Selective Lithium Recovery From Complex Brines Using Crown-Functionalized LDH–PEI Hybrid Membranes

Almogren, Fatema (School: Rowad Alkhaleej International School)

Global lithium demand is projected to increase by nearly 4000% by 2040 due to expansion of electric vehicles and energy storage systems; however, current extraction methods are water-intensive and lack selectivity in complex multi-ion brines. This study developed a dual-mechanism membrane integrating molecular recognition and electrostatic exclusion for selective lithium recovery. The membrane consisted of layered double hydroxide (LDH), hydroxymethyl-12-crown-4 for lithium-specific binding, and post-quaternized polyethyleneimine (PQ-PEI). Membranes containing 2, 5, and 10 wt% PEI were fabricated, along with control membranes without crown ether functionalization. Membrane performance was evaluated using synthetic brines under 1 bar dead-end filtration and validated using real desalination brine under cross-flow conditions. Screening and real brine experiments, including controls, were conducted in duplicate. The optimized 5 wt% PEI membrane achieved 98% lithium passage with high rejection of competing ions (Na? 98%, Mg²? 90%, Ca²? 98%, K? 99%), resulting in Li/Na and Li/Mg selectivity values of 49 and 9.8. The membrane exhibited high initial flux (~190 LMH) with stable performance. Control membranes showed reduced lithium selectivity. Scale-up testing was performed in triplicate and demonstrated consistent selectivity and stable flux, indicating good scalability. This system demonstrates high-selectivity lithium recovery with strong potential for scalable extraction from realistic brine conditions.

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