Screening Life Cycle Assessment of beneficiation processes for Rare Earth Elements recovery from secondary sources

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Abstract. Rare Earth Elements (REEs) are identified as critical raw materials for the EU economy. The ENVIREE project aims at contributing to REE supply in Europe, by addressing the exploitation of tailings and mining waste. Two secondary sources: tailings from New Kankberg (Sweden) and Covas (Portugal) are promising and rich in REEs. Beneficiation is one of the main step of REE recovery from secondary sources. The aim of this paper is to report the Life Cycle Assessment modelling with SimaPro software of beneficiation processes developed for tailings from New Kankberg and Covas. For the REE concentrate production from New Kankberg tailings, which come from an operating gold production plant, the highest environmental impact is caused by the disposal of the final residues after beneficiation. For the REE concentrate production from Covas tailings, which represent 30 years underground mining activity focused in tungsten, the highest environmental impact is caused by the electricity consumption for gravity and magnetic separation.

1. Introduction

In the EU Raw Materials Initiative [1] Rare Earth Elements (REEs) - the group of lanthanides as well as yttrium and scandium, have been identified as critical raw materials for the development and competitiveness of the EU economy. The variety of applications of REE are ranging from super magnets in electrical engines in cars and generators in wind turbines, computer hard drives, flat screens for computers and TVs, to NiHM batteries in hybrid electric cars, catalysts for the automotive and petroleum industry, fuel cells and hydrogen storage devices [2]. While REE are highly relevant for high-tech industries and products, there is a significant risk of supply, as the China's monopoly on the production and refining of REE is undeniable.

On the other hand, there are several REE containing materials still being not properly addressed. The ENVIREE project, funded by the ERA-MIN programme, aims at contributing to REE supply in Europe, by addressing the exploitation of specific secondary sources: tailings and mining waste. The ENVIREE project goal is the complete extraction process proposal for secondary sources. Within the project two secondary sources: tailings from New Kankberg (Sweden) and Covas (Portugal) were identified as very promising and rich in REEs. Beneficiation is one of the main step of REE recovery from secondary sources.

In order to have a complete holistic view of the developed processes, Life Cycle Assessment (LCA) methodology is employed to be sure that recovery processes are environmentally feasible and
friendly. The aim of this paper is to report the LCA modelling with SimaPro software of\textsuperscript{1} beneficiation processes developed for two secondary sources: tailings from New Kankberg and Covas.

2. Life cycle assessment methodology

LCA is an effective method of environmental management. It quantifies the relevant emissions and resources consumption and the resulting environmental and health impacts as well as resource depletion issues, throughout a product or a process life cycle [3]. LCA is a structured, internationally standardized method - ISO 14040 [4]. The ISO standard instructs to perform an LCA study in four steps: 1) goal and scope, 2) inventory analysis, 3) impact assessment and 4) interpretation.

The goal and scope define and describe the product, process or activity. This stage establishes the context in which assessment is to be made as well it identifies the system boundaries and functional unit [5]. The primary goal of an LCA is to choose the best product, process, or service in terms of the lowest effect on human health and the environment. Conducting an LCA can help guide the development of new products, processes, or activities toward a reduction of emissions and resource consumption [6]. The functional unit is a quantitative description of the service performance, which means the needs fulfilled, of the evaluated product system. The system boundaries define the processes and the parts of the life cycle, which belong to the analysed system, which processes and life cycle stages are required for providing the function [3].

Life Cycle Inventory (LCI) is a process of identifying and quantifying environmental exchanges: energy and raw materials consumption, emissions and other releases (to air, water, soil) and solid wastes for the entire life cycle of a product or process [6]. The inventory of environmental exchanges is related to the functional unit and has to be done in line with the goal and scope definition [3, 5].

Life Cycle Impact Assessment (LCIA) aims to establish a linkage between the product or process and its potential environmental impacts [6]. The inputs and outputs of elementary flows that have been reported in the LCI are translated into impact indicator results related to three damage areas: human health, natural environment and resource depletion [3].

Interpretation occurs at every stage in an LCA. It is a systematic technique to identify, quantify, and evaluate information from the results of LCI and LCIA, and communicate them effectively [6].

