INTEGRATED FLOW-SHEET FOR THE RECOVERY OF RARE EARTHS FROM CRTs AND LAMP PHOSPHOR WASTE

F. Forte[1], L. Yurramendi[2], J.L. Aldana[2], C. Del Rio[2], R. Banda[2], L. Gijsemans[1] and K. Binnemans[1]

1 KU Leuven, Department of Chemistry, Celestijnenlaan 200 F, Heverlee, Belgium

2 TECNALIA, Parque Tecnológico de San Sebastián, Mikeletegi Pasealekua 2, San Sebastián, Spain

ABSTRACT

In this work REEs recovery from CRTs and lamp phosphor waste was performed through a combination of pyro, hydro and solvometallurgical techniques.

By roasting, the ZnS present in the CRTs is transformed into ZnO which was removed by leaching with acetic acid. Y and Eu recovery from the residue was then investigated by leaching with methanesulfonic acid. REEs recovery from lamp phosphor waste was performed through an integrated process based on roasting the Tb-rich residue (produced after a first leaching step aimed at dissolving Y) with Na_2CO_3; this pretreatment enhanced Tb, La and Ce leaching efficiency. REEs recovery from the leachate was then performed by solvent extraction with D2EHPA in xylene. An alternative approach is the solvometallurgical leaching of the Tb-rich residue with methanesulfonic acid.

INTRODUCTION

The results of this work are part of the experimental activity carried out within the REMAGHIC PROJECT, focused on REEs and Mg recovery from secondary sources to produce new REE-Mg alloys.[1,2,3] The aim is to reduce the European dependency of critical raw materials from non-European countries and to partially mitigate the "balance problem".[4,5] REEs are present in the CRT phosphors as yttrium europium oxysulphides ($Y_2O_2S:Eu^{3+}$) whilst zinc as zinc sulfide (ZnS). By roasting ZnS is transformed into ZnO, thus avoiding H_2S release during leaching.[6] Tb, La and Ce recovery from lamp phosphors was investigated as well. This step is challenging because it involves the attack of the very resistant phosphate and aluminate phosphors.[7] REEs recovery can be enhanced by applying a pyro-treatment before leaching. An alternative approached is the leaching with organic solvents: this is the so-called solvometallurgical leaching, where metal recovery is performed using non-aqueous solutions.[8]

MATERIALS AND METHODS

CRT and lamp phosphors were characterized by ICP-OES, SEM/EDS and XRD. Roasting of the CRT powder was performed at different temperature values (800 to 1000ºC) and residence times.

Acetic acid leaching tests were then performed as a function of several operative parameters. The residue was finally leached with methanesulfonic acid (t=24h, T=90˚C). REEs leaching from the lamp phosphors was performed with H_2SO_4 and HCl. The Tb-rich residue was then roasted in the presence of Na_2CO_3 and subjected to a second leaching step with mineral acids. In the solvometallurgical process the Tb-rich residue was leached with methanesulfonic acid at T=200˚C. REEs recovery from the leachate was then investigated through solvent extraction with D2EHPA.

