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Simultaneous degradation of phenol and paracetamol during photo-Fenton process: Design and optimization

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ABSTRACT

In the present study, the photo-Fenton process was applied for the treatment of phenol and paracetamol in a binary system. The cobalt ferrite nanoparticles synthesized by microwave heating method were applied during photo-Fenton process. The nanoparticles were characterized by XRD and SEM analysis. The response surface methodology based on Box–Behnken design was used to evaluate and optimize the interactive effects of five operating variables including pH, initial concentrations of phenol and paracetamol, H_2O_2 concentration and cobalt nanoferrite dosage on the phenol and paracetamol degradation rates in photo-Fenton process. The results of experimental design indicated that the pH of 3.5, H_2O_2 concentration of 50 mmol/L and cobalt nanoferrite dosage of 0.2 g/L maximize the efficiency of phenol and paracetamol degradation rates at the time in photo-Fenton process. The maximum experimental phenol and paracetamol degradation efficiencies for initial concentrations of 20 mg/L of phenol and paracetamol were found to be 95 and 85%, respectively. These values were in good agreement with the estimated values by the model in optimum conditions (94.5 and 84.4% for phenol and paracetamol degradation efficiencies). The obtained results demonstrated an excellent ability of synthesized cobalt ferrite nanoparticles to remove the phenolic compounds in photo-Fenton process.
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1. Introduction

Phenol is an important industrial raw material frequently used in the manufacture of pharmaceutical compounds such as diclofenac, amoxicillin and paracetamol [1]. Therefore, the presence of phenol in pharmaceutical wastes is inevitable. These compounds, when released in the environment over industrial wastewaters, provided high risk for human and animal health in the long term [2]. Therefore, the removal of pharmaceutical compounds from wastewaters becomes environmentally important. Several methods such as advanced oxidation processes (AOPs) [3], membrane filtration [4], biological treatment [5], photocatalytic degradation [6] and adsorption [7] have been used for the treatment of phenolic compounds from pharmaceutical wastewaters. Among mentioned techniques, AOPs due to its higher efficiency and short reaction time could be considered as an alternative method in industrial applications [8–11]. Among AOPs, photo-Fenton process due to its higher efficiency was found to be an efficient process compared to other AOP techniques [12–15]. Furthermore, the degradation rate of organic compounds is accelerated by presence of spinel-structured ferrite nanoparticles in photo-Fenton process [16–18]. For this, we synthesized the cobalt ferrite nanoparticles by microwave heating method and the potential of synthesized nanoparticles was investigated for degradation of phenol and paracetamol during photo-Fenton process.

Different parameters such as pH, catalyst dosage, initial concentration of phenolic compounds, reaction time, initial concentration of H_2O_2 and temperature could be affected on the phenolic compounds degradation in photo-Fenton process [9]. Investigation

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of each factor separately on the phenolic compounds removing would be very time consuming. Furthermore, the development of mathematical model predicting the degradation of phenolic compounds is one of the most important issues to be solved prior to the large scale for treatment of pharmaceutical compounds from wastewaters. Recently, response surface methodology (RSM) has been used to optimize the performance of complex systems. By RSM, it is possible to investigate the interactions of influencing variables on the treatment efficiency [19–21]. In recent researches, the factor space central-composite design (CCD) and Box–Behnken design (BBD) are commonly selected as experimental design techniques [20,21]. However, for a quadratic response surface model with three or more factors, the BBD technique is much more advantageous compared with CCD method [19,21]. In the present study, BBD was used to evaluate and optimize the simultaneous degradation rates of phenol and paracetamol during photo-Fenton process in a binary system. The effects of operational parameters of photo-Fenton process including initial pH (3–4), H_2O_2 concentration (30–70 mmol/L), phenol and paracetamol initial concentrations (20–100 mg/L), and cobalt ferrite dosage (0.1–0.3 g/L) for degradation of phenol and paracetamol were evaluated and verified by the means of five-factor at three-level of BBD.

2. Experimental

2.1. Materials

$FeSO_4{\cdot}7H_2O$, $CoCl_2{\cdot}6H_2O$, fumed silica (7 nm) and $NaAlO_2$ were purchased from Sigma-Aldrich (Sigma Aldrich, USA). NaOH and ethanol were obtained from Merck (Merck, Darmstadt, Germany). Phenol from (analytical grade; Fluka) was used. Paracetamol ($C_8H_9NO_2$) was provided from *Jalinous* pharmaceutical company of Iran.

The microwave equipment used in this study was a commercial microwave oven (CE1110C, Sumsung, Korea) with 900 W output power at wavelength of 2.45 GHz. The oven was equipped with an electronic system in order to control the temperature, accurately.

