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Efficient removal of naphthalene-2-ol from aqueous solutions by solvent extraction

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ABSTRACT

Naphthalene-2-ol is a typical biologically recalcitrant pollutant in dye wastewater. Solvent extraction of naphthalene-2-ol from aqueous solutions using mixed solvents was investigated. Various extractants and diluents were evaluated, and the effects of volume ratio of extractant to diluent, initial pH, initial concentration of naphthalene-2-ol in aqueous solution, extraction time, temperature, volume ratio of organic phase to aqueous phase (O/A), stirring rate and extraction stages, on extraction efficiency were examined separately. Regeneration and reuse of the spent extractant were also investigated. Results showed that tributyl phosphate (TBP) achieved 98% extraction efficiency for naphthalene-2-ol in a single stage extraction, the highest among the 12 extractants evaluated. Extraction efficiency was optimized when cyclohexane and *n*-octane were used as diluents. The solvent combination of 20% TBP, 20% *n*-octanol and 60% cyclohexane (V/V) obtained the maximum extraction efficiency for naphthalene-2-ol, 99.3%, within 20 min using three cross-current extraction stages under the following extraction conditions: O/A ratio of 1:1, initial pH of 3, 25°C and stirring rate of 150 r/min. Recovery of mixed solvents was achieved by using 15% (W/W) NaOH solution at an O:A ratio of 1:1 and a contact time of 15 min. The mixed solvents achieved an extraction capacity for naphthalene-2-ol stably higher than 90% during five cycles after regeneration.

Introduction

Phenolic compounds as pollutants have received close attention due to their toxicity and possible bioaccumulation in the environment (Ku and Lee, 2000). Phenolic compounds are a group of chemicals containing one or more aromatic rings bearing one or more hydroxyl groups (Dai and Mumper, 2010), and common phenolic compounds include phenol, cresols, naphthol, dihydric phenols, *etc.* (Dai et al., 2008).

Phenolic compounds are typical organic components in industrial wastewaters from coal gasification plants, coking plants, oil refineries, and printing and dyeing mills (Wang et al., 2012). Wastewaters containing phenolic compounds can be a threat to public health and the environment when

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released without proper recovery or treatment, and can lead to harmful and lethal effects toward aquatic organisms even at relatively low concentrations of 5 to 25 mg/L (Broholm and Arvin, 2000) as well as objectionable taste in drinking water.

Thus, the discharge of phenolic compounds has been strictly regulated by many governments and organizations. The maximum emission concentration of phenolic compounds in water free from chlorine is 0.1 mg/L, while that in water containing chlorine is 0.001–0.002 mg/L (González-Muñoz et al., 2003). The European Union also lists several phenolic compounds as priority pollutants, and the 80/778/EC directive regulates that the total level of phenolic compounds in drinking water should be below 0.0005 mg/L (Jiang et al., 2003).

Naphthalene-2-ol is a dye intermediate, and is generally present in industrial wastewater. Naphthalene-2-ol, whose toxicity is similar to that of phenol, can strongly irritate human skin and mucus membranes. When entering the human body through percutaneous absorption, naphthalene-2-ol is toxic to blood circulation and the liver, and can also induce cancer and abnormalities (Dai et al., 2008). Unfortunately, few publications on the removal of naphthalene-2-ol from aqueous solutions are available. Therefore, investigations on the treatment of aqueous solutions containing naphthalene-2-ol are needed.

However, naphthalene-2-ol is a naphthalene homolog of phenol, and a number of treatment processes for the phenolic compound removal from aqueous solutions have been reported due to the toxicity and poor biodegradability of phenolic compounds (Bokhove et al., 2012). The treatment processes can be classified as destruction methods and recovery methods according to the final destination of the phenolic compounds. Among the destruction methods, there are biological treatments (Broholm and Arvin, 2000; Pinto et al., 2002), incineration, and wet air oxidation. The recovery methods include liquid–liquid extraction (Palma et al., 2010; Yang et al., 2006), adsorption (Li et al., 2003a; Wang et al., 2014; Zhao et al., 2014), solvent-impregnated resins (Burghoff et al., 2008), ionic exchange with resins, and membrane processes, such as extraction with membranes (Kujawski et al., 2004) or supported liquid membranes (Zidi et al., 2010, 2011).