LCA is usually conducted with a dedicated software and database. For the purpose of ENVIREE project the LCA study has been performed with SimaPro 8.3 software and Eco-invent database v.3. As the method for LCIA, the ReCiPe midpoint and endpoint have been chosen, which is commonly used and recommended by European authorities. ReCiPe comprises two sets of impact categories with associated sets of characterisation factors [7]. Eighteen impact categories shown in table 1 are addressed at the midpoint level.

| No | Impact category                   | No | Impact category                   |
|----|----------------------------------|----|----------------------------------|
| 1  | Climate Change (CC)              | 10 | Freshwater Ecotoxicity (FET)     |
| 2  | Ozone Depletion (OD)             | 11 | Marine Ecotoxicity (MET)         |
| 3  | Terrestrial Acidification (TA)   | 12 | Ionising Radiation (IR)          |
| 4  | Freshwater Eutrophication (FE)   | 13 | Agricultural Land Occupation (ALO)|
| 5  | Marine Eutrophication (ME)      | 14 | Urban Land Occupation (ULO)      |
| 6  | Human Toxicity (HT)             | 15 | Natural Land Transformation (NLT)|
| 7  | Photochemical Oxidant Formation (POF) | 16 | Water Depletion (WD)           |
| 8  | Particulate Matter Formation (PMF) | 17 | Mineral Resource Depletion (MRD)|
| 9  | Terrestrial Ecotoxicity (TET)    | 18 | FossilFuel Depletion (FD)        |

\textsuperscript{1}The method for LCIA, the ReCiPe midpoint and endpoint have been chosen, which is commonly used and recommended by European authorities.
At the endpoint level the midpoint impact categories are further converted and aggregated into the following three endpoint categories (damage categories): Damage to Human Health (HH), Damage to Ecosystem (ED), Damage to Resource Availability (RA).

At the characterisation step, mandatory for LCIA, the impact categories indicator values are calculated on basis of the LCI and they are express in different units. This step of LCIA allows the quantitative evaluation of individual processes contributions within individual impact categories. At the normalisation step all categories values are recalculated - divided by the reference value. The reference value is the total impact that is all emissions and resource uses, caused by one person, during one year, in a certain geographical region - Europe. At this step impact categories can be compared to each other. It allows identifying significant impact categories with the highest indicator values. At the weighting step indicator values are recalculated using numerical factors based on value-choices - weights. The weighting expresses the relative importance of different environmental indicators for the decision making. The single score is calculated by summing up the results of all category indicators multiplied with the normalization factor and multiplied with the weighting factor for each category.

Graphical presentation of modelling is a process tree. The line thickness represents the environmental impact – the thicker the line, the higher the impact. Red colour indicates negative environmental impact, green colour indicates positive environmental impact - so called avoided impacts. Bars on the righted of the blocks present contribution to the total environmental impact of the system.

3. Goal and scope
The primary goal and scope of the LCA is the environmental assessment of the innovative processes. Several stages of processes of REE recovery to be developed within the ENVIREE project were proposed in the work [8]. These stages included: Excavation and transport of the secondary raw material; Physical and chemical pre-treatment (beneficiation); Leaching and purification; Hydrometallurgical separation; Ionic liquid separation; Membrane separation; Post-treatment stage: residues disposal and reclamation.

However, for the purpose of this study, the system boundaries include: excavation and transport of secondary sources as well as beneficiation with disposal and land reclamation of the residues after beneficiation. The system boundaries do not include leaching or separation processes.

The functional unit is defined as 1000 kg of a secondary source, to be excavated and processed, as the input for all subsequent processes of REE recovery. It means a certain quantity of pre-treated / beneficiated material, obtained from 1000 kg of the input material, enters the leaching, and then another quantity of liquid enters the separation.

The goals of this study are to identify environmental aspects associated with:
- REE concentrate production from New Kankberg tailings.
- REE concentrate production from Covas tailings.

The system boundaries for REE concentrate production from New Kankberg tailings include transport of gold tailings to REE processing facility, 3 stages of beneficiation: flotation, magnetic separation, filtration, then residues deposition and reclamation. The figure 1 shows simplified beneficiation process proposed for New Kankberg tailings.

