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Analyzing Impacts of Gas Evolution within a Batch-Mode Electrodialysis of Lithium Sulfate using Two-Phase Flow CFD Simulation

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Electrodialysis (ED) technology relies on the selective permeability of ion exchange membranes, which allows for the separation of ions in a solution under the influence of an electric field [1,2]. ED has been frequently used for recovery of acids and bases [3,4]. Due to the growing demand for electric vehicles and the crucial need for recycling the lithium-ion batteries (LIBs), this study investigates the ED of Li_2SO_4 solution, aimed at comprehending the efficacy and requisite control measures for effective closed-loop recycling of spent LIBs. The enhanced flexibility and energy efficiency of batch-mode EDs highlight the significance of time-dependent models. The gaseous molecules evolved from electrochemical reactions induce localized turbulence, significantly impacting the velocity, potential, and concentration distributions. This study presents a two-phase model that analyzes the dynamic behavior of a batch-mode Li_2SO_4 ED and investigates the bubbles'impact employing an Euler-Euler model. The finite element method solves time-dependent hydrodynamic, mass conservation, and electrochemical equations. The excellent agreement between the model and experimental data shows the accuracy and validity of this model, emphasizing its applicability to analogous electrochemical processes such as electrolysis. According to the results of this two-phase flow model, the maximum volume fraction of H_2 and O_2 bubbles at the surface of the cathode and anode are 9% and 5%, respectively. As illustrated in Fig. 1, It is observed that the buoyant and drag forces induced by bubbles lead to a significant increment in liquid velocity and circulation of electrolyte solutions in concentrate channels. The bubble-induced turbulence disrupts the uniform velocity distribution near electrodes and increases the liquid velocity magnitude. This circulation of electrolyte solution declines the average concentration polarization across IEMs and increases ionic fluxes, particularly on the concentrate side of IEMs, which leads to the improvement of ED performance, as observed in Fig. 2. The liquid circulation leads to a substantial 60% reduction in the average Li^+ concentration polarization across the CEM. In this case, the average total Li^+ flux enhances by 12%, resulting in the significant increment of average Li^+ concentration from 551 $mol \cdot m^{-3}$ to 768 $mol \cdot m^{-3}$ at t=2 hours. Furthermore, the enhancement of current density and reduction of bubbles diameter results in the increment of gas volume fraction. Increasing the flowrate causes a decrease in the average gas volume fraction and eliminates the impact of bubbles on the liquid velocity, as shown in Fig. 3. The effect of bubbles on ohmic overpotential is also discussed, and it is understood that it is highly dependent on inlet flowrate and liquid circulation. The presence of bubbles increases the ohmic overpotential by approximately 0.5 V in high inlet flowrate, $F_{in}=0.3$ $L \cdot s^{-1}$, that no circulation is observed. However, when electrolyte solutions circulate at low inlet flowrates, $F_{in}=0.03$ $L \cdot s^{-1}$, a higher ionic concentration and consequently lowers ohmic overpotential of electrolyte solutions are observed.

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