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Enhanced removal ability of phenol from aqueous solution using coal-based carbon membrane coupled with electrochemical oxidation process

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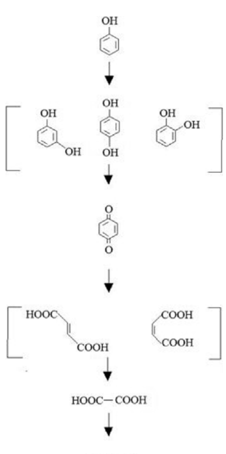
GRAPHICAL ABSTRACT

During treatment, phenol was firstly transformed into catechol, hydroquinone and resorcinol. And then, hydroquinone was degraded to benzoquinone, which was further decomposed to organic acids such as maleic, fumaric, and oxalic acids. Finally, these organic acids were oxidized into CO_2 and H_2O.



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ABSTRACT

A treatment system combining the coal-based carbon membrane with electrochemical oxidation process was designed for the enhanced phenol removal ability. The effects of various parameters including electric voltage, phenol concentration, solution pH, rotate speed, electrolyte type and concentration, and electrode distance on the permeate flux and removal efficiency of the treatment system were carried out. The degradation intermediates of phenol were detected by gas chromatography-mass spectrometry (GC–MS). The results showed the phenol removal ability of carbon membrane was significantly improved by coupling with electrochemical oxidation process. High phenol concentration usually increased the load of the treatment system, resulting in low permeate flux and removal efficiency because of limited decomposition ability. When the initial pH increased from 4.0 to 10.0, the enhanced permeate flux and removal efficiency were observed. The permeate flux displayed a decrease trend with the increase of electrode distance, while the removal efficiency showed an opposite trend. The optimum operation parameters of 2.0 V electric voltage, 7.5 r/min pump rotate speed, and 2.50 g/L Na_2SO_4 were recommended. During the degradation process, the reactive free radicals and H_2O_2 attacked the

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benzene ring of phenol to produce catechol, hydroquinone and resorcinol. And then, hydroquinone was degraded to benzoquinone, which was further decomposed to organic acids such as maleic, fumaric, and oxalic acids. Finally, these organic acids were oxidized into CO_2 and H_2O.

1. Introduction

Phenol, generated from many industrial processes such as steel, petrochemical, and pharmaceutical, is recognized as priority toxic organic pollutant by the US Environmental Protection Agency (USEPA) due to its high carcinogenic, toxic and mutagenic [1–3]. Up to now, numerous methods, such as adsorption, membrane separation, liquid-liquid extraction, advanced oxidation processes, biological treatment, etc., have been conducted to treat phenol wastewater [4–10]. Among them, membrane technologies are becoming increasingly important method for wastewater treatment because of many benefits including high efficiency, simple operation, compact design, and low energy consuming [11]. Recently, Ehtash et al. used a liquid membrane to treatment phenol solution [12]. Cui et al. explored forward osmosis (FO) for the removal of phenol from wastewater [6].

However, as for microfiltration or ultrafiltration membranes, they usually demonstrate poor separation performance on the removal of pollutants, such as phenol, whose sizes are smaller than membrane pores. Thus combined membrane separations with other processes have been implemented to tackle this challenging issue. Mukherjee and De fabricated mixed matrix hollow fiber membrane by doping activated carbon into polysulfone. They found the novel membrane could effectively treat small-sized pollutants (phenol, benzene, and toluene) by combining adsorption and filtration [13]. Dosta et al. reported their works on using membrane biological reactors (MBR) to efficiently remove phenol ($>$ 98.5%) in the industrial hypersaline wastewater [14]. Osegueda et al. developed the catalytic membrane reactors by depositing the palladium uniformly on the surface of the ceramic fibers, which demonstrated high phenol degradation ability [15]. Estrada-Arriaga et al. coupled an ultrafiltration hollow fiber membrane with photo-ferrioxalate and Fenton's reaction for the efficient treatment of phenol [16]. Wang et al. constructed the membrane electro-bioreactor (MEBR) by combining membrane filtration with electrokinetic phenomena and biological processes to treat phenol wastewater, indicating high removal efficiency than the sum of these individual processes [17]. Rueda-Márquez et al. designed the multi-barrier treatment (MBT) system by integrating micro-filtration, H_2O_2/UVC and CWPO for the treatment of organic pollutants [18]. Among them, coupling membrane technology with advanced oxidation processes (AOPs) is one of the most promising combination strategies for the removal of smaller sized organic molecules from wastewater because AOPs are eco-friendly chemical, photochemical or electrochemical processes that utilize highly reactive radicals as strong oxidants for the degradation of organic pollutants [19–22].

