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Optimization of the microwave roasting extraction of palladium and rhodium from spent automobile catalysts using response surface analysis

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ABSTRACT

The optimum conditions for the extraction of palladium and rhodium from spent automobile catalysts by microwave roasting were determined using response surface methodology (RSM). A central composite design (CCD) was used to investigate the effects of three independent variables (namely proportion (m/m, reagent/material), roasting temperature (°C) and holding time (min)) on the leaching rate of palladium and rhodium. The independent variables were coded at three levels and their actual values based on experimental results. The CCD consisted of 20 experimental points and six replications at the center point. Data were analyzed using design expert and statistical analysis system software. A second-order polynomial model was used for predicting the response. The model explained 53% more variation. Canonical analysis of surface responses shows that the stationary surface was a saddle. The optimal conditions for the leaching rate of palladium and rhodium obtained using ridge analysis were 9.75, 550 °C, 60 min; 9.00, 540 °C, 50 min; and 9.96, 550 °C, 53.84 min, respectively. The predicted leaching rate of palladium is more than 99.29% and the predicted leaching rate of rhodium is more than 95.34% under three optimum conditions. In order to obtain the highest leaching rate, the conditions for the extraction of palladium and rhodium were tuned to 10, 550 °C, 60 min. The actual leaching rates of palladium and rhodium were 99.33% and 95.48% at the tuned conditions, which are very similar to those predicted by RSM models.

1. Introduction

Palladium and rhodium were widely applied in the automobile catalytic converter to convert the emission of gaseous pollutants such as carbon monoxide, hydrocarbons and nitrogen to carbon dioxide and moisture (Kim and Kim, 2009; Parthasarathi and Hegde, 2010). Palladium and rhodium are currently the essential elements as an automobile catalyst.

Natural resources of palladium and rhodium are limited and their demand is increasing because of their extensive uses in automobile catalyst. So, it is necessary to recycle them from the spent automobile catalytic converter (Faisal et al., 2008; Christian, 2012). The recovery methods included a range of hydrometallurgical or pyrometallurgical processes. The potential of pyrometallurgical processes is high recovery rates as well as coping with the impurities found in catalysts (Kayanuma et al., 2004a, 2004b; Yoo, 1998; Benson et al., 2000a, 2000b). The pyrometallurgical processes were to fuse palladium and rhodium from spent automobile catalytic converters with a collector metal such as copper or active metal with high temperature (Benson et al., 2000a, 2000b; Angelidis, 2001). During hydrometallurgical processes, the dissolution of palladium and rhodium is an important procedure, because palladium and rhodium are chemically stable and hardly dissolve in most acids (Kayanuma et al., 2004a, 2004b). The recovery of palladium and rhodium requires a large amount of energy and acids, and generates much wastewater. It is necessary to develop an efficient process for palladium and rhodium recovery.

Microwave heating technique as a novel application was used in many fields. Microwave-assisted leaching has been used to improve the extraction yield of metal and to reduce process time, especially with the environmental-friendly processes (Jafarifar et al., 2005). Microwave heating technologies have many advantages including selective and volumetric heating, low processing time, and controllable heating process. Microwave energy was widely applied to mineral extraction (Haque, 1999; Kingman and AL-Harahsheh, 2004).

Response surface methodology (RSM), described originally by Box and Wilson (1951), enables evaluation of the effects of several process variables and their interactions on response variables. Thus, RSM is a collection of statistical and mathematical techniques that has been successfully used for developing, improving and optimizing processes (Somasundaram et al., 2014; Chandrika and Fereidoon, 2005).

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The report was to enhance the leaching rate of palladium and rhodium by microwave roasting and to optimize the main influencing factors (roasting temperature, holding time, the proportion of reagent and material) using central composite design (CCD) and response surface methodology (RSM) analysis.

2. Materials and methods

2.1. Materials

The raw materials used in this work were the spent binary catalyst of palladium and rhodium. The spent catalyst was cracked by the bill mill and the main particle size is 2.12 um. The main chemical composition is cordierite, palladium and rhodium located at the surface of the cordierite substrate.

2.2. Reagent

The leaching reagents consisted of sodium chlorate ($NaClO_3$) and sodium bisulfate monohydrate ($NaHSO_4 \cdot H_2O$). They were purchased from Guangdong Project Technology Research Exploitation Center, Guangdong, P.R. China. The ratio of sodium chlorate to sodium bisulfate monohydrate is 1:1.

