PF7) 펜톤산화에 의한 ciprofloxacin 과 cyclophosphamide의 제거: 반응표면기법을 이용한 최적화

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1. Introduction

The main objective of this study was to investigate the removal of ciprofloxacin and cyclophosphamide from hospital wastewater by Fenton oxidation. We also aim to optimize the experimental conditions using a Box-Behnken design analysis. These two chemicals were selected as target compounds because they are frequently used in cancer therapeutic treatments and have been detected in surface waters in South Korea(NIER, 2007). For this study, we hypothesized that the organic matter in wastewater would have a negative effect on the Fenton oxidation of pharmaceuticals, and that a higher ratio of H_2O_2/Fe^{2+} would show better removals of the pharmaceuticals. In addition, it was also hypothesized that a lower pH would promote the Fenton reaction, resulting in increased removal of the chemicals.

2. Experimental Methods

Ciprofloxacin and cyclophosphamide were purchased from Sigma-Aldrich. Acetonitrile (LC/MS grade) and methanol were obtained from J.T. Baker. The Fenton oxidation experiments of the two chemicals were carried out with a jar tester at room temperature, and the reaction time was 1 h. The pH of the solution was adjusted with H_2SO_4 and NaOH solutions. After the addition of H_2O_2 , the solution was stirred for 1 min at 250 rpm and then mixed more slowly for 59 min at 100 rpm. The treated samples were with drawn after 60 min and quenched by adding 30 mL of a supersaturated sodium thiosulfate solution. These were then filtered with a 0.22-mm cellulose acetate syringe filter for LC-MS/MS analysis. To determine the effect of the molar ratio of H_2O_2/Fe^{2+} , the concentration of H_2O_2 was varied from 6.54 to 32.71 mg/L (corresponding to 19.23 to 96.15 μ M), and the concentration of Fe^{2+} was set to 10 mg/L (equivalent to 38.46 μ M). Experimental design, mathematical modeling, and optimization were performed using Design-Expert 7.0 software.

3. Results and Discussion

In this study, Fenton oxidation was investigated to treat ciprofloxacin and cyclophosphamide under different conditions of initial concentration of NOM (0-3 mg/L), initial pH (2-10), and molar ratio of $H_2O_2/Fe^{2+}(0.5-2.5)$. The design matrix of coded values for the factors and the response in terms of the percent removal of pharmaceuticals for all 17 experimental runs include the amount of organic matter (NOM), the ratio of H_2O_2/Fe^{2+} , and the effect of pH are denoted as X_1 , X_2 , and X_3 , respectively. The results obtained in the experimental design for the removal of pharmaceuticals by Fenton oxidation were used to establish the following models (equation 1 and 2) for each chemical.

$$\begin{array}{l} Y_{1}(ciprofloxacin,\%) = +90.43 - 0.53X_{1} + 4.20X_{2} - 21.46X_{3} + 17.55X_{1}X_{2} + 3.83X_{1}X_{3} + 0.47X_{2}X_{3} - 9.69X_{1}^{2} - 3.81X_{2}^{2} - 12.38X_{3}^{2} \\ (1) \\ Y_{2}(cyclophosphamide,\%) = +93.56 - 5.08X_{1} + 5.40X_{2} - 16.64X_{3} + 13.85X_{1}X_{2} + 2.11X_{1}X_{3} + 2.66X_{2}X_{3} - 8.54X_{1}^{2} - 3.88X_{2}^{2} - 10.30X_{3}^{2} \\ (2) \end{array}$$

4. References

National Institute of Environmental Research, 2007, Development of Analytical Method and Study of Exposure of Pharmaceuticals and Personal Care Products in Environment (II), Korea.

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