Supporting Information

Adsorption of Cu(II), Zn(II), and Pb(II) from aqueous single and binary metal solutions by regenerated cellulose and sodium alginate chemically modified with polyethyleneimine

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Synthesis of adsorbents

To achieve the proposed goals, the following experimental details were conducted: firstly, cotton linter was dissolved in 7 wt.% NaOH/12 wt.% urea aqueous solution pre-cooled to -12° C to obtain a 4 wt.% transparent cellulose solution under vigorous mechanical agitation for 3 min at ambient temperatures (below 25° C)¹; SA was dissolved in the same weight ratio of NaOH/urea aqueous solution without pre-cooled to form a 4 wt.% homogeneous SA solution. Secondly, the cellulose solution and SA solution were mixed together by the weight ratio of 1:1 under agitation to generate a homogeneous dispersion. Subsequently, 30% (w/v) PEI aqueous solution was added into the above mixtures (100 g) with a blend ratio of 1:5 (v/w) under gentle agitation to prepare a mixed solution as the water-phase; paraffin oil (250 mL) containing 5% (v/v) span 80 was used as oil-phase. And then, the prepared water-phase and oil-phase were transferred into a three-necked round-bottomed flask (500 mL) successively. To obtain the adsorbent particles with suitable size, the two-phase mixture was stirred under a vigorous speed of 1000 rpm at the room temperature. After 30 min, 6 mL of GA (25 wt.% aqueous solution) was dropwise-added to this reaction system within 10 min, and the system was further vigorously stirred for 6 h at 60°C. Finally, the resultant adsorbent particles were rinsed thoroughly with acetone and double deionized water and freeze-dried (-52°C, 20 Pa) for 48 h in a freeze drier. The drying samples were marked as PEI-RCSA and stored in a desiccator for sample characterization and further adsorption experiments. Fabrication of the samples RC was the same process as PEI-RCSA, except adding SA solution and PEI aqueous solution into water-phase.



Fig. S1 (a) SEM image of native cellulose, (b) SEM image of SA, (c) SEM image of PEI-RCSA after the adsorption of Cu(II), (d) EDS pattern of PEI-RCSA after Cu(II) adsorbed.



Fig. S2 Nitrogen adsorption-desorption isotherm of the PEI-RCSA.



Fig. S3 (a), (b), (c), and (d) were digitized photographs of the PEI-RCSA before adsorption and the PEI-RCSA loaded with Cu(II), Zn(II) and Pb(II), respectively.



Fig. S4 pH variation during the adsorption of Cu(II), Zn(II) and Pb(II) in their respective single-component aqueous solutions.



Fig. S5 Pseudo-first-order kinetic model and pseudo-second-order kinetic model for the adsorption of Cu(II), Zn(II) and Pb(II) in their respective single-component aqueous solutions. (Conditions: $25 \pm 1^{\circ}$ C; pH 5.5; adsorbent dosage: 1g L⁻¹; shaking speed: 130 rpm; C_0 : 200 mg Cu L⁻¹, 200 mg Zn L⁻¹ and 200 mg Pb L⁻¹).



Fig. S6 Intraparticle diffusion model for adsorption of Cu(II), Zn(II) and Pb(II) in their respective single-component aqueous solutions. (Conditions: $25 \pm 1^{\circ}$ C; pH 5.5; adsorbent dosage: 1g L⁻¹; shaking speed: 130 rpm; C_0 : 200 mg Cu L⁻¹, 200 mg Zn L⁻¹ and 200 mg Pb L⁻¹).



Fig. S7 Langmuir model and Freundlich model for the adsorption of Cu(II), Zn(II) and Pb(II) in their respective single-component aqueous solutions. (Conditions: $25 \pm 1^{\circ}$ C; pH 5.5; adsorbent dosage: 1 g L⁻¹; contact time: 8 h; shaking speed: 130 rpm).



Fig. S8 Adsorption behavior of the prepared adsorbent PEI-RCSA in the Pb(II)-Cu(II), Pb(II)-Zn(II) and Cu(II)-Zn(II) binary aqueous system (i,j is Pb(II), Cu(II), Zn(II), $i\neq j$).



Fig. S9 Possible adsorption mechanism between heavy metal ions, nitrogen atoms in the PEI polymer chains and carboxyl groups on SA (M²⁺ represents divalent metal ion).



Fig. S10 Adsorption efficiency of the prepared adsorbent PEI-RCSA for removal of Cu(II), Zn(II) and Pb(II) in their respective single-component aqueous solutions at different regeneration recycles. Significant differences between cycle 0 and cycle 3 were indicated by stars; *: 0.01 < p-value < 0.05, n = 3.

References

1 J. Cai and L. Zhang, *Biomacromolecules*, 2006, 7, 183–189.