

Review

Microwave-enhanced catalytic degradation of phenol over nickel oxide

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Abstract

A mix-valenced nickel oxide, NiO_x, was prepared from nickel nitrate aqueous solution through a precipitation with sodium hydroxide and an oxidation by sodium hypochlorite. Further, pure nickel oxide was obtained from the NiO_x by calcination at 300, 400 and 500 °C (labeled as C300, C400 and C500, respectively). They were characterized by thermogravimetry (TG), X-ray diffraction (XRD), nitrogen adsorption at –196 °C and temperature-programmed reduction (TPR). Their catalytic activities towards the degradation of phenol were further studied under continuous bubbling of air through the liquid phase. Also, the effects of pH, temperature and kinds of nickel oxide on the efficiency of the microwave-enhanced catalytic degradation (MECD) of phenol have been investigated. The results indicated that the relative activity affected significantly with the oxidation state of nickel, surface area and surface acidity of nickel oxide, i.e., NiO_x (>+2 and $S_{\text{BET}} = 201 \text{ m}^2 \text{ g}^{-1}$) \gg C300 (+2 and $S_{\text{BET}} = 104 \text{ m}^2 \text{ g}^{-1}$) > C400 (+2 and $S_{\text{BET}} = 52 \text{ m}^2 \text{ g}^{-1}$) > C500 (+2 and $S_{\text{BET}} = 27 \text{ m}^2 \text{ g}^{-1}$). The introduction of microwave irradiation could greatly shorten the time of phenol degradation.

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1. Introduction

The content of the organic pollution is serious in industrial wastewater. Aromatic pollutants, in particular phenol and

phenolic derivatives have been considered on the EPA's priority pollutants list since 1976 [1]. These compounds are considered as hazardous pollutants because of their potential to harm human health. Indeed, it is necessary to eliminate them from industrial wastewater before it is discharged. A number of methods such as sonochemical degradation [2–4], photocatalytic degradation [5–9], advanced oxidation process with UV/H₂O₂ (AOP) [10], catalytic oxidation [11] and the microwave-enhanced advanced

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oxidation processes [12–15] have been used for removal of phenol and phenolic derivatives. One of the most promising technologies is to utilize microwave-enhanced advanced oxidation procedure for the degradation of 4-chlorophenol. Microwave irradiation technology has already been applied to industry, family, medical science and environmental organic pollution for polycyclic aromatic hydrocarbons (PAHs) [16–18] and polychlorinated biphenyls (PCBs) [19], etc.

Due to the properties of internal and volumetric heating (dipole rotation and/or ionic conduction), thermal gradients during microwave processing are avoided, providing a uniform environment for reaction. Therefore, microwave heating has shown advantages over the conventional heating method in terms of energy efficiency, higher reaction rates and shorten in reaction times [20]. The use of microwaves as a source of energy is rapidly growing economized and getting convenient advantage.

Using solid materials as catalysts is of great interest and important in purifying waste waters containing phenol. Transition metal oxides have proved to be active in the catalytic reactions of the degradation of phenol and its derivatives [12,21]. Among them, the catalysts based on the oxides of nickel and cobalt appears to be especially efficient. Various factors can influence the activity, such as the oxidation state of metal ions and their bond strength of metal–oxygen, the content of active oxygen and the morphology of the material. From this point of view, adopting nickel oxide as active components in this article and combining the microwave irradiation technology to promote the degradation of phenol might be the possible path for practical application.

2. Experimental

2.1. Preparation of nickel oxide

The as-prepared nickel oxide (marked as NiO_x) with high valence state of nickel was synthesized by the precipitation–oxidation method in an aqueous solution. The process was carried out at 70 °C with dropwisely added 50 mL of 0.6 M $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ solution to the 100 mL of 3.2 M NaOH solution to obtain as-prepared $\text{Ni}(\text{OH})_2$. Further, a 100 mL of NaOCl (12 wt.%) was introduced drop by drop under a constant stirring. The precipitate was then filtered, washed with deionized distilled water and dried in an oven at 110 °C for 20 h. The dried product was ground and put in a desiccator as fresh sample (as-prepared NiO_x). The fresh sample was calcined separately at 300, 400 and 500 °C for 3 h to obtain the pure nickel oxide (labeled as C300, C400 and C500, respectively).

