Utilisation of factorial experiments for the UV/H_2O_2 process in a batch reactor

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Abstract

Factorial experiments provide a comprehensive understanding of the impact of operational variables on process performance. Utilisation of the Hadamard matrix taking into account all interaction effects, appeared to be efficient for giving a mathematical model that conformed to criteria validity. The predictions given by the factorial experiments model were confirmed by the experiments. Phenol oxidative degradation kinetics were not significantly influenced by pH or hardness of the solution to be treated, as is predicted by factorial experiments. On the other hand, initial H_2O_2 concentration, initial phenol concentration and temperature significantly influenced the efficiency of the process. Optimal values were determined: a temperature of about 20°C and a C_{H2O2}/C_{phenol} ratio of 120 (mg/mg).

Introduction

Over the last decade several studies have been conducted on the performance of hydrogen peroxide (H_2O_2) and ultraviolet (UV) light in an advanced oxidation process (AOP) which has been successfully utilised for the treatment of various refractory (Doré, 1989; Sundstrom et al., 1990; Legrini et al., 1993; Sapach and Viraghan, 1998; Ho and Bolton, 1998; Von Gunten and Oliveras (1997); Shiraishi et al., 1999; Wenzel et al., 1999; Huston and Pignatello, 1999; Ince, 1999; Wang and Hong, 1999; Andreozzi et al., 2000a; Chiron et al., 2000). These previous studies have given the effect of initial H_2O_2 concentration, initial concentration of the recalcitrant pollutants, and of time and UV light on the performance of the AOP process. Alternatively, various kinetic models for the AOP process using hydrogen peroxide and ultraviolet irradiation have been developed (Turchi and Ollis, 1990; Beltran et al., 1999; Crittenden et al., 1999; Andreozzi et al., 2000b).

The main purpose of this work was to utilise a factorial type experimental design, to determine the effect of each experimental parameter on the performance of the AOP process using H_2O_2 and UV and to establish a mathematical model with a minimum of experiments. This mathematical model may be utilised to explain the phenomenon or to predict the performance of the process without performing the experiment. This last utilisation may be considered to be a simulation (Duea and Girault, 1978). For this purpose, the Hadamard matrix was used. It is an experimental design most frequently used for determining the effect of parameters in an experimental field resulting in a classification with respect to the statistical significance of their influence on the response of the system. This matrix makes it possible to determine the effect of K factors with $K \leq N-1$, where N is the number of the experiments (Perrin and Scharff, 1995).

A synthetic phenol solution was utilised as a standard solution to test the ability of the factorial type experiments to evaluate the degradation of phenol by the UV/H_2O_2 process. For this purpose a relation between the response parameter function Y representing the remaining phenol concentration, and the operational variables X_1, X_2, X_3, X_4, X_5 and X_6 which represent the peroxide concentration, initial phenol concentration, irradiation time, temperature, pH and hardness of the solution to be treated, was determined.

The first step of the method consisted of the elaboration of the Hadamard matrix, which makes it possible to determine the influent parameter on the response Y. In the second step, a complete Hadamard matrix was applied which provides information about the model and possibilities for its improvement (Goupy, 1988).

The validity of the mathematical model represented by the linear regression was assessed by addressing the following issues: the meaning of the coefficients, analysis of the coefficient of determination (r^2), analysis of the residuals, the regression and the confidence interval (Lagrade, 1983). The results obtained with the factorial experiments were then confirmed by an experimental study. Optimum conditions were attained.

Experimental

The experiments were carried out in a cylindrical double-walled reactor with water circulating through the walls to maintain the temperature at the chosen value. The photocatalytic reactor was equipped with a mercury lamp (Kadatyn®, France) with an output of 14 W (Fig. 1), which mainly emits irradiation at 253.7 nm and can be considered to be monochromatic. The solution was stirred by a magnetic stirrer and by a centrifugal pump during the run. The characteristics of the reactor are presented in Table 1.

TABLE 1
Characteristics of the photocatalytic reactor

Parameter	Value
Volume (mℓ)	150
Flow rate (mℓ/s)	14
Reactor space time (s)	10.71

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Received 4 July 2000; accepted in revised form 7 May 2001.