



E-waste mining and the transition toward a bio-based economy: The case of lamp phosphor powder

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ABSTRACT

Replacement of conventional hydrometallurgical and pyrometallurgical process used in E-waste recycling to recover metals can be possible.

The metallurgical industry has been considered biohydrometallurgical-based technologies for E-waste recycling. Biorecovery of critical metals from phosphor powder from spent lamps is an example of transition to a bio-based circular economy.

E-waste contains economically significant levels of precious, critical metals and rare-earth elements (REE), apart from base metals and other toxic compounds. Recycling and recovery of critical elements from E-waste using a cost-effective technology are now among the top priorities in metallurgy due to the rapid depletion of their natural resources. This paper focuses on the perceptions of recovery of REE from phosphor powder from spent fluorescent lamps regarding a possible transition toward a bio-based economy. An overview of the worldwide E-waste and REE is also demonstrated to reinforce the arguments for the importance of E-waste as a secondary source of some critical metals. Based on the use of bio-processes, we argue that the replacement of conventional steps used in E-waste recycling by bio-based technological processes can be possible. The bio-recycling of E-waste follows a typical sequence of industrial processes intensely used in classic pyro- and hydrometallurgy with the addition of bio-hydrometallurgical processes such as bioleaching and biosorption. We use the case study of REE biosorption as a new technology based on biological principles to exemplify the potential of urban biomining. The perspective of transition between conventional processes for the recovery of valuable metals for biohydrometallurgy defines which issues related to urban mining can influence the mineral bioeconomy. This assessment is necessary to outline future directions for sustainable recycling development to achieve United Nations Sustainable Development Goals.

Keywords absorption · circular economy · critical materials · waste management · recycling · recovery

Discussion

- In the coming years, both efficient legislation on the E-waste management and the intensification of measures to reduce dependence on rare-earth elements imports will be essential to obtain autonomy over the critical raw material essential for economic and technological development. To this end, it implies measures to diversify the supply of primary and secondary sources through sustainable and efficient processes, which must be based on clean and green technologies, especially bio-based technologies.
- In recent months, the most powerful countries have established an ambitious plan to recover from the pandemic and increase strategic autonomy, which will promote the transition to an ecological economy where the bioeconomy will play a fundamental role in meeting most industrial demands through the implementation of bioprocesses, even in urban mining to reduce dependence on imported mineral commodities.

Introduction

Almost 53.6 million tons of waste of electric and electronic equipment (WEEE or E-waste) was produced in 2019. This fastest-growing fraction of municipal waste has a value estimated to be US\$ 57 billion due to the presence of metals with known economic value such as copper, nickel, iron, aluminum, and zinc; as well as precious metals like silver, gold, platinum group metals (PGM), and rare-earth elements (REE).¹⁻⁴

REE group consists of seventeen elements and is divided into two categories, namely the light rare-earths (LREE), including cerium (Ce), lanthanum (La), neodymium (Nd), praseodymium (Pr), samarium (Sm); and the heavy rare-earths (HREE) group including gadolinium (Gd), europium (Eu), terbium (Tb), dysprosium (Dy), thulium (Tm), ytterbium (Yb), lutetium (Lu), yttrium (Y), holmium (Ho), and erbium (Er).⁵

The five most common REE phosphors in fluorescent lamps are $Y_2O_3:Eu^{3+}$ (red phosphor) $BaMgAl_{10}O_{17}:Eu^{2+}$; (Sr,Ca,Ba)₅(PO₄)₃Cl: Eu²; LaPO₄:Ce³⁺, Tb³⁺; GdMgB₅O₁₀:Ce³⁺, Tb³⁺; (Ce,Tb)MgAl₁₁O₁₉ and (Sr,Ca,Ba,Mg)₅(PO₄)₃Cl:Eu²⁺. The oldest LF may still be composed of a strontium calcium halophosphate fraction ((Sr, Ca)₁₀(PO₄)₆(C1,F)₂: Sb³⁺, Mn²⁺).⁶ Red phosphor has the highest intrinsic value among phosphors for recyclers because it contains higher concentrations of Y and Eu and is often presented in higher proportions in recycled phosphorus fractions (up to about 20% by weight).⁷

