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Methods on the recovery of rare earth metals from electronic waste: a patent landscape

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Abstract

Waste and electrical and electronic equipment (WEEE), generally disposed of in landfills, contains valuable elements, called rare earth metals, 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.

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Introduction

Rare earth elements (REE - Rare Earth Elements) 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]

Five are the most critical elements regarding supply and industrial use: 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-of-life 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]

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 derives from permanent magnets, in particular 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][4][5][6][7]}

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][9][10]}

What is the technological trend of methods for 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; in none of these documents a comprehensive patent prior art landscape was carried out.

Documents by year

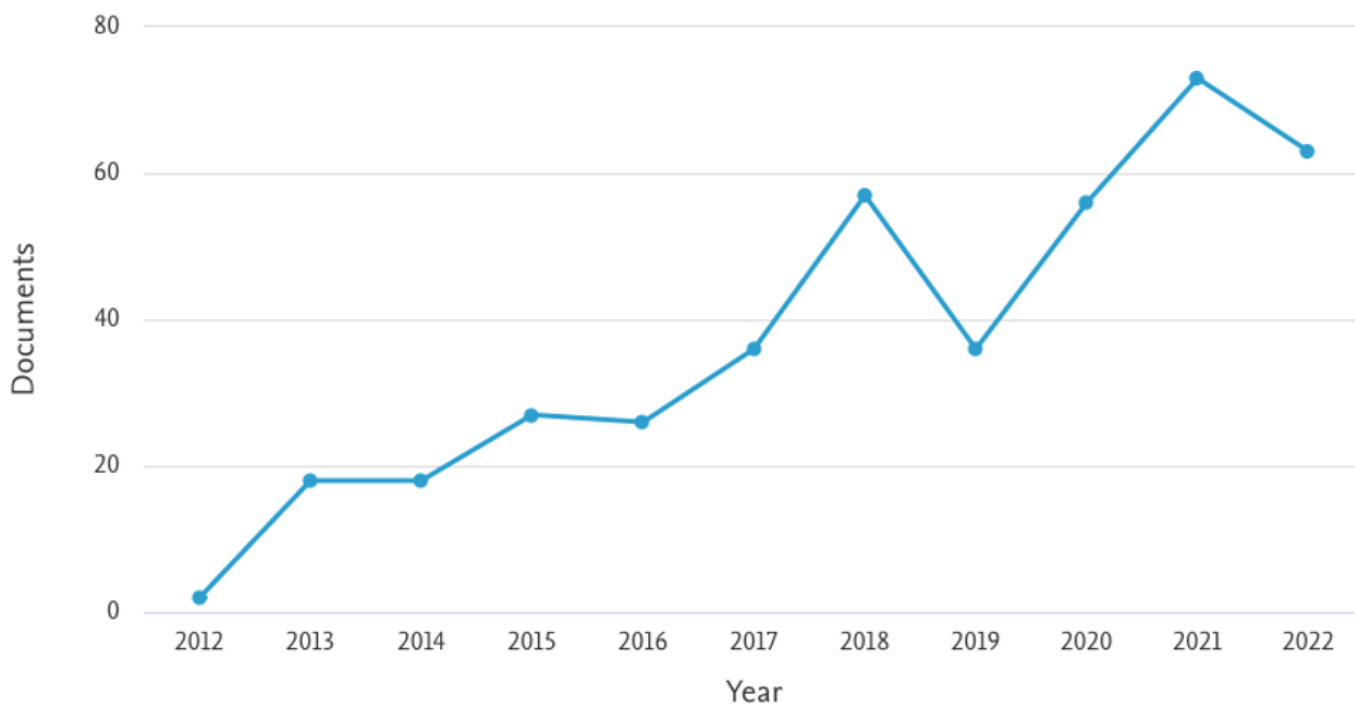


Figure 1. Number of scientific publications per year (Source: Scopus; accessed 19 March 2023)

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 recovery of rare earth metals from WEEE was carried out on the Espacenet (<https://worldwide.espacenet.com>), Google Patents (<https://patents.google.com>), Octimine (<https://app.octimine.com/auth/login>), and Orbit Intelligence platform (FamPat database) (<https://www.orbit.com>) using a combination of keywords and IPC and CPC classification codes

To retrieve patent documents, specific strategies should be planned, with the understanding that patent databases are all 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 year 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) of rare earth metals are classified in the main group C01F 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 originate limited results.

