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Searching patent information on the recovery of rare earth metals from electronic waste

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Abstract

Waste and electrical and electronic equipment (WEEE), which is generally disposed of in landfills, contains valuable elements, called rare earth metals, and they are applicable in many industrial sectors.

Only 1% of the rare earth elements are recycled from the residues mentioned above.

This study aims to show a patent landscape in recovering rare earth metals from end-of-life electronic equipment.

The patented methods were searched on the Orbit Intelligence platform (FamPat database), Espacenet, Google Patents and Octimine, using keywords and classification codes (IPC and CPC).

China is the country with the greatest number of priority patent applications filed in this technical field, followed by the USA and Japan.

The different techniques developed for recovering rare earth metals are hydrometallurgy, pyrometallurgy and biometallurgy or bioleaching. Hybrid treatments have been also patented.

A focus on the biological treatment technologies for recovery of rare earth metals from e-waste was carried out.

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Introduction

Rare earth elements (REEs) are a group of metals that include 17 elements, namely yttrium, scandium, and 15 lanthanides. These elements are generally divided into two categories, based on their electronic configuration: heavy (europium, gadolinium, terbium, dysprosium, holmium, erbium, thallium, ytterbium, lutetium, and yttrium) and light rare earth metals (lanthanum, cerium, praseodymium, neodymium, promethium, and samarium). ^{[\[1\]](#page-14-0)}

The most critical elements regarding supply and industrial use are neodymium, dysprosium (for permanent magnets), europium, terbium, and yttrium (superconductors and lasers).

It would therefore be desirable to intensify the methods and processes for the recovery of rare earth metals from end-oflife products/devices, also in consideration of the significant increase in the amount of electronic waste (WEEE), which will reach 74.4 million tons in 2030.^{[\[2\]](#page-14-1)}

There are three types of recovery adopted, starting from production residues (for example, during the manufacture of NdFeB magnets), from devices deriving from the urban separate collection (computers, telephones,...), or industrial waste.

Most of the rare earth recycling activity currently comes from permanent magnets, from those present in electronic waste, using hydrometallurgical, and pyrometallurgical techniques, biometallurgy, biological treatment technologies (bioleaching, biosorption, phytoremediation, bioelectrochemical systems, or by gas phase extraction. ^{[\[3\]](#page-14-2)[\[4\]](#page-14-3)[\[5\]](#page-14-4)[\[6](#page-14-5)[\]\[7\]](#page-14-6)}

Electronic waste is defined as all those devices (and their components) that have a plug, a cable, or a battery and have reached the end of their life. These residues can be classified as heat exchange equipment (air conditioners, refrigerators, stoves, and electric fans), small (calculators, video cameras, electric razors, coffee machines, etc.), or large devices (washing machines, dishwashers, photocopiers, photovoltaic panels), fluorescent lamps, screens (TVs, monitors, laptops, or tablets) and communication devices (telephones and navigators).

Rare earth metals are present above all in the latter two product categories (a smartphone contains less than a gram) and can be recycled using, for example, an oxidation-reduction protocol for the recovery of neodymium, praseodymium, and dysprosium from permanent magnets. ^{[\[8\]](#page-14-7)[\[9\]](#page-14-8)[\[10\]](#page-14-9)}

What is the technological trend of methods regarding the recovery of rare earth?

From a search of the publications on the Scopus database, the trend has been growing since 2012, with a peak in 2018 and 2021 (see Figure 1), with a total of 438 papers.

To find the scientific articles, the following search string on Scopus was used:*(TITLE-ABS-KEY (recover* OR recuperat** OR retriev* OR regain* OR extract* OR recycl*)) AND ((TITLE-ABS-KEY (rare AND earth AND metal? OR rare AND earth *AND element?)) OR (TITLE-ABS-KEY (europium OR gadolinium OR terbium OR dysprosium OR holmium OR erbium OR thallium OR ytterbium OR lutetium OR yttrium)) OR (TITLE-ABS-KEY (lanthanum OR cerium OR praseodymium OR neodymium OR promethium OR samarium))) AND (TITLE-ABS-KEY (e-waste OR electronic AND waste OR (electric AND electronic AND waste) OR WEEE)) AND (LIMIT-TO (SUBJAREA, "ENVI") OR LIMIT-TO (SUBJAREA, "ENGI") OR LIMIT-*

TO (SUBJAREA, "MATE") OR LIMIT-TO (SUBJAREA, "CHEM") OR LIMIT-TO (SUBJAREA, "CENG") OR LIMIT-TO (SUBJAREA, "EART"))

In 23 documents the keyword "patents" occurs; a comprehensive patent prior art landscape was not carried out in any of these documents.

