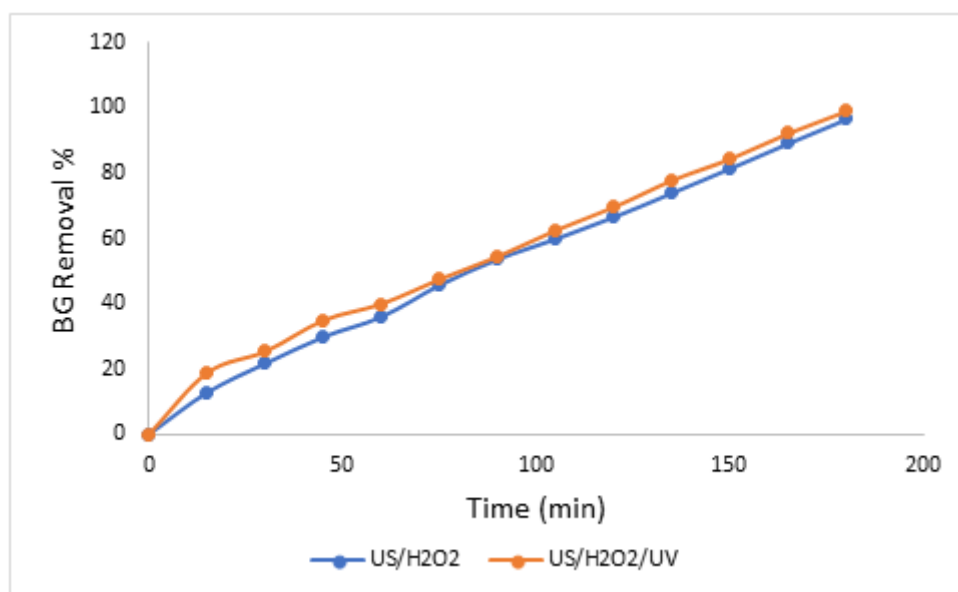


However, in some cases, radical recombination occurs, as follows [36]:



Many factors influence the efficiency of the reactions described, including the concentration of hydrogen peroxide, the source of UV radiation, and the physicochemical characteristics of the fluid involved in the oxidation process. The ultrasonic technique is employed to either promote or modify chemical reactions. Compared to other advanced oxidation processes (AOPs), ultrasonic treatment is considered a green and eco-friendly method. The ultrasonic waves play a crucial role in the oxidative degradation process in the presence of H<sub>2</sub>O<sub>2</sub>, as their cavitation effect can significantly enhance the decomposition of hydrogen peroxide [37].

The effect of UV radiation on the degradation of Brilliant Green (BG) dye by H<sub>2</sub>O<sub>2</sub> under ultrasonic radiation is illustrated in Fig. 3, with an initial dye concentration of 2 g/L and 40 mM H<sub>2</sub>O<sub>2</sub>. As shown in the figure, approximately 95.6% of the dye was removed after 180 minutes of treatment. This removal percentage increased slightly to reach 99.8% under UV irradiation over the same period.



**Fig. 3.** Sonication of BG dye in  $\text{H}_2\text{O}_2$  solution in the presence and absence of UV radiation. [BG = 500 mg/L,  $\text{H}_2\text{O}_2$  = 40 mM].

### 3.3. *US/catalyst and US/catalyst/UV systems*

The combination of catalysts and ultrasonication creates a heterogeneous sonocatalytic process that significantly enhances the degradation efficiency of organic pollutants. The presence of sonocatalysts improves mass transfer due to their larger surface area and acts as nucleation sites, increasing the number of cavitation bubbles. Moreover, the catalysts can elevate the production of free radicals, further accelerating the degradation rate of organic pollutants [38-40].