It is recommended to verify 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)

| Query No. # | Results No. # | Search query |
|-------------|---------------|--|
| 1 | 145 | ctxt = "rare earth" AND (ftxt = "recover*" OR ftxt = "recuperar*" OR ftxt = "recycl*" OR ftxt = "extract*") AND (ftxt any "WEEE" OR ftxt any "smartphone") |
| 2 | 6 | (ftxt = "recover*" OR ftxt = "recuperar*" OR ftxt = "recycl*" OR ftxt = "extract*") AND (ftxt any "WEEE" OR ftxt any "smartphone") AND cl any "C01F17" |

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

| Classification code | Definition |
|---------------------|--|
| C22B 59/00 | Obtaining rare earth metals (IPC/CPC) |
| H01F 1/053+ | Alloys containing rare earth metals (IPC/CPC) |
| H01F 1/15325 | Amorphous metallic alloys containing rare earth (CPC only) |
| Y02P 10/20 | Recycling (CPC only) |
| Y02W 30/50 | Reuse, recycling, or recovery technologies (CPC only) |
| Y02W 30/82 | Recycling of waste of electrical or electronic equipment [WEEE] (CPC only) |
| B09B 2101/15 | Electronic waste (CPC, indexing code) |
| B09B 2101/15 | Electronic waste (IPC, indexing scheme) |

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 EARTHES" while in version 2 and in the following ones it is renamed to "OBTAINING RARE EARTH METALS").

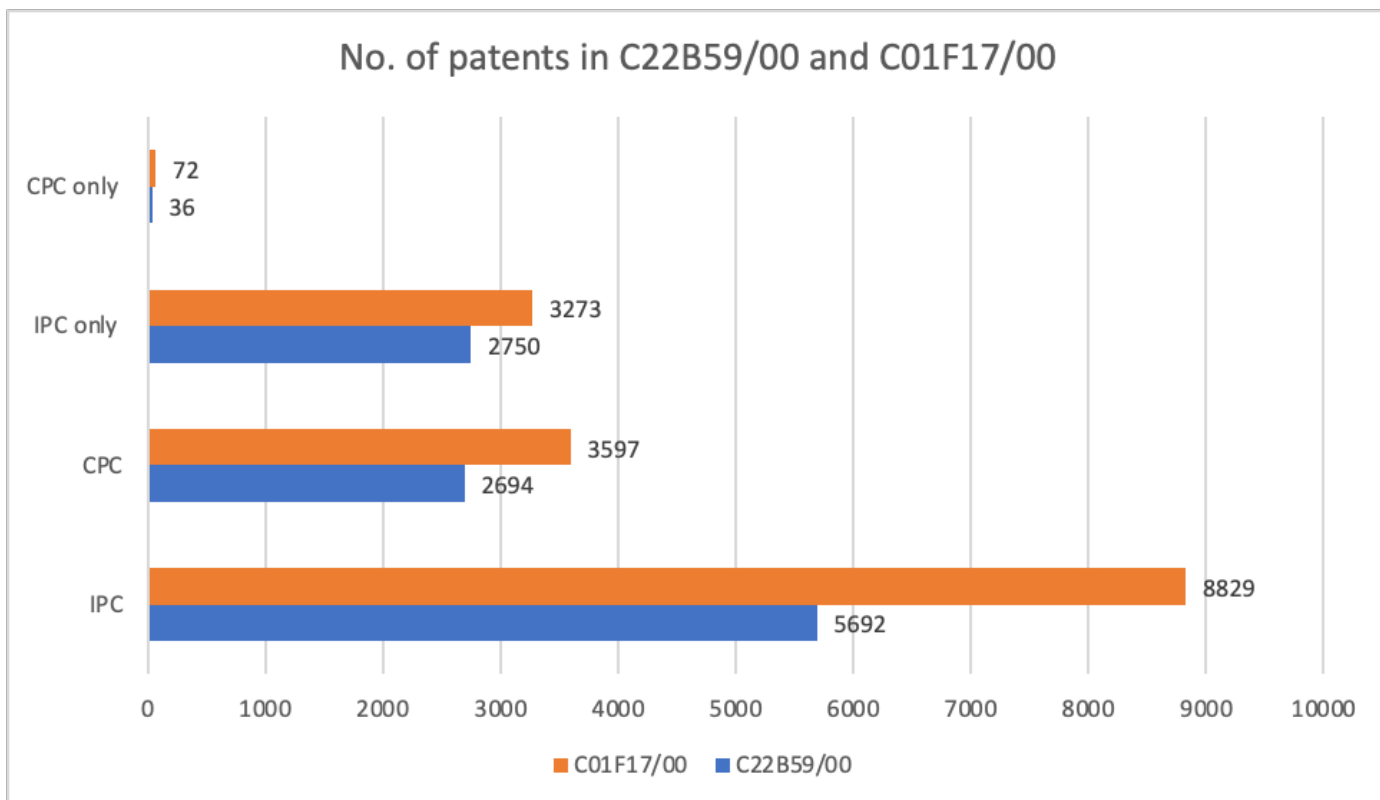
IPC and CPC codes do not contain the same number of documents (see Graph 1), because it is mandatory for patent examiners to use the IPC classification systems.

