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Vanadium(V) Adsorption and Reduction on *Shewanella oneidensis* Bacterial Surfaces: The Role of Amine Sites

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Bacterial reduction of V(V) represents a potentially effective and efficient method for removing vanadium from contaminated waters and for recovering vanadium from secondary resources. However, a limited understanding of the underlying reduction mechanisms limits optimization of bacterial vanadium removal and recovery. V(V) reduction occurs on bacterial cell surfaces, requiring V(V) adsorption onto the cell as the first step in but the overall reduction reaction. However, the mechanism that controls V(V) oxyanion adsorption onto negatively charged bacterial cell surfaces under environmentally relevant conditions is unknown. Here, we investigated the interaction between V(V) and two bacterial species, *Bacillus subtilis* and *Shewanella oneidensis*, measuring bulk removal and using X-ray absorption spectroscopy to measure the extent of V(V) reduction and to constrain the binding mechanisms. We found that both species can remove significant concentrations of dissolved V(V) under non-metabolizing conditions, with removal strongly dependent on pH and bacterial species. At near-neutral pH (5–7), where V(V) is primarily present as anionic $H_2VO_4^-$, *S. oneidensis* removed substantially more V(V) than *B. subtilis*. Selective site-blocking experiments indicate that cell surface amine sites control $H_2VO_4^-$ removal from solution, whereas sulfhydryl sites do not play a significant role. X-ray absorption spectroscopy and surface complexation modeling indicate that adsorbed V(V) on the *S. oneidensis* surface was entirely reduced to V(IV), followed by transfer of cationic V(IV) from positively charged amine sites to negatively charged carboxyl sites. This process likely regenerates amine sites and facilitates the enhanced V(V) adsorption and reduction by *S. oneidensis*. These findings provide new mechanistic insights into V(V) oxyanion adsorption onto bacterial cells, and highlight the importance of cell surface amine sites in controlling V(V) adsorption and reduction under environmentally relevant conditions. We anticipate that other metal-reducing bacterial species exhibit similar enhanced capacities to those observed for *S. oneidensis* for V(V) removal and reduction, suggesting that these species could be especially effective in V removal and recovery from aqueous sources.