

## Development of Lithium Separation Technique Based on the Solubility of Salts in Deep Eutectic Solvents

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### Summary

In lithium production from salt lake brine, the separation of lithium from abundant alkali and alkaline earth metals is a decisive factor in the success of the process. Solvent extraction is one of the industrially viable techniques for rapid metal separation; however, conventional extraction systems struggle with the separation of lithium and magnesium. In response to this challenge, the authors have proposed a lithium separation method based on solid-liquid extraction. By employing appropriately molecularly designed deep eutectic solvents (DES) for the poorly water-soluble phosphate salts of lithium and magnesium, it becomes possible to selectively extract lithium from mixed salts. DESs, which are mixtures of hydrogen bond donors (HBDs) and hydrogen bond acceptors (HBAs), have gained attention as third-generation solvents that can serve as alternatives to water and organic solvents. This study investigates the effect of DES molecular structure on the mutual separation of lithium and magnesium. DESs were prepared using various  $\beta$ -diketone compounds as HBDs and alkylphosphine oxides as HBAs. The  $^{31}\text{P}$  nuclear magnetic resonance spectra confirmed that the intermolecular interactions between HBD and HBA vary depending on the Brønsted acidity of the HBD. Among the  $\beta$ -diketones, the strength of intermolecular interactions with the HBA followed the order: furyl > thenoyl > benzyl substituents.

In liquid-liquid extraction tests using aqueous solutions containing lithium and magnesium, divalent metal ions such as magnesium were preferentially extracted depending on the pH of the aqueous phase, making selective lithium extraction challenging. On the other hand, when phosphate salts of each metal were directly contacted with the DES for solid-phase extraction, it was found that DESs containing  $\beta$ -diketones with thenoyl groups enabled preferential dissolution of lithium over magnesium. These results highlight that, in addition to the cooperative effect of  $\beta$ -diketones and phosphine oxides, substituent design plays a crucial role in enhancing lithium/magnesium separation performance.