

RECOVERY OF RARE EARTH ELEMENTS FROM FLUORESCENT LAMP PHOSPHORS

C Dorfling
Stellenbosch University, South Africa

KEY FINDINGS

The project investigated the recovery of rare earth elements (REEs) from phosphor powder recovered from end-of-life fluorescent lamps using a hydrometallurgical process route. A multiple leaching stage approach was tested for metal dissolution, where after solvent extraction tests were performed for metal recovery from the pregnant leach solutions. The proposed process yields Y_2O_3 and mixed rare earth oxide (Y_2O_3 , Eu_2O_3 , Tb_2O_3 , CeO_2) product streams at an overall REE recovery exceeding 90%.

INTRODUCTION

Rare earth elements (REEs) are strategically important resources given their widespread use in manufacturing of high technology equipment and consumer goods. Recovery of REEs from secondary resources will play an integral part in the REEs market. Manufacturers of fluorescent lamps consume a large portion of the world's Europium (Eu), Terbium (Tb) and Yttrium (Y) production; recycling of these lamps is important to maintain economic balance, and creates the opportunity for local production of REEs and its associated products.

South African lamp recyclers focus on dismantling and physical separation of the different components (e.g. glass, end caps and fluorescent powders), but very limited processing capacity exists for complete value recovery from the phosphor powders. This project identified technologies / processes potentially suitable for metal recovery from fluorescent lamp phosphors, and experimentally evaluated the performance of the respective unit operations in an integrated manner; a conceptual flow sheet yielding various rare earth oxide product streams at an overall recovery exceeding 90% was proposed. The project has led to significant capacity development in the research field related to REE recovery from electronic waste as well as treatment of end-of-life fluorescent lamps.

APPROACH

Based on a literature review of existing processes and unit operations, a conceptual flow sheet for REE recovery from lamp phosphors was proposed (Figure 1). Bench-scale tests were performed to evaluate the effect of key process variables on the behaviour of REEs in the process and the performance of the respective key unit operations, namely first stage leaching, first stage solvent extraction, alkali fusion, second stage leaching, and second stage solvent extraction.

Leaching

Waste fluorescent phosphor powder was obtained from a lamp recycling and mechanical processing facility in South Africa. The first leaching stage was aimed at dissolution of Y and Eu, which are present in the readily soluble red phosphors ($Y_2O_3:Eu^{3+}$); sulfuric acid was preferred as lixiviant because of the expected suppression of calcium dissolution. The optimal conditions for the first stage leach were determined to be 2 M H_2SO_4 at 25°C and a solid-to-liquid ratio of 10 % (w/v). At these conditions, 98% Y and 89% Eu dissolution with insignificant Ce and Tb leaching were achieved after 6 hours.

The solid residue recovered from the first leaching step was subjected to alkali fusion: the leach residue and sodium hydroxide were mixed in the ratio 1:1.5 (mass basis), and sintering performed in a muffle furnace at a temperature of $800 \pm 3^\circ C$ for 120 minutes. The washed and dried alkali fusion product was pulverized before being leached; at the optimal conditions of 5 M HCl at 60°C and a solid-to-liquid ratio of 5 % (w/v), more than 96% Ce and 98% Tb leaching were achieved after 45 minutes.

Solvent extraction

Solvent extraction tests showed that DEHPA could be used to recover rare earths from aqueous solutions obtained after the first and second leaching stages. In order to achieve more than 95% Y recovery from the first stage sulphuric acid leach solution, 11 extraction stages are required when operating at a pH of -0.25, O/A ratio of 1 and a temperature of 25°C. Eu and residual Y could only be extracted after pH adjustment to 0.5; more than 92% Eu and 99% Y extraction was achieved at this pH using an O/A ratio of 1.5 and 1 M extractant concentration. A multiple stage stripping process can then be used to recover more than 95% of the rare earths from the organic phase prior to precipitation and calcination.