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 ipc = "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 ipc = "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).

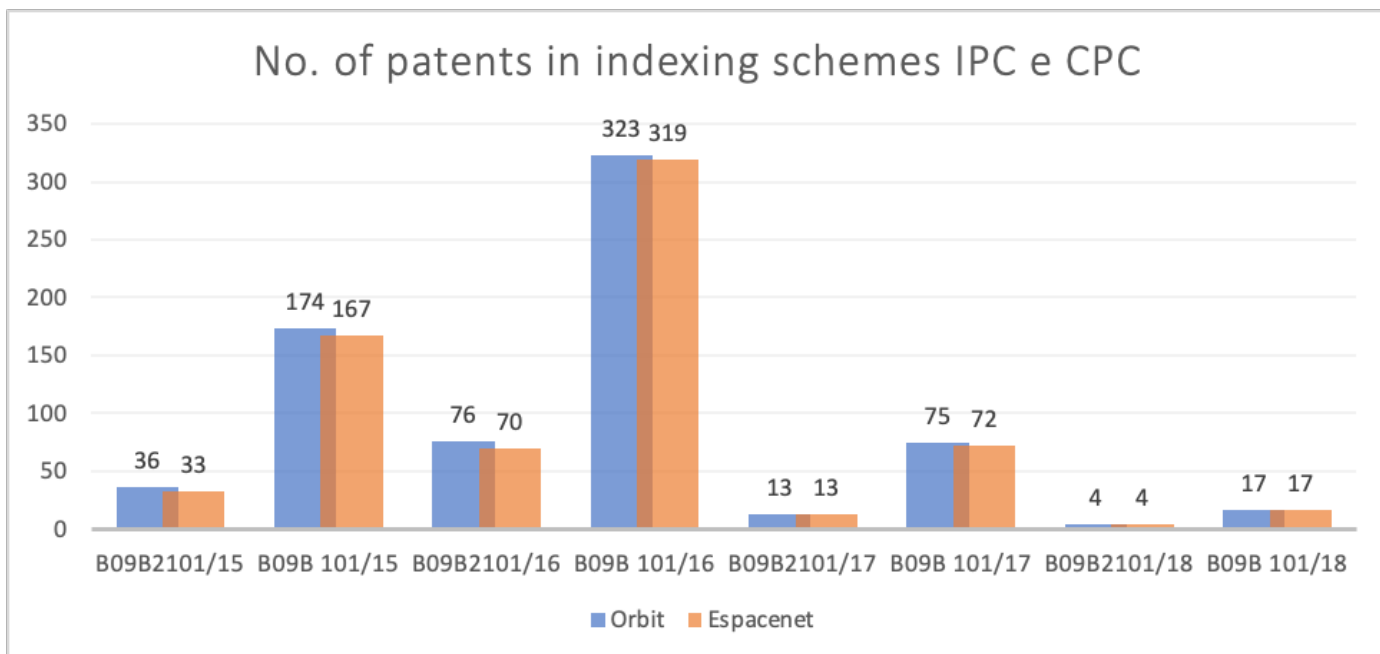
Although the number of documents present in these two schemes is different (see Graph 2), this factor did not affect the final result of the research.

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 above-mentioned code on Espacenet (see Table 3).

