

Theoretical and Experimental Study of Electrochemically Mediated Adsorption Processes

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There is a major water crisis in the modern world due to the shortage in clean water availability and concerns over potable water quality, which necessitates the development of more energy efficient and cost effective means for water deionization and waste water remediation. Water treatment by electrosorption has been a topic of increasing interest due to its high efficiency, small footprint and no secondary treatment requirement, compared with traditional water treatment methods. Electrochemically mediated adsorption is one of the emerging electrosorption techniques and has the advantage of superb selectivity (>100), which allows selective removal of toxic or high-valued target ions, and is difficult to achieve by other water treatment techniques.

Firstly, to understand electrochemically mediated adsorption, a theoretical approach has been used to develop models based on the underlying physics with hierarchical complexity that range from an equivalent circuit model to a process-level lumped parameter model and eventually to a full two-dimensional time dependent transport model. These models enable focus on different aspects of the process (thermodynamics, kinetics and mass transport) and shed light on the mechanism of competitive electrosorption processes from the microscale to the macroscale. The theoretical framework also provides guidelines for the design of redox active materials and optimization of operation conditions to achieve high salt adsorption capacity as well as selectivity towards target micropollutants.

Secondly, we study the electrochemically mediated adsorption experimentally. With a continuous flow experimental platform equipped with inline sensors, we developed methodologies to allow real-time measurement of the salt adsorption performance as well as quantification of the selectivity of the competitive electrosorption process. Asymmetric redox active electrodes are designed, synthesized and further characterized in the flow system. The experimental study has demonstrated high salt adsorption capacity and ultra-high salt adsorption rate using the asymmetric redox active electrodes for brackish water deionization; and more importantly, selective removal of an organic acid, which is chosen as a model micropollutant, for wastewater remediation applications.

Finally, we extend the concept and methodology of electrochemically mediated adsorption from surface based (heterogeneous) electrosorption to bulk adsorption/regeneration with (homogeneous) complexation reactions. A theoretical framework coupling thermodynamics, kinetics and mass transport with bulk complexation reactions is developed for a flow-through configuration. The model has been applied to a particular case study of carbon dioxide capture/release, which is of great practical interest. A novel reactor design has been implemented and tested under various current densities and flow rates, which demonstrates the feasibility of utilizing the flow-through reactor for electrochemically mediated carbon capture and regeneration.

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