2.3. Experimental apparatus

The microwave roasting equipment used in the study is an industry microwave with automatic control devices. The microwave power is 1.5 kW, the microwave frequency is 2450 MHz, and the top temperature is 1200 °C.

2.4. Procedure

The catalytic converter was crushed and milled using a ball mill. A main particle size of 2.12 μm was achieved. The chemical composition of the catalytic converter samples was confirmed by X-ray fluorescence (XRF) using a Rigaku ZSX100e diffractometer. The amount of sample used for every experiment was 20 g. After the spent catalysis sample was uniformly blended with reclaiming reagent according to the ratio, the mixture was put into the ceramic crucible. Then, the ceramic crucible was coated by thermal insulation materials and put into a microwave reactor. The microwave power is 300 W–330 W. After reaction, the sinter was cooled, dispersed into water, and separated. The concentration of palladium and rhodium in the spent automobile catalysts was confirmed by fire assaying. The concentration of metals in the leaching solution was measured with an ultraviolet spectrophotometer using a SHIMADZU UV2550.

2.5. Selection of appropriate roasting conditions

The appropriate roasting conditions, including roasting temperature, holding time, the proportion of reagent and material, for spent automobile catalyst sample were investigated. Firstly, the leaching rate was tested when the holding time varied from 30 to 150 min, the roasting temperature is 300 °C and the proportion of reagent and material is 8 (m/m, reagent/material). The second step was to determine the roasting temperature**.** Palladium and rhodium were extracted using the best holding time based on the first step. The roasting temperature varied from 300 to 700 °C while holding the proportion of reagent and material constant at 8. The final step was to select the appropriate proportion between reagent and material. Using the chosen best roasting temperature and holding time, the proportion of reagent and material varied from 8 to 14. Based on the results, the three levels (lower, middle, upper) of each process variables were determined for RSM.

Table 1
Chemical composition of spent automobile catalysts.

Composition	Al	O	Si	Al	Mg	Ce	Zr	C	Pd	Rh
Content(%)	30.97	28.27	16.66	4.70	7.68	3.82	1.20	1.43	1698[a]	242[a]

[a] $g \cdot t^{-1}$.

2.6. Experimental design

Optimization for the microwave roasting extraction technique of palladium and rhodium from spent automobile catalysts was carried out using RSM (Somasundaram et al., 2014; Chandrika and Fereidoon, 2005; Narges et al., 2014). A three-factor and a three level face-centered cube design consisting of twenty experimental runs were employed including six replicated at the center point. The design variables were the proportion of reagent and material (X_1, m/m, reagent/material), roasting temperature(X_2, °C)and holding time (X_3, min) and the response variables were the leaching rate of palladium and rhodium. In the design, the effect of unexplained variability comes from extraneous factors which were minimized by randomizing the order of experiments.

2.7. Data analysis

The response surface regression (RSREG) procedure of statistical analysis system (SAS) and design expert (version 8.0.5) software were used to analyze the experimental data (Somasundaram et al., 2014). Experimental data were fitted to a quadratic model. The generalized second-order polynomial model used in the response surface analysis was as follows:

$$Y = \beta_0 + \sum_{i=1}^{3} \beta_i X_i + \sum_{i=1}^{3} \beta_{ii} X_i^2 + \sum \sum_{i<j=1}^{3} \beta_{ii} X_i X_j,$$

whereβ_0, β_i, β_j and β_{ij} are the regression coefficients for intercept, linear, quadratic and interaction terms, correspondingly, and X_i and X_j are the independent variables.

2.8. Verification of model

Optimal conditions for the microwave roasting process of spent automobile catalysts depended on the proportion of reagent and material, the roasting temperature and the holding time which were obtained using the predictive equations of RSM. The leaching rate of palladium and rhodium was determined under optimal conditions. The predicted and experimental values were compared in order to determine the validity of the model.

3. Results and discussion

3.1. Characterization of spent automobile catalysts

The chemical composition of the catalytic converter samples was confirmed by X-ray fluorescence (XRF). The chemical composition of the spent catalytic converter was showed in Table 1.

Table 2
Independent variables and their coded and actual values used for optimization.

Independent variables	Units	Symbol	Coded levels		
			−1	0	1
Proportion of reagent and material	(m/m)	X_1	9	10	11
Roasting temperature	°C	X_2	450	500	550
Holding time	min	X_3	40	60	80