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

| Query No. # | Results No. # | Search query |
|-------------|---------------|--|
| 1 | 0 | (ftxt = "recover*" OR ftxt = "recuperar*" OR ftxt = "recycl*" OR ftxt = "extract*") AND cctx any "rare earth" AND ipc any "H01F1/04" AND (ftxt any "smartphone" OR ftxt any "tablet" OR ftxt any "laptop") |
| 2 | 6 | (ftxt = "recover*" OR ftxt = "recuperar*" OR ftxt = "recycl*" OR ftxt = "extract*") AND ftxt any "rare earth" AND ipc any "H01F1/04" AND (ftxt any "mobile phone" OR ftxt any "tablet") |



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 have to combine in the following way:

- R1 = KW1f1 OR KW2f1 OR KW3f1 OR CL1f1 OR CL2f1 OR CL3f1
- R2 = KW1f2 OR KW2f2 OR KW3f2 OR CL1f2 OR CL2f2 OR CL3f2
- R3 = KW1f3 OR KW2f3 OR KW3f3 OR CL1f3 OR CL2f3 OR CL3f3
- R = R1 AND R2 AND R3

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

Table 3. Specification table for patent landscape

| Concepts | Keywords | Classification symbols |
|--------------------------|--|--|
| Rare earth metals | Rare earth metals, rare earth elements, REEs, europium, gadolinium, terbium, dysprosium, holmium, erbium, thallium, ytterbium, lutetium, yttrium, lanthanum, cerium, praseodymium, neodymium, promethium, samarium | C01F 17+ C22B 59/00 H01F 1/053+ H01F 1/15325 |
| Recycling | Recover*, recycl*, recuper*, regain*, retriev*, extract* | Y02P 10/20 Y02W 30/50 Y02W 30/82 |
| Electronic waste | WEEE, E-waste, Waste electrical electronic equipment, consumer electronic scrap, electronic waste | B09B 2101/15 B09B 2101/16 B09B 2101/17 B09B 2101/18 |

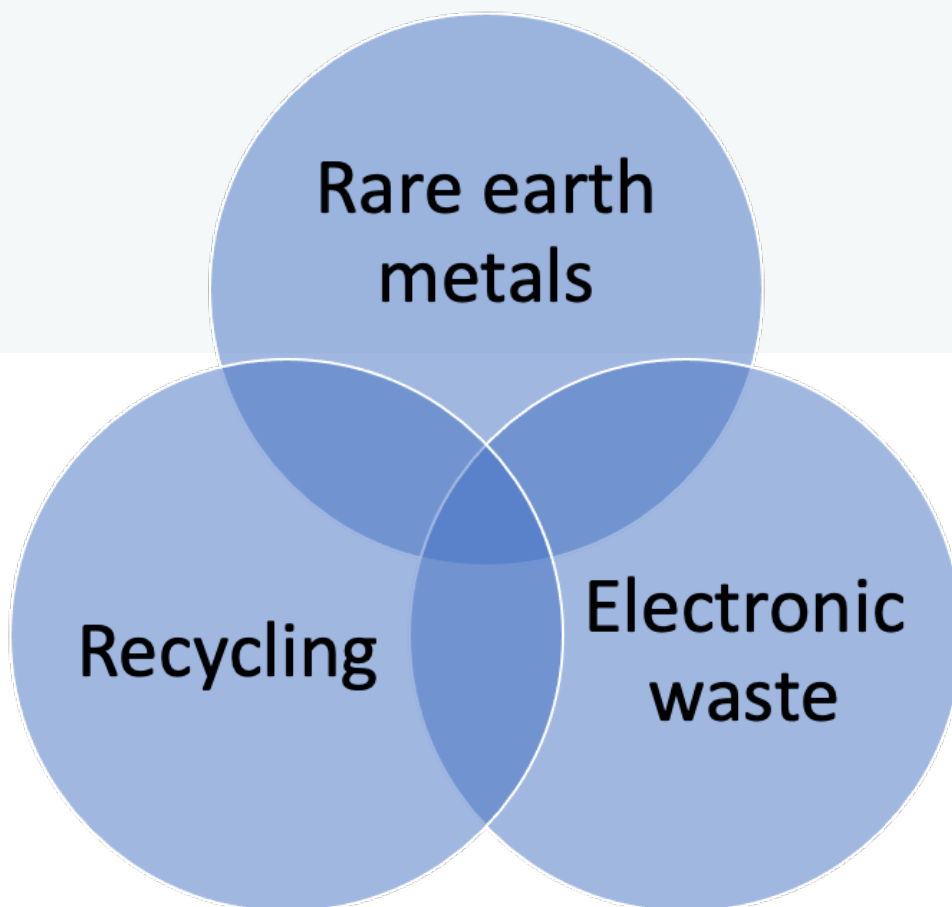


Figure 3. Concepts to be combined in prior art searches.