The system boundaries for REE concentrate production from Covas include excavation and transport of old tailings to the processing facility, 4 stages of beneficiation: size reduction and classification, gravimetric separation, magnetic separation, filtration, waste deposition and reclamation. The figure 2 shows the simplified beneficiation process proposed for Covas tailings.
4. Life cycle inventory for tailings beneficiation

4.1. Life cycle inventory for the beneficiation of New Kankberg tailings

New Kankberg gold mine is located in the Boliden Area. The ore is currently mined underground and transported, around 10 km, by truck to the Boliden concentrator. The ore is crushed to below 250 mm then ground in two stages. Gold is floated with tellurium. The gold is recovered by hot cyanide leaching from the flotation concentrate. The cyanide leaching residue is going to the tellurium plant where tellurium is recovered. The residue from the tellurium plant is going back to cold cyanide leaching together with the flotation tailing. The tailings are going to the tailing pond [9].

Samples from the flotation tailings from gold production were delivered to be further processed within the ENVIREEE project. After their homogenisation, samples were analysed. The results are shown in tables 2 and 3.

Table 2. Mineral characterization of samples from New Kankberg using Rietveld method [10].

| Mineral | K feldspar | Quartz | Muscovite | Kaolinite | Albite | Sum |
|---------|------------|--------|-----------|-----------|--------|-----|
| Content % | 3.15 | 49.47 | 39.41 | 5.76 | 2.21 | 100 |

Ammonium-sulfate leaching of samples was investigated, in order to test the availability of REE extraction. The results are presented in Table 3.

Table 3. ICP-MS analysis of samples from New Kankberg washed with ammonium sulphate [10].

| Element | Sc | Y | La | Ce | Pr | Nd | Sm | Eu | Gd | Tb | Dy | Ho | Er | Tm | Yb | Lu | SUM REE |
|---------|----|---|----|----|----|----|----|----|----|----|----|----|----|----|----|----|---------|
| Content [ppm] | 0.41 | 3.27 | 12.19 | 23.89 | 2.4 | 9.83 | 1.75 | 0.45 | 1.28 | 0.14 | 0.49 | 0.08 | 0.22 | 0.03 | 0.17 | 0.02 | 56.62 |

New Kankberg tailings come from the operating plant, therefore no excavating from a tailing pond is needed. In an industrial practicethe tailings will be directly pumped to a separate plant for further processing - REE recovery, instead of being sent to a tailing pond. A facility for REE recovery should be located in the near proximity of the gold/tellurium recovery plant. The fuel consumption for transport of the material to the REE processing plant it is assumed to be 0.2 kWh/ton.

The proposed beneficiation scheme of New Kankberg includes flotation followed by magnetic separation. Flotation allows recovering 70% of the total phosphate content and 50% of REE. Following the flotation stage, the concentrate that contains a mix of phosphates (apatite and monazite) is further enriched through magnetic separation [11]. The final step is dewatering - filtration. A pilot was operated on 1 ton of New Kankberg tailings. The mass balance of the pilot is following [11]:

- Input - 1000 kg tailings from New Kankberg,
- Concentrate (after flotation and magnetic separation) - 10 kg (water content = 10%, output of press filter),
- Residues - 990 kg (water content = 15%, output of belt filter).

The simplified beneficiation process for New Kankberg tailings is shown in figure 1. Electricity consumption for the whole beneficiation is calculated in table 4.
Table 4. Energy consumption calculated on the basis of the mass balance of circuit for New Kankberg.

| Step of beneficiation processes                      | Unit energy consumption [kWh/Mg] | Input material [kg] | Calculated energy consumption [kWh] |
|-------------------------------------------------------|----------------------------------|---------------------|-----------------------------------|
| Flotation                                             | 5.0                              | 1000                | 5.0000                            |
| Magnetic separation                                   | 2.6                              | 95.4                | 0.2480                            |
| Filtration - dewatering (concentrate)                 | 0.25                             | 10                  | 0.0025                            |
| Filtration - dewatering (residues)                    | 0.5                              | 990                 | 0.4950                            |

In the flotation process following reagents are consumed: Depressant - Water glass (Na₂SiO₃), 700 g/Mg of tailings; Conditioner - Aero 845 (C₂₆H₄₃N₄Na₄O₁₀S), 160 g/Mg of tailings; Collector - Resinoline BD2, 160 g/Mg of tailings.

