

Degradation of Basic yellow 28 by heterogeneous photocatalysis in Suntest in aqueous suspension of TiO_2 , ZnO and Fe_2O_3

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Abstract

This study aims to evaluate the effectiveness and applicability of a new advanced oxidation process (UV / semiconductor) for the treatment of waste water. Photocatalysis is a very robust technology that is capable of destroying a wide range of recalcitrant organic contaminants and is equally effective on highly reduced contaminants such as basic yellow 28 (BYE28) cationic dyes. In Suntest CPS+, an attempt has been made to optimize the process parameters. The efficiency of the degradation of Basic yellow 28 (BYE28) dye in aqueous suspensions of TiO_2 , ZnO and Fe_2O_3 strongly depends on the operating conditions. The rate of photodegradation increased with the increase of the quantity of catalysts (TiO_2 , ZnO and Fe_2O_3) from 0.5 to 3g/L and after that rate decreased in the case of the three catalysts, with an optimum quantity found to be 1g/L. The photodegradation rate increased with the increasing of the initial concentration of Basic yellow 28 (BYE28) dye in aqueous suspensions of TiO_2 , ZnO and Fe_2O_3 from 1 to 50 mg/L after that it decreased in the range from 50 to 150 mg/L. The better degradation of Basic yellow 28 (BYE28) dye was found at basic pH range for the three catalysts. The degradation rate of Basic yellow 28 increased with increasing temperature in the range considered range 15 to 55 °C in the case of the three catalysts. The photodegradation rate of Basic yellow 28 (BYE28) dye in aqueous suspension of TiO_2 P25 increased with the increasing of radiation intensity from 250 to 500 W/m² after that, it remained constant from 500 to 750 W/m². TOC decreased with treatment, confirming that the photocatalytic process led to a decrease in organic matter content.

Keywords: Photocatalysis, Basic yellow 28, TiO_2 , ZnO , Suntest.

I. Introduction

Pollution from dye wastewater has become a serious environmental problem due to the large and increasing uses of a variety of dyes [1-5]. The reports are alarming that 15% of the total world production of dyes is lost in wastewater stream during the dyeing and finishing operations [6-8]. Among them Basic yellow 28 synthetic colors are very abundant in wastewater in the north east of Algeria. The main sources of Basic yellow are chemical and textile industries, where it is

mainly used for dyeing acrylic fibers. These processes require large volumes of water and consequently generate huge amounts of wastewaters with high loads of contaminants [9, 10]. The impact of these dyes on the environment is a major concern because of its high toxicity, stability to many oxidizing agents and possible accumulation in the environment [1-3]. The presence of dyes even at very low concentration in water can reduce the light penetration, which leads to inhibiting photosynthesis and oxygen dissolution, which are essential for aquatic life [11, 12]. Therefore, it is

necessary to reduce dyes concentration in the wastewater before it is released into the environment [13, 14].

For the removal of dye from industrial wastewaters, various conventional methods such as biological treatment, coagulation, flotation, electrochemical techniques, ultrafiltration, adsorption and oxidation are applied. Nevertheless, these methods are usually non destructive, inefficient, costly and resulted in the production of secondary waste products which needs to be processed further [6, 9, 10]. An alternative to these classical methods includes the advanced oxidation processes (AOPs) which are based on the generation of very reactive species, such as hydroxyl radicals (OH) that oxidize a broad range of organic pollutants quickly and non-selectively. Among them, top priority goes to semiconductor assisted photocatalytic degradation. This technique has confirmed its efficiency in degrading a wide range of dyes present in water [15].

Metal oxides are widely present in nature and some of them are of great practical use. Many of them have been tested so far as photocatalysts. Among them, TiO_2 in the anatase form seems to have the most interesting required attributes such as non-toxicity, chemical stability, good performance and low cost [12, 16]. TiO_2 -based photocatalysts are often studied for dye degradation [18, 19]. However, recently some studies have been carried out to evaluate the priority of other metal oxides. Among the other semiconductors, ZnO , Fe_2O_3 appear to be a highly promising photocatalysts [19-21]. ZnO has been increasingly important and intensively studied. ZnO is an n-type semiconductor with 3.37 eV wide band gap (E_g) and 60 meV large excitation binding energy [22] which is found to be a suitable alternative to TiO_2 . Moreover, ZnO has been found to be more economic than TiO_2 , it has the potential for using as photocatalysts to treat organic wastes by photocatalysis [23].

In this article, we report the photodegradation performance of nanoparticles in Suntest CPS+. The photocatalytic degradation of basic yellow 28 has been investigated for three photocatalyst TiO_2 , ZnO and Fe_2O_3 .

