

Terephthalic Acid (TPA)의 분해를 위한 고도산화공정 (AOP)공정의 검토 및 실험적 접근

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Review and Experimental Analysis of the Performance of Advanced Oxidation Technologies for Terephthalic Acid (TPA) Removal

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1. Introduction

Terephthalic acid (TPA) is widely used as a raw material for the manufacture of poly-ester and polyethylene terphthalate (PET) compounds. With our ever growing dependence on plastics, the production of TPA is also increasing at about 3.2 % annually. In 2004, the production of TPA in Korea alone was about 500 million tonnes. Recently, it is evident that TPA is classified as one of the toxic chemicals and is linked to endocrine disruption ability[1,2]. Considering the widespread application of TPA with its high risk of exposure and health hazard, a simple and effective wastewater treatment method is very important.

TPA wastewater contains high amount of BOD and COD values and generally biological treatment method is adopted[3]. However, biological treatment may not be the only option always, especially when at high initial concentrations. Recently, Advanced Oxidation Processes (AOPs) with UV radiation and photocatalyst titanium dioxide (TiO_2) is gaining growing acceptance for the treatment of organic materials. Among many semiconductors, TiO_2 is frequently used as the photocatalyst because it is non-toxic, chemically stable, and possesses relatively high photocatalytic activity. Several oxidants like hydrogen peroxide (H_2O_2) is also adopted to aid in the mineralization of organic pollutants[4,5,6]. However, very limited information is currently available about the TPA destruction using AOP treatment.

In this study, a detailed investigation was made in studying the performance of TPA degradation using AOP treatment (UV/ TiO_2). The effectiveness of H_2O_2 individually and in combination with UV/ TiO_2 system (as hybrid process) was also analyzed.

2. Experimental Materials and Method

TPA used in this study was obtained from Sam Nam Petrochemical Co., Ltd.(Korea) and H₂O₂ (35%) solution was purchased from Yakuri Pure Chemicals Co., Ltd.(Japan). TPA aqueous solutions were prepared with ultra pure water from reverse osmosis treatment. P-25 TiO₂ particles (Degussa, Germany) were used as photocatalyst. Average diameter, BET surface area, and density of TiO₂ particles were 205 nm, 5015 m²/g, and 3.89 g/cm³ at 20°C, respectively.

A Cylindrical shape, batch type photocatalytic reactor was used for this study. Philips TUV 36 SP T5 UV lamps (6×40W) were mounted in the reactor, as UV light source. The irradiation intensity was 144 W/cm² and the wavelength was 253.7 nm. 1 L of TPA solution from 25 to 100 ppm was used for the evaluation of destruction efficiency by the several photo oxidation process such as UV/TiO₂, UV/H₂O₂ and UV/TiO₂/H₂O₂ systems. It was found that TPA remained soluble in solution up to pH 6. Especially at high concentrations (>50 ppm), further decrease in pH was not possible.

The amount of TPA in the aqueous solution was measured by a high-performance liquid chromatography HPLC (Shimadzu LC-10 VP, Japan) equipped with a UV detector (Shimadzu SPD-10A VP) and a Shim-pack CLC-ODS column. The elution was monitored at 275nm. The elutant used was a solvent mixture of 1% acetic acid and acetonitrile (85:15, v/v). The flow rate of the mobile phase was 1 ml/min. The TiO₂ photocatalyst was removed from the solution by filtration, and the resulting solution was analyzed with HPLC.

3. Results and discussion

The effect of photocatalytic efficiency based on the solution pH indicated that the decrease in pH was found to increase the degradation efficiency (Figure 1). Considering the trend in degradation pattern and the practical application, pH 8 was chosen for further experimentation. TPA removal was significantly affected based on the initial concentration (Figure 2). The effect of UV/H₂O₂, at different concentration of H₂O₂ is shown in Figure 3. Based on our results it was evident that UV/H₂O₂ was far more efficient in TPA degradation. Considering the TPA concentration of 50 ppm, UV/TiO₂ (TiO₂ = 1 g/L) would require about 10 hours for complete destruction. However, 1.5 mM of H₂O₂ would require 3 hours for the destruction of same amount of TPA concentration. It was also found that the hybrid system (Figure 4) with the combination of UV/H₂O₂/TiO₂ had a synergic effect when compared to UV/TiO₂ alone. The effect of TiO₂ concentration when applied in the combined process with H₂O₂, must be carefully

chosen. Further experiments are underway to examine, in detail, the performance of the hybrid system and to study the mechanism of TPA removal.

