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Cradle-to-cradle recycling of spent NMC batteries with emphasis on novel Co^{2+}/Ni^{2+} separation from HCl leached solution and synthesis of new ternary precursor

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ABSTRACT

Herein, a cradle-to-cradle recycling of post-consumer NMC batteries has been demonstrated with a focused separation of cobalt over nickel by applying the ionic liquid, trihexyl(tetradecyl)phosphonium bis-2,4,4-(trimethylpentyl)phosphinate. At the optimized condition of: 0.8 mol·L^{-1} ionic liquid, 3.0 mol·L^{-1} Cl^- ions, equilibrium pH value ~5.0, and organic-to-aqueous ratio 2/3, > 99% cobalt was extracted in the organic phase with a separation factor of 1097. Approximately 99% cobalt from the loaded organic was stripped back in 2.0 mol·L^{-1} HCl solution, yielding high-pure $CoCl_2 \cdot xH_2O$ crystals after crystallizing the stripped solution. Subsequently, ~99% of nickel from the Co-depleted raffinate was extracted over lithium using 0.32 mol·L^{-1} acetophenone at an organic-to-aqueous ratio of 1 and equilibrium pH ~5.3. Nickel stripped in 2.0 mol·L^{-1} H_2SO_4 was crystallized to yield high-pure $NiSO_4 \cdot 6H_2O$ crystals. Further, Li-bearing raffinate was subjected to carbonate precipitation at a higher pH (~12) and CO_3^{2-}:Li^+ ratio of 1.2. All the recycled products were further employed to the stepwise synthesis of a new ternary precursor, exhibiting similar electrochemical behaviour (with 149 mAh·g^{-1} capacity) and found compatible with the precursor prepared using virgin materials. The sustainable process index value determined to be 0.0006 cap·t^{-1} for the overall recycling process indicated that the process is suitable for sustainable development.

1. Introduction

The electric vehicles (EVs) and portable electronic devices with long durability are currently the main niches of application of lithium-ion batteries (LIBs) (Mayyas et al., 2018; Takahashi et al., 2020; Zhang et al., 2022). As an estimation, the global LIBs manufacturing capacity has increased by four to six folds by 2021–2022 compared to the 2017 level (Sommerville et al., 2020; Tsiropoulos et al., 2018). In order to achieve the industrial development with carbon neutrality by 2050 (Fei et al., 2022), the shifting of major economies towards EVs has projected about 56% annual growth of this industry by 2025 and 24.4 million EVs on road by 2030 (Anon, 2020), thereby resulting in increased LIBs consumption over 700 GWh (Natarajan and Aravindan, 2018; Porvali et al., 2019; Anon, 2018). A similar growth trend has been projected by Taskforce 40 on Critical Raw Materials for EVs, assuming a 30% annual growth worldwide (Anon, 2020). Despite the aforementioned estimations that do vary, all are depicting an upward trajectory. Therefore, the need for safe disposal of post-consumer spent LIBs are more apparent and become a determining factor for the sustainable economic growth of society (Chabhadiya et al., 2021; Nayaka et al., 2018; Shaw-Stewart et al., 2019). Notably, many of the substances in LIBs chemistries are toxic, flammable, and irritant, causing the leakage of dozens of toxic gases (that includes CO) potentially fatal to human health (Loughran, 2016). Due to these characteristics, the post-consumer LIBs eventually meet the definition of hazardous waste under the Resource Conservation and Recovery Act (RCRA) (Anon, 2022). Moreover, the uncertainty on whether the LIBs were (partially/fully) charged/discharged before their disposal, make them consider to be the "waste" under "the federal universal waste" regulation in Title 40 of the Code of Federal Regulations part 273 (Anon, 2016).

Besides the issues related to hazardous waste management, a

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