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Selective removal of 2,4-dichlorophenol in aqueous solutions by nanoparticles modified yeast using miniemulsion imprinting polymerization

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ABSTRACT

This research reports a surface molecularly imprinted polymers (SMIPs) which is synthesized by miniemulsion polymerization based on yeast as substrate material. Then, the obtained SMIPs were characterized by several techniques including Fourier transmission infrared spectrometry (FT-IR), scanning electron microscope (SEM), transmission electron microscope (TEM), thermogravimetric analysis (TGA) and elemental analysis. The results demonstrated that the obtained elliptical-shaped molecular imprinting polymer was coated with thickness imprinting layer. Then, SMIPs were used as sorbents to selective removal of 2,4-dichlorophenol (2,4-DCP) from aqueous solutions. Batch mode of binding experiments was carried out to determine the equilibrium isotherm, kinetics, regeneration and selectivity recognition of SMIPs. The results indicated that the selective adsorption behaviors of SMIPs were well described by the Langmuir isotherm model and the pseudo-second-order kinetics model. The SMIPs exhibited outstanding specific recognition capacity for 2,4-DCP (29.25 mg g^{-1} at 298 K). SMIPs also possessed excellent selective recognition for 2,4-DCP in the presence of other competitive compounds (such as 2,4,6-TCP, 3-CP and 2,6-DCP). Finally, the SMIPs were successfully adopted to the selective adsorption of 2,4-DCP from the environmental samples.

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Introduction

Chlorophenols (CPs) and its derivatives are widely employed in various kinds of industrial processes, such as herbicides, antiseptics, insecticides, wood preservatives and chlorination of drinking water [1]. Due to their widespread use, these organochlorine compounds are widely distributed in the surface and groundwaters. However, because of high toxicity, CPs has attracted much more attention in last decades. CPs will enriched in the human body which will induce inhibit central nervous, viscera function damage and even cause cancer. CPs have been listed as priority pollutants by both Environmental Protection Agency (EPA) and European Union Priority Substances (EUPS). Thus, it is of great significance in developing more efficient methods for the selective removal of CPs from aqueous solutions.

Molecular imprinted technique (MIT) is a promising and facile method to synthesize molecularly imprinted polymers (MIPs) with specific molecular recognition properties [2]. In order to fabricate the specific binding sites, target molecule and functional monomers are firstly incorporated to creating a three-dimensional polymeric matrix. Then the target molecule is removed from the polymeric matrix, which resulted in leaving specific binding sites in MIPs that are complementary in geometry, size and spatial distribution to the target molecule [3]. Owing to the favorable thermal, mechanical and chemical stability, preparation simplicity and low cost together with tailor-made molecular recognition binding sites [4], MIPs have been utilized in a wide range of fields, such as antibody mimics, drug development, solid phase extraction, separation, chemical sensing [5] and many other applications. Dai et al. [6] synthesized MIPs by atom transfer radical emulsion polymerization and selective removal tetracycline from aqueous

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solutions. Li et al. [7] prepared MIPs for 2,4-DCP separation based on reverse microemulsion polymerization. Although the above MIPs can selective removal of target molecular, they still have some inherent shortcomings, such as incomplete template removal, small binding capacity, poor accessibility of the binding sites and heterogeneous binding site distribution [8]. These limitations lead to the lower mass transfer rate and poor binding capacity towards the target molecule.

To solve these issues, surface molecularly imprinted polymers (SMIPs) have been developed in recent years. It takes certain measures to fabricate and situated specific binding sites on the surface of substrate materials. Thus, all the imprinted template molecules can be thoroughly removed from the cross-linked polymers and the binding sites obtained on the surface are all valid for the target [6]. Wei et al. [9] prepared SMIPs based Mn-doped ZnS quantum dots for separation of 2,4,5-trichlorophenol from wastewater. Pan et al. [10] used Fe_3O_4/halloysite nanotube magnetic composites as substrate materials to prepared SMIPs and the SMIPs were used as adsorbent to selective removal 2,4,5-trichlorophenol from aqueous solutions. However, the traditional substrate materials are of expensive and preparation complex. Yeast is well-known as an important and interesting group of microorganisms in biotechnology fields. Nowadays, yeast was also recognized as a kind of economic and feasible substrate materials. Compared with the other substrate materials, yeast not only has the advantage of low cost, but also the available source is easy. In addition, the active biomolecule on the yeast cell wall was abundant, so that yeast can be used directly without further modification process. Li et al. [11,12] prepared SMIPs used yeast as substrate material for selective adsorption cefalexin.