II. Material and methods

A. Reagents :

Basic yellow 28 (known as) (abbreviation): (BYE28); chemical class: cationic dye; molecular formula: $C_{21}H_{27}N_3O_5S$, molecular weight: 433, 16 g mol⁻¹). The molecular structure of basic

yellow 28 was shown in Fig. 1. TiO_2 P25 was supplied from Degussa, ZnO was supplied from Prolab and Fe_2O_3 from Sigma. All reagents are used without further purification.

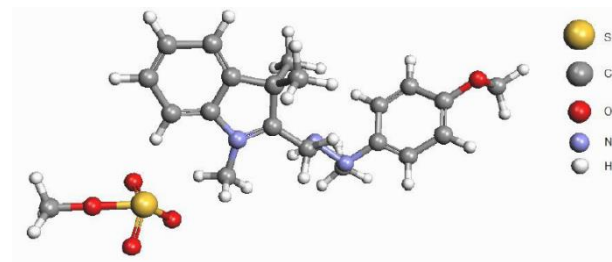


Figure 1. Basic yellow 28 structures



Figure 2. Semiconductor used in this study, ZnO , Fe_2O_3 ; TiO_2 P25.

B. Experimental setup Suntest CPS +

The experimental device Suntest CPS + (Fig.3) is a chamber equipped with a lamp allows irradiation simulating the best solar radiation thanks to its xenon lamp for wavelengths ranging from 290 to 800 nm. The xenon arc lamp comprises a quartz filter with an IR coating and an additional filter that adapts the spectral distribution functions of the specific intended application. Suntest is equipped with a ventilation system and a cry-thermostat to keep its temperature at 20 ± 5 ° C. Photochemical reactor at double envelope of Pyrex glass of 250×10^{-3} l capacity was placed in the Suntest (Fig.3). The temperature in the reactor is kept constant at 25 °C using a thermostated bath (Lauda R6). An appropriate amount of TiO_2 is added to the Basic yellow 28 solution and the resulting suspension is left for 20 minutes in the dark with agitation and recirculation to ensure complete adsorption equilibrium between the Basic yellow 28 and the catalyst particles. Once equilibrium is reached, the Suntest lamp is on, and this time is taken as the initial reaction time. Periodic samples are taken, filtered through Millipore filter (0.45 microns) and then analyzed by UV-Visible Spectrophotometer. Each test was repeated twice or more.



Figure 3. Suntest CPS+

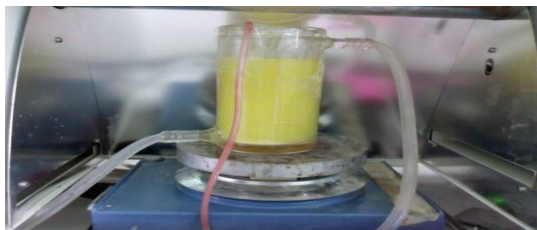


Figure 4. Experimental apparatus

III. Results and discussion

A. Photocatalytic degradation

The study of photocatalytic degradation of Basic yellow 28 dye was done, in aqueous suspension of TiO_2 P25, ZnO and Fe_2O_3 under the irradiation of light (in Suntest CPS+). A 0.2 g of TiO_2 , ZnO and Fe_2O_3 sample was taken in a 250 ml batch reactor containing 200 ml of Basic yellow 28 dye solution having concentration 10^{-2} g/L. After 1 hour equilibrium of adsorption, this solution was then kept inside the chamber of photoreactor with light ON. Sample was pipette out. The taken samples are then analyzed in the *JASCO V-630* UV-Visible Spectrophotometer to determine the absorption spectrum. The experimental results are illustrated in Fig.5.

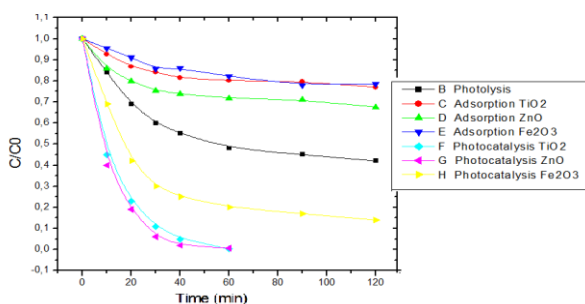


Figure 5. Degradation kinetics of BYE28, in aqueous slurry of TiO_2 P25, ZnO and Fe_2O_3 in a Suntest CPS+ , $(BYE28)_0 = 10$ mg/L, $[Photocatalyst]_0 = 1$ g/L, $pH \approx 7$, $T = 25$ °C, Intensity of radiation=500 W/m²).

Figure.5 reveals the comparative study of systems on the elimination of Basic yellow 28 dye. The degradation of BYE28 by photolysis (Complete absence of TiO_2 , ZnO and Fe_2O_3) has been examined in order to check the contribution of this phenomenon in the process of photocatalysis. The results, shown in the Fig.5, shows that the degradation rate does not exceed 51.5% after 1 hour.