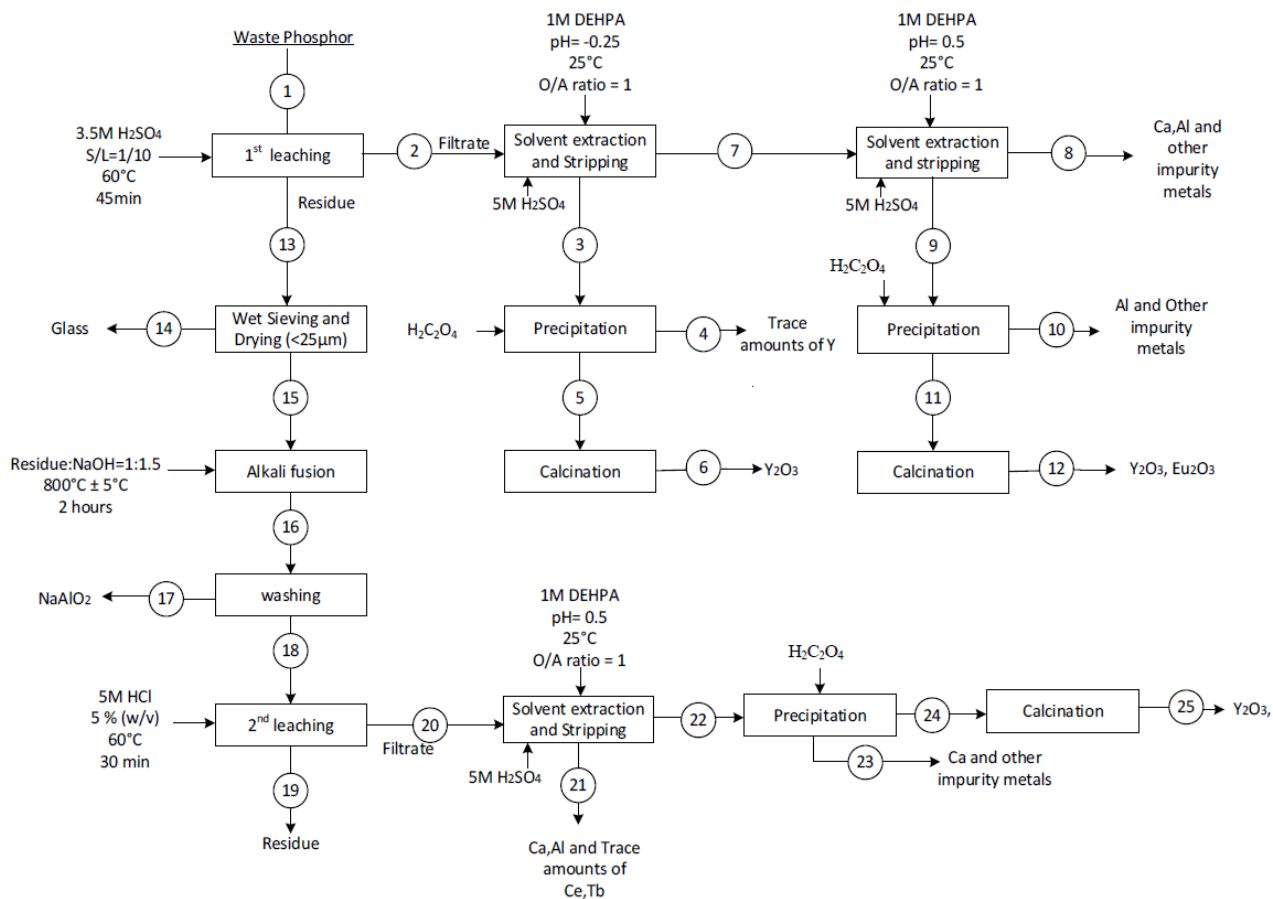


Figure 1. Conceptual flow sheet for rare earth element recovery from fluorescent lamp phosphors.

For the second stage hydrochloric acid leach solution, Ce and Tb solvent extraction results showed that all the targeted rare earths could only be recovered as a mixed REE product at the range of conditions investigated in this study. A mixed rare earth product was produced using 1 M DEHPA, a pH of 0.5, O/A ratio of 1 and a temperature of 25°C. A single extraction stage was sufficient to extract all Y and Eu as well as more than 95% and 98% Ce and Tb, respectively.

CONCLUDING REMARKS

The project has contributed significantly to capacity development in the field of REE recovery and phosphor powder treatment. The results highlighted key fundamental and technical aspects of the REE recovery process that require further investigation. For example, costs associated with solvent extraction steps are high and renders the proposed process uneconomical; a

current final year research project investigates the use of precipitation instead of solvent extraction to produce mixed REE products from the pregnant leach solution. The results from these projects can be combined in a more detailed economic analysis and environmental impact study to move towards a sustainable processing solution.

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