

# Electrodialytic recovery of lithium from secondary mining resources with lepidolite

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Lithium (Li) is an alkali metal widely applied in chemical, energy, medicine and metallurgical sectors. The highest sublimation energy, electronegativity and ionization energy and the lowest ionic radius for the alkali groups promotes a high charge density for Li ions. These features turned Li into a strategic source to produce rechargeable batteries in, e.g., mobile phones, laptops, digital cameras, and electric vehicles (Bao et al. 2023).

Nevertheless, Li supplies are raising awareness across the globe, once Li demand is expected to quadruplicate in 8 years, largely driven by the diffusion of green technologies and electric-vehicle battery use. From 2022 to 2030, 56% of known reserves are foreseen to be consumed, which means approximately 11,108,000 metric tons. The world Li top suppliers are Australia, Chile, China and Argentina, with 96% of global Li production. In Portugal, Li resources are estimated in 270,000 and reserves in 60,000 metric tons (U.S. Geological Survey 2022).

The European Union has recognized the Li dependency of Europe outside countries and has included Li in the European critical raw material list due to the high risk of scarcity and current economic relevance (European Commission 2020a). Lithium resources mainly exist in brines and minerals. Regarding the hard rock exploitation, granite pegmatites are important sources of Li. Pegmatites are composed by abundant quartz, feldspar and mica, and valuable minerals. The main mineral sources of Li are spodumene, lepidolite and petalite. For instance, lepidolite,  $K(Li, Al)_3(Si, Al)_4O_{10}(F, OH)_2$ , is a Li-rich mica mineral with Li contents of 3 to 7.7 % (Liu et al. 2022).

On the other hand, mining activities cause adverse environmental impacts and tend to intensify vulnerabilities in the local ecosystems and social livelihood. In fact, during mine ore's extraction, significant amounts of residues are generated, where Li can be found in lower contents. Therefore, active research on the extraction of Li is being conducted to find alternatives to conventional mining exploitation and alleviate the impacts from Li production. European Commission standards towards circular economy are also pushing the development of innovative technologies to recover critical raw materials from secondary resources and to reuse them in a safe manner (European Commission 2020b).

Electro-based technologies entail the application of a direct low-level current density ( $\text{mA}/\text{cm}^2$ ) between two electrodes, to promote the removal of substances from a wide variety of environmental substrates. Anion (AEM) and cation exchange membranes (CEMs) are commonly placed in the reactor configuration to separate the contaminated matrix from the electrolyte compartment. This step allows the control of the pH conditions and the selectivity of the removal of contaminants (Ribeiro and Rodríguez-Maroto 2006).

The electrodialytic (ED) process was successfully applied during the recovery of critical raw materials from solid samples, such as phosphorus from sewage sludge (Guedes et al. 2016) and tungsten from mining residues (Almeida et al. 2020). In this sense, the ED technology is being optimized to empower the recovery of Li from lepidolite present in secondary mining sources. Also, the combination of organic and inorganic acids and bases with the ED treatment was evaluated to enhance Li extraction ratio. In this work, the ED treatment was tested, combining acids and bases, to assess the feasibility to recover Li from secondary mining resources with lepidolite contents. The selection of the optimal system design and the improvement of the ED reactor configuration was achieved through experimental tests performed at a bench scale.

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