

Photodegradation of C.I. Acid Yellow 23 Using Immobilized ZnO under Sunlight Irradiation

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ABSTRACT/RESUME

Abstract: The present study explored the possibility of using immobilized ZnO to photodegrade C.I. Acid Yellow 23 (tartrazine). Through this immobilization method, the separation process of fine zinc oxide (ZnO) photocatalyst at the end of the treatment can be avoided. The photocatalytic degradation of both dyes was investigated under sunlight irradiation. The results showed that the percentage of dye removal increased with decreasing initial dye concentration and increasing irradiation time. For the dye solution studied, higher removal efficiency was obtained under acidic solution medium. The findings have shown the potential of this application in dye removal from aqueous solution.

I. Introduction

Wastewaters generated by textile industries are known to contain considerable amounts of toxic aromatic dyes [1,2]. The colored wastewater released into the ecosystem is a dramatic source of pollution and perturbation in the aquatic life [3]. Because of the high stability of textile dyes and their toxicity, conventional biological processes involving activated sludge do not allow complete removal. Physical processes are nondestructive, since they only consist of a transfer of the pollution to another phase, and hence pollution post-treatment is then necessary to perform degradation [4,5]. Moreover, regeneration of the adsorbent materials and post-treatment of solid wastes, which are expensive operations, are needed. Recently, there has been a considerable interest in the utilization of advanced oxidation processes (AOPs) to remove organic compounds. AOPs are based on the production of hydroxyl radicals as oxidizing agent to mineralize organic compounds [6]. Semiconductor photocatalysis is a newly developed AOP, which can be conveniently applied for the degradation of dye pollutants. Semiconductors (such as TiO₂, ZnO, Fe₂O₃, CdS, and ZnS) are important due to the electronic structure of the metal atoms in chemical combination, which is characterized by a filled valence band, and an

empty conduction band [7]. The biggest advantage of ZnO in comparison with TiO₂ is that it absorbs over a larger fraction of UV spectrum and the corresponding threshold of ZnO is 425 nm.

The aim of the present work is to study the photocatalytic degradation of a mono azo anionic dye in the presence of immobilized ZnO photocatalyst under solar light. The effect of various parameters, such as concentration, flowrate of tartrazine solution and mass of catalyst on the decay of pollutant was investigated using a new reactor design.

II. Materials and methods

II.1. Chemical and reagents

II.1.1. Pollutant

C.I. Acid Yellow 23 (AY23), a mono azo anionic dye was obtained from ACROS organics (USA)

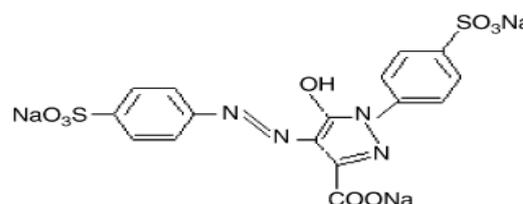


Figure 1. Chemical structure of Acid Yellow 23.

II.1.2. photocatalysts

A commercial available ZnO was used as a photocatalyst produced by Aldrich Chemical Company. This photocatalyst has a structure of 100% Zincite form with BET specific surface area of 9 m².g⁻¹ and 1.5 μm average size.

II.2. Photoreactor

A photocatalytic experiments were carried out in a pilot plan specially developed for photocatalytic application installed at the Solar Equipment Development Unit (UDES) on the north of Algeria (latitude 36°39'; longitude 2°42') using natural sun-light irradiation. Solar ultraviolet radiation (UV) was measured by global UV radiometer (KIPP&ZONZN, CMP11).

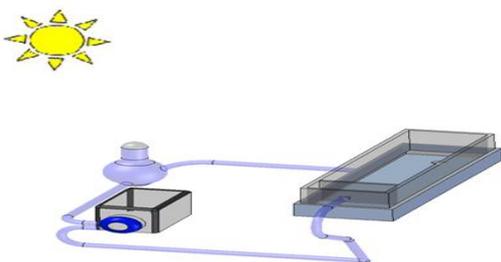


Figure 2. The schematic diagram of thin film immobilized surface reactor setup.