The adsorption of Basic yellow BYE28 dye on TiO_2 P25, ZnO and Fe_2O_3 has been also examined to test and to quantify the quantity of BYE28 dye eliminated via adsorption. The percentage of removal by adsorption was found to be equal to 23, 22 and 32% for TiO_2 P25, ZnO and Fe_2O_3 , respectively. The equilibrium is reached after about 60 min, so after then and in all experiments of photocatalysis that following, light turn on after adsorption equilibrium.

The results illustrated in Fig.5 shows that Basic yellow BYE28 dye degrades well in the three systems $UV + TiO_2$, $UV + ZnO$ and $UV + Fe_2O_3$ compared to the photolysis, total degradation 100% for TiO_2 and ZnO , while 80% for Fe_2O_3 is obtained after 1 hour for the same time photolysis only 51.5% are degraded. Thus, the presence of photocatalyst significantly increases the rate of degradation of Basic yellow BYE28 dye.

B. Effect of catalyst dose

In heterogeneous photocatalysis, the quantity of the suspended catalyst is an important parameter because of cost of photocatalyst used and for an effective degradation of Basic yellow BYE28 dye. Too few particles will absorb a small amount of UV light, the amount of electron/hole pairs is insufficient consequently that of $HO^\bullet$ radicals photogenerated. While too many particles diffuse UV light (screen effect and increase turbidity) [23, 24], on the other hand, particle agglomeration is favored reducing the interfacial area available for reaction [25].

In our case, we have experimentally attempted to obtain this optimum concentration. The amount of catalyst (TiO_2 P25, ZnO and Fe_2O_3) is varied from 0.5 up to 4 g/L keeping all of such others constant operating conditions, namely, $T = 25$ °C, $pH \approx 7$, For an initial concentration of Basic yellow BYE28 dye equal to 10^{-2} g/L. The experimental results are illustrated in the Fig.6.

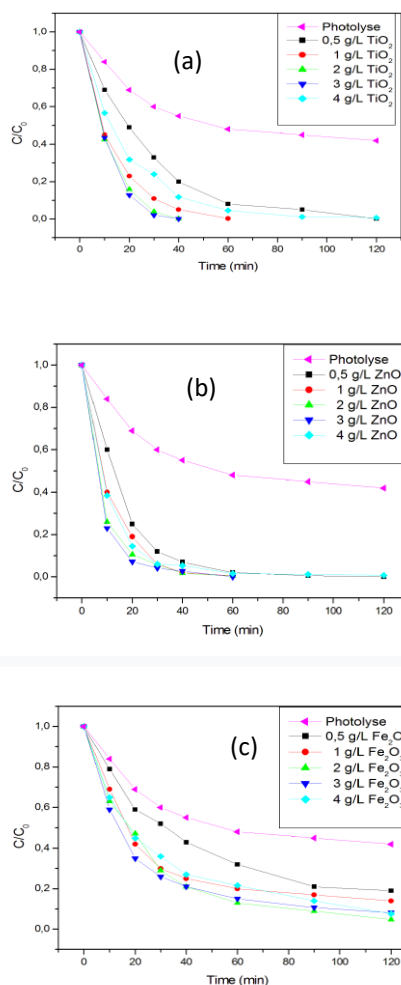


Figure 6. Effect of the photocatalyst mass on the photocatalytic oxidation kinetics of the dye (BYE28) in an aqueous suspension of: (a) TiO_2 , (b) ZnO and (c) Fe_2O_3 . $(\text{BYE28})_0 = 10^{-2} \text{ g/L}$, $\text{pH} \approx 7$, $T = 25^\circ\text{C}$, Intensity of radiation = 500 W/m^2 .

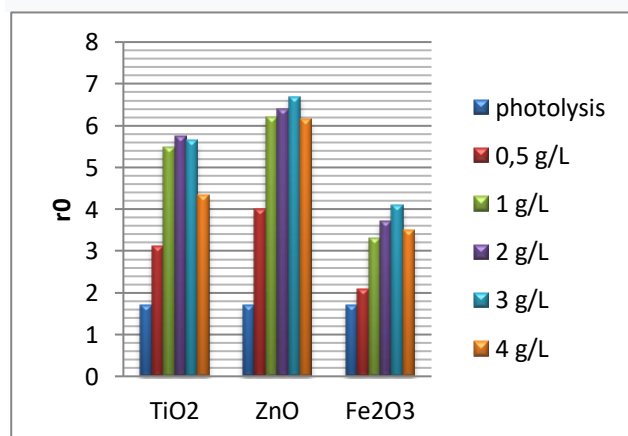


Figure 7. Variation of initial rate of photodegradation of BYE28 dye versus of quantity of TiO_2 , ZnO and Fe_2O_3 . $(\text{BYE28})_0 = 10^{-2} \text{ g/L}$, $\text{pH} \approx 7$, $T = 25^\circ\text{C}$, Intensity of radiation = 500 W/m^2 .