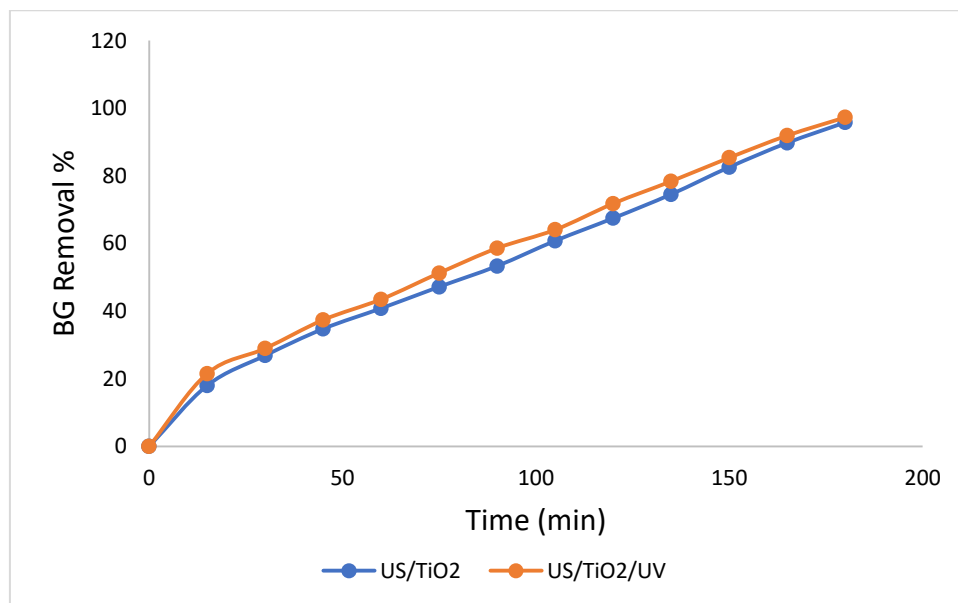
To evaluate the catalytic activities of two different metal oxides,  $\text{TiO}_2$  and  $\text{ZnO}$ , their degradation efficiencies under ultrasound (US) were compared. As shown in Figs. 4 and 5,  $\text{TiO}_2$  demonstrates a higher degradation efficiency than  $\text{ZnO}$ ,

achieving a 94.85% removal of the dye compared to 83.12% for  $\text{ZnO}$ . This difference can be attributed to the stronger adsorption of dye particles on  $\text{TiO}_2$  compared to  $\text{ZnO}$ . The figures also indicate that the presence of UV radiation slightly enhances the degradation rates of BG dye with both catalysts, suggesting that the ultrasonic effect is more efficient than photocatalysis alone.

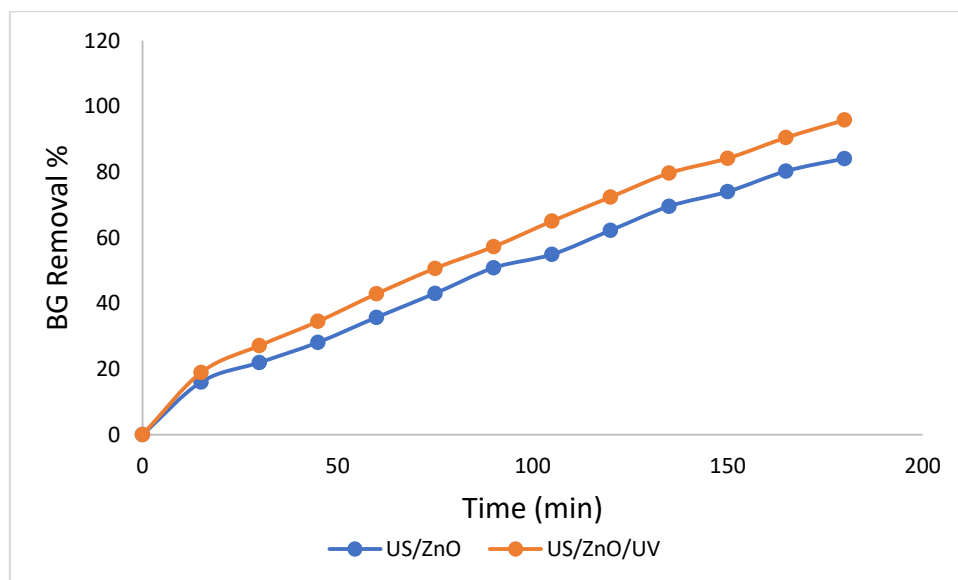
To investigate the effect of catalyst dose on degradation efficiency, various amounts of  $\text{TiO}_2$  and  $\text{ZnO}$  were added to the dye solution (40 mM) during the ultrasonication process, both in the presence and absence of UV radiation. Figs. 6 and 7 present the relationship between removal percentage and catalyst doses for  $\text{TiO}_2$  and  $\text{ZnO}$ , respectively. The results indicate that the optimal dose for

TiO<sub>2</sub> is 0.1 g/L, while for ZnO, it is 0.4 g/L in both sonocatalysis and sono-

photocatalysis processes.

