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RESEARCH ARTICLE

THE PREPARATION AND PHOTOCATALYTIC PERFORMANCE OF FE-DOPED TIO_2/GO PHOTOCATALYSTS.

Huiming Zhang, Bin Xu and He Bian.
Department of Chemical Engineering and Safety, Binzhou University, Binzhou 256603, PR China.

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Abstract

Fe-doped TiO_2/GO photocatalysts were synthesized with tetrabutyl titanate, acetylacetone, ferric chloride and graphite powder by hydrothermal method. The photocatalysts were characterized by UV–visible spectroscopy, FTIR and BET. The photocatalytic efficiencies of the catalysts were studied by degradation with rhodamine B(Rh B) as a representative organic under visible light irradiation. The results indicated that ferric acetylacetonate improved the RhB absorption of TiO_2 and the graphene accelerate the electron conduction. The synergistic effect of the two improved the photocatalytic performance of TiO_2.

***Corresponding Author:* Huiming Zhang.**

Introduction:

The environment faces more and more challenges of increasing organic pollutants. As we know, the organic pollutants can't be decomposed themselves. Several methods have been used for decomposing pollutants, such as biological treatment, ozonation and advanced oxidation processes. Photocatalytic degradation attracted researchers' interest because it can make good use of solar energy to decompose organics [1-3]. Semi-conductors were studied in photocatalysis field.

Comparing with other semiconductors, TiO_2 has many advantages, such as richness, high photocatalytic activity, nontoxic and stability. However, there are two difficulties we face when the TiO_2 were used. Firstly, only under the ultraviolet that wavelengths of $\lambda<376$nm, the electron and valence can be generated to degrade the pollutants, while the ultraviolet account for only 3%-4% of total incident sunlight. Secondly, the combination of electron and hole reduces the efficiency of the catalysts. Therefore, efforts have been made to improve the charge separation efficiency and the visible absorption of TiO_2[4-5].

Several investigators have attempted doping with metal ions to shorten the band gap to absorb the visible lights[6], metal-ion dopant may influence the photo reactivity of TiO2 by acting as electron traps and by altering the electron-hole recombination rate. For example, Zn-doped TiO_2 was prepared and Zn^{2+} promote TiO_2 to capture hole and electron, so that it can decrease the recombination of hole-electron[7] . Fe doped TiO_2 was synthesized by hydrolysis of titanium butoxide and it's more active than bare TiO_2 in degrading organic molecules[8]. Fe was used to decrease the band gap of TiO_2 and to extend the photo response range of the TiO_2 matrix to the visible region.

Graphene is a two-dimensional atomic sheet of sp(2) hybridized carbon with exceptional properties, such as high charge mobility, optical transparency, and mechanical flexibility[9,10]. Because of its high electron transition and larger specific surface area, graphene is used for exploring new composite photocatalysts. Khalid synthesized new type graphene-TiO_2 composite photocatalysts and the result shows that the composite material can degrade Methylene orange effectively[11].

One of the problems is that how to improve the lipophilicity of TiO_2 in degradation the organic compounds. So that we used Iron acetylacetonate (FeAA) as Fe resource to prepare Fe doped TiO_2. We also added graphene oxide to enhance the electron transmission speed.

Experiment:

Preparation of TiO_2 modified by Iron(III) acetylacetonate(FeAA)

Synthesis of Iron(III) acetylacetonate(FeAA)

10 mmol of $FeCl_3$ was dissolved into 20 ml of deionized water, and then 10 ml of acetylacetone was added to get two-phase of oil and water. 10ml of ethanol was dropped into the forenamed mixed solution until the solution became deep red. Subsequently 8 ml of ammonia was added slowly with red precipitate generating in the process. The solution was stirred for another 15 min. Solids were got by the way of vacuum filtration and washed with deionized water several times, then the solids were dried at 343K for 2h to get FeAA.