Post-consumer fluorescent lamps are not reused. Generally, the recycling process starts with the receiving stage, where the lamps are unloaded and stored. Then, the lamps are sorted and classified according to size, type, and model. Lamps are also separated according to their integrity, considering whether the material is whole or broken. Only phosphor powder has not been adequately recycled;⁶ and some pyro- and hydrometallurgical processes have been used for this purpose.⁸⁻¹⁰

In recent years, the use of bio-based methods replacing traditional extraction has been gained attention.^{8,11,12} However, due to the scarcity of primary resources and the growing need to extract metallic values from complex mineral phases, low-grade reserves, or secondary sources such as solid waste and effluents, microbial activity-based processes are emerging as emerging technologies for the supply of strategic metals and minerals.^{13,14}

Methodology

Our conceptual approach involved the critical literature of global policy documents related to E-waste generation, the raw materials initiative focusing on REE and EOL fluorescent lamps recycling. We reviewed the existing academic literature and other studies that review REE recycling technologies using pyro-, hydro-, and bio-hydrometallurgical methods and online articles and market reports. The complete list of references is provided at the end of the article. Our case study provides an analysis of the generation of global E-waste and its potential as REE secondary sources, emphasizing the use of bio-based technologies to recover REE from phosphor powder from spent fluorescent lamps.

Rare-earth elements: Critical raw materials in E-waste

REE has been referred to as the key to the "Industry 4.0" revolution and has currently become very critical to several modern technologies ranging from cell phones and televisions to lightemitting diodes (LED) lamps, wind turbines, and electric car batteries.^{5,15} The global demand for the REE market was valued at over USD 8.8 billion in 2019 and is expected to reach above USD 15.7 billion in 2026, anticipated to grow at a compound annual growth rate (CAGR) of above 8.6% between 2020 and 2026.¹⁶ China (61.2%), United States (US) (12.2%), Myanmar (10.3%), and Australia (9.9%) are the leading producers of REE.¹⁷ Asia's market dominance has greatly increased the REE prices, causing tension and uncertainty among the world's hi-tech markets. Also, REE natural reserves are concentrated in a small number of countries (China, Brazil, Vietnam, Russia, India, Australia, Greenland, and the US).¹⁸

In 2008, the United Nations Environment Program (UNEP) identified REE as critical raw materials (CRM) in a specific list where global risks were prioritized according to the growth in demand, supply risks, and restrictions on recycling.¹⁹ Since 2011, the European Commission placed REE in the European list of CRM due to economical importance and low availability. In 2020, LREE and HREE appeared on the list with other CRM such as cobalt, niobium, and PGM.²⁰

In 2018, the US also added REE to its list of 35 CRM.²¹ For 31 CRM, the US imports more than half of its annual consumption. Thus, from October 2020, during a commercial crisis caused by the COVID-19 pandemic, the US initiated an action plan to reduce its vulnerability to adverse actions by foreign governments, natural disasters, or other disruptions in supply. The order is to develop secure supply chains for critical minerals that do not depend on resources or processing from other countries.²²

Among the global initiatives to decrease the import of REE and its dependence on foreign markets, the investment in extraction and recovery of REE and other CRM from E-waste was initiated in the early 2010s. An example is the initiatives dedicated to mineral raw materials within the framework of the European Research Area (ERA MIN) and Horizon 2020, which are significant financiers of RD&I projects that have sought solutions to the issue of supplying critical mineral raw materials since 2014.²³ These initiatives are aligned with circular economy guidelines based on CRM recovery from secondary sources.^{1,3}

Table 1 shows a parallel between the production of annual REE, annual E-waste, and the estimated amount of REE contained in this waste, based on data collected from 2019.

The US and Australia are the leading world producers of REE after China (132,000 t in 2019).²⁴ However, their demands are not met by their production. The US, for example, imported 14,000 t of REE in 2019. With public policies to reduce dependence on the foreign market and strengthen national production, one of the fronts being evaluated by countries is the extraction of REE from their E-waste.