2.2. Characterization of nickel oxide

X-ray diffraction (XRD) measurements were performed using a MAC Science MXP18 diffractometer with $\text{Cu K}\alpha_1$ radiation ($\lambda = 1.5405 \text{ \AA}$) at 40 kV and 30 mA with a scanning speed in 2θ of 4° min^{-1} . The crystallite sizes of nickel oxide were estimated using the Scherrer equation.

Thermal gravimetric analysis (TG/DTG) was carried out using a Seiko SSC5000 TG system. The rate of heating was

maintained at $10^\circ \text{C min}^{-1}$ and the mass of the sample was $\sim 10 \text{ mg}$. The measurement was carried from RT to 800 °C under air flowing with a rate of 100 mL min^{-1} . The temperature-programmed desorption of pyridine (py-TPD) experiments was also detected by TG method that compared the surface acidity of nickel oxides.

Nitrogen adsorption isotherms at -196°C were determined volumetrically with Micromeritics ASAP 2010. The nickel oxides were pre-outgassed at 5×10^{-5} Torr for 3 h at 110 °C. The surface area was determined from the nitrogen adsorption isotherm.

Reduction behavior of nickel oxide was studied by temperature-programmed reduction (TPR). About 50 mg of the sample was heated in a flow of 10% H_2/N_2 gas mixture at a flow rate of 10 mL min^{-1} . During TPR, the temperature was increased by $7^\circ \text{C min}^{-1}$ increment from room temperature to 600 °C.

The infrared spectra were obtained by a Nicolet 5700 FT-IR spectrometer in the range of $400\text{--}2000 \text{ cm}^{-1}$. One milligram of each powder sample was diluted with 200 mg of vacuum-dried IR-grade KBr and subjected to a pressure of 8 tonnes.

The chromatographic experiment were performed using high performance liquid chromatograph Agilent 1100 Series equipped with diode array detector and a column oven. A $125 \text{ mm} \times 4 \text{ mm}$ reverse-phase C-18 column (chrompack) was used for separation. The injection volume was 20 μL , flow rate was 1.0 mL/min, UV detector wavelength was 270 nm and column oven temperature maintained 25 °C. The compounds were eluted with CAN/ H_2O (v/v, 50/50). Qualitative of the degradation intermediates was carried out on an Agilent 1100 Series LC/MS liquid chromatograph–mass spectrometer (LC/MS) system.

2.3. Degradation of phenol

The microwave-enhance catalytic degradation (MECD) experiments were carried out in a controllable temperature units microwave apparatus (CEM Discover, USA) that can program the reaction temperature and time. The system operated at 2450 MHz and worked at 100 W. The temperature can be controlled at the variation within $\pm 2^\circ \text{C}$ from the set temperature by an IR sensor. The reaction vessel was conical flask that connected with a simple refluxing device. A 20 mL of aqueous phenol solution (initial concentration of 200 ppm) was used for each experimental run. Air was bubbled in the solution for 30 min before adding the catalyst. Then, a 40 mg of catalyst was suspended in the solution. The air was continuously bubbled during the runs. The initial and products concentration of phenol in the solution was determined by UV–vis spectroscopy (Varian, Cary 5000). Absorbance at 268.4 nm was used to measure the concentration of phenol.

3. Results and discussion

3.1. Characterization of the prepared high valence nickel oxide— NiO_x

Fig. 1 shows the TG/DTG curves of the as-prepared NiO_x under a dynamic air (100 mL min^{-1}) environment. The TG

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