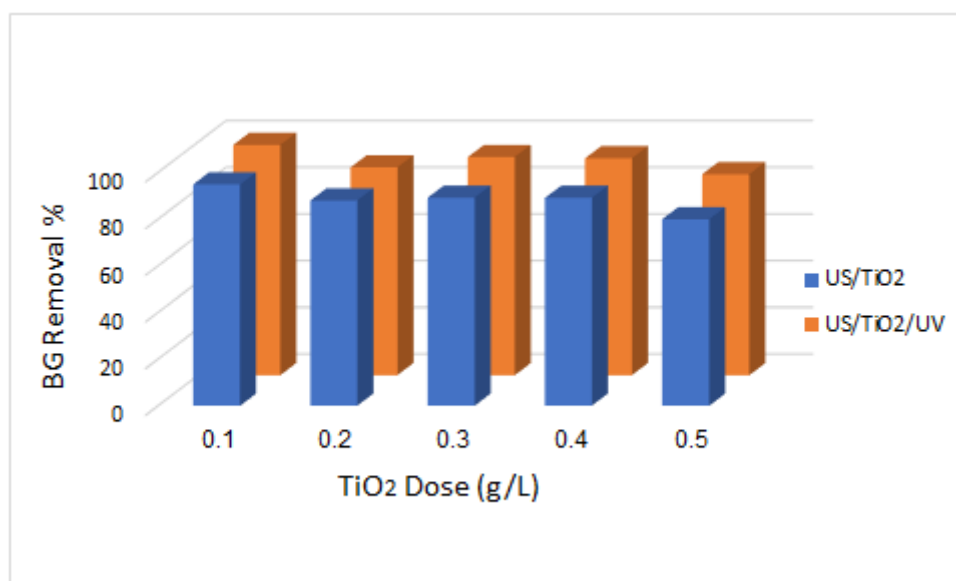


**Fig. 4.** Effect of TiO<sub>2</sub> during sonolysis of BG dye solution in the presence and absence of UV radiation.

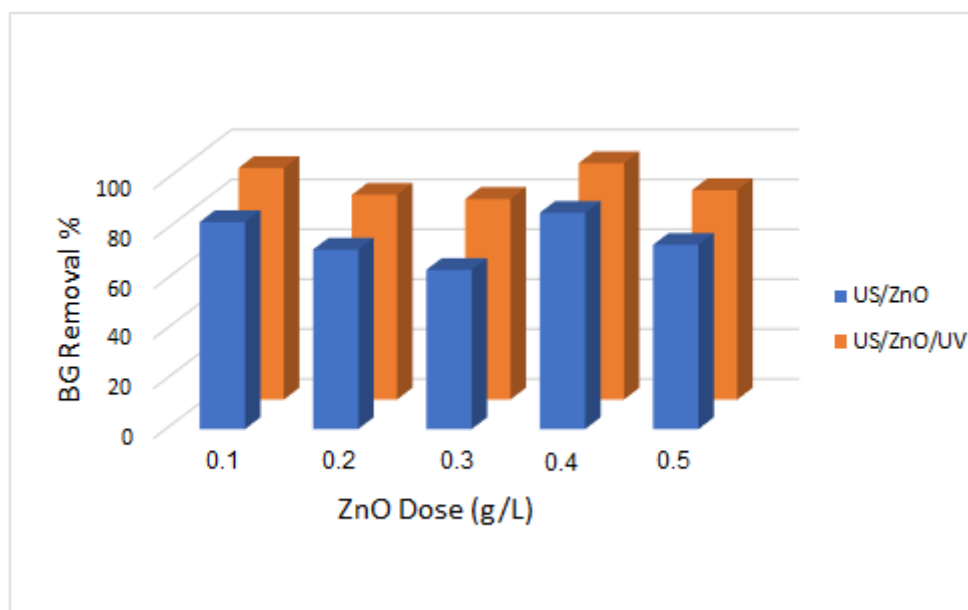


**Fig. 5.** Effect of ZnO during sonolysis of BG dye solution in the presence and absence of UV radiation.





**Fig. 6.** Effect of TiO<sub>2</sub> dose on BG removal under sono-catalysis and sono-photocatalysis processes.



**Fig. 7.** Effect of ZnO dose on BG removal under sono-catalysis and sono-photocatalysis processes.

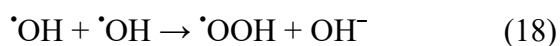
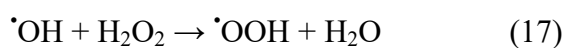
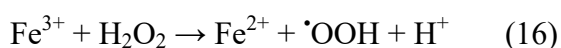
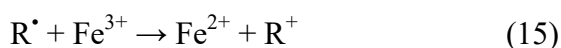
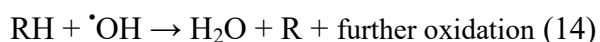