The largest generators of E-waste in the world are China (10,129 kt), the US (6918 kt), India (3230 kt), Japan (2569 kt)

 Table 1. REE and WEEE production in the 5 countries representing their continents.

Country	REE prod. in 2019 (t) ^a	WEEE prod. in 2019 (kt) ^b	REE content in WEEE (t) ^d	Au content in WEEE (t) ^e
US	26,000	6918	1.52	25.8
Australia	21,000	554	0.12	2.1
India	3000	2230	0.71	12.1
Brazil	1000	2143	0.47	7.99
UE	-	12,000 ^c	2.64	44.78

^aU.S. Geological Survey, Mineral Commodity Summaries, January 2020.²⁴

^bThe Global E-waste Monitor 2020.²

^cEstimated to Europe. The Global E-waste Monitor 2020.²

^dEstimated REE content based on European Commission.⁶³ It was reported that the concentration of REE (Sc, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu) was 220 ± 11 mg/kg in British landfill.

^eEstimated Au content for each country based on The Global E-waste Monitor 2020.² It was reported that the concentration of Au was 0.2 kt in 53.6 Mt of global E-waste produced.

and Brazil (2143 kt). REE recycling is estimated at < 1% worldwide.^{25,26} One of the main obstacles to recycling these elements is that REE in EOL products varies from < mg to several kg.²⁷

Iron (20,466 kt), aluminum (3046 kt), and copper (1808 kt) represent the majority of the total weight of raw waste materials that can be found in E-waste in 2019.² We have estimated the REE contents and have compared them to gold, the precious primary metal found in E-waste. The quantities of REE that can be mined from E-waste as a secondary source are low but are highly justified by high demand and a lack of reserves.

Fluorescent lamp powder: Is it an obsolete treasure?

From total E-waste produced in 2019, about 0.9 Mt corresponded to lamps, an increment of 4% from 2018.² These data corresponded to fluorescent lamps (FL), high-intensity discharge (HID) lamps, and LED lamps. Generally, FL are composed by glass (88%), metals (5%), plastics (4%), powders (3%), and mercury (0.005%).²⁸ The composition of phosphor powder consists in halophosphate (45%), glass (20%), REE (20%), alumina (12%), and others (5%).^{6,28}

REE-based phosphor powder uses varying amounts of REE, resulting in various powder compositions, as shown in Table 2.

Primarily phosphor powder contains some proportion of Y, Eu, and Tb to generate red, green, and blue phosphors.

For example, there are five main REE phosphors found in FL: the red phosphor Y_2O_3 : Eu^{3+} (YOX), the green phosphor CeMgAl₁₀O₁₇: Tb³⁺ (CMAT) or LaPO₄: Ce³⁺, Tb³⁺ (LAP), and the blue phosphor BaMgAl₁₁O₁₇: Eu²⁺ (BAM).

Each company manufactures the lamp with a characteristic composition. For example, Toshiba brand commercial phosphor materials (SPD series) have a mixture of 85% white (halophosphates), 5% red, 5% green, and 5% blue phosphors (17:1:1:1 mixture ratio).²⁹ The YOX has the highest intrinsic value because it contains large concentrations of Y and Eu (up to about 20 wt%), whereas in lamps with trichromatic phosphors, the concentration of REO can be as high as 27.9 wt%.²⁷

The REE contents in LF phosphor powder are over 23% (230 kg/t),³⁰ corresponding to 15 times the content found in ores considered REE primary sources. The fact that E-waste has higher concentrations of metals than primary sources is one of the advantages that makes it a potential secondary source in the supply chain within the concept of circular mineral economy and urban mining.³

The phosphor powder, which exists at approximately 2–5% of the total FL weight, has had its neglected recycling, and few projects have left the laboratory scale because no practical way to manage them has been established, although the tiny particles of phosphor may pose a risk to human health.³¹

Solvay was the first company to develop an industrialized spent FL recycling process for REE marketing.²⁸ The process consisted of removing the mercury, glass, and other components to physically liberate REE concentrate halophosphate free, then sent to the solvent extraction process for the individual REE chemical separation. The final yield of REE was at about 80%.³² Solvay constructed a demonstrative plant to process 3000 tons of spent FL/year,^{28,33} resulting in 90% waste stream valorization corresponding to 10-20% of REO besides glass and phosphate as by-products.

