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Photocatalytic Degradation of Yellow-GCN dye using C-N-codoped TiO₂ Thin Film in Degradation Reactor Using Visible-Light Irradiation

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ABSTRACT

Yellow-GCN (C₂₈H₁₄N₂O₂S₂) textile dye was successfully degraded by photolysis process under solar and visible light irradiation using C-N-codoped TiO₂ thin film photocatalyst. Yellow-GCN [120 mg/L] was irradiated by using 2 visible lamps (philips LED 7 watts 600 lumen and 14 watts 1400 lumen) and solar intensity around 25.000 lux with concentration and time variation. Degradation efficiency was measured by spectrophotometer UV-Vis (λ=300-800 nm). The optimum C-N-codoped TiO₂ thin film used under solar irradiation was 5 coatings. From both different light sources can be concluded that degradation under solar irradiation was better than visible light irradiation due to its degradation percentage during 120 minutes was 30,25%.

Key words : yellow-GCN, solar light, visible light, C-N-codoped TiO₂, thin film, photocatalyst.

INTRODUCTION

Yellow-GCN can be found in industrial wastewater which has carcinogenic effect. It is synthetic organic dye, stable with aromatic heterocyclic chains. Its stable structure can produce cotton with a longer lifetime dye, unfortunately effluent consumption contribute negative impact to water ecosystem. The dye influences water characteristic just by impede solar, penetration and decrease photosynthesis cycle process [1]. Thus, it is very important to degrade the dyes into environmentally friendly simple compounds.

Technologies have been developed to reduce or even remove synthetic dyes from wastewater, such as membrane filtration [2], absorption technique [3], coagulation-floucculation [4], biology technique by using microba or pure enzyme [5,6]. As promising alternative, heterogeneous photocatalysis using semiconductor which one of AOPs (Advanced Oxidation Processes) method could be used for dyes wastewater treatment.

Titania (TiO₂) is one of semiconductor catalyst which has great potential in organic wastewater treatment [7,8] due to its high catalytic activity and high biological and chemical stability [9]. However, the main deficiency of TiO₂ that active only under UV light irradiation because of its large band gap (anatase ≈ 3,2 eV) [10]. To increase photocatalytic performance of TiO₂, doping TiO₂ with other elements is the best way.

TiO₂ doping with non metal elements (C, N and S) is the most important method to raise utilization of visible light irradiation in dye wastewater degradation [11]. TiO₂ doped carbon and nitrogen show amazing photocatalytic activity than using another TiO₂ modification (N-doped TiO₂, La-doped TiO₂, Fe-doped TiO₂, Bi₂WO₆/TiO₂) [12-15]. Mostly, application of photocatalytic processes are widely carried out in slurry system operating using TiO₂ or TiO₂ modification powder. The major problem in these systems is needed much time and difficult process to

separate and recycle the catalyst powder [16]. Thus, to solve this problem semiconductor catalyst powder are coated on supporting substrate to form thin film. Supporting substrate, including glass (2011, Fang Li), glass sphere (2015, V.Vaiano), hollow glass microsphere (2014, Lei sun), and stainless steel (2003)

C-N-codoped TiO₂ was synthesized by using peroxo sol-gel method. The advantage of this method are using water as solvent and lack of chemical which beneficially low cost, environmental friendly, and simple steps [17]. Meanwhile, immobilization of C-N-codoped into glass substrate is formed by dip-coating method.

In previous research, powder of C-N-codoped TiO₂ photocatalyst was used to degrade dyes in aqueous medium and irradiated by solar light without degradator [18,19]. Based on its great photocatalytic activity under solar-light irradiation, in this research C-N-codoped TiO₂ in two dimensional system as thin film which is mobilized into glass substrate. With the result that, it could be applied to larger wastewater volume using degradation reactor which was planned. This application became effective, efficient, simple and low cost technology to solve environmental problems.

MATERIALS AND METHODS

Equipments

Spectrophotometer UV-Vis (S.1000 Secomam Sarcelles, French), Degradator reactor (SFN-Deg 001), Visible light (Phillips LED, 21 watt), analytical balance, and glasses equipments.

Materials

Yellow-GCN dyes (C₂₈H₁₄N₂O₂S₂, Mr = 474.56 g/mol from Silungkang Industry, distilled water, C-N-codoped TiO₂ thin film.

