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Characterization of Arsenic Sorption on Siderite Spherules under anoxic Environments: Effects of Siderite Crystal Size and Surface Modification by Citrate

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Arsenic (III and V) uptake by different sized siderite spherules was characterized under anoxic and neutral pH conditions. Adsorption isotherm studies showed that small-sized siderite has stronger arsenic sorption affinity rather than large-sized siderite, and depends more on the arsenic species. The small-sized siderite has the highest sorption affinity for As(V) due to the additional precipitation of symplesite ($Fe_3(AsO_4)_2 \cdot 8H_2O$). In contrast, it has the lowest sorption affinity for As(III) due to the competition with dissolved bicarbonates generated by its faster dissolution. Extended X-ray absorption fine structure (EXAFS) revealed that As(V) adsorbed on both siderites forms inner-sphere complexes through bidentate-binuclear corner-sharing, whereas As(III) can be adsorbed on these siderites through outer-sphere as well as inner-sphere surface complexations. For enhanced arsenic uptake, a novel citrate/$FeCO_3$ nanocomposite (CF-NC) was synthesized by surface modification of the small-sized siderite using citrate. As a result of the modification, citrate was incorporated into the surface structure of siderite (up to 17.94%) through Fe(II)-citrate complexes. In addition, the crystal morphology of rhombohedral siderite was changed into hierarchically nanostructured spherical aggregates composed of several sheet-like crystals. As(III) uptake increased from 15.2% (without citrate) up to 88.2% after the modification. The maximum sorption capacities (q_m) of the CF-NC for As(III) and As(V) were 189.0 and 290.2 mg/g, respectively, which are comparable or superior to other siderite-based adsorbents. Our findings suggest that both arsenic species (III and V) can be effectively attenuated by siderite under anoxic aqueous environments. In addition, the sorption behaviors and mechanisms are strongly dependent on the arsenic species but also the surface properties of siderite associated with dissolution character. Therefore, siderite with smaller crystals or/and further modified surface using citrate can lead to the advanced arsenic remediation even in natural anoxic environments.