III. Results and discussion

III.1. Adsorption, direct photolysis, and photocatalysis

Preliminary experiments were carried out to determine the photocatalysis performance (Fig. 3). An initial three successive stages experiment was undertaken: the first stage took place at ambient temperature in the presence of ZnO without radiation; the second stage is in the presence of radiation without ZnO; and the third under UV radiation with ZnO catalyst. Adsorption in Fig. 3 shows the sorption kinetics of the active substance over time. Equilibrium was reached after several hours and only 9% of the active molecule was adsorbed. In the absence of ZnO, no significant decrease in the concentration of the active molecule was observed during the 3 h of illumination, the amount of Acid Yellow removed is less than 3%. In photocatalysis, the presence of ZnO with solar radiations has increased the removal efficiency up to 90%. Thus, in comparison to the mechanism of photocatalytic degradation, the kinetics of adsorption and photolysis were negligible [8].

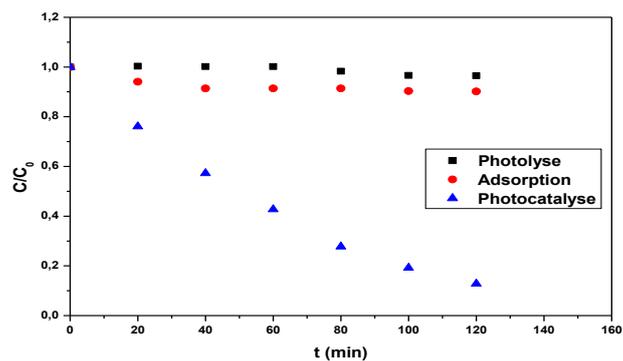


Figure 3. Temporal evolution of Acid Yellow 23 – reduced concentration for different processes.

III.2. Effect of pH

The interpretation of pH effects on the efficiency of dye photodegradation process is a very difficult task since it has multiple roles. Because of the amphoteric behavior of most semiconductor oxides, an important parameter governing the rate of reaction taking place on semiconductor particle surface is the pH of the dispersions, since, it influences the surface-charge properties of the photocatalysts. Further, industrial effluents may not be neutral. Therefore, the effect of pH on the rate of degradation needs to be considered. Experiments were carried out at pH values of (4–11) using 20 mg l⁻¹ AY23 Fig. 4 shows the time–concentration profiles of degradation of AY23 at various pHs. The extent of photocatalysis increased with increasing pH. The decrease in the photocatalytic degradation at acidic pH may be due to dissolution of ZnO at low pH. The presence of Zn²⁺ was measured at three different pHs. The results indicate that at pH of 7.5 there was no trace of Zn²⁺. This clearly confirms that at acidic medium the dissolution of ZnO would occur. At higher pHs, there were excess of hydroxyl anions, which facilitate photogeneration of hydroxyl radicals [9–10].

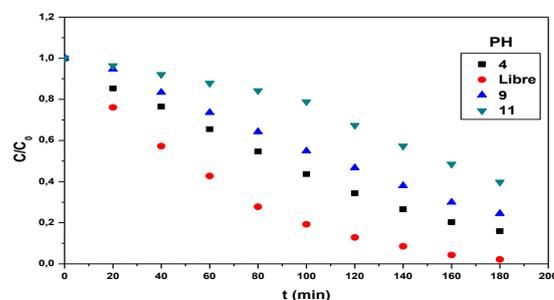


Figure 4. Effect of initial pH on photocatalytic degradation of AY23

III.3. Effect of the flow rate

The effect of the flow rate on the degradation kinetics of Methylene Blue was investigated employing different flow varying from 8 to 38 L/min. The curves in Fig.5 shown that for flow 8 L/min the degradation is slightly lower than for flows 26 and 38 L/min. This is due to a low speed of ZnO particles, due to low traffic volume which limits the effectiveness of treatment. With the flow 26 and 38 L/min we got close degradation rates as shown in Fig.4.

