

Adsorption of rare earth ions using carbonized polydopamine nano carbon shells

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Abstract: Herein we reported the structure effects of carbon nano-shells prepared by the carbonization of polydopamine for the adsorption of rare earth elements (REEs) for the first time. Solid carbon spheres, 60 nm carbon shells and 500 nm carbon shells were prepared and evaluated for adsorption and desorption of REEs. The adsorption performance of carbon nano-shells for REEs was far superior to the solid carbon spheres. In addition, the effect of acidity on the adsorption and desorption properties was discussed. The good adsorption performance of the carbon nano-shells could be attributed to their pore structure, specific surface area, and the presence of both amine and carbonyl groups from the grafted dopamine.

Keywords: carbon shells; rare earths; adsorption; separation

Rare earth elements (REEs) represent an important material resource for many high-tech applications. Moreover, the separation of REEs from fission products is a challenging procedure in the nuclear fuel cycle process. Because of the importance of REEs, a number of technologies have been developed for preconcentration and separation, including solvent extraction^[1,2], ion exchange^[3,4], adsorption^[5,6], and liquid membranes^[7]. Of these separation methods, adsorption has become an effective strategy to improve separation efficiency for REEs. We have reported a variety of advanced adsorption materials such as sol-gel materials^[8,9], imprinted sorbents^[10-13], and ordered mesoporous materials^[14-17]. Because of the ability to achieve well-defined structures and composition, interest in adsorption materials for REE separation has recently expanded as the critical need for these materials has grown. For example, poly (acrylamide-expanded perlite) has been synthesized to absorb Tb³⁺ using free-radical polymerization; the composite was able to bind Tb³⁺ ions with strong chemical affinity^[18]. A novel hybrid gel was prepared by crosslinking calcium alginate (ALG) and γ -poly glutamic acid (PGA). Doping PGA molecules into the calcium alginate beads was used to enhance the adsorption capacity and selectivity of REEs from non-rare earths^[19]. Polymeric composite beads encapsulating di-2-ethyl hexyl phosphoric acid have been developed for REEs separation and the encapsulated polymeric composite beads are quite stable

and reusable even after 10 cycles of operation^[20]. Solvent-impregnated resin containing 2-ethylhexyl phosphonic acid mono-2-ethylhexyl ester, coated with polyvinyl alcohol and crosslinked by either glutaraldehyde or divinyl sulfone was investigated for the separation of lanthanum and cerium. Coating of the resin contributed to suppressing the leakage of extractant during the adsorption process^[21].

Porous carbon materials are ubiquitous and indispensable in many scientific applications, such as fuel cells, supercapacitors, separations, and catalysis^[22]. Many novel synthetic methodologies have been developed to create unique porous carbon materials^[23-25]. Some studies on the adsorption ability of carbon nanomaterials for organics and inorganics in aqueous phase have been reported, e.g., adsorption of polycyclic aromatic hydrocarbons onto single-walled and multiwalled carbon nanotubes were investigated^[26]. For different carbon nanomaterials, adsorption seems to be related to their surface area, micropore volume, and the volume ratios of mesopores to micropores. The adsorption inter-relationship between the surface oxidation of multi-walled carbon nanotubes (MWCNTs) and the polarity of organic solutes indicated that pristine MWCNTs exhibited a stronger affinity toward non-polar compounds compared to that of ozone-oxidized MWCNTs. In contrast, the adsorption of polar compounds on MWCNTs occurred to a much lesser extent compared to that on ozone-oxidized

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MWCNTs^[27]. The adsorption features of MWCNTs with the magnetic properties of iron oxides were combined in a composite to produce a magnetic adsorbent. The composite exhibited a good adsorption capability for Cr(III). In addition, the magnetic composite could be used as an adsorbent for contaminants in water and be controlled and removed from the medium by a simple magnetic process^[28]. Dopamine can self-polymerize at alkaline pH values and spontaneously deposit polydopamine conformal films on virtually any surface, the thickness of such a conformal coating can be precisely controlled. The single-step deposition technique has demonstrated direct benefits in the construction of polymeric nanocapsules^[29,30]. A versatile and facile method for synthesizing hollow carbon spheres using dopamine as the carbon source has been developed recently in our laboratory. Uniform carbon capsules were obtained easily by a simple immersion of the template in a dopamine aqueous solution followed by carbonization and template removal^[31]. The carbon shells prepared by this approach reveal satisfactory surface area and high structural stability. Moreover, the size of the carbon shells can be effectively adjusted by changing the size of the templating silica spheres. To our knowledge, no investigation concerning the application of carbon nano-shells prepared by dopamine on REE adsorption has been addressed in the literature. In this paper, we reported the adsorption/desorption properties of solid carbon spheres and carbonized polydopamine nano shells with varying diameters for REEs for the first time.