Documents by year

Figure 1. Number of scientific publications per year (Source: Scopus; accessed 19 March 2023)

A patent landscape could help scientists to improve their knowledge in this technical field.

The aim of this paper is to provide a search strategy to retrieve a specific patent document on the recovery of rare earth elements from electronic scraps.

The following paragraph describes the methodology used to plan a search for patent information retrieval.

Materials and Methods

The patent analysis on methods for the recovery of rare earth metals from WEEE was carried out on the Espacenet, Google Patents, Octimine, and Orbit Intelligence platform (FamPat database) using a combination of keywords and IPC and CPC classification codes.

To retrieve patent documents, specific strategies should be planned, with the understanding that all patent databases are

incomplete and that using keywords only (intuitive but subjective activity) might not produce relevant results.

The classification codes are independent of the language used and can increase the number of relevant retrieved documents.

The classification systems have been designed to organize patents according to the technical field of a claimed invention and to simplify their retrieval.

The two most used systems are IPC (International Patent Classification) and CPC (Cooperative Patent Classification). Both are hierarchical systems, with eight/nine sections respectively and several classes, subclasses, groups, and subgroups. The IPC system, introduced in 1968, is used by more than 120 national patent offices and is updated every ear in January (the version currently in force is 2023.01). The CPC system is an advanced version of the IPC, having the same structure but with a greater number of subdivisions. The classification codes are assigned according to the function or application described in the claims and refer to the inventive information. The indexing codes are used to classify aspects not covered by the classification scheme and to classify additional information only.

Results and Discussion

The preparation and treatment (separation and [purification\)](https://ipcpub.wipo.int/?notion=scheme&version=20230101&symbol=C01F0017000000&menulang=en&lang=en&viewmode=f&fipcpc=no&showdeleted=yes&indexes=no&headings=yes¬es=yes&direction=o2n&initial=A&cwid=none&tree=no&searchmode=smart) of rare earth metals are classified in the main groupC01F 17/00 and in subgroups up to C01F 17/38.

Version 1 of the IPC scheme did not include subgroups, which were introduced in version 2020.01. There are no differences in the titles of the IPC and CPC codes, except in the number of documents categorized in the two classification schemes.

Searching using only the C01F 17/00 classification and/or compound names might produce limited results.

We recommended verifying that there are no other relevant codes, which could make the prior art search more complete. Using Espacenet as a reference database and some simple search queries (see Table 1), a series of classification codes listed in Table 2 were obtained and then used in the subsequent search on Orbit.

Table 1. List of search queries used on Espacenet to find other classification codes (Accessed 27 March 2023)

Table 2. List of classification codes (IPC/CPC) resulting from Espacenet search.

The IPC code C22B 59/00 has also been present since IPC version 1; what changes is the title (in version one it is "METALS OF THE RARE EARTHS" while in version 2 and in the following versions, it has been renamed to "OBTAINING RARE EARTH METALS").

IPC and CPC codes do not contain the same number of documents (see Graph 1), because it is mandatory for patent examiners to use the IPC classification systems.