In 2011, China was the largest producer of LF and REE phosphors globally, and approximately 7 billion LF units were produced.³⁴ For this purpose, in 2011, 8000 tons of REE phosphors were produced,³⁴ which meant more than 80% of the global production of these strategic metals,^{35,36}

In 2013, REE phosphors were the second application market for REE (USD 616 million), representing 20% of the global REE market (USD 3 billion).³⁷ The illuminating industry used almost 95% of manufactured phosphors. China is the largest producer and exporter in the REE phosphor industry with the raw material advantage. In 2015, Chinese production accounted for 63.62% of the global REE phosphor market, while Japan, the secondlargest producer of REE phosphors, had a 23.74% share.³⁸

In 2016, the demand for FL was still high, although the LED segment started to gain a significant market share (35%) due to technological and economic advances, such as high efficiency and high brightness. As a result, the segment accounted for USD 5.12 billion in 2016 and was projected to grow with a 6.8% CAGR from 2017 to 2025.³⁹

Eu and Tb prices had felled in 2015, and this could be attributed to the replacement of compact FL lamps with LED lamps
 Table 2.
 Price of REE (2020) and concentrations of REO in different FL phosphor powder samples.

	Price	REO concentration (wt%)							
REO	(USD/ kg) ^a	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
Y ₂ 0 ₃	2.75	6.29	8.1	17.8	26.8	15.5	6.8	26.4	28.0
La ₂ 0 ₃	1.38	0.9	2.1	4.1	ND	ND	0.3	ND	3.0
Ce ₂ O ₃	1.38	0.74	1.0	2.4	2.88	0.7	0.4	3.9	3.6
Eu ₂ 0 ₃	30.53	0.43	0.51	1.1	1.68	0.9	0.4	2.2	2.0
Tb ₂ O ₃	718.48	0.32	0.62		1.86	ND	0.2	2.2	2.1
Gd ₂ 0 ₃	25.91	0.14	ND	ND	ND	ND	0.2	ND	ND

REO rare-earth oxides, ND not detected.

^aRef. 60.

(1) Phosphor powder from RELIGHT, Italy.³¹

(2) Phosphor powder from typical tricolor-type fluorescent lamps after removing mercury supplied by Japan Recycling Light Technology and System, Kitakyushu, Japan.⁶⁴

(3) Phosphor powder from the waste fluorescent lamps recycling plant Itomuka Mining Laboratory, Nomura Kohsan Co., Ltd, Hokkaido, Japan.²⁹

(4) Phosphor powder from EOL compact fluorescent lamps from a local electric warehouse in Uttarakhand, India. 65

(5) Phosphor powder from Baogangxinli RE Co., Ltd, Ganzhou, China.⁷⁴

(6) Phosphor powder from a company located in Sweden.⁶⁶

(7) Phosphor powder from a waste fluorescent lamp, supplied by Zhejiang Chenhui Lighting Co., Ltd., collected from the market.⁶⁷

(8) Phosphor powder from T5 super 80 linear fluorescent lamps collected from the electrical maintenance department of Indian Institute of Technology (IIT) Bhubaneswar and CSIR-Institute of Minerals and Materials Technology (IMMT), Bhubaneswar (India).⁶⁸

that contains mainly Y and Ce as its REE phosphor contents. As a result, LED lamps have been rapidly replacing other lighting technology, and its market was valued at USD 58.91 billion in 2018 and was expected to reach USD 127.97 billion by 2024, with a CAGR of 13.75% from 2020 to 2025.⁴⁰ In Canada, the cumulative waste from LED lamps is expected to reach 12,000 tons in 2021.⁴¹