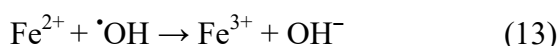
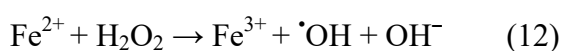
### 3.4. *US/Fenton and US/Fenton/UV systems*

The Fenton oxidation reaction is widely utilized for treating toxic organic pollutants in industrial wastewater. This process involves the reaction of Fenton's reagent, which consists of iron salts in the

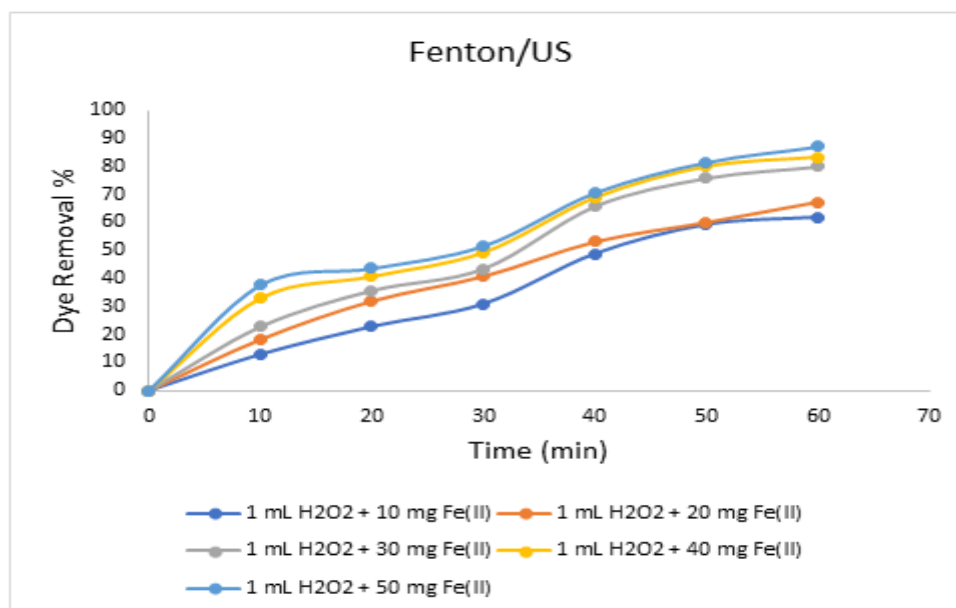
Fe<sup>2+</sup> form, with hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) to generate hydroxyl radicals (•OH). The reaction is typically conducted under acidic conditions, which are essential for optimal performance. The effectiveness of the Fenton process stems from the strong

oxidizing power of the hydroxyl radicals produced.