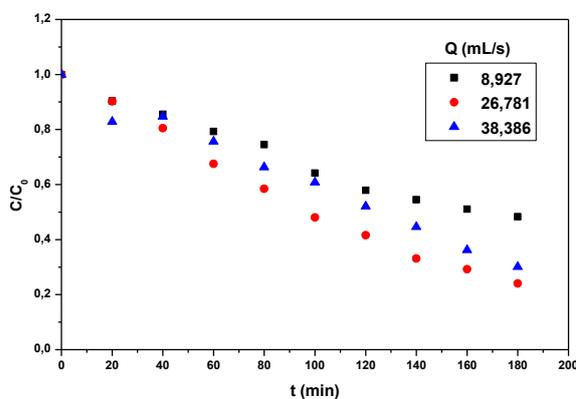


Figure 5. Kinetics of the photocatalytic degradation of AY23 at different flow.

III.4. Effect of the initial dye concentration

The influence of initial concentration of the dye solution on the photocatalytic degradation is an important aspect of the study. Initial concentrations of AY23 were varied in the range of 20–40 mg l⁻¹ and the photodegradation percent decreased with increasing initial concentration of the dye solution as shown in Fig. 6. An explanation to this behavior is that as initial concentration increases, more and more organic substances are adsorbed on the surface of ZnO, therefore, the generation of hydroxyl radicals will be reduced since there are only a few active sites for adsorption of hydroxyl ions and the generation of hydroxyl radicals. Further, as the concentration of a dye solution increases, the photons get intercepted before they can reach the catalyst surface, hence the absorption of photons by the catalyst decreases, and consequently the degradation percent is reduced [11–12].

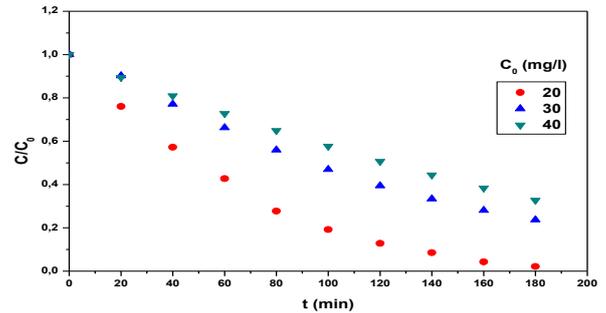


Figure 6: Temporal evolution of AY23 elimination rate for different initial AY23 concentration.

Table 1 show that the rate constant decreases with increasing the initial pollutant concentration; this is due to molecular interactions occurring during the reaction. An optimal degradation rate of 78.2 % is obtained for an initial concentration of 2.5 mg/L. The kinetic constants increase when the concentration of methomyl decreases. The rate constants for each concentration were determined from the linear regression of $-\ln(C/C_0)$ versus time for reactions of order 1 (fig 7).

Table 1. Value of kinetic constants, medium solar flux and AY23 elimination rate for different initial AY23 concentration

	C ₀ (mg/L)	k _{app} (min ⁻¹)	r ₀ (mg/L.min)	t _{1/2} (min)	R ²	R (%)	Solar flux (W/m ²)
TRZ	20	0,017	0,3382	40,990	0,997	97,863	853,151
	30	0,008	0,2723	89,093	0,998	76,298	792,988
	40	0,006	0,2348	118,083	0,997	67,276	666,876

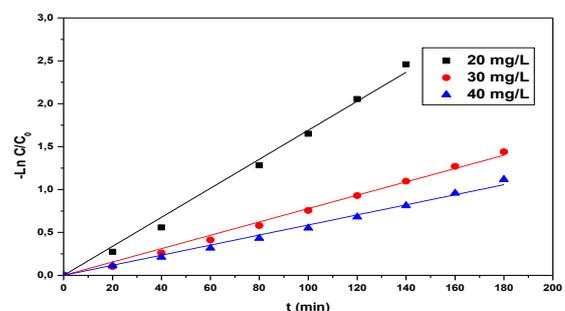


Figure 7. Kinetics of AY23 dye degradation for different initial concentrations.

IV. Conclusion

The degradation of C.I. Acid Yellow 23 (AY23) in the presence of immobilized ZnO photocatalyst was investigated under solar irradiation. The results show that the photocatalytic process UV/ZnO seems to be very efficient in the removal of dyes resistant to conventional techniques. The solar photocatalysis can be recommended for its low energy cost and very good efficiency.

Acknowledgements

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