The degradation of Basic yellow BYE28 dye by photolysis (complete absence of catalyst) has been examined in order to

check the contribution of this phenomenon in the process of photocatalysis. The degradation rate in the absence of catalyst (photolysis) equal to 45% at $t = 0.66 \text{ h}$, observed at the same time that the rate of degradation increases by 80 to 100%, 93 to 100% and 57 to 79% with the increase in the amount of TiO_2 , ZnO and Fe_2O_3 from 0.1 to 3 g/L, respectively.

According to Fig.7 is shown the variation of the initial rate of photodegradation a function of the quantity of catalyst. For the period of 1 h, the rate increases with increasing of TiO_2 photocatalyst dosage, and it reaches to an optimum value of catalyst dose equal to 1 g/L (0.2g) at which the initial rate of dye degradation was maximum ($r_0 = 0.30 \text{ g/L.h}$). After, increasing the dose of TiO_2 , the initial degradation rate remains constant. This observation may be explained by the availability of active sites on the surface of TiO_2 and the possibility of penetration of light in the entire surface of the catalyst in the solution.

The rate of photodegradation increases with the increasing of ZnO and Fe_2O_3 photocatalyst dosage, and it reaches to an optimum value of catalyst dose equal to 1 g/L. at which the initial rate of dye degradation was $r_0 = 0.36 \text{ g/L.h}$ for ZnO photocatalyst and $r_0 = 0.186 \text{ g/L.h}$ for Fe_2O_3 photocatalyst after that, a low degradation rate of dye photodegradation was found with increasing of catalyst dose from 1 to 3 g/L.

The optimum dose of catalyst for photodegradation of Basic yellow BYE28 dye was found to be 1 g/L with the three catalysts. Also, all our experiments were performed with this dose. Any time, the results published in the literature concerning the dose of photocatalyst are between 1 and 3 g/L according to the experimental device (light power, reactor type, pH , type of TiO_2) [23-26].

D- Effect of pH

The pH of the solution is the most important parameter affecting in a considerable way the efficiency of the photodegradation of dyes. It can change the structure of the dye and affect the reactivity of the photocatalyst. In this context, the effect of pH on the photocatalytic degradation of Basic yellow BYE28 dye was carried out with a pH value equal to 3, 6 and 11, by adding whether sulfuric acid (H_2SO_4) for acids pH range or sodium hydroxide (NaOH) for the basics values, keeping also constant all other operating conditions, namely, $T = 25^\circ\text{C}$, $[\text{BYE28}]_0 = 10^{-2} \text{ g/L}$,

[Photocatalyst] = 1 g/L. The experimental results obtained are shown in the Fig.8.

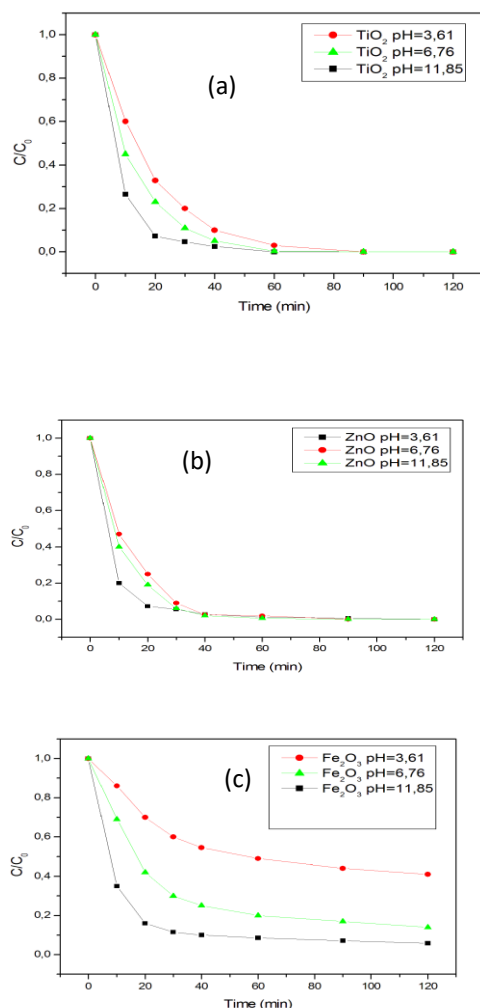


Fig.8 Effect of pH on the photocatalytic oxidation kinetics of the dye (BYE28) in an aqueous suspension of : (a) TiO_2 , (b) ZnO and (c) Fe_2O_3 . (BYE28)₀ = 10^{-2} g/L, [Photocatalyst]₀ = 1 g/L, T = 25 °C, Intensity of radiation = 500 W/m²).