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).

| Query No. # | Results No. # | Search query |
|--------------------------------------|---------------|--|
| 1 [recycling] | 15829561 | ((RECOVER+ OR RECYCL+ OR RECUPER+ OR REGAIN+ OR RETRIEV+ OR EXTRACT+)/TI/AB/CLMS/DESC/ODES/ICLM OR (Y02P-010/20 OR Y02W-030/50 OR Y02W-030/82)/CPC) |
| 2 [electronic waste] | 14084 | ((WEEE OR E_WASTE OR (WASTE ELECTRICAL ELECTRONIC EQUIPMENT) OR (CONSUMER ELECTRONIC SCRAP) OR (ELECTRONIC SCRAP) OR (ELECTRONIC WASTE))/TI/AB/CLMS/DESC/ODES/ICLM OR (B09B-2101/15 OR B09B-2101/16 OR B09B-2101/17 OR B09B-2101/18)/CPC) |
| 3 [rare earth metals] | 2812985 | ((((RARE 1W EARTH 1W METAL?) OR (RARE 1W EARTH 1W ELEMENT?) OR (RARE 1W EARTH) OR (REE?) OR EUROPIUM OR GADOLINIUM OR TERBIUM OR DYSPROSIUM OR HOLMIUM OR ERBIUM OR THALLIUM OR YTTERBIUM OR LUTETIUM OR YTTRIUM OR LANTHANUM OR CERIUM OR PRASEODYMIUM OR NEODYMIUM OR PROMETHIUM OR SAMARIUM)/TI/AB/CLMS/DESC/ODES/ICLM OR (C01F-017+ OR C22B 59/00 OR H01F-001/053+ OR H01F+001/15325)/IPC/CPC) |
| 4 | 1001 | 1 AND 2 AND 3 |
| 5 [selection of relevant results] | 114 | *MEM "PPSTATMEM_PKIDS" /PKID |

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 e-waste” (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 above-mentioned classification codes.

