

# RECOVERY OF RARE EARTHS FROM E-WASTE RESIDUES BY AN INTEGRATED APPROACH

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## ABSTRACT

The objective of the REMAGHIC project is the recovery of rare-earth elements (REEs) and magnesium (Mg) from several waste streams to produce new REE-Mg alloys of high performance and low costs. This poster presents the selection procedure of the REE-containing waste residues, the characterization results and the general description of the REEs recovery processes. TOPSIS and LCA methodology, which were used as tools for the identification of the best process routes, are introduced as well.

## INTRODUCTION

This work describes the first outcomes from the research activities performed within the REMAGHIC project, focused on the recovery of REEs and Mg from secondary sources to produce new REE-Mg alloys with improved mechanical properties, reduced weight and low cost which can find application in the automotive, aeronautic and biomedical industries.<sup>1,2</sup> The aim is to reduce the European dependency of critical raw materials from non-European countries and to partially mitigate the "Balance Problem".<sup>3,4</sup> The selection of the residues was performed by taking into account the REEs concentration and the relevance from the balance problem point of view.<sup>5-8</sup> The residues were then characterized by ICP/OES, SEM and XRD to gather information about their chemical and mineralogical composition (Tables 1-3). For each waste stream, combinations of several processes (pyrometallurgical, hydrometallurgical and iono/solvometallurgical processes) have been applied. The balance between the resources needed and the emissions to the environment was performed based on the Life Cycle Assessment (LCA). TOPSIS methodology (Technique for Order of Preference by Similarity to Ideal Solution) was then applied as decision making method to support the selection. The criteria were defined according to seven Key Performance Indicators (KPI): (1) TRL [1-9], (2) Process efficiency [%]; (3) Waste generation [kg]; (4) Toxicity (CTUe)], (5) Global Warming Potential [CO<sub>2</sub>e]; (6) Material cost [€]; (7) Income [€].

## **RESIDUES CHARACTERIZATION**

Table 1. Lamp phosphors composition, wt.%

Si	Ca	Y	La	Се	Eu	Tb	Gd
18.9	7.1	6.3	0.9	0.7	0.4	0.3	0.1





Table 2. CRT phosphors composition, wt.%								
Zn	S	Y	Si	Al	Pb	Eu		
30.6	16.5	15.5	7.2	2.0	1.7	1.0		

Table 3. NiMH electrode powder composition, wt.%

Ni	La	Се	Со	Zn	Fe	Mn
56.9	12.3	4.0	5.2	1.1	0.5	0.4



**RECOVERY ROUTES** 

### LAMP PHOSPHORS

REEs recovery (*Figure 1*) was performed by a leaching step with  $H_2SO_4$  followed by precipitation with oxalic acid in the HydroWEEE process.<sup>9,10</sup> Y/Eu separation from the obtained mixed oxide was then investigated by leaching and solvent extraction with a phosphonium ionic liquid. Tb, La and Ce recovery from the residue was enhanced by applying a pyro-treatment before leaching (roasting in the presence of  $Na_2CO_3$ ). The obtained leachate was then subjected to a solvent extraction step with D2EHPA. An alternative approach is the leaching of the Tb-rich residue with organic solvents; this is the so-called *solvometallurgical leaching*, where metal recovery is performed using non-aqueous solutions.<sup>11</sup>



Figure 2. REEs recovery flow-sheet from NiMH electrode powder

#### **CRT PHOSPHORS**



A pyrometallurgical treatment was investigated for the recovery of La and Ce from NiMH batteries (*Figure 2*). This pretreatment allowed obtaining a metal phase rich in nickel and other minor metallic compounds, whilst the REEs are concentrated in the slag phase as oxides. After leaching with mineral acids, REEs are recovered by solvent extraction with ionic liquids ("split-ion extraction")<sup>12</sup>. La and Ce recovery was also investigated by direct leaching of the black electrode powder with hydrochloric acid; REEs recovery from the leachate was performed by split anion extraction as well.





A thermal pretreatment was investigated to convert the ZnS to ZnO, thus avoiding  $H_2S$  release during the leaching step (*Figure 3*). Zinc oxide was then selectively removed from the roasted phosphors by leaching with acetic acid (AcOH), giving access to the REE content. Y and Eu recovery from the residue was then studied by leaching with methanesulfonic acid (MsOH), followed by precipitation with oxalic acid.

### CONCLUSIONS



Yttrium, lanthanum and cerium recovery from a selection of three waste residues (lamp phosphors, CRT phosphors and NiMH batteries electrode powder) was investigated by a combination of different techniques within the REMAGHIC project (pyrometallurgical, hydrometallurgical and iono/solvometallurgical processes). The selection of the best process routes was performed by employing TOPSIS and LCA methodology. The recovered rare-earths can find new applications in REE-Mg alloys, thus offering a unique opportunity to partially mitigate the Balance Problem.

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