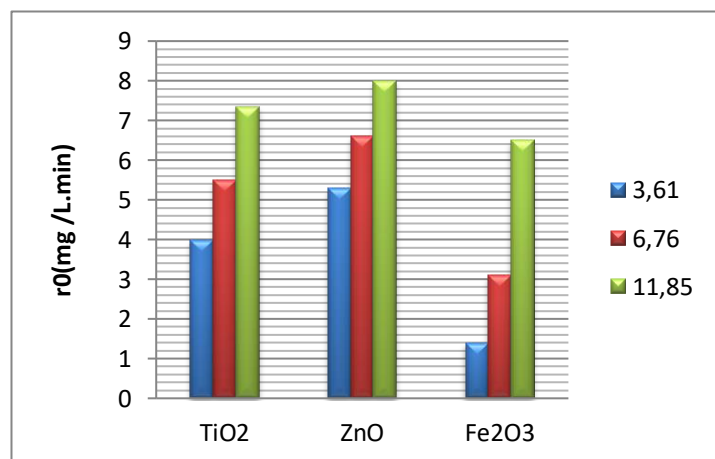
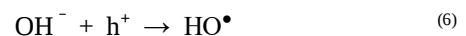
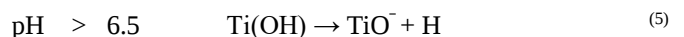
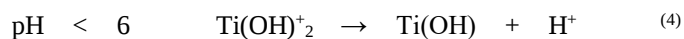


Fig.9 Variation of initial rate of degradation versus the pH.

(BYE28)₀ = 10^{-2} g/L, [Photocatalyst]₀ = 1 g/L, T = 25 °C, Intensity of radiation = 500 W/m²).

Fig.9 shows that the initial rate of photodegradation of Basic yellow *BYE28* dye increases with increasing pH, the modifications of the charges present at the surface of the catalysts at each pH, play on the adsorption of reactive molecules on the surface of photocatalyst and consequently on heterogeneous photocatalysis [27, 28].

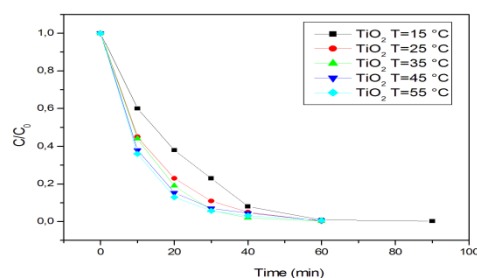


Since the Basic yellow *BYE28* dye is a cationic structure, the degradation of the dye is favored at basic pH range.

At acidic pH range, the degradation efficiency of Basic yellow *BYE28* dye was less; this is due to repulsive interactions between the Basic yellow *BYE28* dye and the charge of the surface of the photocatalyst decreasing the probability of adsorption of dye, thereby decreasing the degradation. Furthermore, the initial rate of Basic yellow *BYE28* dye was less because of the probability of dissolution of TiO_2 particles at low pH [29]. While, at basic pH range, the degradation of Basic yellow *BYE28* dye found to be increased, due to the pH influenced the surface properties of photocatalyst as well as it influenced the dissociation of the dye. Moreover at basic pH range, the dominant species are the hydroxyl ions OH^- that easily converts to $\text{HO}^\bullet$ radicals according to reaction 6.

E. Effect of Temperature

The study of the effect of this parameter on the photocatalytic degradation process of Basic yellow *BYE28* dye was carried out with varying the temperature in the range from 15 to 55 °C at interval of 10 °C. The other parameters (pH, initial concentration, dose of the photocatalyst) are held constant i.e., pH ≈ 7, (BYE28)₀ = 10^{-2} g/L, [Photocatalyst]₀ = 1g/L. the experimental results obtained are shown in Fig.10:



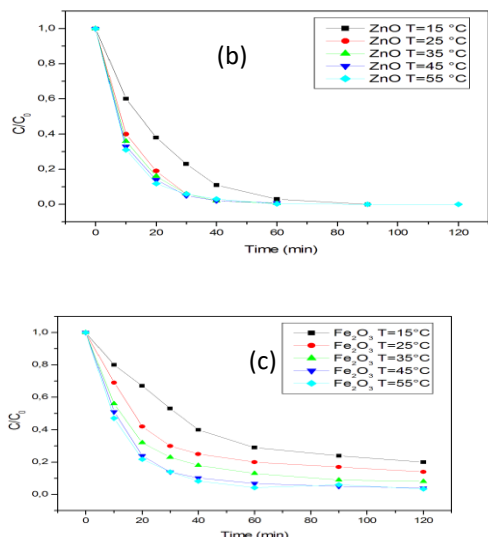


Figure 10. Effect of temperature on the photocatalytic oxidation kinetics of the dye (BEY28) in an aqueous suspension of: (a) TiO_2 , (b) ZnO and (c) Fe_2O_3 . $[BEY28]_0 = 10^{-2}$ g/L, $[Photocatalyst]_0 = 1$ g/L, $pH \approx 7$.

