Removal of Selenium Ions from Aqueous Media by Magnetic Graphene Oxide

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Abstract

Selenium is an essential nutrient element for life at trace concentrations, but extremely toxic at higher concentrations.[1] Selenium is widely distributed in sulfide minerals and coal ashes. During mining and mineral processing, selenium ions can be released into water and soils, and then further transferred to the plants, animals and life cycles. A guideline value of less than 40 ppb selenium in drinking-water is given by the World Health Organization (WHO).[2] Excessive selenium can be normally accumulated through mine drainage water, industrial wastewater and agricultural drainage water. The removals of selenium from these water systems are crucial to reduce its environmental impact and enhance the sustainable development of natural resources. Among all the valence states (i.e., Se^{2-}, Se^{0}, SeO_3^{2-}, and SeO_4^{2-}), Se (IV) (selenite) and Se (VI) (selenate) are more mobile and toxic.

Among the different methods developed for the removal of selenium[3] (such as ion exchange, air flotation, chemical precipitation, emulsion liquid membranes, nanofiltration, biological reduction, reverse osmosis, solvent extraction and adsorption), the adsorption method is considered to be more efficient and economic due to its fast removal rate and minimum pretreatment of samples. However, conventional adsorbents, such as silica gel, clay, activated carbon, molecular sieve, ferrihydrite, ferric oxyhydroxide/peat/resins, and activated aluminium oxide, only show limited removal capacity for Se (IV) and perform poorly in removing Se (VI).[3]

In this work, a water-dispersible magnetic graphene oxide (MGO) was synthesized using a two-step method: (1) preparation of graphene oxide (GO) by a modified Hummers method[4], and (2) in situ growth of magnetic iron oxide nanoparticles. The chemical and surface properties of GO and MGO were characterized by various techniques such as X-Ray Diffraction (XRD), Fourier Transform Infrared Spectroscopy (FTIR), Atomic Force Microscope (AFM), X-ray Photoelectron Spectroscopy analysis (XPS), Transmission Electron Microscopy (TEM), Scanning Electron Microscope (SEM), Quantum Design 9T-PPMS magnetometer and Thermo Gravimetric Analysis (TGA). The XRD patterns manifested that the magnetic particles were mainly Fe_3O_4 and γ-Fe_2O_3 particles. FTIR and XPS spectra explained the bonding mode of Fe with O. AFM image illustrated the thickness of monolayer GO. Together, TEM (Figure 1.) and SEM images showed the uniform distribution of magnetic particles on monolayer GO sheets. Thermo stability was proved according TGA results. Magnetic hysteresis loop showed about equal to zero value for Mr (remanence) and Hc (coercivity), indicating MGO is superparamagnetic.

MGO was applied to remove selenium ions (both Se (IV) and Se (VI)) in industrial wastewater. The systematic adsorption tests showed high removal ratio of ~99% for Se (IV) and ~85% for Se () in wastewater within 10 seconds. Adsorption capacity (Q (mg/g)) under different initial (a) Se (IV) and (b) Se (VI) concentrations were well fitted both Freundlich and Langmuir Models (Figure 2.), indicating a large potential saturated adsorption capacity (64.10 mg/g for Se (IV) and $1.44*10^{6}$ mg/g for Se (VI)). The synthesis and adsorption mechanisms of the MGO were proposed.[5] Additionally, the MGO could be separated effectively under an external magnetic field and reused after recycling. It is evident from this work that MGO is a novel and very promising adsorbent for selenium removal, and our results provide new insights into the development of novel graphene-based nanomaterials for water treatment and environmental applications.[6]

