

# Heterogeneous photo-Fenton oxidation for paracetamol removal using iron containing TiO<sub>2</sub> hollow spheres as Catalyst

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## Abstract:

Fe-TiO<sub>2</sub> hollow microspheres were prepared by stage solvothermal method and substituted Fe<sup>2+</sup> for photo-Fenton oxidation degradation of acetaminophen. Fe-TiO<sub>2</sub> microspheres characterization of specific surface area and pore structure analysis SEM, TEM, XRD and XPS, etc. Therefore this work examines the performance and stability of Fe-TiO<sub>2</sub> microspheres as catalyst for this reaction. In the experiments, the concentration change of Fe<sup>2+</sup> were 10、15、20、25 mg/L and the concentration change of H<sub>2</sub>O<sub>2</sub> were 300、450、600、900、1200、1500 mg/L. After about 1 hour, the degradation rate of acetaminophen at Fe<sup>2+</sup> concentration of 10 mg / L and H<sub>2</sub>O<sub>2</sub> concentration of 1200 mg / L demonstrated 2 times better than the conventional Fenton method.

**Keyword:** Photo-Fenton、Fe-TiO<sub>2</sub>、Acetaminophen

## 1. Introduction

Paracetamol is commonly found in wastewaters, as a consequence of its incomplete elimination by conventional treatment methods. Fenton oxidation uses the decomposition of hydrogen peroxide with dissolved ferrous salt as a catalyst to generate HO•, showing excellent degradation rate.<sup>1</sup> However, the main drawback of this homogeneous process comes from the necessity of a posttreatment to control iron ions discharge. Solid-state catalyst Fe-TiO<sub>2</sub> hollow microspheres instead of the same phase Fe<sup>2+</sup>, to improve shortcomings that the traditional photo-Fenton oxidation process cannot recover the catalyst and produces a lot of sludge. Therefore, we replaced Fe<sup>2+</sup> with Fe-TiO<sub>2</sub> hollow microspheres, in the same conditions, photo-Fenton oxidative degradation of acetaminophen is very similar to that of traditional photo-Fenton, and the Fe embedded in the TiO<sub>2</sub> hollow microspheres did not dissolve out of the solution, the effluent did not contain Fe ions and did not produce sludge. The study used solid catalyst Fe-TiO<sub>2</sub> hollow microspheres can prove the traditional Photo-Fenton oxidation process which cannot recover the shortcomings of the catalyst and produce the large amount of sludge.

## 2. Experimental

Ethanol, ether, glycerol, FeCl<sub>3</sub> and TiOSO<sub>4</sub> were mixed and agitated in a specific ratio until completely miscible. The solution was sealed in a high-temperature autoclave and placed in an oven at 110 ° C for 4 hours. Filtered and dried to obtain a powder, and then sealed in a

high-temperature autoclave at a moisture content of 10% hydrolysis reaction placed in an oven at 70 ° C for 48 hours, and finally filtered and dried. The powder is the desired Fe-TiO<sub>2</sub> material. Which named 1Fe-TiO<sub>2</sub> is 1wt% Fe-TiO<sub>2</sub> hollow microspheres. The pH of the solution containing 50 ppm of acetaminophen contaminant was controlled at 2.8, Fe-TiO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub> were added then set up the light source to complete the Photo-Fenton oxidation system, different iron content of Fe-TiO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub> were added to detect the concentration by spectrophotometer to understand its optimal degradation conditions.

### 3. Results and discussion

In this study, Fe-TiO<sub>2</sub> instead of Fe<sup>2+</sup> added to the solution, because Fe is contained in the material, reduce the formation of iron hydroxide precipitates in solution, achieve zero-sludge results. The degradation efficiency shown in Fig.1, we learned that 5Fe-TiO<sub>2</sub> hollow microspheres in Photo-Fenton have the best degradation of acetaminophen effect.

In the photo-Fenton oxidation process, Fig.2a shows the sludge produced by the reaction of Fe<sup>2+</sup>. And we use the solid catalyst Fe-TiO<sub>2</sub> instead of Fe<sup>2+</sup> added to the solution reaction results are shown in Fig.2b, we can clearly see (b) compared to (a) is more clear and can effectively improve shortcomings that the traditional Photo-Fenton oxidation process can not recover the catalyst and produces a lot of sludge.

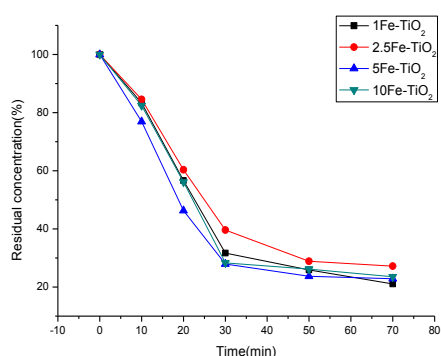


Figure 1. Fe-TiO<sub>2</sub> degradation efficiency

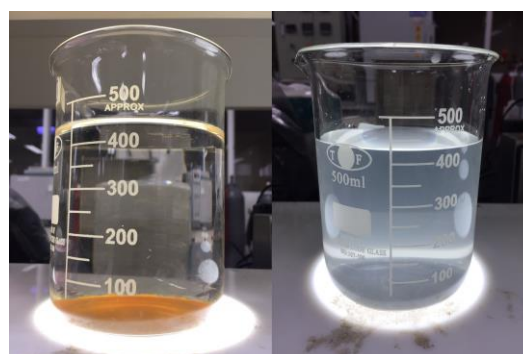


Figure 2. Photo-Fenton with different catalysts

(a) Fe<sup>2+</sup>

(b) Fe-TiO<sub>2</sub>

### 4. Conclusions

In the traditional Photo-Fenton oxidation method, the degradation efficiency of acetaminophen was the best when H<sub>2</sub>O<sub>2</sub> concentration was 1200mg / L and Fe<sup>2+</sup> concentration was 10mg / L. We use Fe-TiO<sub>2</sub> hollow microspheres substituted Fe<sup>2+</sup> are added to the solution in the same conditions, because Fe is contained in the material, so reducing the formation of iron hydroxide precipitate in the solution, the results show that 5Fe-TiO<sub>2</sub> has the best degradation trend and can achieve zero sludge. The 5Fe-TiO<sub>2</sub> can improve the defect that the traditional Photo-Fenton oxidation procedure can not recover the catalyst and the large amount of sludge.

### References

- [1] Stackelberg, P.E., Furlong, E.T., Meyer, M.T., Zaugg, S.D., Henderson, A.K., Reissman, D.B.. Sci. Total Environ. 2004; 329 (1–3), 99–113.