RESULTS AND DISCUSSION

CRT PHOSPHORS TREATMENT

$$ZnS + 3/2\ O_2 \rightarrow ZnO + SO_2$$

By roasting ZnS is transformed into ZnO *(Figure 1)*

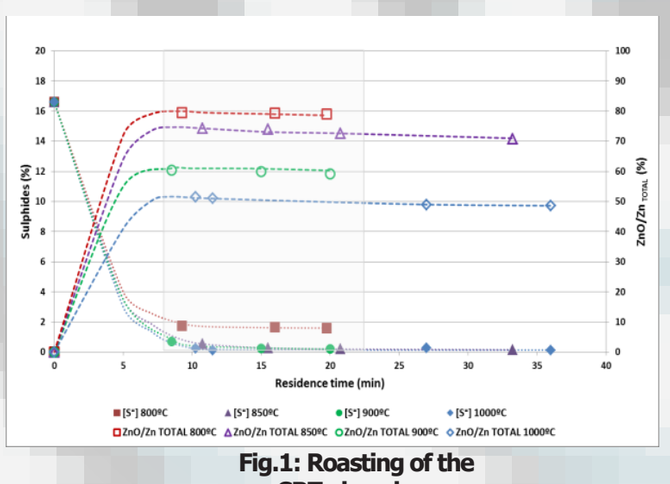

Fig.1: Roasting of the CRT phosphors

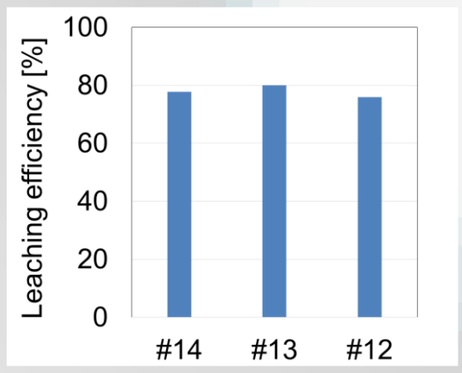

Fig.2: Zn leaching with acetic acid

75-80% zinc can be removed from the roasted phosphors by leaching with acetic acid *(Figure 2)*.

The leaching residue can be treated with methanesulfonic acid for Y and Eu recovery.

LAMP PHOSPHORS TREATMENT

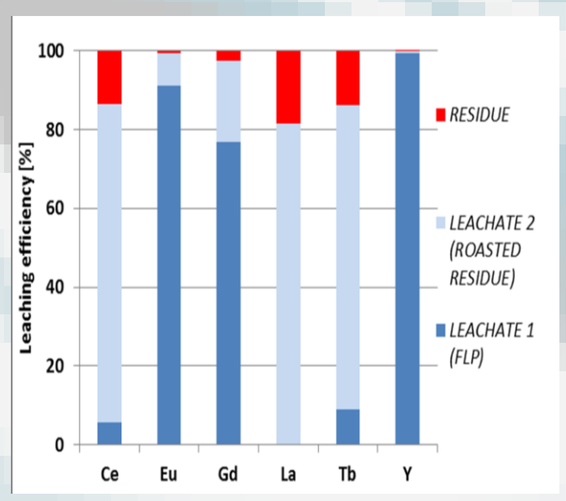

Fig.3: Leaching of the lamp phosphors and the roasted residue

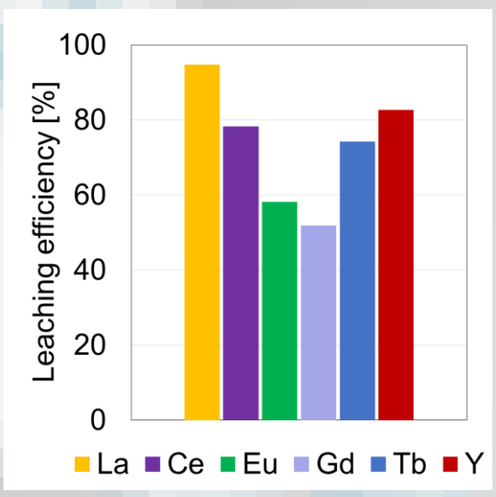

Fig.4: Solvent leaching of the Tb-rich residue

Tb, La and Ce can be dissolved by applying a roasting step prior to leaching *(Figure 3)*. Alternatively they can be leached with strong organic acids, such as methanesulfonic acid *(Figure 4)*.

CONCLUSIONS

- by roasting at 850 ˚C more than 70% of ZnS in the CRT phosphors is converted into ZnO which was then removed by acetic acid leaching
- Y and Eu recovery from the residue is achieved by methanesulfonic acid leaching
- Tb, La and Ce leaching from the lamp phosphor is enhanced if a roasting step with Na_2CO_3 is applied beforehand
- leaching with methanesulfonic acid allows high REEs leaching efficiency from the Tb-rich residue

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