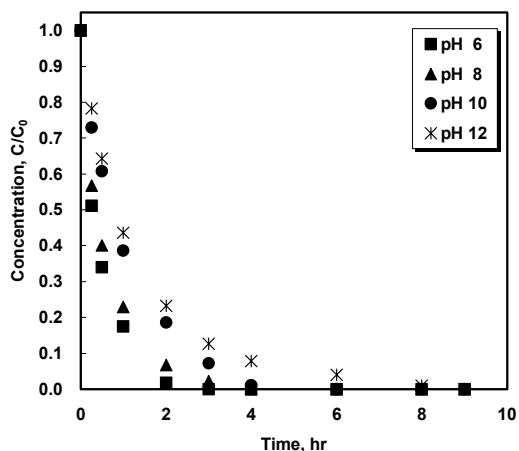


Fig. 1. Effect of initial solution pH on the destruction of TPA by UV/TiO₂ photocatalytic reaction.

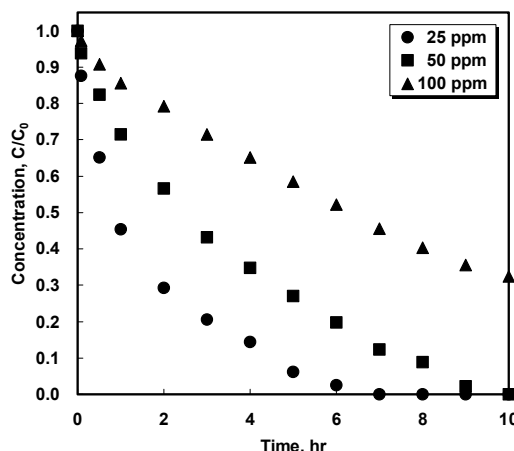


Fig. 2. Effect of initial TPA concentration on the destruction by UV/TiO₂ photocatalytic reaction.

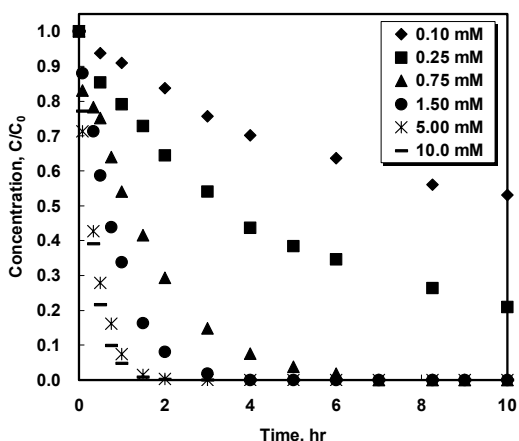


Fig. 3. Effect of H₂O₂ concentration on the destruction of TPA by UV/H₂O₂ photocatalytic reaction.

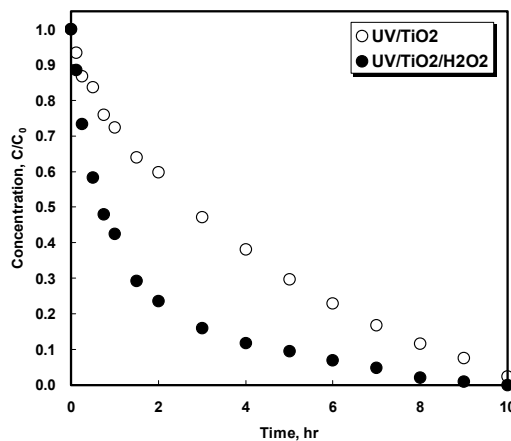


Fig. 4. Performance of the hybrid system (UV/TiO₂/H₂O₂) compared to UV/TiO₂ process

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References

1. Moore, N. P., "The oestrogenic potential of the phthalate esters", *Reproductive Toxicol.* 14: 183-192(2000).
2. Penalver, A., E. Pocurull, F. Borrull, and R. M. Marce, "Determination of phthalate esters in water samples by solid phase microextraction and gas chromatography with mass spectrometric detection", *J. Chromatogr. A* 872: 191-201(2000).
3. Afrin, R. P. and B. F. Taylor, "Aerobic and anaerobic catabolism of phthalic acid by a nitrate respiring bacteria", *Arch. Microbiol.*, 130, 101-104(1981).
4. Tae-Joon Park, Jong-Sung Lim, Youn-Woo Lee, Sung-Hyun Kim, "Catalytic supercritical water oxidation of wastewater from terephthalic acid manufacturing process", *J. of Supercritical Fluids*, 26, 201-213(2003).
5. Rodriguez, M., Sarria, V., Esplugas, S. and Pulgarin, C., "Photo-Fenton treatment of a biorecalcitrant wastewater generated in textile activities: biodegradability of the photo-treatment solution", *Journal of Photochemistry and Photobiology A: Chemistry*, 151, 129-135(2002).
6. Yeber, M.C., Rodriguez, J., Freer, J., Baeza J. and Duran N., "Advanced oxidation of a pulp mill bleaching wastewater", *Chemosphere*, 39, 1679-1688(1999).