Selective Recovery of Cobalt from Spent Lithium-Ion Batteries using Green Deep Eutectic Solvent

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The rapid growth in lithium-ion batteries (LIBs) demand for various emerging applications, such as electric vehicles and energy storage systems, will result in waste and disposal problems in the next few years as these batteries reach end-of-life (EoL). The spent LIBs also include 5−20 wt % cobalt (Co), 1−7 wt % lithium (Li), 5−15 wt % nickel (Ni) and 10-15 wt % manganese (Mn), the composition varying slightly with different manufacturers (Shin et al., 2005). Co is considered a rare and strategic metal as it is relatively expensive and the natural resources are primarily limited to the Democratic Republic of the Congo and Zambia. There is a unique opportunity to utilize these EoL batteries as a secondary source to recover this valuable metal. Over the years, various recycling processes, including pyrometallurgy, hydrometallurgy, and bioleaching, have been developed to effectively recover metals from spent batteries. The hydrometallurgy method has been considered as a facile, safe, and efficient process to extract and recover valuable metals from spent LIBs. However, conventional inorganic acids (HCl, H_2SO_4 and HNO_3) as leaching reagents are not considered environment-friendly. The current focus is to use greener solvents for metal recovery in order to reduce secondary pollution and negative impact on the environment.

Deep eutectic solvents (DES) have emerged as green solvents for metal recovery from spent LIBs/waste material. DES mixtures of choline chloride and ethylene glycol (ChCl : EG) and ChCl : urea have been used for Co recovery from spent LIBs. Although good extraction efficiencies (> 90%) were achieved, the process temperature and time were very high (220 ºC, 60 h and 180 ºC 30 h, respectively) (Tran et al., 2019; Wang et al., 2020). In a recent study, Peeters et al. showed that the presence of reducing agents in DES mixture of ChCl : citric acid increased the extraction of Co by reducing Co(III) into Co(II), which is more water-soluble. A higher extraction of 98% was achieved at moderate process conditions of 40 ºC and 1h (Peeters et al., 2020). In this context, ascorbic acid (a naturally occurring organic compound known for its exceptional leaching efficiency) has been used as a reducing agent to assist the leaching process by using various leaching agents, such as sulfuric acid, citric acid and tartaric acid.

The present study will therefore focus on developing a closed-loop recycling process for the recovery of critically-rare metals from spent lithium-ion batteries using a greener approach. The research will provide a comparative study for Co metal extraction from spent LIBs using different DES mixtures of ChCl : EG, ChCl : urea and ChCl : citric acid. The effect of various operating parameters such as temperature, pH, liquid-solid ratio and reaction time will be optimized using response surface methodology (RSM). An insight into the kinetic modelling of the DES-assisted extraction process will also be provided. Various physico-chemical characterization techniques such as XPS, XRD and UV-Vis spectroscopy will be used to investigate the plausible mechanism for the extraction process. The effect of ascorbic acid as a reducing agent on the extraction of Co from spent LIBs effect will also be explicitly discussed. Such research will pave way for replacing the traditionally used acid leaching extraction processes with a viable greener process using a suitable DES solution.

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