Among these methods, solvent extraction has received close attention. It has been regarded as a promising, feasible and widely used technique for the concentration, separation and purification of various phenolic compounds from aqueous solutions in terms of its ease of use, high selectivity, high yield, high productivity, and wide-ranging applicability (Wang et al., 2012; Stalikas, 2007).

Many solvents such as organophosphorus solvents, hydrocarbon solvents, and aliphatic amines have been used for the extraction of phenolic compounds (Burghoff et al., 2009; Li et al., 2003b). Organic solvents used in the extraction process can be divided into three major types: conventional oxygen-bearing and hydrocarbon extractants, organophosphorous compounds, and high molecular weight aliphatic amines (Yang et al., 1991). Organophosphorous compounds and aliphatic amines are effective extractants, and provide higher distribution coefficients than those of conventional oxygen-bearing and hydrocarbon extractants (Dai et al., 2008). When other factors including costs and physicochemical properties of extractants are taken into consideration, organophosphorous compounds are usually preferred. In particular, the chemical stability of organophosphorous compounds plays an important role in process selection (Yang et al., 2006).

Therefore, it was reasonable to hypothesize that solvent extraction using organophosphorous compounds could probably be feasible for naphthalene-2-ol removal from aqueous solutions.

In this study, the extraction of naphthalene-2-ol by organophosphorous solvents was investigated. The main objectives of this work were to examine the feasibility of the removal of naphthalene-2-ol from aqueous solutions by solvent extraction and to evaluate the effects of extractant and diluent type, volume ratio of extractant to diluent, extraction time, initial concentration, temperature, pH, stirring rate, volume ratio of organic phase to aqueous phase (O/A), and extraction stages on the extraction efficiency and distribution coefficient. In addition, the feasibility of back extraction with alkali to regenerate the spent extractant was also assessed. These results could serve as a reference for further investigations and process designs.

1. Materials and methods

1.1. Chemicals and wastewater samples

All the chemicals were of analytical-reagent grade. Tributyl phosphate (TBP), a phosphorus-bonded oxygen donor, is a colorless liquid with the molar mass of 266.32 g/mol and density of 976.6 kg/m^3, and was produced by Shanghai Chemical Reagents Co. Ltd. (China) with purity of 98%. Naphthalene-2-ol was from Tianjin Kemiou Chemical Reagent Co. Ltd. (China) with purity of 98%. The main properties of naphthalene-2-ol, including some toxicity data, are shown in Table 1. Dimethyl carbonate (DMC) was purchased from Aladdin Industrial Corporation (China) with purity of 99%. Cyclohexane, *n*-hexane, kerosene, xylene, MIBK, toluene, *n*-butylacetate, benzene, 1,2-dichloroethane, and *n*-octanol were from Shanghai Chemical Reagents Co. Ltd. (China). All of the solvents were washed with distilled water to remove water-soluble impurities. The main properties of the solvents used in this study are shown in Table 2.

In a typical experimental procedure, naphthalene-2-ol was dissolved in ultra-pure water to prepare aqueous solution samples. The initial concentrations of naphthalene-2-ol solutions were all 100 mg/L except as noted. All the aqueous solution samples were prepared before the experiments.

Table 1 – The main properties of naphthalene-2-ol.

Property	Description
Stability	Stable (Combustible. Dust may form explosive mixture with air. Incompatible with strong oxidizing agents, phenol.)
Appearance	White or off-white powder with a slight
Toxicity	ORL-RAT LD$_{50}$ 1960 mg/kg, IPR-MUS LD$_{50}$ 98 mg/kg
Safety	Warning. Safety glasses, adequate ventilation.
Melting point	120–124°C
Boiling point	285.5 ± 0.0°C at 760 mmHg
Flash point	144.0 ± 10.6°C
Density	1.2 ± 0.1 g/cm^3
Water solubility	1512 mg/L