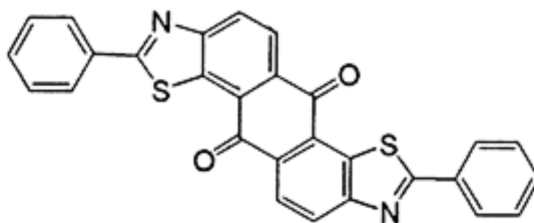


Figure 1. Structure of Yellow-GCN dyes

Photocatalytic Activity of C-N-codoped TiO₂ Thin Film

The photocatalytic activity was evaluated by the degradation of 120 mg/L yellow-GCN aqueous solution. The solution (40 mL) were put into beaker glass which is the container of the degradation reactor. Then, thin film catalyst is inserted into the solution and set spinning at constant speed by the reactor system. The reactor was enclosed in a cabinet during visible irradiation to avoid interference from natural light. While, irradiation under solar light the reactor was open widely. The experiment was carried out under visible light using 2 visible lamps (philips LED 7 watts 600 lumen and 14 watts 1400 lumen) and solar intensity around 25.000 lux. Degradated dyes solution was analyzed at $\lambda_{\max} = 419$ nm by spectrophotometer UV-Vis (S.1000 Secomam Sarcelles, French).

The degradation percentage of dye from solution at different time interval and condition is shown as :

$$\% \text{ deg} = \frac{A_0 - A_t}{A_0} \times 100\%$$

Where A_0 is the initial absorbance of yellow-GCN and A_t is absorbance of yellow-GCN at different condition. The effect of C-N-codoped TiO₂ amount on glass substrate and irradiation time on photodegradation of yellow-GCN was tested.

RESULTS AND DISCUSSION

Effect of C-N-codoped TiO₂ amount on glass substrate

In order to define the best C-N-codoped TiO₂ amount deposited on glass substrate, number of coating was varied into 1, 3, and 5 coating and each was irradiated under solar light for an hour. Figure 2 shown the effect of C-N-codoped TiO₂ amount on glass substrate. The percentage removal of dyes with 1, 3, and 5 coatings about 8,42; 7,55; 11,24%, respectively. From 3 coatings into 5 coatings evidenced that up number of coating increased the amount of

C-N-codoped TiO₂ deposited on glass substrate. Thus, photocatalytic activity increased and the dyes in aqueous solution will be more degraded[19].