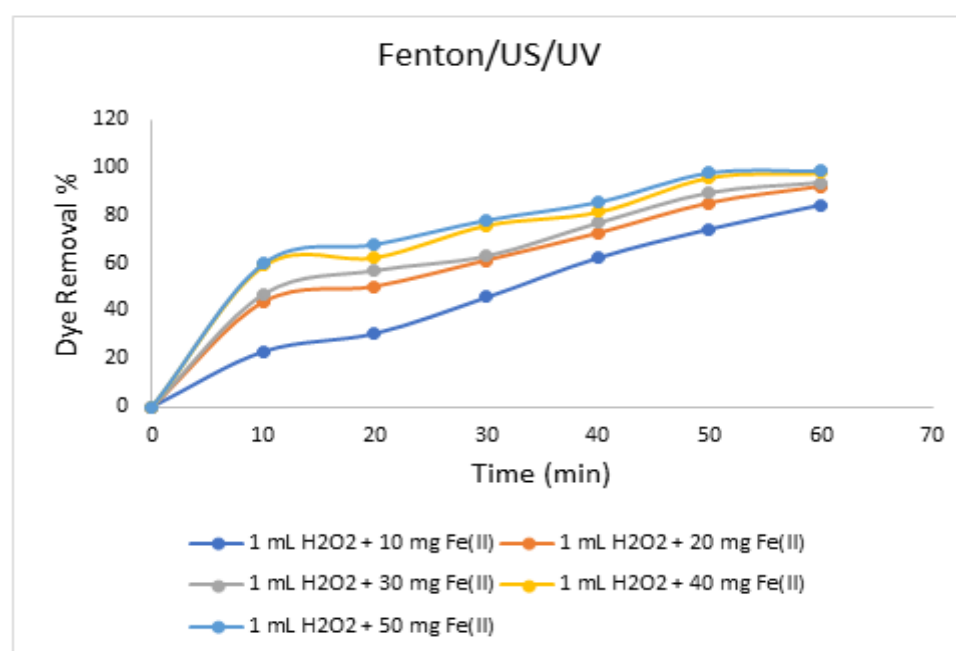
Various adaptations of the Fenton reaction have been developed for water treatment, including electro-Fenton, photo-Fenton, and sono-Fenton oxidation methods [41]. The fundamental mechanism of the Fenton reaction involves the oxidation of  $\text{H}_2\text{O}_2$  in the presence of  $\text{Fe}^{2+}$  as a catalyst, leading to the generation of hydroxyl radicals. This reaction proceeds in the presence of the organic pollutant (RH), as illustrated in the following equation [42]:



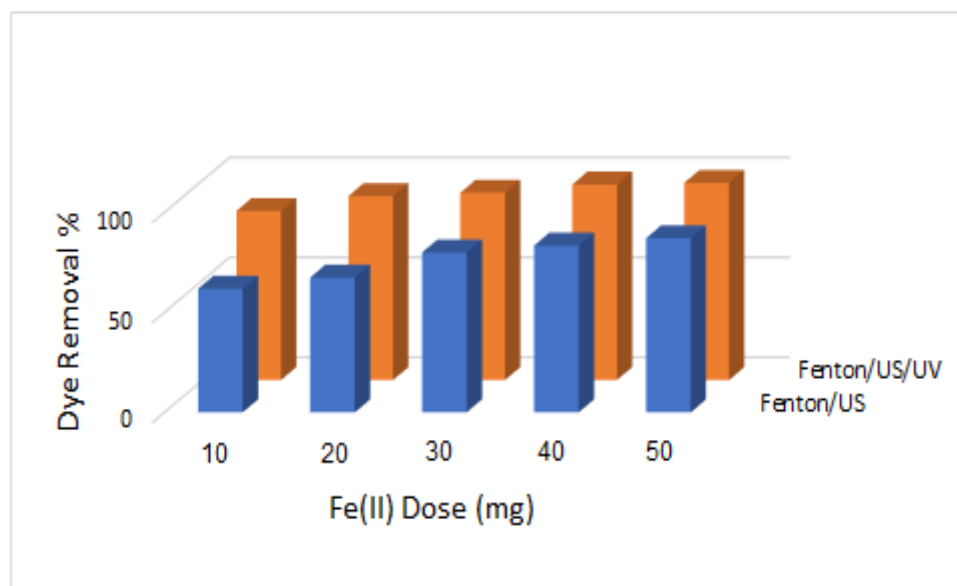
To examine the effect of Fe(II) dose on dye removal efficiency, different doses of ferrous ion (10-50 mg/L) were added to the dye solution (2 g/L) in the presence of a constant amount of  $\text{H}_2\text{O}_2$  (1 mL) under the effect of US radiation. As shown in Fig. 8, the removal percent increased with increasing ferrous ion content. This result confirm the role of Fe(II) ions as a catalyst for enhancing the production of  $\cdot\text{OH}$  radicals. Maximum dye removal was observed at 50 mg Fe(II) using 1 mL of  $\text{H}_2\text{O}_2$ . The same experiments were done under the effect of UV radiation to evaluate the effect of UV radiation on dye removal. As shown in Fig. 9, the removal of BG dye increased with increasing iron content, but the extent of removal is higher than that in case of absence of UV radiation. For comparison of these results, Fig. 10 was constructed to detect the differences in the efficiency of dye removal in both cases.