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

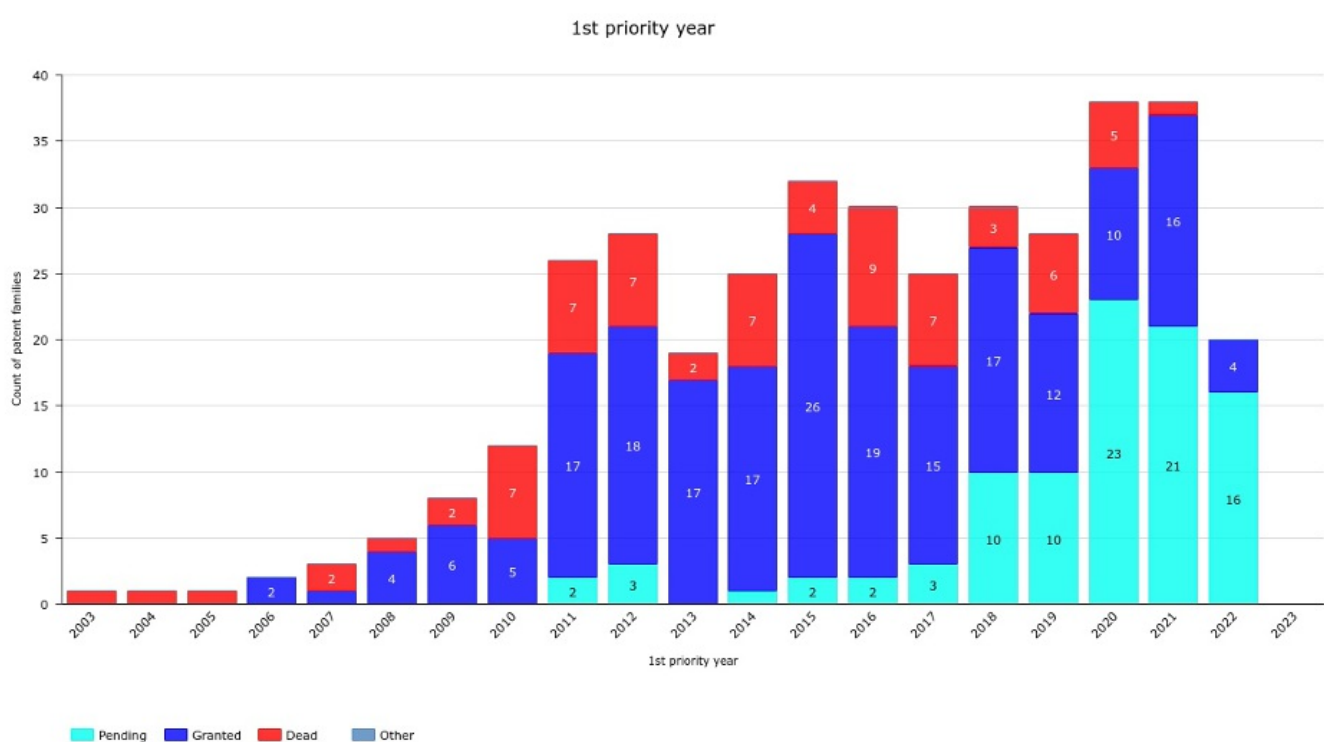
| Query No. # | Results No. # | Search query |
|-------------|---------------|---|
| 6 | 78 | ((RARE 1W EARTH 1W METAL?) OR (RARE 1W EARTH 1W ELEMENT?) OR (RARE 1W EARTH) OR (REE?) OR EUROPIUM OR GADOLINIUM OR TERBIUM OR DYSPROSIUM OR HOLMIUM OR ERBIUM OR THALLIUM OR YTTERBIUM OR LUTETIUM OR YTTRIUM OR LANTHANUM OR CERIUM OR PRASEODYMIUM OR NEODYMIUM OR PROMETHIUM OR SAMARIUM)/TI/AB/CLMS/DESC/ODES/ICLM AND (RECOVER+ OR EXTRACT+ OR REGAIN+ OR RECYCL+)/TI/AB/DESC/ODES/CLMS AND ((ELECTRONIC 1W WASTE) OR (E_WASTE) OR (ELECTRONIC 1W SCRAP) OR WEEE)/TI/AB/CLMS/DESC/ODES/ICLM) AND (Y02P-10/20 OR C22B-059/00)/IPC/CPC |
| 7 | 30 | (C22B-059/00 AND Y02W-030/82)/IPC/CPC |
| 8 | 15 | (C01F-017+ AND Y02W-030/82)/IPC/CPC |
| 9 | 163 | 5 OR 6 OR 7 OR 8 |

The patent search strategy was enhanced using a semantic engine. The results, obtained using the abstracts of articles [1], [3] and [7], were filtered by query no. 10 (see Table 6).