Commonly, SMIPs are prepared by bulk polymerization, emulsion polymerization, miniemulsion polymerization and suspension polymerization [13]. Since it first discovery in 1973 [14], miniemulsion polymerization has attracted much more attention because of its excellent performance in the synthesis of SMIPs. In miniemulsion system, the oil phase, which contains the monomers and the hydrophobe, is dispersed in the water phase, which consists of the surfactant. Miniemulsion are dispersed into a relative stable droplets under high shear device, which the size is in the range of 50–500 nm. When combining surface molecularly imprinted technique with miniemulsion polymerization, the resultant SMIPs could be the promising multifunctional adsorbent with excellent selective recognition ability. To our knowledge, there are few reports on selective recognition of CPs using yeast surface molecularly imprinted polymers by miniemulsion polymerization.

In this research, surface molecularly imprinting polymers were prepared based on yeast as substrate material via miniemulsion polymerization. Then, the SMIPs were used as adsorbents for selective removal 2,4-dichlorophenol (2,4-DCP) from aqueous solutions. The SMIPs were synthesized by using 2,4-DCP as template molecule, methacrylic acid and styrene as monomer, divinyl benzene as cross-linking agent, sodium dodecyl sulfate as emulsifier, potassium persulfate as initiator. The preparation of SMIPs via miniemulsion polymerization is illustrated in Fig. 1. The obtained SMIPs were characterized by FT-IR, SEM, TEM, TGA and elemental analysis. The adsorption properties, such as equilibrium isotherm, kinetics, regeneration and selectivity of SMIPs were investigated through the batch mode adsorption experiments. Furthermore, the SMIPs were also used as adsorbents for the extraction of 2,4-DCP from the vegetable samples.

Experimental

Materials

Styrene (St), ethanol, sodium dodecyl sulfate (SDS), potassium persulfate (KPS) and HPLC-grade methanol were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Divinyl benzene (DVB), methacrylic acid (MAA) was obtained from Aladdin reagent Co., Ltd. (Shanghai, China). 2,4-DCP, 2,4,6-trichlorophenol (2,4,6-TCP), 3-chlorophenols (3-CP) and 2,6-dichlorophenol (2,6-DCP) were purchased from Tianda Chemical Reagent Factory

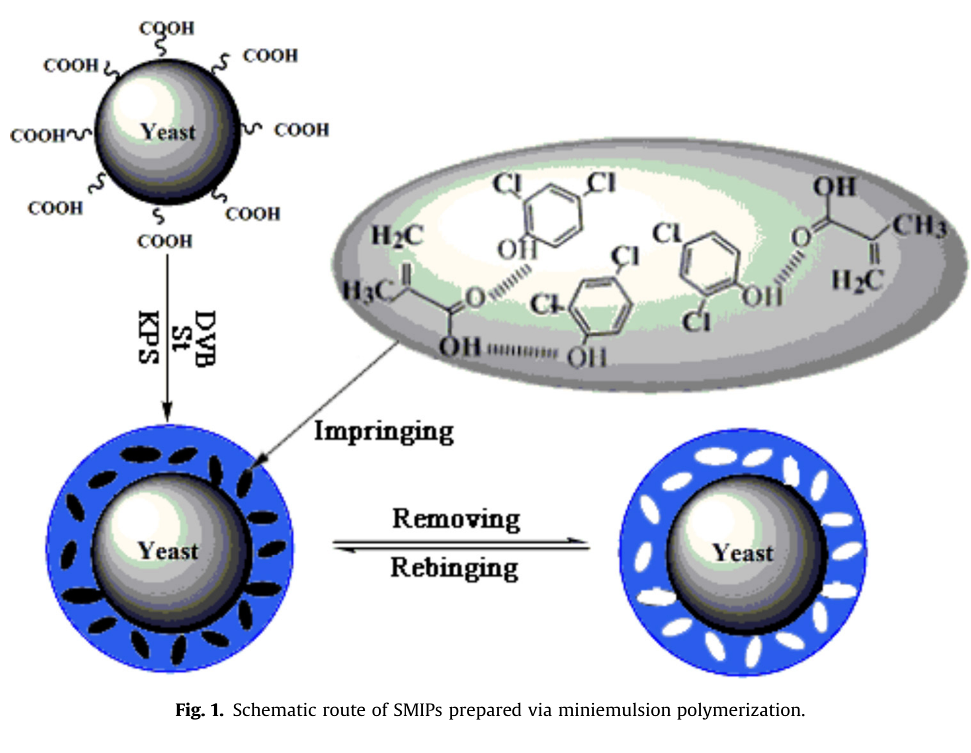

Fig. 1. Schematic route of SMIPs prepared via miniemulsion polymerization.

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