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A three-electrode dual-power-supply electrochemical pumping system for fast and energy efficient lithium extraction and recovery from solutions

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The demand for Li-ion batteries (LIBs) for use in electric vehicles, which is key to realizing a decarbonized society, is accelerating. However, the supply of Li resources has recently become a major issue, thereby necessitating the development of economical and sustainable technologies of brine/seawater-based Li extraction and recycling Li from spent LIBs. This paper presents an innovative electrochemical pumping technology based on a new cell structure for Li extraction/recovery. This system can provide large electrochemical driving forces while preventing the occurrence of electronic conduction due to electrolyte reduction. This electrochemical pumping system allows extraction/recovery of Li ions from the anode side to the cathode side, rather than the diffusion of other ions, due to the ion-diffusion-bottleneck size of the electrolyte material. Using this system, high-purity Li can be collected with high energy efficiency and at least 464 times faster than that via conventional electrochemical pumping, even with a commercially available Li-ion electrolyte plate.

The use of rechargeable batteries has accelerated in the 21st century. In particular, the market share of Li-ion batteries (LIBs) is significantly expanding owing to their high performance, characterized by their high energy and power densities and long life[1–5]. Furthermore, Li resources are anticipated to be used as fuel for thermonuclear fusion power generation after 2035[6–9]. Until 2017, most Li resources were extracted from brine via long-term evaporation. However, the majority of the recent supply is derived from mines for temporarily satisfying the rapidly increasing demand. The supply from mines can tackle the rapid increase in demand owing to the short construction period of this approach. However, problems such as a large environmental impact and high extraction and refining costs are encountered in this strategy. Thus, to satisfy the rapidly growing demand for Li resources, the development of innovative Li extraction technologies is crucial for enabling an economical, safe, and sustainable supply of Li. The European Union is considering mandating the extraction, recovery, and recycling of Li resources as battery materials[10]. However, the recovery of battery-grade high-purity Li using conventional refining methods is difficult. Therefore, technologies that can permit the recovery of Li from spent LIBs must be urgently developed. New Li-extraction/recovery technologies, such as adsorption/dissociation (ion sieving)[11–22], ion exchange[23–28], precipitation[29–33], and liquid–liquid extraction[34–40], have been proposed in this regard. However, these technologies are unsuitable for industrial processes because of their small extraction volumes, low recoveries, and inferior element selectivity.

Electrochemical pumping using Li-ion conductors shows promise as an alternative Li-extraction/recovery technology[41–48]. In this method of resource extraction/recovery, the desired ions in a tank—which are fractionated by a diaphragm with ionic conductivity—are transferred and extracted to another tank owing to a difference in electrochemical potential. Li-containing solutions, such as aqueous solutions of the Li ions from spent LIBs dissolved in acid, brine, or seawater, can be used in this regard. Densely sintered Li-ion conductors, such as lithium lanthanum titanates (LLTO; $La_{2/3-x}Li_{3x}TiO_3$) and ionic liquids, are typically used as diaphragms. When a sufficiently large potential difference for the electrolysis of water is

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