Recently, carbon membranes fabricated from various carbonaceous materials have received great attention in many industrial areas. In the past several years, we successfully fabricated the tubular microfiltration carbon membrane derived from coal, which not only demonstrated considerable separation performance, but also possessed good electrical conductivity [23,24]. Based on this idea, we constructed a wastewater treatment system combining the carbon membrane with electrochemical oxidation process. The effects of main operating conditions including electric voltage, phenol concentration, solution pH, rotate speed, electrolyte type and concentration, and electrode distanceon the separation performance of the treatment system were investigated in detail. Moreover, the possible degradation mechanism of phenol in this system was proposed.

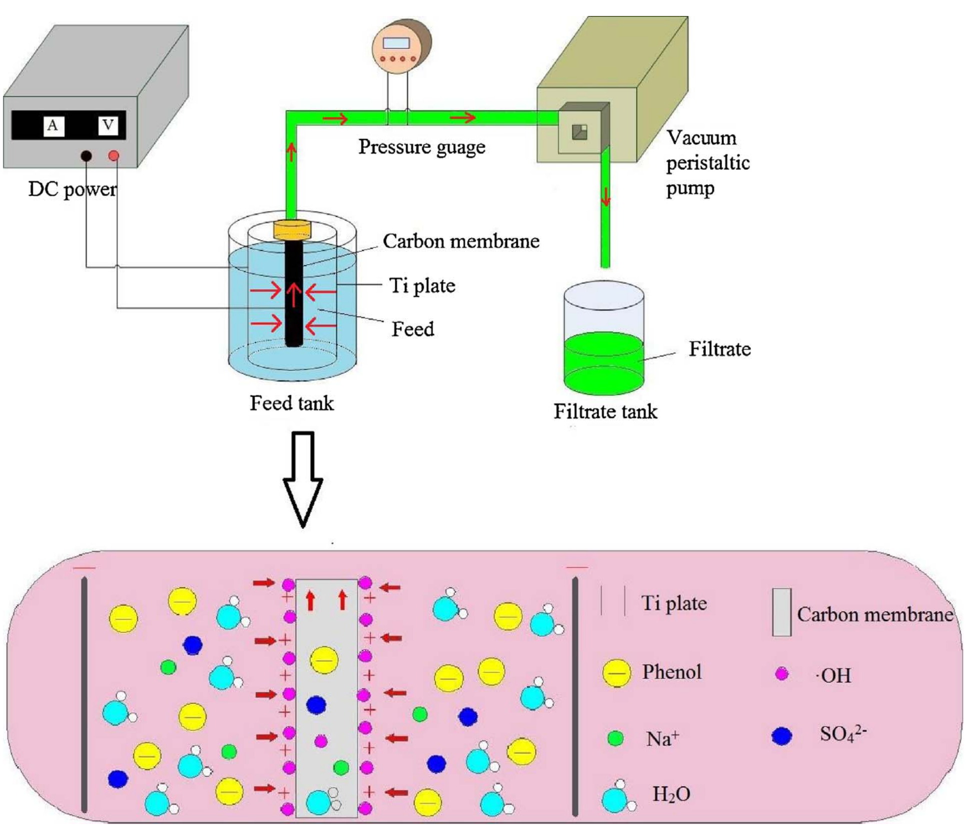

Fig. 1. Flow schematic diagram of the carbon membrane coupling with an electric field.