In 2018, only 7% of global REE production was destined for phosphor powder manufacture, against 21% to permanent magnets, 20% to fuel cracking catalysts, and 18% to alloys.⁴² As a result, the world market for REE phosphor is expected to grow at a negative CAGR of approximately – 1.2% over the next 5 years, reaching USD 390 million in 2024.⁴³

Bio-recycling of phosphor powder: Viability and the relationship to the bio-based economy

Currently, the FL recycling rate for REE recovery is minimal. In pyrometallurgical methods, the REE can be oxidized easily, however, with complicating recovery.⁴⁴ Hydrometallurgy is the process of solubilizing metals by using large quantities of chemical agents (acids). The disadvantages of this method include pretreatment, long time, high operating cost, and large amounts of acidic wastewater effluent.^{45,46}

Biohydrometallurgy is already an established route to process low-grade primary ores of many metals¹⁵ and may play an essential role in the sustainable urban mining of critical raw materials in the future.^{3,7,9,11,12,47} Bioleaching is a bio-hydrometallurgical process performed by different microorganisms (fungus and bacteria) to secrete inorganic or organic acids or cyanide which enhances enzymatic oxidation-reduction, proton-promoted mechanisms, and ligand and complex formation.^{48,49} Europium, for example, was preferably solubilized from LF phosphor powder by acetic and gluconic acid mixture produced by *Zygosaccharomyces lentus* and *Komagataeibacter hansenii* species, a symbiotic mixed culture from Kombucha.⁵⁰

Biosorption is a physicochemical and metabolically-independent process to remove metals selectively that includes adsorption, ion exchange, precipitation, and surface complexation mechanisms on biosorbents.^{11,51-53} In a recent study, the operating cost of a hydrometallurgy plant for the American project Bear Lodge (which includes two REE mines) was estimated at USD 10.78 / kg of REO, while the biosorption process would cost USD 15/kg, with results of comparable quality (95% *vs.* 97% purity).⁵⁴

These bio-hydrometallurgical processes can be called biobased technologies that can be involved in primary source mining and E-waste recycling, as observed in Fig. 1.

Biohydrometallurgy is present in the 2030 Agenda for bioeconomy proposed by the Organization for Economic Cooperation and Development OECD,⁵⁵ with actions for implementing new bioleaching and biosorption industrial plants being conducted in the short term within a mineral bioeconomy agenda. The bioeconomy is based on the innovative use of sustainable biological resources to develop efficient bioprocesses to support sustainable production and promote the integration of biotechnology applications between different sectors.^{56,57}

There is little evidence in the literature on bioprocesses to recycle phosphor powder. Our case study was based on a recent publication where an acid leachate liquor from LF phosphor powder was subjected to the biosorption process using the yeast *Saccharomyces cerevisiae* embedded in a cellulose matrix. The adsorption of 55% Eu from an FL leachate containing 40.2 mg/L of Eu was considered selective due to the presence of high concentrations of other metals as Y, La, Ce, Ca, and Al.⁵⁸

From the data presented in the literature related to the leaching and biosorption processes, 57,59 the costs of the Eu recovery process by *S. cerevisiae*-cellulose were partially estimated, as shown in Table 3.

The integrated hydro-biotechnological process presented an estimated cost of USD 22,861 per kg of Eu recovery from 250 kg



Figure 1. Flowchart of the bio-based process of E-waste disposal.

of LF phosphor powder. However, this price may be lower if we consider the regeneration and reuse of the biosorbent. For example, if the biosorbent material is reused at least four cycles with 95% efficiency, the calculated material cost would decrease to USD 5429.

Considering only the gross calculation of the process just for the recovery of Eu, it seems impracticable that these operations are carried out commercially, as the value of 1 kg of Eu is estimated at USD 30.53.⁶⁰ Furthermore, it is still necessary to account for the costs of desorption from the Eu and regeneration of the biosorbent material.