1 Experimental

1.1 Reagents and methods

All chemicals and solvents were of reagent grade and used without further purification unless noted otherwise. The solid carbon spheres were prepared according to a previously-reported method^[32], while the hollow carbon shells were prepared based on the work of Liu et al.^[31]. The specific surface area was measured using a TriStar surface area and porosity analyzer at 77 K and calculated

using the Brunauer-Emmett-Teller (BET) equation. Scanning transmission electron microscope (STEM) images were collected on a VG Microscope (HB501UX). The aqueous solutions containing 0.035 mmol/L for each REE ion were prepared by dissolving their nitrate salts in an aqueous solution. The deionized (D.I.) water used for preparing the aqueous solutions has a specific resistance of 18 M Ω -cm or greater; pH values of the REEs aqueous solutions was 5.3 except for the acidity experiment.

2.2 Preparation and characterization of nanosorbents

To compare the structure effects of carbon-based nanosorbents on rare earth element separations, three different types of nanosorbents were prepared in our investigations, that is, solid carbon spheres, 60 nm carbon shells and 500 nm carbon shells. The 60 nm carbon shells and 500 nm carbon shells were prepared by varying the size of silica spheres used as the template^[31]. Silica spheres (80 mg) were mixed with dopamine (80 mg) in tris-buffer (25 mL, 10 mM; pH=8.5) for 24 h. The polymer/silica nanocomposites obtained were collected by centrifugation, then carbonized under N₂ atmosphere at 400 °C for 2 h with a heating rate of 1 °C/min, which was followed by further treatment at 800 °C for 3 h with a heating rate of 5 °C/min. After washing in 5% HF aqueous solution for 24 h, the carbon shells were obtained. STEM images of the carbon shells are given in Fig. 1 revealing clearly their structure difference. In addition to the obvious difference in diameter, the shell thickness of the 500 nm carbon is significantly larger than the shell of the 60 nm carbon.

2.3 Adsorption and desorption experiments

The adsorption experiments were performed by contacting 0.01 g carbon-based nanosorbents with 5 mL of an REE-containing aqueous solution for 60 min in a vibrating mixer. After 1 h, the suspensions were filtered for determination of REE concentrations in the aqueous phase. For desorption experiments, the nanosorbents loaded with REEs were placed in nitric acid with different concentrations and vibrated for 60 min. The REE

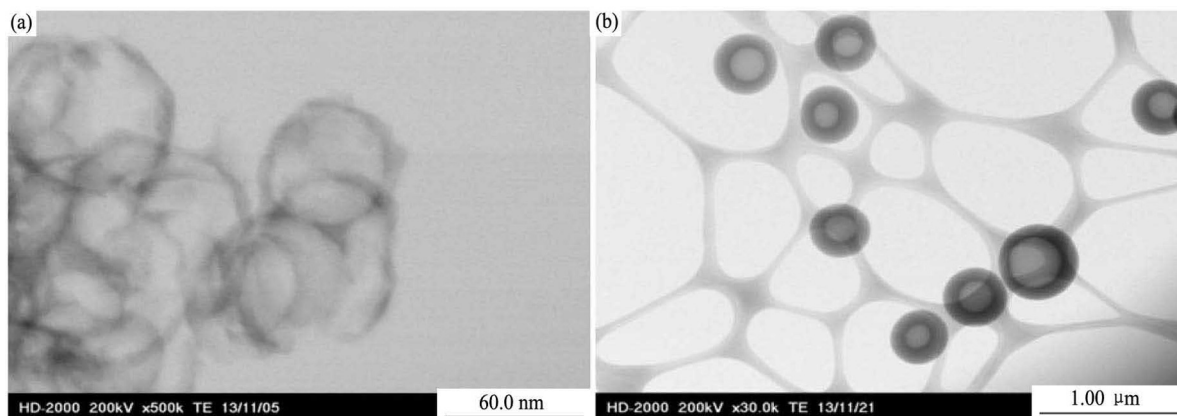


Fig. 1 STEM images of the 60 nm carbon shell (a) and 500 nm carbon shell (b)

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