References

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Figures

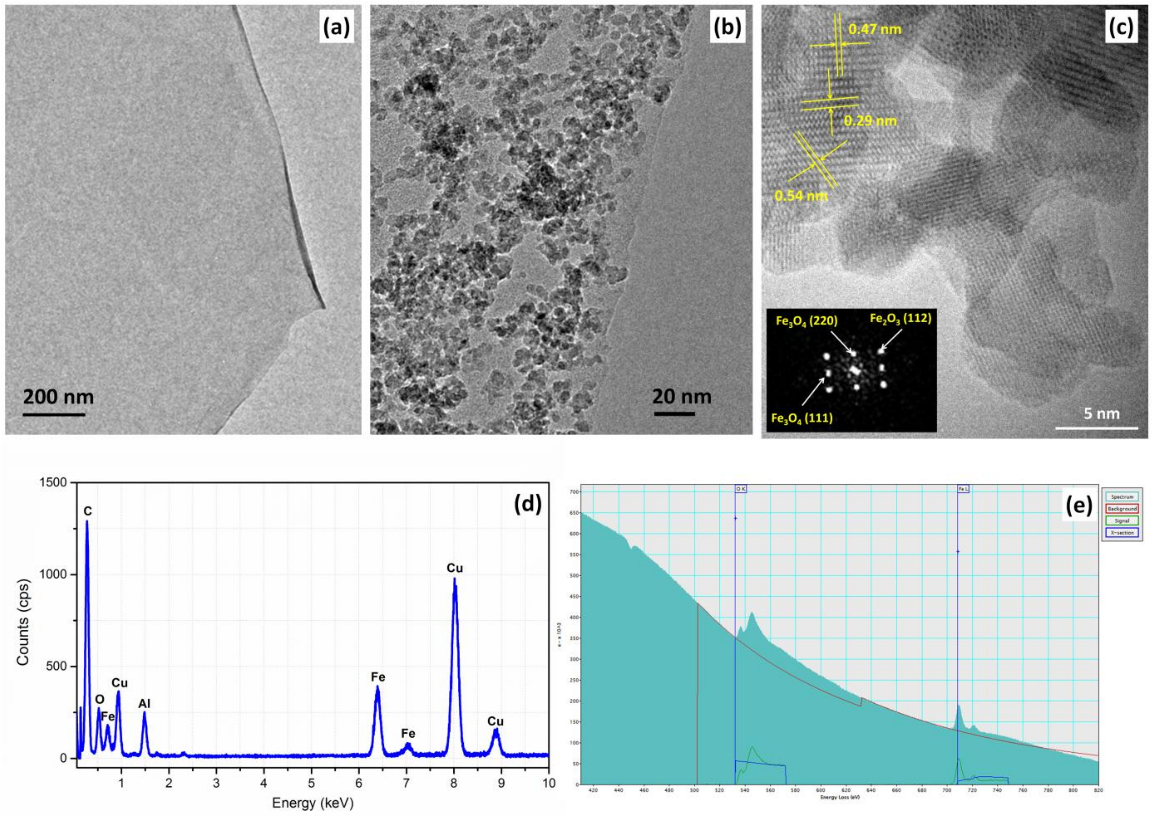

Figure 1. TEM images of (a) GO, (b) MGO and (c) HRTEM image of selected area of magnetic particles, insert: FFT image of selected area electron diffraction pattern of magnetic particles; (d) EDS spectrum indicates the presence of elements O, C, and Fe; (e) EELS spectrum of iron oxide nanoparticles in MGO with an atomic percentage of Fe (17.77 %) and O (82.23 %).

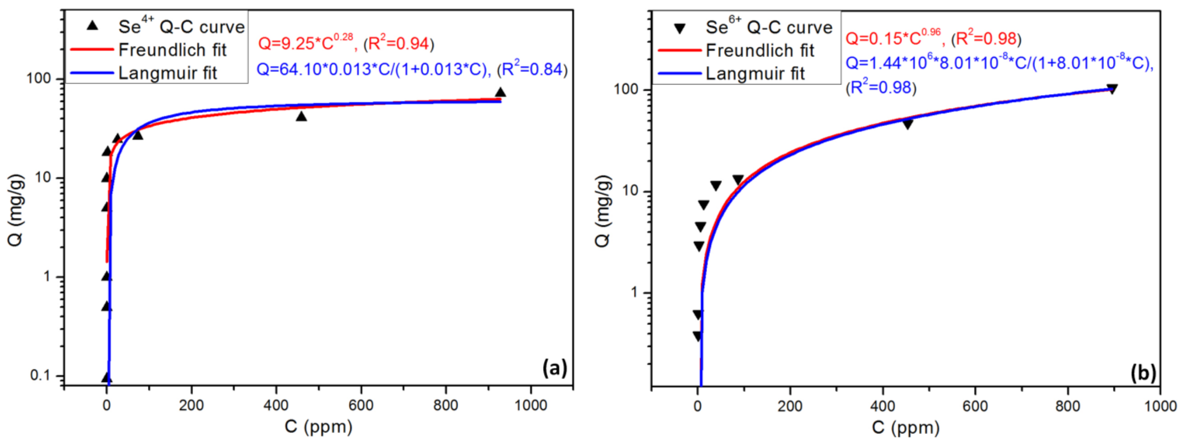

Figure 2. Fitting curves using Freundlich and Langmuir Models under different initial (a) Se (IV) and (b) Se (VI) concentrations.