The degradation kinetics of Basic yellow 28 (BEY28) dye increases with increasing temperature. Total degradation of BEY28 in TiO_2 P25 and ZnO suspension is obtained after: 1.5 hours for the temperature value equal $15^\circ C$ and 1 hour for the temperature range from 25 to $55^\circ C$. While, Total degradation of BEY28 in Fe_2O_3 suspension isn't obtained even after 2 hours.

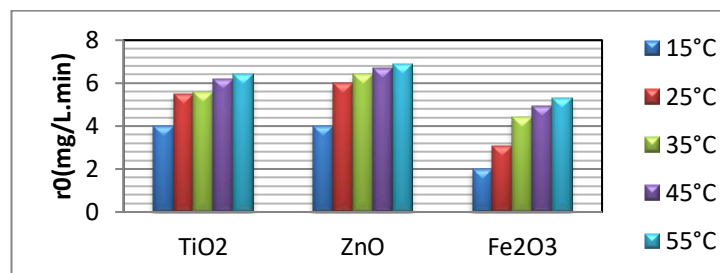


Figure 11. Variation of initial rate of photocatalytic degradation of Basic yellow 28 (BEY28) dye versus the Temperature. $[BEY28]_0 = 10$ mg/L, $[Photocatalyst]_0 = 1$ g/L, $pH \approx 7$, Intensity of radiation = 500 W/m^2 .

So as it can be seen in Fig.11 that the initial rate of photocatalytic degradation of Basic yellow 28 (BEY28) dye increases with the temperature increasing. Regarding photocatalysis, this result matched well with the results published in the literature. The average operating temperature during a photocatalytic reaction is between 20 and $80^\circ C$. A reactor containing TiO_2 suspension, an increase of the rate of degradation of atrazine dye while the temperature increases from 20 to $70^\circ C$ [30].

F-Effect of initial concentration of Basic yellow 28 (BEY28)

To investigate the effect of the initial concentration of Basic yellow 28 (BEY28) dye on photodegradation process was carried out at varying initial concentration of Basic yellow 28 (BEY28) dye as 1×10^{-3} , 5×10^{-3} , 10×10^{-3} , 50×10^{-3} , 110×10^{-3} , 150×10^{-3} g/L prepared from stock solution of 1g/L, with fixed TiO_2 , ZnO and Fe_2O_3 photocatalyst dose of 1 g/L and the same operating conditions as above namely, $T = 25^\circ C$, $pH \approx 7$. The experimental results obtained are shown in the Fig.12.

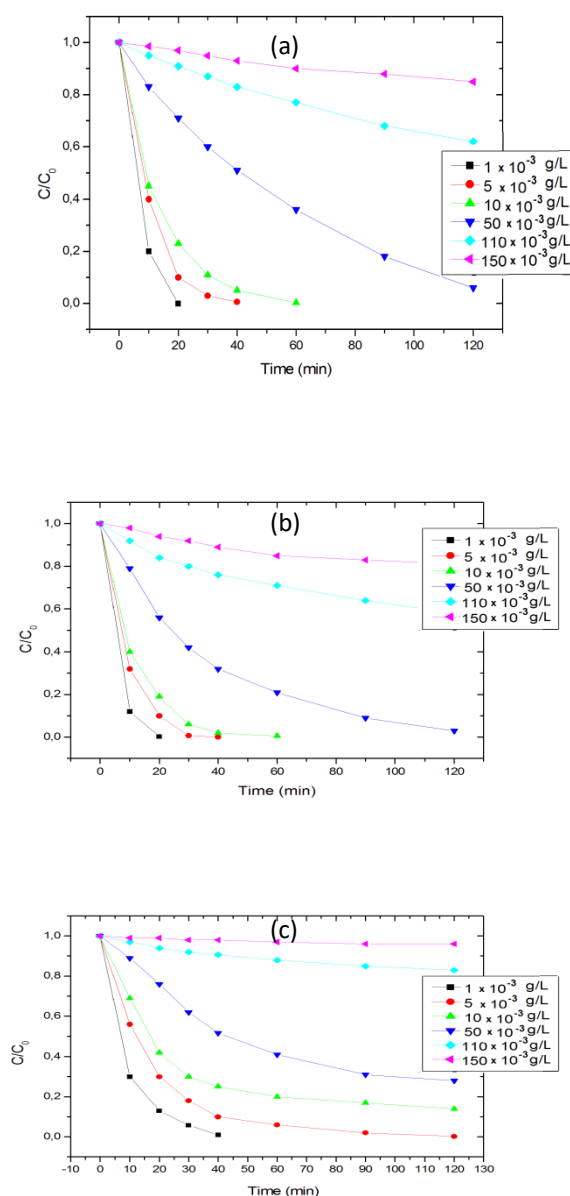


Figure12. Effect of initial concentration of BEY28 on kinetic degradation of the dye (BEY28) in an aqueous suspension of : (a) TiO_2 , (b) ZnO and (c) Fe_2O_3 . $(Photocatalyst)_0 = 1$ g/L, $pH \approx 7$, $T = 25^\circ C$, Intensity of radiation = 500 W/m^2 .