**Fig. 8.** Effect of Fe(II) on BG removal in the presence of constant amount of H<sub>2</sub>O<sub>2</sub> using sono-Fenton reaction.



**Fig. 9.** Effect of Fe(II) on BG removal in the presence of constant amount of H<sub>2</sub>O<sub>2</sub> using sono-Photo Fenton reaction.

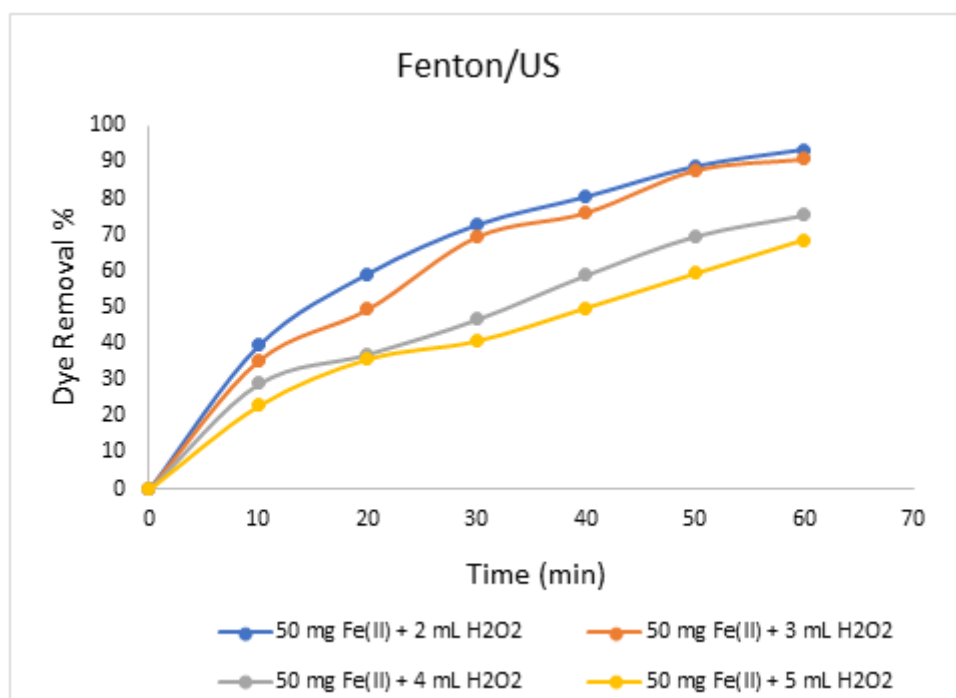


**Fig. 10.** Fenton Reagent [1 mL H<sub>2</sub>O<sub>2</sub>/Fe(II), BG = 500 mg/L]

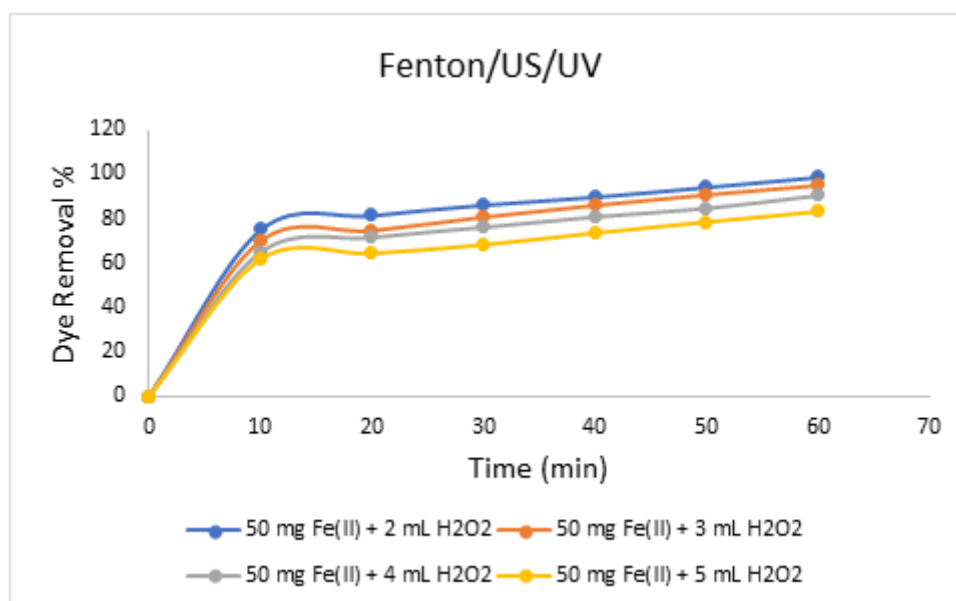
In another set of experiments, constant dose of Fe(II) was combined with varying doses of H<sub>2</sub>O<sub>2</sub> (2-5 mL) to treat 2 g/L Brilliant Green (BG) dye under ultrasonic radiation, in absence and presence of UV radiation (Figs. 11 and 12, respectively). The results indicate that UV has a remarkable effect on removal efficiency. Interestingly, while increasing the H<sub>2</sub>O<sub>2</sub> concentration at a fixed Fe(II)

