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Magnetic Solid-Phase Separation of Uranium from Aqueous Solution Using High Capacity Diethylenetriamine Tethered Magnetic Adsorbents

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Abstract : The magnetic solid-phase extraction is a relatively new method among the other solid-phase extraction techniques for the separating of metal ions from aqueous solutions, such as mine water and groundwater, contaminated wastes, etc. However, the bare magnetic particles (Fe3O4) exhibit poor selectivity due to the absence of target-specific functional groups for sequestering the metal ions. The selectivity of these magnetic particles can be remarkably improved by covalently tethering the task-specific ligands on magnetic surfaces. The magnetic particles offer a number of advantages such as quick phase separation aided by the external magnetic field. As a result, the solid adsorbent can be prepared with the particle size ranging from a few micrometers to the nanometer, which again offers the advantages such as enhanced kinetics of extraction, higher extraction capacity, etc. Conventionally, the magnetite (Fe3O4) particles were prepared by the hydrolysis and co-precipitation of ferrous and ferric salts in aqueous ammonia solution. Since the covalent linking of task-specific functionalities on Fe3O4 was difficult, and it is also susceptible to redox reaction in the presence of acid or alkali, it is necessary to modify the surface of Fe3O4 by silica coating. This silica coating is usually carried out by hydrolysis and condensation of tetraethyl orthosilicate over the surface of magnetite to yield a thin layer of silica-coated magnetite particles. Since the silica-coated magnetite particles amenable for further surface modification, it can be reacted with task-specific functional groups to obtain the functionalized magnetic particles. The surface area exhibited by such magnetic particles usually falls in the range of 50 to 150 m2.g-1, which offer advantage such as quick phase separation, as compared to the other solid-phase extraction systems. In addition, the magnetic (Fe3O4) particles covalently linked on mesoporous silica matrix (MCM-41) and task-specific ligands offer further advantages in terms of extraction kinetics, high stability, longer reusable cycles, and metal extraction capacity, due to the large surface area, ample porosity and enhanced number of functional groups per unit area on these adsorbents. In view of this, the present paper deals with the synthesis of uranium specific diethylenetriamine ligand (DETA) ligand anchored on silica-coated magnetite (Fe-DETA) as well as on magnetic mesoporous silica (MCM-Fe-DETA) and studies on the extraction of uranium from aqueous solution spiked with uranium to mimic the mine water or groundwater contaminated with uranium. The synthesized solid-phase adsorbents were characterized by FT-IR, Raman, TG-DTA, XRD, and SEM. The extraction behavior of uranium on the solid-phase was studied under several conditions like the effect of pH, initial concentration of uranium, rate of extraction and its variation with pH and initial concentration of uranium, effect of interference ions like CO32-, Na+, Fe+2, Ni+2, and Cr+3, etc. The maximum extraction capacity of 233 mg.g-1 was obtained for Fe-DETA, and a huge capacity of 1047 mg.g-1 was obtained for MCM-Fe-DETA. The mechanism of extraction, speciation of uranium, extraction studies, reusability, and the other results obtained in the present study suggests Fe-DETA and MCM-Fe-DETA are the potential candidates for the extraction of uranium from mine water, and groundwater.

Keywords : diethylenetriamine, magnetic mesoporous silica, magnetic solid-phase extraction, uranium extraction, wastewater treatment
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