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

| Query No. # | Results No. # | Search query |
|-------------|---------------|---|
| 10 | 4197 | (C22B-059/00 AND (Y02W-030/82 OR Y02P-010/20))/IPC/CPC |
| 11 | 5749 | ..SEMSEARCH "CIRCULAR ECONOMY" 20 "DISPOSAL RATE" 61 "ELECTRONIC EQUIPMENT" 29 "ELECTRONIC INDUSTRIAL MARKET" 81 "ENVIRONMENTAL IMPACT" 48 "FAUNA" 22 "FLORA" 31 "FUTURE PERSPECTIVE" 18 "HIGH TECHNOLOGICAL APPLIANCE" 100 "HITECH EQUIPMENT" 71 "RAPID" 44 "RARE EARTH ELEMENT" 92 "RECOVERY" 40 "REE RECOVERY" 33 "REE SOURCE" 48 "REE SOURCE DEPLETION" 62 "REES" 63 "REES RECOVERY" 79 "REGULATORY FRAMEWORK" 16 "RESEARCH ROADMAP" 12 "RISING CONCERN" 56 "SUSTAINABILITY" 72 "WASTE ELECTRICAL AND ELECTRONIC EQUIPMENT" 100 "WIDE RANGE" 26 |
| 12 | 4028 | ..SEMSEARCH "ADOPTION" 45 "AID" 27 "ARTICLE PUBLICATION" 31 "BIBLIOMETRIC STUDY" 32 "BIOLEACHING" 20 "BIOMETALLURGY" 58 "BIOSORPTION" 18 "COMPREHENSIVE SUMMARY" 41 "COUNTRY ECONOMIC GROWTH" 17 "COUNTRY POLICY MAKER" 7 "E WASTE" 52 "E WASTE MANAGEMENT" 100 "ECONOMIC GROWTH" 16 "END OF LIFE" 63 "END OF LIFE RATE" 77 "ENORMOUS INCREASE" 72 "ENVIRONMENT" 90 "FUTURE RESEARCH" 40 "GLOBAL CONCERN" 72 "GREEN TECHNOLOGY PERSPECTIVE" 28 "HUMAN HEALTH" 55 "HYDROMETALLURGY" 62 "INDUSTRIALIST" 14 "KEYWORD" 28 "LIMITATION STILL" 57 "METAL EXTRACTION" 59 "METAL RECOVERY" 52 "POLICY MAKER" 9 "PYROMETALLURGY" 63 "RARE EARTH" 49 "RECOVERY" 42 "STUDENT" 15 "UNDERSTANDING E WASTE" 9 "VALUABLE KNOWLEDGE" 12 "WASTE EEE" 53 "WASTE EEE DISPOSAL" 68 |
| 13 | 7121 | ..SEMSEARCH "AIR CONDITIONER" 69 "BATTERY" 21 "BIOLEACHING" 28 "BIOMETALLURGY" 31 "BIOSORPTION" 27 "CABLE" 20 "CALCULATOR" 50 "CERIUM" 37 "COFFEE MACHINE" 45 "COMMUNICATION" 23 "CRITICAL ELEMENT" 61 "DISHWASHER" 35 "DYSPROSIUM" 69 "ELECTRIC FAN" 55 "ELECTRIC RAZOR" 48 "ELECTRONIC CONFIGURATION" 100 "ELECTRONIC WASTE" 62 "END OF LIFE" 39 "ERBIUM" 48 "EUROPIUM" 71 "FLUORESCENT LAMP" 31 "GADOLINIUM" 66 "GAS PHASE EXTRACTION" 30 "GRAM" 19 "HEAT EXCHANGE" 71 "HEAT EXCHANGE EQUIPMENT" 80 "HEAVY" 57 "HOLMIUM" 53 "INDUSTRIAL WASTE" 35 "LANTHANIDE" 47 "LANTHANUM" 43 "LAPTOP" 20 "LASER" 32 "LATTER" 30 "LIFE END" 26 "LIGHT RARE EARTH METAL" 61 "LUTETIUM" 48 "METAL" 55 "METHOD TECHNOLOGICAL TREND" 17 "NAVIGATION" 17 "NDFEB MAGNET" 32 "NDFEB MAGNET MANUFACTURE" 40 "NEODYMIUM" 59 "NEODYMIUM RECOVERY" 17 "OXIDATION REDUCTION" 27 "PERMANENT MAGNET" 60 "PHOTOCOPIER" 34 "PHOTOVOLTAIC PANEL" 35 "PHYTOREMEDIATION" 29 "PLUG" 20 "PRASEODYMIUM" 45 "PRODUCT CATEGORY" 34 "PRODUCT DEVICE" 42 "PRODUCTION RESIDUE" 41 "PROMETHIUM" 42 "PYROMETALLURGICAL" 41 "RARE EARTH" 10 "RARE EARTH ELEMENT" 82 "RARE EARTH METAL" 76 "RARE EARTH METAL RECOVERY" 46 "RARE EARTH RECOVERY" 12 "RARE EARTH RECYCLING" 50 "RECOVERY" 33 "REFRIGERATOR" 61 "RESIDUE" 76 "SAMARIUM" 38 "SCANDIUM" 43 "SCREEN" 22 "SMARTPHONE" 24 "STOVE" 46 "SUPERCONDUCTOR" 42 "TABLET" 18 "TECHNOLOGICAL TREND" 19 "TELEPHONE" 34 "TERBIUM" 65 "THALLIUM" 51 "TON" 26 "URBAN SEPARATE COLLECTION" 44 "VIDEO CAMERA" 49 "WASHING MACHINE" 43 "YTTERBIUM" 51 "YTTRIUM" 69 |
| 14 | 4459 | ..SEMSEARCH "ELECTRONIC WASTE" 92 "RARE EARTH" 81 "RARE EARTH RECOVERY" 100 |
| 15 | 280 | 10 AND 11 AND 12 AND 13 AND 14 |
| 16 | 414 | 9 OR 15 |

The patent search provided a total of 414 results (208 granted and 93 pending). The trend of priority filings is reported in Figure 2.