dose initially improved dye degradation, a further increase in H<sub>2</sub>O<sub>2</sub> resulted in decreased dye removal.

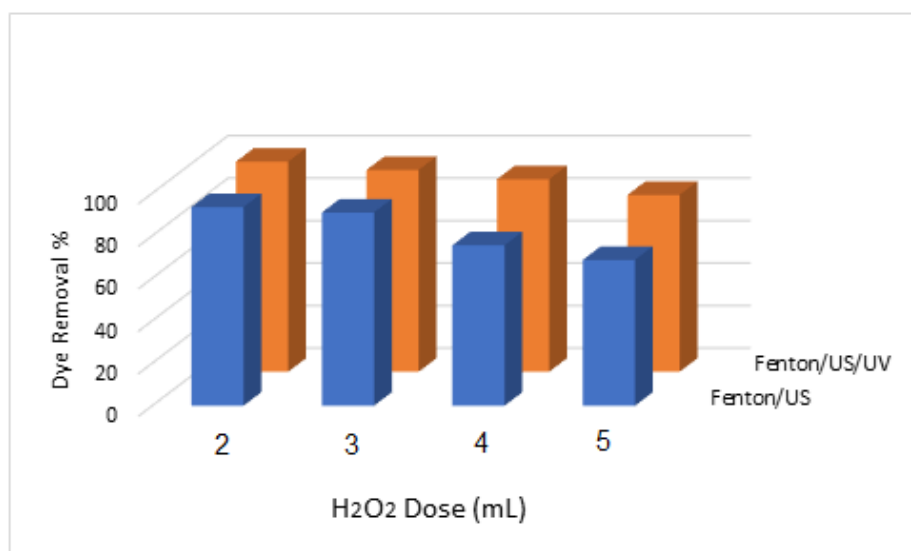
Comparative analysis of the Fenton and photo-Fenton processes under sonolysis, effect in absence and presence of UV radiation was illustrated in Fig. 13, revealing that an optimal ratio of Fe(II) to H<sub>2</sub>O<sub>2</sub> is crucial for maximizing dye removal efficiency.



**Fig. 11.** Effect of H<sub>2</sub>O<sub>2</sub> concentration on BG removal at constant Fe(II) dose on BG dye removal under the effect of US.



**Fig. 12.** Effect of H<sub>2</sub>O<sub>2</sub> concentration on BG removal at constant Fe(II) dose on BG dye removal under the effect of US and UV.