2.2. Synthesis of cobalt ferrite nanoparticles

The cobalt ferrite nanoparticles were synthesized using microwave heating method. For synthesis of nanoparticles, firstly, 0.56 g of $FeSO_4{\cdot}7H_2O$ and 0.24 g of $FeCl_3{\cdot}6H_2O$ were dissolved in 20 mL of de-ionized water by intensive stirring to obtain the homogeneous solution. Then, NaOH was added to the solution and the stirring was continued at room temperature for 1 h. Then, the homogenous solution was applied into the microwave at temperature of 160 °C for 10 min. Then, the solid products were collected by magnetic filtration and were washed by de-ionized water and ethanol. Finally, the samples were dried in a vacuum oven at 100 °C for 6 h.

2.3. Photo-Fenton process

The performance of the synthesized catalyst was evaluated in the photo-Fenton process of phenol and paracetamol degradation. The experiments were carried out under 4 UV lamps (15 W, λ_{max} = 365 nm) in a 500-mL Pyrex-glass cell wrapped in aluminum foil.

2.4. Analytical measurements and methodology

The powder's X-ray diffraction (XRD) patterns were recorded at 25 °C on a Philips instrument (X'pert diffractometer using Cu-$K\alpha$ radiation) with a scanning speed of 0.03° (2θ) min^{-1} to confirm the cobalt ferrite nanoparticles structure. The morphology and particle size of nanoparticles were characterized using a scanning electron microscopy (SEM, TESCAN, VEGA 3SB).

The concentrations of phenol and paracetamol were determined using a UV–vis spectrophotometer (JAS.CO V-530, Japan) at wave lengths of 271 and 248 nm for phenol and paracetamol, respectively.

The degradation rates of phenol and paracetamol were calculated as follows:

$$D_e = \frac{(C_0 - C_t)}{C_0} \times 100\% \quad (1)$$

where D_e is the degradation rate of phenol and paracetamol after t min of reaction, C_t is the concentration of compound after t min of reaction, and C_0 is the initial concentration of compound.

2.5. Design of photo-Fenton experiments

Forty one experiments based on Box–Behnken design (BBD) for five factors at three levels were used to determine the effect of key parameters including pH (3–4), H_2O_2 concentration (30–70 mmol/L) phenol and paracetamol initial concentrations (20–100 mg/L), and cobalt nanoferrite dosage (0.1–0.3 g/L) on the removal of phenol and paracetamol in a binary system. Contact time of 1 h and temperature of 45 °C were considered constant in the experiments. The statistical calculation levels associated with each variable were summarized in Table 1. The polynomial models for the phenol and paracetamol removal percentage with respect to the photo-Fenton variables were expressed as follows:

$$Y = \beta_0 + \sum_{i=1}^{5}\beta_i x_i + \sum_{i=1}^{5}\beta_{ii}x_i^2 + \sum_{i=1}^{5}\sum_{j=1}^{5}\beta_{ij}x_i x_j \quad (2)$$

where Y is the predict response by the model and β_0, β_i, β_{ii}, β_{ij} are the constant regression coefficients of the model. X_i, X_{ii} and X_{ij} represent the linear, quadratic and interactive terms of the uncoded independent variables, respectively. The coefficient of determination (R^2) was used to evaluate the accuracy of the full quadratic equation. The experimental design and results of phenol and paracetamol removal percentages during photo-Fenton process are presented in Table 2.

3. Results and discussion

3.1. Characterization of cobalt ferrite nanoparticles

The X-diffraction pattern and SEM image of the synthesized cobalt ferrite nanoparticles are shown in Fig. 1. As shown in Fig. 1a, the diffraction of prepared nanoparticles indicated the expected peaks for the pure inverse spinel structure of $CoFe_2O_4$ (JCPDS 221086) [22]. The crystallite sizes (D) of nanoparticles are calculated from XRD peak broadening of the (3 1 1) peak using Scherrer formula: $D = 0.9\lambda/\beta\cos\theta$, where λ the wavelength of Cu

Table 1
Experimental ranges and levels of independent variables used in photo-Fenton process.

Variable	Code	Range of levels		
		−1	0	1
pH	X_1	3.0	3.5	4.0
Phenol concentration (mg/L)	X_2	20	60	100
Paracetamol concentration (mg/L)	X_3	20	60	100
H_2O_2 concentration (mmol/L)	X_4	30	50	70
Cobalt ferrite dosage (g/L)	X_5	0.1	0.2	0.3

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