Synthesis of TiO_2:

Firstly, 17 ml of tetrabutyl titanate was added into 30ml of ethanol (under stirring condition), followed by adding hydrochloric acid and deionized water (the pH of the solution was kept between 2-3), and then the solution was transferred to a hydrothermal reaction kettle. The reaction kettle was put into muffle furnace at 453K for 8 h. The kettle was removed from the oven and cooling to room temperature to get the raw materials, which should be washed by deionized water and anhydrous ethanol and dried at 353K for 24h. Finally, the dried powder was calcined at 673K for 4h and the TiO_2 powder was synthesized.

Preparation of TiO_2 modified by FeAA:

TiO_2 and FeAA were dissolved into 40ml of ethanol under continuous ultrasonic stirring for 30min. The former solution was put in constant temperature water bath for 2h. The solid was washed with deionized water and ethanol, then dried at 70℃ for 24h, obtained FeAA modified TiO_2 catalyst(Fe-TiO_2).

Preparation of TiO_2/GO:

Graphene oxide (GO) was prepared from graphite powder according to the modified method reported by Hammers[12]. In brief, 1.0 g of Fe-TiO_2 and a certain amount of GO were dispersed on 80 ml of distilled water, stirring and treated by supersonic for 30 min. The solution was transferred to hydrothermal reactor which maintained at 130℃ for 24h and cooled down to room temperature. The solid product was separated by centrifugation and washed by water and ethanol, respectively. Then TiO_2/GO was obtained.

Catalytic degradation test:

The catalytic activities of the samples (500mg) were evaluated by the degradation of rhodamine B (Rh B) in liquid phase. In the absent of light, the samples and the degradation solution coexist for 1 hour to achieve the adsorption equilibrium with the help of ultrasonic wave. Then the Xe lamp would be turned on. At irradiation time intervals of 20 min. 5 ml of the suspensions were collected and then centrifuged to remove the particles. The obtained solutions were analyzed by UV-vis spectrophotometer and the absorbance at 464 nm was monitored.

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Results and Discussions:-

Characterizations of Catalysts Fe-T:

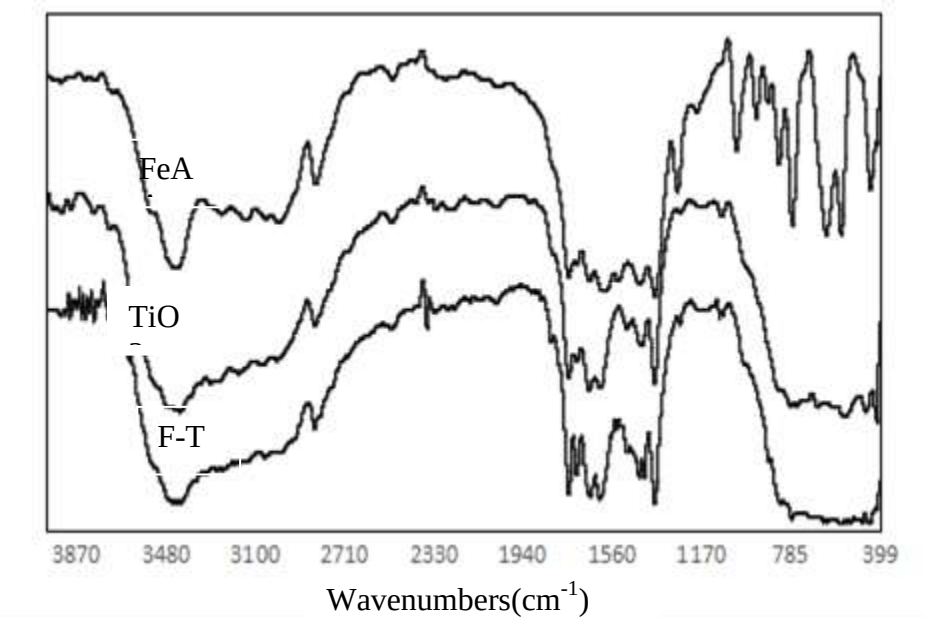

Fig. 1: FT-IR spectra of catalysts

Fig. 1 shows the FT-IR spectroscopy of FeAA, TiO_2 and Fe-doped TiO_2(F-T). The absorption peak from 2900 $cm^{-1}$to 3000 cm^{-1} in the spectrum of FeAA can be attributed to the stretching vibrations of C-H of CH_3^-, and the peak from 1500 cm^{-1}-1600 cm^{-1} can be attributed to C=O. From the spectrum of TiO2, the bending vibrations of Ti-O is at 500 cm^{-1}-700 cm^{-1}, and the featured peak of O-H is at 3400 cm^{-1} and 1600 cm^{-1}, mainly caused by the absorption of water on the surface of TiO_2. The feature peak of FeAA isn't found in the spectrum of F-T, maybe caused by the low content of FeAA.

Photocatalytic Activities of Fe-T:

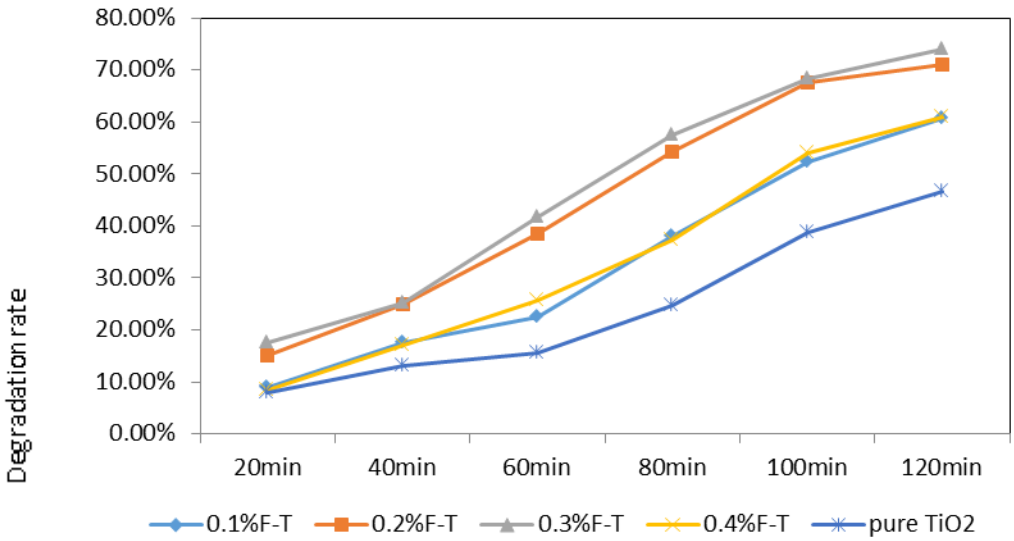

Fig. 2:- Degradation rate curves of FeAA with different Fe content over time

Fig. 2 shows the degradation of TiO_2 with variety content of FeAA. The degradation rate grows with the degradation time increasing. Among all the catalysts, the best degradation happened when the FeAA contend is 0.3%.

Photocatalytic Activities of Fe-T/GO:

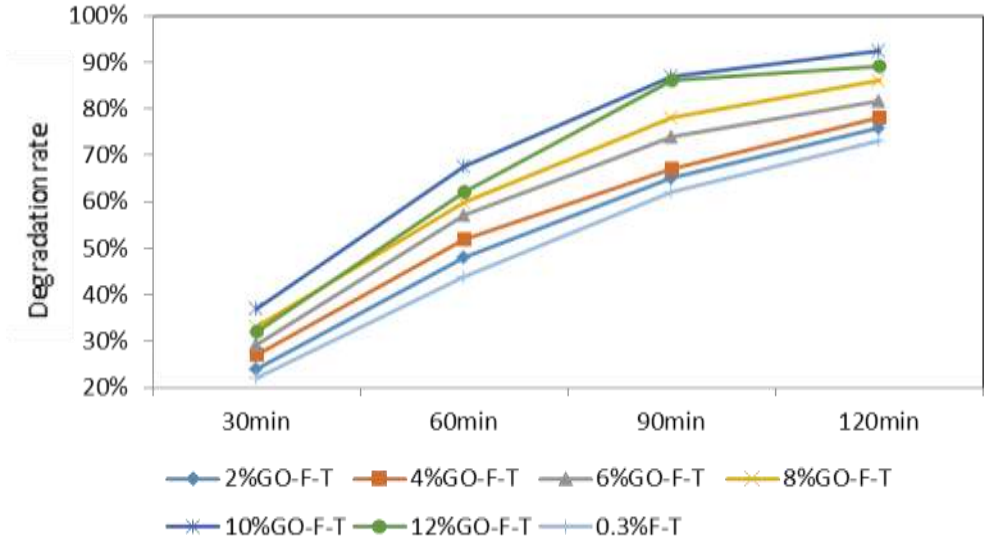

Fig. 3:- Photocatalytic degradation under visible light irradiation using various catalysts

The photocatalytic degradation of GO-F-T exposed to visible light irradiation was used as a test reaction to evaluate the catalytic activity of the various GO/F-T samples. As shown in Fig.3, the degradation rate of the F-T doping with GO were higher than that without GO. The catalyst with 10% GO contents showed the best performance that the degradation rate reached 95.2% under the visible light for 120 min.

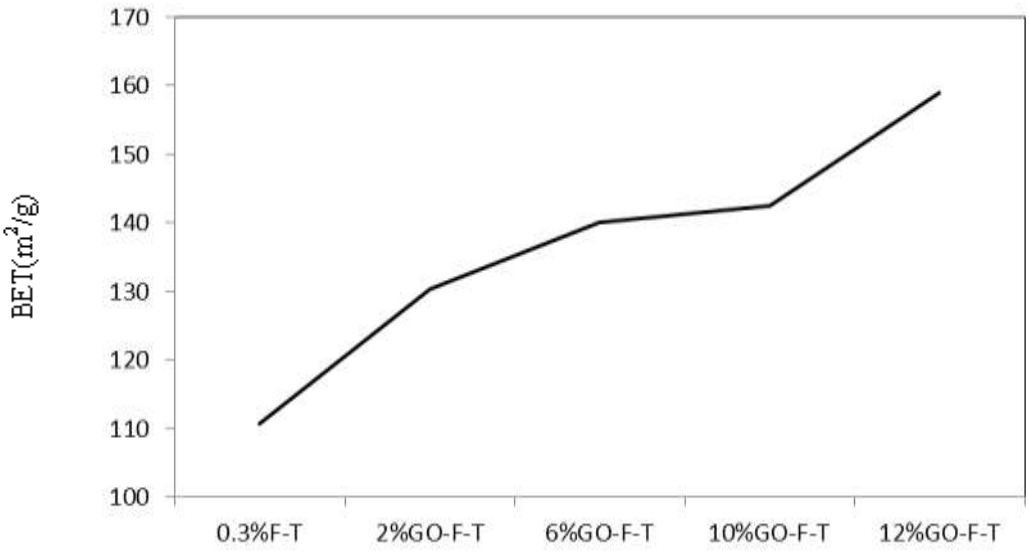

Fig.4:- specific surface area of various catalysts with different GO content

Fig.4 shows that the specific surface area increases with the increase of GO content. The degradation rate showed in Fig.3 didn't increase with the increase of GO content, because the active sites were covered with excessive GO.

Conclusions:-

This work has investigated the effects of FeAA and GO contents on the photocatalytic performance of Fe-doped TiO_2/GO photocatalysts for Rh B degradation. The results showed that the best degradation rate happed when FeAA and GO contents are 0.3% and 10%, respectively.

Acknowledgments:-

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