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Mechanochemical-Assisted Leaching of Lamp Phosphors: A Green Engineering Approach for Rare-Earth Recovery

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ABSTRACT

Rare-earth elements (REEs) are essential metals for the design and development of sustainable energy applications. Recycling these elements from waste streams enriched in them is crucial for securing an independent future supply for sustainable applications. This study compares the mechanisms of mechanical activation prior to a hydrometallurgical acid-leaching process and a solvo-metallurgical mechanochemical leaching process for the recovery of REEs from green lamp phosphor, $LaPO_4:Ce^{3+},Tb^{3+}$. The REE leaching rates showed a significant enhancement of 60% after mechanical activation, and 98% after the combined mechanochemical leaching process. High-resolution transmission electron microscopy (HR-TEM) imaging disclosed the cause for the improved REE leaching rates: The improved leaching and leaching patterns could be attributed to changes in the crystal morphology from monocrystalline to polycrystalline. Reduction of the crystallite size to the nanoscale in a polycrystalline material creates irregular packing of chemical units, resulting in an increase in defect-rich grain boundaries in the crystals, which enhances the leaching process. A solvo-metallurgical method was developed to combine the mechanical activation and leaching process into a single step, which is beneficial for operational cost. This results in an efficient and simple process that provides an alternative and greener recycling route for lamp phosphor waste.

1. Introduction

Rare-earth elements (REEs) are essential metals for the design and development of sustainable energy-related applications such as renewable energy technologies (e.g., solar, wind, and thermoelectric converters), lighting, and magnetic materials [1,2]. Recycling critical REEs from end-of-life consumer goods (e.g., permanent magnets, lamp phosphors, and Ni-metal hydride (MH) batteries) and recovering these elements from industrial residues (e.g., bauxite residue and phosphogypsum) are the most prominent pathways to ensure an independent supply for future applications, aside from primary mining [3–5]. Recycling has major advantages over primary supply, including a smaller environmental footprint, shorter lead times, and cheaper sources of material [6]. Moreover, recycling may provide a solution for the over-supply of "unwanted" REEs, and it can contribute to a steady supply of the more critical REEs (neodymium (Nd), dysprosium (Dy), and terbium (Tb)), thus mitigating the so-called "balance problem" [7].

Several studies have been conducted on recycling REEs from waste, with a particular focus on permanent magnets and lamp phosphors [4,8–14]. Phosphors in compact fluorescent lamps (CFLs) are a rich source of one of the most critical and highly valued elements, Tb, which is retained in the green phosphor ($LaPO_4:Ce^{3+},Tb^{3+}$ or $CeMgAl_{11}O_{19}:Tb^{3+}$) [15]. Hydrometallurgy is a traditionally established and easy method for the recovery of valuable metals. $LaPO_4:Ce^{3+},Tb^{3+}$ can be dissolved using hot concentrated sulfuric acid (H_2SO_4) at temperatures ranging from 120 to 230 °C [16]. Pyrometallurgical methods that apply strongly alkaline conditions (35 wt% NaOH, 150 °C) in an autoclave or that involve the presence of molten alkali (e.g., Na_2CO_3 at 1000 °C or $Ba(OH)_2$ at 950 °C) are used to transform rare-earth phosphates into oxides, which can be leached under mild conditions [4,13,14,16,17]. However, the use of an alkali flux leads to the

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