The search was carried out on the Espacenet smart search field, using the following queries:

- 1. ipc=C22B59/00 NOT (cpc=A OR cpc=B OR cpc=C OR cpc=D OR cpc=E OR cpc=F OR cpc=G OR cpc=H)
- 2. cpc = "C22B59/00" NOT (ipc = "A" OR ipc = "B" OR ipc = "C" OR ipc = "D" OR ipc = "E" OR ipc = "F" OR ipc = "G" OR $\text{inc} = \text{"H"}$
- 3. ipc = "C01F17/00/low" NOT (cpc = "A" OR cpc = "B" OR cpc = "C" OR cpc = "D" OR cpc = "E" OR cpc = "F" OR cpc = "G" OR cpc = "H")
- 4. cpc = "C01F17/00/low" NOT (ipc = "A" OR ipc = "B" OR ipc = "C" OR ipc = "D" OR ipc = "E" OR ipc = "F" OR ipc = "G" OR $ipo = "H"$)

3273 and 2750 documents are classified in IPC codes only, respectively C01F17/00 and C22B59/00.

Therefore, both classification systems should be used in a landscape or prior art search.

Graph 1. No. of documents classified in IPC/CPC codes (C22B59/00 and C01F17/00)

The IPC/CPC indexing schemes (B09B 2101/15) used to classify the various types of electronic waste are relatively recent [IPC: 2022.01]. They include three subgroups: B09B 2101/16 (batteries), B09B 2101/17 (printed circuits), and B09B 2101/18 (smartphones and tablets).

Although the number of documents present in these two schemes is different (see Graph 2), this factor did not affect the results of the research.

The IPC code H01F 1/053 has undergone a revision since version 5 of the IPC scheme [in the previous one (1985.01)], where the reference code was H01F 1/04 - and generically defined as "metals or magnetic alloys").

No significant results have been obtained using the aforementioned code on Espacenet (see Table 3).

Table 3. Search queries used on Espacenet to find classification codes.

Graph 2. No. of patent documents in indexing schemes B09B 20101/15 – 18

The strategy used involves combining all the keywords (and synonyms) and classification codes corresponding to each technical feature to be searched.

Imagine a planned list of keywords (KWs) and classification codes (CL) for three technical features (f). They must be combined in the following way:

- R_1 = KW_{1f1} OR KW_{2f1} OR KW_{3f1} OR CL_{1f1} OR CL_{2f1} OR CL_{3f1}
- R_2 = KW_{1f2} OR KW_{2f2} OR KW_{3f2} OR CL_{1f2} OR CL_{2f2} OR CL_{3f2}
- R_3 = KW_{1f3} OR KW_{2f3} OR KW_{3f3} OR CL_{1f3} OR CL_{2f3} OR CL_{3f3}
- $R = R_1$ AND R_2 AND R_3

In this case the main concepts to be combined in the patent search are illustrated in Figure 2. The creation of a specification table may be helpful (Table 3).

Table 3. Specification table for patent landscape

The search queries used to retrieve the patent data on the Orbit database are summarized in Table 4.

Table 4. List of search queries used on the Orbit database (first step).

1001 results were obtained but only 114 were deemed relevant (the analysis was performed by reading each abstract of query No. 4).

To increase the number of relevant results, a search on Espacenet was carried out, using the "Smart search" field and the following queries: "Recovery of earth metals from electronic waste" (2624 results); "Recovery of rare earth metals from ewaste" (128 results). After the analysis of the search results, it was found that two classification codes are recurring: Y02P10/20, and C22B59/00.

Another set of search queries was planned (Table 5), comprising the aforementioned classification codes.

Table 5. List of search queries used on the Orbit database (second step).

The patent search strategy was enhanced using a semantic engine. The results, obtained using the abstracts of

articles ^{[\[1\]](#page-14-0)}, ^{[\[3\]](#page-14-2)} and ^{[\[7\]](#page-14-6)}, were filtered by query no. 10 (see Table 6).

10 4197 (C22B-059/00 AND (Y02W-030/82 OR Y02P-010/20))/IPC/CPC

Table 6. List of search queries used on the Orbit database (third step).

No. #

11 5749

12 4028

13 7121

Query Results Search query

No. #

The patent search provided a total of 414 results (208 granted and 93 pending). The complete list of patents is available in the Supplementary file.

The trend of priority filings is reported in Figure 3.

China is the country with the greatest number of priority patent applications filed in this technical field, followed by the USA and Japan.