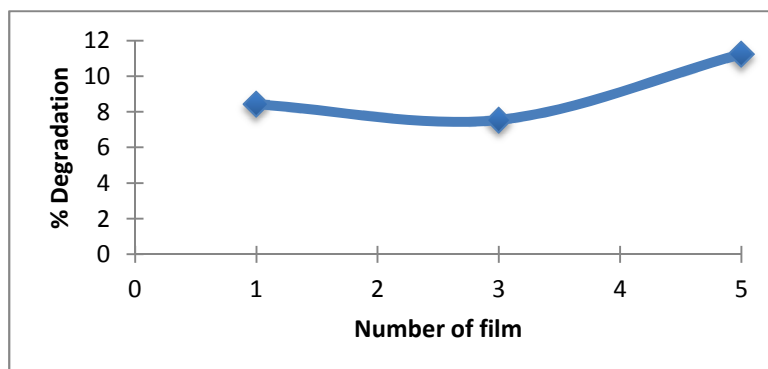


Figure 2. Effect of C-N-codoped TiO₂ amount on glass substrate

Effect of irradiation time under visible and solar light

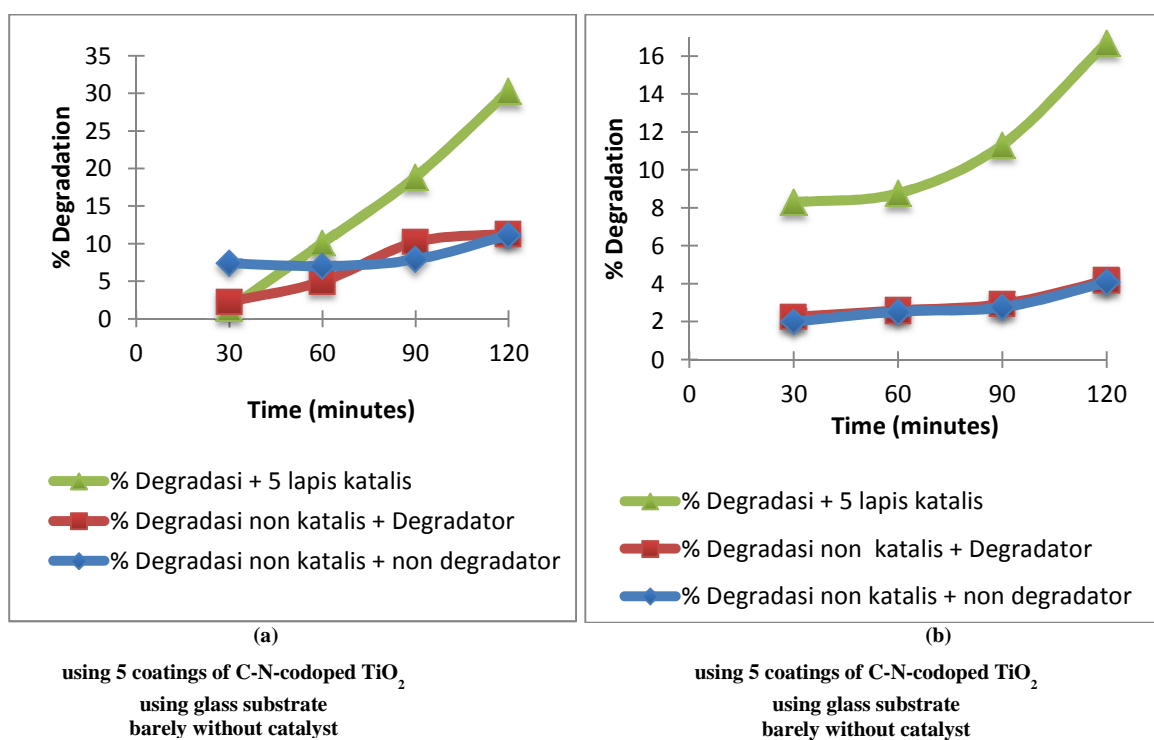


Figure3. Effect of irradiation time under (a) solar light (b) visible light

Figure 3 shown the effect of irradiation time under both of solar light and visible light irradiation. The percentage removal of dyes increasing by increment of irradiation time. During the photocatalytic reaction, C-N-codoped TiO₂ absorbs light to produce electron-hole pair which migrates to the catalyst surface to react with absorbed O₂ and H₂O, to produce strong oxidizing agents in the form of O₂⁻ and HO• radicals, respectively, which are the main species responsible for the degradation of organic pollutants[20]. By increasing the irradiation time, C-N-codoped thin film photocatalyst will more produce strong oxidizing agents. Thus, the photocatalytic degradation will increase. From figure 3, we can see that glass substrate has no photocatalytic activity due to its low degradation percentage. The structure compound of yellow-GCN dyes is also very difficult to be degraded naturally. It is shown in figure 3 that degradation percentage without catalyst is only 4,07% and 11,15% under visible light and solar light irradiation, respectively. Highest degradation percentage of yellow-GCN is achieved about 30.25% under solar light irradiation for 120 minutes.

Recyclability of C-N-codoped TiO₂ thin film 5 coating

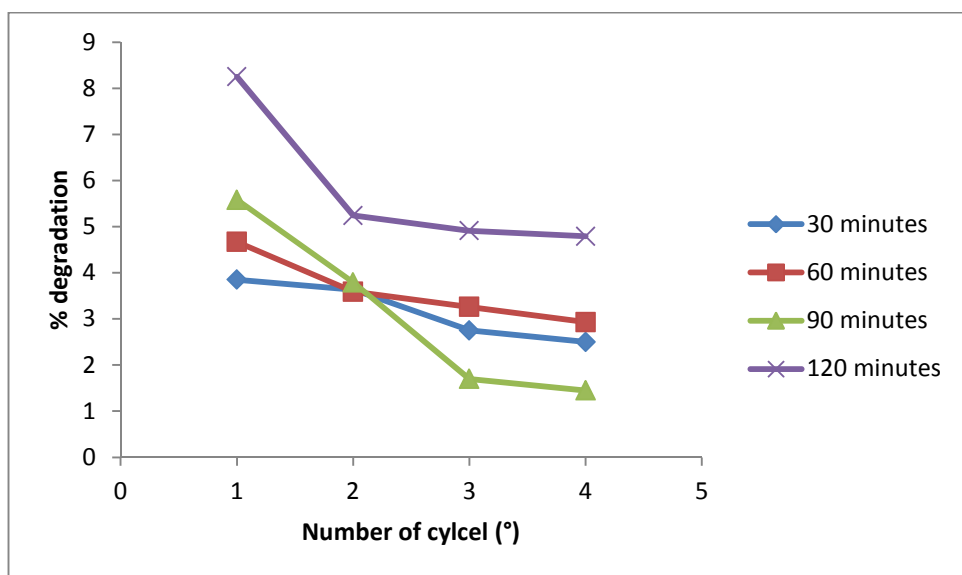


Figure 4. Recyclability of C-N-codoped TiO₂ thin film after 4 recycling experiments