The biosorption process is only advantageous if the biosorbent material is cheap, presents a high capacity for biosorption $(q_{\max} \operatorname{mg} \operatorname{REE}/\operatorname{g} \operatorname{biosorbent})$, and also a high capacity for reuse. However, the required amount of chemicals and displacement from the entire process were relatively small compared with the other process as ion-exchange resins. In the REE recycling system suggested in this study, large quantities of acid and base substances were unnecessary. This is a significant advantage of the biosorption process.⁶¹

Table 4 shows the maximum adsorbent capacity of different adsorbents and the amount required for 1 kg of Eu recovery described in literature data. The greater the REE's adsorption capacity, the smaller the amount of biosorbent required, which reduces material and process costs, as it requires less liquor volume and operational costs. If the adsorbent is based on industrial waste such as microbial or agro-industrial biomass, the costs will be even lower and make the process feasible.

For example, it was estimated that REE mining from ore costs ~ 0.42 USD/kg (in-situ REO), while the REE concentrate costs ~ 27.6 USD/kg (in-situ REO). The process for recovery REE from LF phosphor proposed by Solvay would cost ~ 6 USD/kg³.⁶² In addition to the few studies that leave the bench environment for industrial plants, it is also necessary to calculate the costs of the economic viability of biotechnological processes, something that is not well described.

Table 3.	Estimated	cost perf	formance o	f Eu rec	overy from	ı LF phos	phor	pow-
der using	acid leachi	ing and t	biosorption	process	es.			

Chemicals	Price of chemicals	Required chemi- cals amount for 1 kg of Eu recovery	Price of chemicals required for 1 kg of Eu recovery (USD) ^h
	Acid le	eaching	
Nitric acid	18 USD/l ^a	51.9 I	
	Biosorbent	preparation	
<i>S. cerevisiae</i> lyophilized	238 USD/kg ^b	24.87 kg	
Cellulose	162 USD/kg ^c	49.75 kg	
Glutaralde- hyde	156 USD/I ^d	49.75 I	
	Bioso	rption	
Biosorbent	174 USD/kg ^e	124.3 kg	21,74
Leachate	0.91 USD/I ^f	1243 1,12	

^aNitric acid 70%, purified by redistillation, \geq 99.999% trace metals basis. Price based on currently available prices from MERCK (official site).

^bPrice based on currently available prices from Amazon (official site).

^cMicrocrystalline powder. Price based on currently available prices from MERCK (official site).

^d50wt% in water. Price based on currently available prices from MERCK (official site).

eEstimated from biosorbent preparation⁵⁹ focus only on the material cost.

^fEstimated from acid leaching process⁵⁸ focus only on the material cost.

^gEstimated from Eu biosorption process described in Ref. 58.

^hEstimated from data obtained from Refs. 58, 59.

Conclusions

Despite the high content of REE in the LF phosphor powder, a group of authors considers that recycling this material is not economically viable. However, the collection and recycling of FL have a high social value in contamination avoided by mercury, which is difficult to quantify in economic terms. Besides, there is the fact that the spent FL are already recycled at least Table 4. Eu uptake capacity $({\tt q}_{max})$ and the required amount of adsorbent for 1 kg of Eu recovery.

Adsorbent	Eu uptake capacity (g/kg)	The required amount of adsor- bent for 1 kg of Eu recovery (kg)
S. cerevisiae	14.2 ⁵⁸	70.42
Cellulose	18.5 ⁵⁸	54.05
M. smegmatis	19.2 ⁶⁹	52.08
Palygorskite	24.3 ⁷⁰	41.15
Chitosan	48.3 ⁷¹	20.70
Fe ₃ O ₄ @MnOx	138.172	7.24
A. acuminatus	174.2 ⁷³	5.74

partially, and adding processing steps to make better use of their phosphoric components can add significant economic value to the process.

It is necessary to reconcile the different costs and benefits of pollution avoided through collection and recycling, in addition to evaluating the different stages and processes required for industrial recycling, be it phosphor powder or other types of E-waste. Despite the current dominance of the LED market in the FL case, it is necessary to end the environmental liability formed by the thousands of FL discarded in the world.

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Declarations

Conflict of interest

The corresponding author states that there is no conflict of interest on behalf of all authors.

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