The different techniques developed for recovering rare earth metals are hydrometallurgy, pyrometallurgy and biometallurgy or bioleaching. Hybrid treatments have been also patented (see Graph 3).

Graph 3. Number of patent applications/granted patents per type of treatment (the calculations are based on the results of query no. 16).

A more focused and precise search was carried out on biometallurgical and biological methods, using Espacenet as a reference patent database.

The search query used is the following:

 $\left($ cl = "C22B59/00/low" OR cl = "C01F17" OR ftxt= $\left($ "rare " prox/distance<3 "earth") OR ftxt all "REE") AND $\left($ ctxt =

"biometal*" OR ctxt = "biosorp*" OR ctxt = "bioleach*" OR ctxt = "algae" OR ctxt = "microb*" OR ctxt = "microorg*" OR ctxt = "bacter*" OR cl = "C22B3/18") AND (ftxt = "WEEE" OR ftxt=("electronic " prox/distance<3 "waste") OR ftxt=("electronic " prox/distance<3 "scrap") OR ftxt=("e" prox/ordered "waste") OR cpc all "Y02W30/82").

A list of 33 patent applications was retrieved.

All applications are recent and published between 2014 and 2022 (see Figure 4).

Figure 4. Publication range of biometallurgical patent applications (Source: Espacenet, accessed 16 April 2023).

In biometallurgy, microorganisms (bacteria, archaea, fungi) are used to extract REEs from a source. Biological treatment technologies for the recovery of rare earth metals from e-waste are characterised by different aspects: bioleaching, biosorption, phytoremediation, and bioelectrochemical systems. ^{[\[3\]](#page-14-2)}

The most recent published patent applications are shown herein.

Patent No. CA2915318C describes a process for isolating and enriching REEs (Scandium) using biomass comprising at least one organism selected from genera Pseudomonas and Bacillus, pre-treating the particulate material with autotrophic or heterotrophic bacteria.

In patent application No. WO2022182599A1, engineered microorganisms were used to improve the bioleaching of REEs.

Patent application No. US2022106667A1 discloses a method of algae and microbial screening for the recovery of specific heavy metals and/or rare earth elements (REEs).

EP3127874A1claims the use of a fungus of genus Cladosporium isolated from mine mud, for bioaccumulating and recovering precious metals and/or rare-earth elements from waste electrical and electronic equipment containing the same.

In WO2022266120A2, the claimed method uses a protein that can selectively bind one or more REEs, forming one or more protein-REE complexes. The disclosed protein is lanmodulin (LanM), which, in some embodiments, may include a C-terminal cysteine residue, an N-terminal cysteine, or an internal cysteine. In other embodiments, the cysteine is linked to the LanM via a glycine-serine-glycine amino acid linker or a hydrophilic linker.

The method disclosed in US2020277684A1 provides for using genetically engineered microbes encoding at least one REE binding ligand with the REE-containing material to form a microbe REE-complex. The REEs are separated from the microbe-REE complex using a solution that comprises an oxalate, an inorganic acid, an organic acid, a carbonate salt, a buffer, or any combination thereof.

In WO2021217148A1, Arthrobacter Nicotianae microbes are used to separate scandium from other rare earth elements.

In WO2020257702A1 a new method for recovering rare earth metals using a binding protein is disclosed.

Concluding Remarks

The patent analysis concerning methods for the recovery of rare earth metals from waste electronics was carried out on different patent databases, but especially on the Obit Intelligence platform, using a combination of keywords and precise classification codes (IPC/CPC systems).

To increase the number of relevant results, a semantic search was also implemented.

The top three countries of origin for patent applications are China, the USA, and Japan. The trend of filings remains essentially stable in 2020 and 2021, with a slight dip in 2022 (data are not complete since patent applications are published 18 months after their filing).

Data were also analysed from a technical point of view. Hydrometallurgical methods are the most patented, followed by pyrometallurgical and bioleaching treatments. Hybrid methods (above all hydro-pyrometallurgical) have also been patented.

A focus on the biological treatment technologies for recovery of rare earth metals from e-waste was carried out.

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Declaration of Interests

The author declares that he has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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