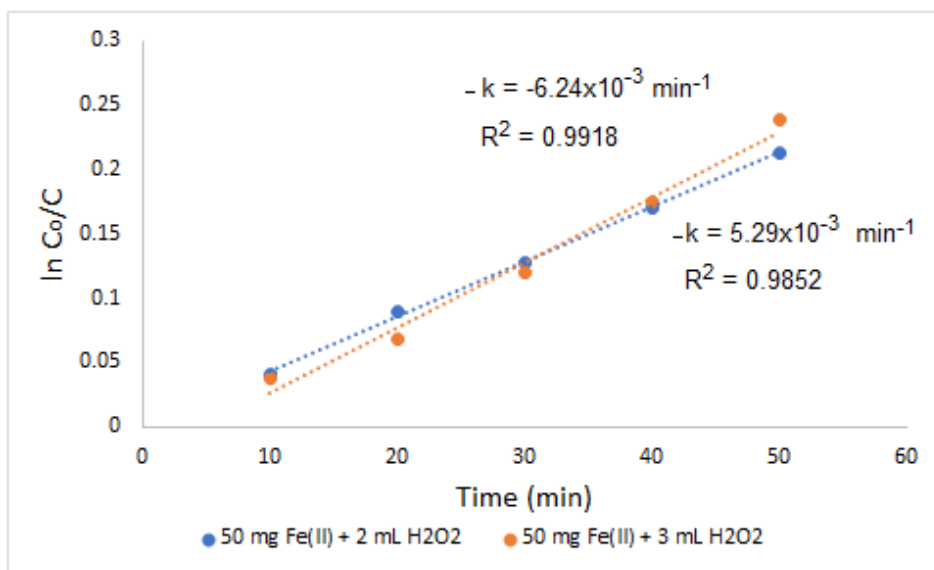
**Fig. 13.** Comparison between the effects of H<sub>2</sub>O<sub>2</sub> dose in Fenton reaction by the effect of US and US/UV radiation.

The kinetic parameters of dye removal by sono- photo-Fenton reaction were studied using the first-order kinetic Equation (19) as follows:

$$\ln \frac{C_0}{C} = kt \quad (19)$$

where  $C_0$  (mg/mL) and  $C$  (mg/mL) represent the initial BG dye concentration and the dye concentration after time  $t$  of

photo-Fenton reaction under sonolysis, respectively.  $k$  represents the apparent rate constants, and  $t$  represents the reaction time (min) [43]. Thus,  $k$  was used to evaluate the photocatalytic activity in this work. The plot of  $\ln C_0/C$  vs.  $t$  is shown in Fig. 15. The values of  $k$  were calculated to be  $5.29 \times 10^{-3}$  and  $6.24 \times 10^{-3} \text{ min}^{-1}$ .



**Fig. 15.** Plot of first order kinetics of dye removal efficiency under sono- photo-Fenton degradation at two different concentrations of H<sub>2</sub>O<sub>2</sub>.

## 4. Conclusion

The findings of this study underscore the efficacy of advanced oxidation processes (AOPs) in the degradation of Brilliant Green (BG) dye, particularly through the synergistic effects of sonolysis, UV radiation, and the incorporation of catalysts like TiO<sub>2</sub> and ZnO. The results demonstrate that degradation efficiency is significantly influenced by various factors, including pH, catalyst type, and the concentrations of H<sub>2</sub>O and Fe(II).

Notably, a slightly acidic environment was optimal for dye removal, achieving an impressive 81.85% reduction in dye concentration. The combination of UV radiation with H<sub>2</sub>O<sub>2</sub> further enhanced degradation, resulting in up to 99.8% dye removal. Among the catalysts studied, TiO<sub>2</sub> exhibited superior performance compared to ZnO, likely due to its stronger adsorption properties.

The Fenton and photo-Fenton processes highlighted the necessity of an optimal ratio of Fe(II) to H<sub>2</sub>O<sub>2</sub> for maximizing dye removal, with the addition of UV radiation further augmenting the efficiency of these reactions. These insights affirm that AOPs, particularly when optimized through the right combinations of conditions and materials,

represent a promising approach for effectively mitigating the environmental impact of azo dyes in wastewater.

Future research should focus on exploring the scalability of these processes for application in real-world wastewater treatment scenarios, as well as investigating the potential for mineralization of degradation by-products into non-toxic end products.

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