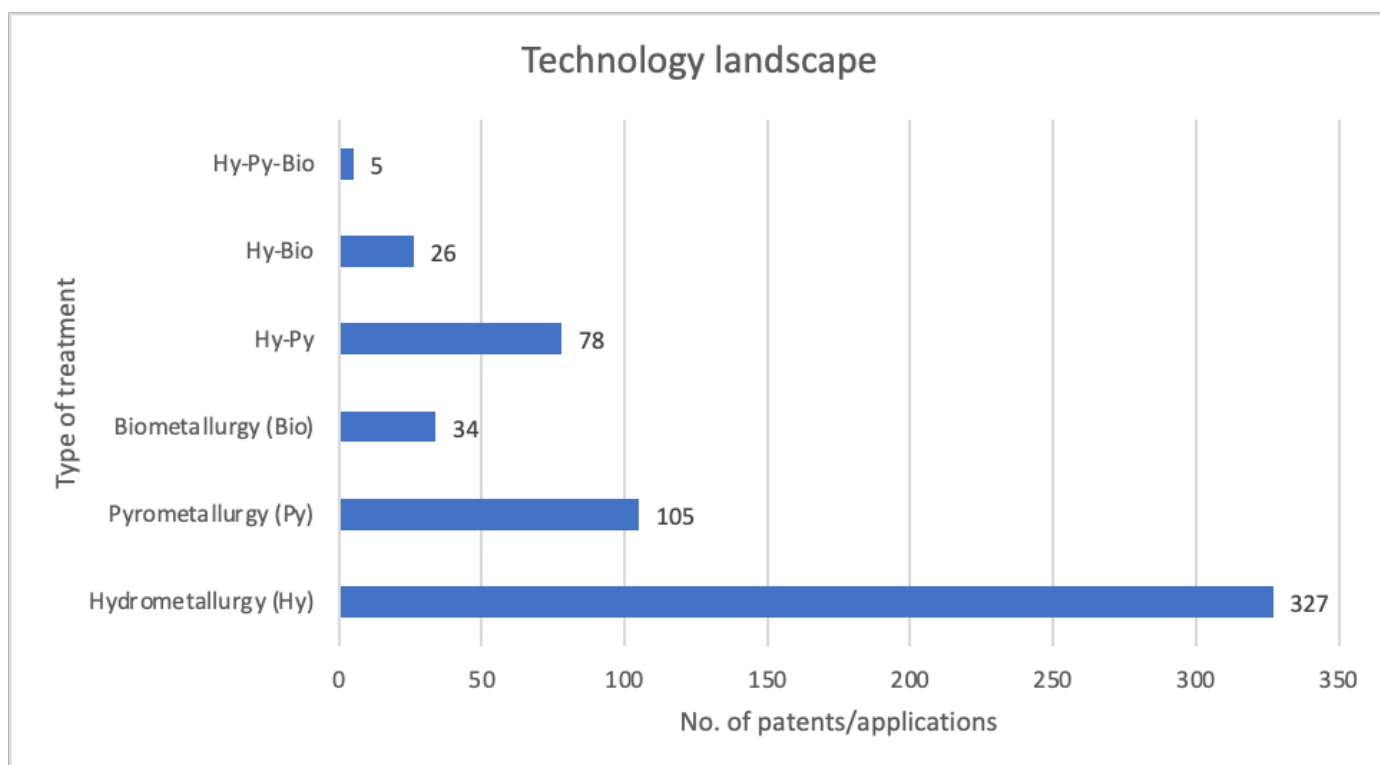


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Figure 2. Trend of priority filings (Orbit data).

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 (Own calculations based on the results of query no. 16)

Concluding remarks

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

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

The top three countries of origin for patent applications are China, USA, and Japan. The trend of filings remains essentially stable in the last period (2020 – 2021), with a slight dip in 2022 (data are incomplete, because patent applications are published 18 months after their filing).

Data were also analyzed from a technical point of view. Hydrometallurgical methods are the most patented, followed by pyrometallurgical and bioleaching treatments.

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