



The new liquid–liquid extraction method for separation of phenolic compounds from coal tar



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HIGHLIGHTS

- Imidazole and its homolog compounds were developed as new extraction agent that can form deep eutectic solvent with phenols.
- This new separation process avoids the use of strongly alkaline and acidic aqueous solutions.
- Hydrogen bond between phenols and extraction agent was confirmed by FT-IR and the experimental conditions were optimized.
- The extraction agent exhibited good recycling property.
- The simulation results of separation process were in a good accordance with experiment values.

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ABSTRACT

Phenolic compounds are mainly derived from coal tar, petroleum, and coal liquefaction oil. The traditional acid–base separation method can cause serious environmental problems. In this research, imidazole and its homolog compounds were designed as new extraction agents for the separation of phenols from coal tar, which can form deep eutectic solvent with phenols with the removal efficiencies more than 90%. The influence of the extraction agent structure was explored, and different substitutes were investigated. The key experimental conditions, such as reaction time, reaction temperature, and mole ratio of extraction agent to phenols were studied. The hydrogen bond between phenols and extraction agent is the reason for the formation of DES, and this was proved based on analyzing the chemical bonds of product by using FT-IR. The extraction agents exhibited good recycling property. Finally, the experimental process was simulated by Aspen Plus, and a specific separation process and technology was proposed.

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1. Introduction

Phenols are widely used to produce phenolic resins, synthetic fibers, and other products, majority of these compounds are mainly derived from coal tar and coal liquefied oil, as well as petroleum. In particular, low-temperature coal tar has higher phenol content than high-temperature coal tar, with a total content of approximately 20–30% [1]. In addition, the existence of phenols affects the storage stability and increases hydrogen consumption in the following oil refining process [2,3]. A new and efficient separation method should be explored to utilize these phenolic compounds.

The traditional method of separating phenols from oil mixture involves the use of strongly alkaline and acidic aqueous solutions through various chemical reactions [4,5]. This process convention-

ally produces a large quantity of phenol-containing waste water. Additionally, the strong alkalis and acids can cause an erosive action on the process equipment. Therefore, these outcomes require the development of a separation method that avoids the use of strongly alkalis and acids and instead uses environment-friendly methods.

A new type of green solvent called deep eutectic solvent (DES) has recently emerged, conventionally used in liquid form. This solvent is composed of two or three compounds with hydrogen bond interactions. The formed eutectic mixture has a lower melting point than each of the individual components, and exhibits physico-chemical properties similar to those of ionic liquids [6–9]. Abbott et al. first defined and formed DES by using chloride and urea with a mole ratio of 1:2 with quaternary ammonium salts and hydrogen bond donors [10–12]. Thereafter, various kinds of DES were synthesized, and their applications were explored.

The novel properties of DES make it a widely used material in synthesis, electrochemistry, nanomaterial, biochemistry, analysis,

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Notations

List of symbols

C_O	original oil concentration (g L^{-1})
V_O	original oil volume (mL)
C_f	oil concentration after reaction (g L^{-1})
V_f	oil volume after reaction (mL)
C_r^E	total phenols concentration in DES phase (mol L^{-1})
C_{IMZ}^E	IMZ concentration in DES phase (mol L^{-1})
Z	loading factor

Abbreviations

IMZ	imidazole
MIMZ	1-methylimidazole
EIMZ	1-ethylimidazole
PIMZ	1-propylimidazole

BIMZ	1-butylimidazole
FT-IR	Fourier transform infrared spectrometer
GC	gas chromatograph
DES	deep eutectic solvent