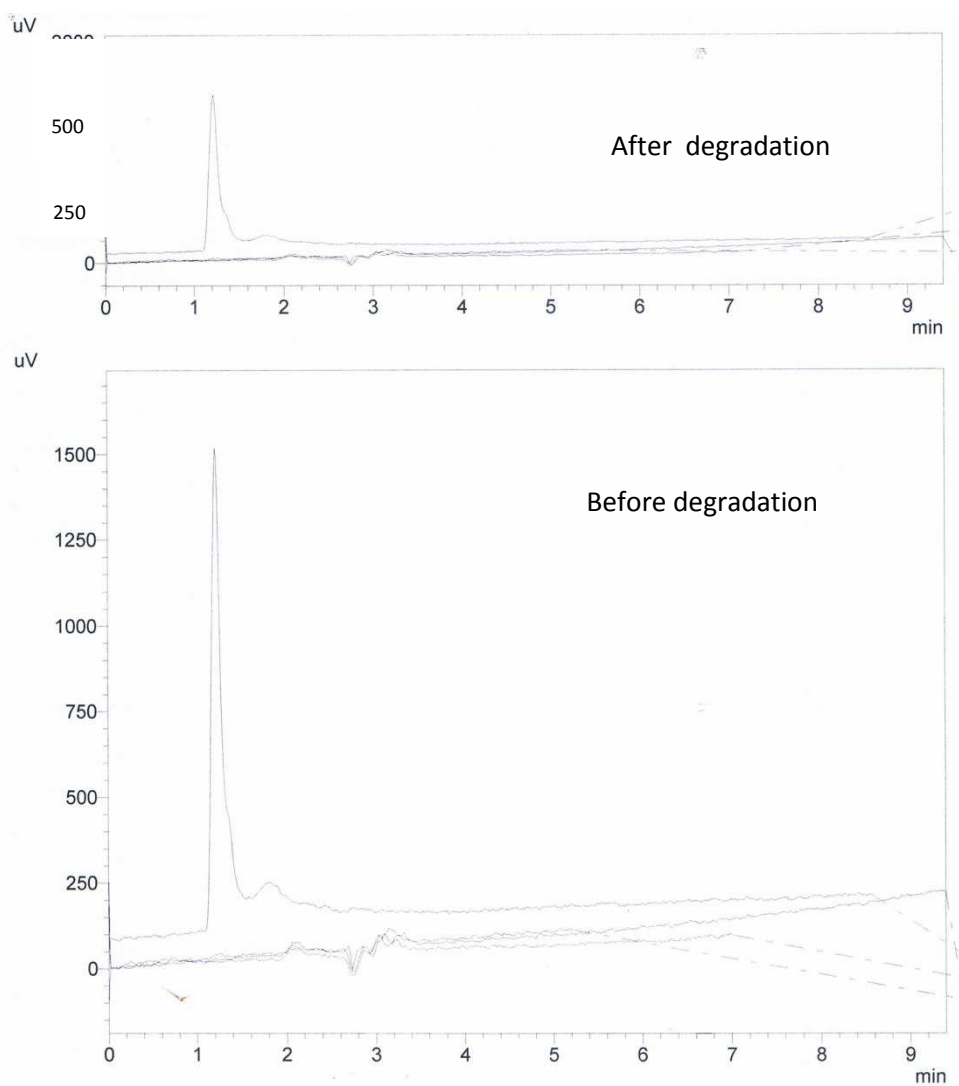


Figure 5. Analysis of Yellow GCN before and after degradation

Recyclability is one of the most important factor in catalysis research. To confirm the recyclability of C-N-codoped TiO₂ thin film with 5 coatings, the photocatalytic decolorization reaction was repeated up to four cycles in different irradiation time under visible light. The results is shown in figure 4. The results demonstrated that there was about 2% reduction of degradation percentage in 30, and 60 minutes of irradiation and about 4% in 90 and 120 minutes of irradiation after four cycles. These results confirm that there is no leaching of C-N-codoped TiO₂ from immobilized on glass substrate.

Analysis using High Performance Liquid Chromatography

Figure 5 describes that peak high and peak areas of Yellow-GCN decrease after degradation using C-N-codoped TiO₂ thin film in degradation reactor under solar-light irradiation, without other peaks appeared.

CONCLUSION

Yellow-GCN dyes in aqueous solution was successfully degraded by C-N-codoped TiO₂ immobilized on glass substrate. The degradation percentage of 120 mg/L yellow-GCN (40mL) is 4,07% and 11,15% without catalyst under visible and solar light irradiation for 120 minutes, respectively. By the addition of C-N-codoped TiO₂ thin film with 5 coatings and set constant spinning speed, the degradation percentage increase to 16,7% and 30,25% under visible and solar light irradiation, respectively. The C-N-codoped TiO₂ thin film photocatalyst confirm that there is no leaching while mineralization of dyes occurred. Photocatalytic degradation of yellow-GCN dyes in aqueous solution is more effective under solar light irradiation.

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