The increase in the initial concentration of Basic yellow 28 (BEY28) dye slow its photocatalytic kinetic. Two main causes

are reported in the literature to explain this phenomenon: the recovery of the catalyst surface by molecules embarrassing the generation of hydroxyl radicals, the screen effect of absorbing molecules part of *UV* radiation and preventing them from reaching the surface of TiO_2 . Both phenomena reduce the efficiency of the photocatalytic reaction, by lower radicals $HO^\bullet$ and electron pairs / holes generated [31, 32]. The kinetics of degradation of the Basic yellow 28 (BYE28) dye is shown in Fig.13.

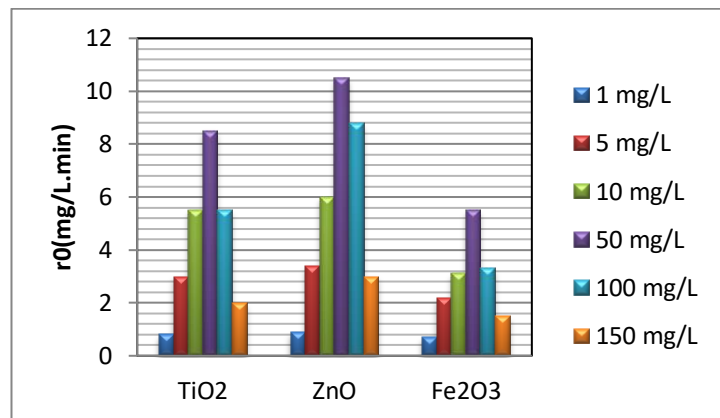


Figure 13. Effect of initial concentration of Basic yellow 28 (BYE28) on initial rate of photodegradation of Basic yellow 28 dye. ([Photocatalyst]₀ = 1 g/L, *pH* ≈ 7, *T* = 25°C, Intensity of radiation = 500 W/m²).

The increase in the initial concentration of Basic yellow 28 (BYE28) dye increases the rate of photocatalytic degradation from 1 to 50 mg/L, after that, with the increase of initial concentration, a decrease of the rate was observed. The highest rate of photocatalytic degradation of Basic yellow 28 (BYE28) dye obtained at 50 × 10⁻³ g/L initial concentration for all catalyst used.

G- Effect of radiation intensity

The effect of radiation intensity on the photocatalytic degradation of Basic yellow 28 (BYE28) dye was studied only in aqueous suspension of TiO_2 P25 and to investigate this effect, process was carried out at varying the radiation intensity at 250, 500 and 750 W/m², with fixed initial dye concentration at 10⁻² g/L; fixed catalyst dose at 1 g/L and with the same operating conditions as above namely, *T* = 25 °C, *pH* ≈ 7. The experimental results obtained are shown in the Fig.14.

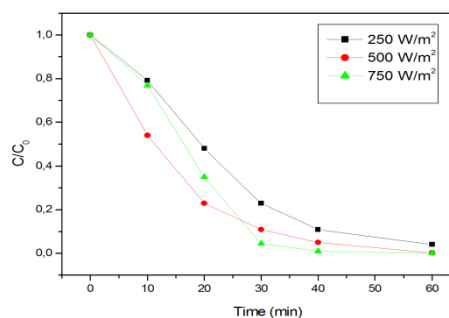


Figure 14. Effect of radiation intensity on the photocatalytic degradation of Basic yellow 28 (BEY28) dye in aqueous suspension of TiO_2 . ($[BYE28]_0 = 10^{-2}$ g/L, [Photocatalyst]₀ = 1 g/L, *pH* ≈ 7, *T* = 25°C).

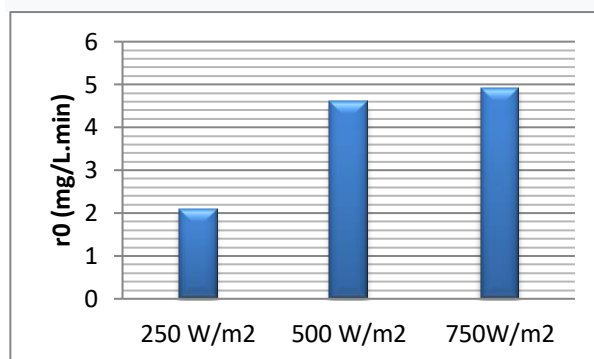


Figure 15. Effect of intensity of lamp radiation on initial rate of photodegradation of Basic yellow 28 dye in aqueous suspension of TiO_2 . ($[BYE28]_0 = 10^{-2}$ g/L, $[TiO_2]_0 = 1$ g/L, *pH* ≈ 7, *T* = 25°C).