Subscripts

o	original data
f	data after reaction
E	DES phase

Superscript

E	DES phase
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and separation. DES can be successfully used as extraction agent in separation processes, such as in the separation of glycerol from biodiesel [13–16], alcohols from esters [17], saponins from jua and sisal [18] and so on [19]. As regards the separation of phenols, liquid–liquid extraction has become increasingly advantageous for its high efficiency and economy. The extraction solvents used in the separation of phenols has changed from organic solvents to green solvents, such as ionic liquids. Khalid [20] used five new solvents to extract phenol and *p*-chlorophenol from wastewater. Liu et al. [21] used cumene to extract phenol from wastewater. Denise et al. [22] used tetraethylene glycol as a high-boiling solvent for the separation of *m*-cresol from neutral oils. Though these solvents could efficiently separate phenols, green solvents needed to be developed because of the series of accompanying environmental problem. Fan et al. [23] used an imidazolium ionic liquid as the extraction agent to separate phenols from aqueous solutions and optimized the experimental parameters, most phenols were extracted quantitatively to the ionic liquid phase. Hou et al. [24] used an imidazolium-based ionic liquid to extract phenols from model oil, with a removal efficiency that exceeded 95%. Egorv et al. [25] used a novel ionic liquid incorporating quaternary ammonium cation to extract phenols and aromatic amines efficiently, and its solute distribution ratio can reach as high as $n \times 10^4$. DES has recently been found to be an efficient separation medium, which is cheaper than ionic liquids. Pang et al. [26] found that choline chloride can react with phenols to form DES, which become a uniform liquid phase to separate phenols from oil. Guo et al. [27,28] studied the phenol removal efficiency of choline chloride and its homologous compounds from model oils and then discussed how different cations and anions affect the removal efficiency of phenol. With the upsurge of green chemistry, the utilization of DES is predicted to draw increasing attention. Therefore, the considerable potential of DES for use in the separation process will be further studied.

From the experiment, imidazole and its homolog compounds were found to be able to form DES with phenols that are immiscible with oil in our research, thereby making phenols separable. Therefore, a series of DES of IMZ homolog compounds and phenols was designed and synthesized, and the separation process was discussed. Additionally, the influence rules of different experimental conditions and the structure of extraction agent on phenols removal efficiencies and Z were studied in this article. The basic reaction conditions were optimized, and the reaction mechanism of the separation process was explored. Finally, the new separation process was proposed.

2. Experiment

2.1. Chemicals

Phenol (>98.0% w), *p*-cresol (>99.0% w), *m*-cresol (>99.0% w), *o*-cresol (>99.0% w), and hexane (>95.0% w) are provided by Sinopharm Chemical Reagent Co., Ltd. Imidazole (IMZ) (>99.0% w), 1-methylimidazole (MIMZ) (>99.0% w), 1-ethylimidazole (EIMZ) (>99.0% w), 1-propylimidazole (PIMZ) (>99.0% w), and 1-butylimidazole (BIMZ) (>99.0% w) are provided by J&K Scientific Co., Ltd. In all cases, the percentage purities mentioned above refer to mass fraction as reported by the suppliers. All the chemical agents were used without further purification. The water was distilled in glass before use.

2.2. Reaction

Phenol, *p*-cresol, *m*-cresol, and *o*-cresol were dissolved in hexane in a beaker with continuous shaking to prepare the model oil, and the mixing ratio by mass was set at 2:1:1:1 in the bulk density as their content ratio in coal tar. In this work, hexane was selected as the solvent for the model oil. First, 80.10 g of phenol, 40.26 g of *o*-cresol, 40.28 g of *p*-cresol, and 40.03 g of *m*-cresol were added to 500 mL of hexane at 303.15 K in a beaker with continuous shaking. The solution was then transferred to a 1000 mL volumetric flask after the phenol and cresols completely dissolved. Hexane was added to the scale mark of the flask. The model oil was prepared as mentioned above. The *m*-cresol model oil with a concentration of 200.0 g/L was prepared in the same way.

The experiments were performed as follows (Fig. 1): a certain amount of model oil was placed in a conical flask, and then the extraction agent with a certain mole ratio to the phenols was added into the model oil. The flask was placed in an incubator shaker with a temperature controller within ± 0.1 K of an appropriate temperature and 400 rpm for a known time. After the reaction, DES was formed, and two layers appeared in the model oil. The system was allowed to stand for more than 30 min. Finally, the upper and lower layers were carefully separated by using a funnel.

2.3. Analysis methods

This article used a gas chromatograph with flame ionization detection to detect the component content of the oils after reaction. The main components of the upper layer were a small amount of phenol, *o*-cresol, *p*-cresol, and *m*-cresol with high hexane content. The chromatographic column used in GC was a

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