



## Augmenting deep eutectic solvent leaching and anti-solvent crystallization with density functional theory (DFT) for recycling of lithium-ion batteries

S. Teimouri<sup>1</sup>, C. Ma<sup>1</sup>, M. Svärd<sup>1</sup>, and K. Forsberg<sup>1\*</sup>  
<sup>1</sup>KTH Royal Institute of Technology, Stockholm, Sweden  
[\\*kerstino@kth.se](mailto:*kerstino@kth.se)

**Keywords:** Deep Eutectic Solvent (DES), Anti-Solvent Crystallization, Recycling, Li-Ion Batteries, Density Functional Theory (DFT)

As the demand for lithium-ion batteries (LIBs) continues to grow, there is a critical need for environmentally friendly and efficient recycling methods for the resulting LIB waste [1]. One novel recycling technique is based on deep eutectic solvents (DESs) — eutectic mixtures formed through hydrogen bonding, typically between two components: one acting as a hydrogen bond donor (HBD) and the other as a hydrogen bond acceptor (HBA). The components of DESs are often inexpensive and derived from biodegradable sources. Favourable DESs properties include them being biodegradable, easy to synthesis, eco-friendly, non-volatile and not requiring additional reducing agents in the leaching process [2, 3]. This study examines several carboxylic acid-based DESs for their ability to dissolve lithium cobalt oxide (LCO), a common cathode material in LIBs. The DESs comprised polyethylene glycol 200 as the HBA and five different organic acids as the HBD: tartaric acid, ascorbic acid, citric acid, oxalic acid, and succinic acid. After identifying the most effective DES for extracting lithium and cobalt from LCO material, the metal can be recovered with anti-solvent crystallization. Anti-solvent crystallization entails adding a water-miscible organic solvent (i.e. acetone, ethanol) to the solution to reduce the solubility of the solute and initiate its precipitation. Because their volatility differs greatly from that of the DESs, the organic solvents can be recovered by evaporation with relatively low energy consumption [4]. Calculating the Gibbs Free Energy of reactions using density functional theory (DFT), a computational modelling technique [5], it is possible to predict which carboxylic acid-based HBDs are most stable and likely to form complexes with lithium and cobalt. By corroborating the experimental results, DFT is proposed as a complementary tool that provides deeper insights into the studied systems, such as the optimal geometry of complexes, enhancing the understanding of reactions at a fundamental level.

- [1] J. Neumann, M. Petranikova, M. Meeus, J.D. Gamarra, R. Younesi, M. Winter, S. Nowak, (2022): Recycling of Lithium-Ion Batteries—Current State of the Art, Circular Economy, and Next Generation Recycling, *Advanced Energy Materials*, 12, 1-26.
- [2] B. Li, Q. Li, Q. Wang, X. Yan, M. Shi, C. Wu, (2022): Deep eutectic solvent for spent lithium-ion battery recycling: comparison with inorganic acid leaching, *Physical Chemistry Chemical Physics*, 24, 19029-19051.
- [3] N. Peeters, K. Binnemans, S. Riaño, (2020): Solvometallurgical recovery of cobalt from lithium-ion battery cathode materials using deep-eutectic solvents, *Green Chemistry*, 22, 4210-4222.
- [4] C. Ma, J.D. Gamarra, R. Younesi, K. Forsberg, M. Svärd, (2023), Antisolvent crystallization from deep eutectic solvent leachates of  $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$  for recycling and direct synthesis of battery cathodes, *Resources, Conservation and Recycling*, 198, 107210.
- [5] S. Mohammadnejad, J. L., Provis, & J. S. J. van Deventer, (2017). Computational modelling of interactions between gold complexes and silicates. *Computational and Theoretical Chemistry*, 110, 113–121.