The increase of radiation intensity from 250 to 500 W/m², increase the rate of photocatalytic degradation of Basic yellow 28 (BYE28) dye in aqueous suspension of TiO_2 P25 after that, it remained constant from 500 to 750 W/m². We can explain this by taking in the count that the number of active site on the surface of catalyst was constant, the increase of radiation intensity excite the total number of active site after that an excess of intensity radiation doesn't have any effect on photocatalytic degradation of Basic yellow 28 (BYE28) dye.

H- Total organic carbon

Mineralization of Basic yellow 28 (BYE28) dye in aqueous suspension of TiO_2 P25, ZnO and Fe_2O_3 was investigated. The experimentations were carried in the same conditions. In this case, $[BYE28]_0 = 10^{-2}$ g/L, [Photocatalyst]₀ = 1 g/L *pH* ≈ 7, *T* = 25°C, Intensity of radiation = 500 W/m². the experimental results obtained are shown in figures below:

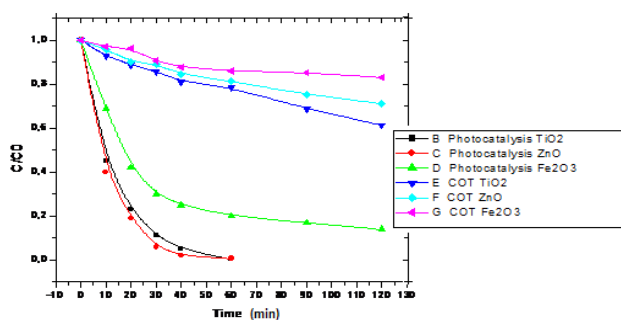


Figure 16. Kinetics of Mineralization of Basic yellow 28 (BYE28) dye by photocatalytic process in aqueous suspension of TiO_2 P25, ZnO and Fe_2O_3 ($[BYE28]_0 = 10^{-2}$ g/L, $[Photocatalyst]_0 = 1$ g/L, $pH \approx 7$, $T = 25^\circ C$, Intensity of radiation = 500 W/m^2).

TOC decreases with treatment but slower than degradation, confirming that the treatment of Basic yellow 28 (BYE28) dye by the photocatalytic process leads to a decrease in organic matter content, however this requires relatively slow processing time.

IV. Conclusion

The kinetics of photocatalytic degradation of Basic yellow 28 (BEY28) dye in an aqueous suspension of TiO_2 P25, ZnO and Fe_2O_3 in a Sun Test CPS+ was studied. All these results show that:

- Basic yellow 28 dye disappearance rate increases with the increasing of the quantity of TiO_2 , ZnO and Fe_2O_3 , from 0.1 to 3g/L; after that, it decreases with the increasing of the quantity of TiO_2 , ZnO and Fe_2O_3 , from 3 to 4g/L. optimum amount of catalyst chosen in all experiments equal to 1g/L.
- pH is a parameter to take into consideration. In the all cases, the efficiency of the degradation of the dye is better in the basic pH range.
- The rate of photocatalytic degradation of Basic yellow 28 (BEY28) dye increases with the temperature increasing for the three catalysts.
- The increase in the initial concentration of Basic yellow 28 (BYE28) dye increases the rate of photocatalytic degradation from 1 to 50×10^{-3} g/L after that decreases from 50×10^{-3} to 150×10^{-3} g/L. The highest initial rate of degradation of Basic yellow 28 (BYE28) dye was obtained at 50×10^{-3} g/L initial concentration for all catalyst used.

- The increase of intensity radiation increases the rate of photocatalytic degradation of Basic yellow 28 (BYE28) dye from 250 to 500 W/m^2 and after that remained constant.
- TOC decreases with treatment, confirming that the photocatalytic process leads to a decrease in organic matter content.

Acknowledgements

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List of symbols

BYE28 :	Basic yellow 28
TiO_2 :	Titanium dioxide
ZnO :	Zinc oxide
Fe_2O_3 :	Ferric oxide
C :	Concentration
C_0 :	Initial concentration
pH :	Potential hydrogen
T :	Temperature
$^\circ C$:	Celsius
h :	Hour
t :	Time
min :	Minute
IR :	Infrared
$[TiO_2]_0$:	Initial concentration of titanium dioxide
$(BYE28)_0$:	Initial concentration of Basic yellow 28
$(BYE28)$:	Concentration of Basic yellow 28
$[Photocatalyst]_0$:	Initial concentration of photocatalyst
TOC :	Total organic carbon.

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