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Multicomponent transport in membranes for redox flow batteries¹

CHARLES MONROE, University of Michigan

Redox flow batteries (RFBs) incorporate separator membranes, which ideally prevent mixing of electrochemically active species while permitting crossover of inactive supporting ions. Understanding crossover and membrane selectivity may require multicomponent transport models that account for solute/solute interactions within the membrane, as well as solute/membrane interactions. Application of the Onsager–Stefan–Maxwell formalism allows one to account for all the dissipative phenomena that may accompany component fluxes through RFB membranes. The magnitudes of dissipative interactions (diffusional drag forces) are quantified by matching experimentally established concentration transients with theory. Such transients can be measured non-invasively using DC conductometry, but the accuracy of this method requires precise characterization of the bulk RFB electrolytes. Aqueous solutions containing both vanadyl sulfate (VOSO_4) and sulfuric acid (H_2SO_4) are relevant to RFB technology. One of the first precise characterizations of aqueous vanadyl sulfate has been implemented and will be reported. To assess the viability of a separator for vanadium RFB applications with cell-level simulations, it is critical to understand the tendencies of various classes of membranes to absorb (uptake) active species, and to know the relative rates of active-species and supporting-electrolyte diffusion. It is also of practical interest to investigate the simultaneous diffusion of active species and supports, because interactions between solutes may ultimately affect the charge efficiency and power efficiency of the RFB system as a whole. A novel implementation of Barnes’s classical model of dialysis-cell diffusion [*Physics* **5**:1 (1934) 4-8] is developed to measure the binary diffusion coefficients and sorption equilibria for single solutes (VOSO_4 or H_2SO_4) in porous membranes and cation-exchange membranes. With the binary diffusion and uptake measurement in hand, a computer simulation that extends the approach of Heintz, Wiedemann and Ziegler [*J. Membrane Science* **137**:1-2 (1997) 121-132] is used to establish Onsager resistances that describe the drag forces VOSO_4 and H_2SO_4 exert on each other as they interdiffuse. The ramifications of these interactions for different classes of membranes – and for RFB applications – will be discussed.

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