The emissions to the air, water and ground are reported in literature [13-16]. Nevertheless, information on emissions from the New Kankberg beneficiation operation is not available. Therefore emissions are neglected in the modelling.

The residues from the beneficiation process are non-toxic and may be used for various purposes. The most suitable way of their disposal is to send them to the tailings disposal facility designed for the tailings from gold/tellurium production. Such a tailing disposal facility can be then reclaimed for forestry or nature restoration. Tailings can be used as a soil substrate in land reclamation. The formation of plant cover is necessary to avoid dust emission from the surface. Organic amendments: sewage sludge or municipal waste compost, supply nutrients for plants and enable rapid development of plant cover [12].

Figure 1. Simplified Beneficiation Process for tailings from New Kankberg (System Boundaries).

4.2. Life cycle inventory for the beneficiation of Covas tailings

The Covas tailings represents 30 years (1954-1984) of mining, focused in tungsten, exploited by underground mining works. These tailings and the mining area were rehabilitated [9].

After collection of samples and their homogenisation they were analysed. The results of these analyses are shown in tables 5 and 6.
Table 5. Mineral characterization of samples from Covas using Rietveld method [10].

| Article I. Mineral | Article II. Feldspar | Quartz | Muscovite | Kaolinite | Arsenopyrite | Albite | Hematite | Clinochlore | Chalcopyrite | Ferberite | Sum |
|-------------------|----------------------|--------|-----------|-----------|--------------|--------|----------|-------------|--------------|-----------|-----|
| Count %           | 17.81                | 16.46  | 23.11     | 13.18     | 4.37         | 3.79   | 3.43     | 11.74       | 5.2          | 0.91      | 100 |

Ammonium-sulfate leaching of samples from Covas is presented in table 6.

Table 6. ICP-MS analysis of samples from Covas washed with ammonium sulphate [10].

| Element | Content [ppm] | Sc | Y  | La  | Ce  | Pr  | Nd | Sm | Eu | Gd | Tb | Dy | Ho | Er | Tm | Yb | Lu | SUM | REE |
|---------|---------------|----|-----|-----|-----|-----|----|----|----|----|----|----|----|----|----|----|----|-----|-----|
|         |               | 0.95| 1.62| 1.63| 2.94| 0.29| 1.2| 0.25| 0.09| 0.25| 0.05| 0.22| 0.04| 0.13| 0.02| 0.02| 9.84 |     |

1 Mg of tailings from Covas was excavated with a backhoe excavator, with fuel consumption of 18 litre. Then the tailings were sent by truck to BRGM (France) for processing. If the tailings are processed in an industrial practice, the pre-treatment facility will be located in the close proximity of the Covas mining area. In this study the transport is modelled for short distance of 20 km by a truck.

The proposed beneficiation scheme of Covas tailings includes gravity separation followed by magnetic separation. Magnetic separation only stands for tungsten concentration purposes. Magnetic separation allows increasing further the REE content and particularly the one of tungsten. Final recovery for REE is 55% and 35% for tungsten [11]. A continuous pilot was operated on 600 kg of Covas tailings to produce a gravimetric concentrate and a magnetic concentrate. After regrinding, tailings were reprocessed using a gravity separator. This stage resulted in the production of a concentrate and tailings (residues). Then, the gravimetric concentrate was treated by magnetic separation producing: a magnetic concentrate, a middling and non-magnetic tailings (residues), followed by dewatering (filtration) [11]. The simplified beneficiation process for Covas tailings is shown in figure 2.

The overall mass balance of the process is following:

- Input - 1000 kg tailings from Covas
- Concentrate (after gravimetric and magnetic separation) - 150 kg (including 130 kg of Middlings and 20 kg of Magnetic fraction; water content = 10%, output of press filter)
- Residues (tailings) - 850 kg (water content = 15%, output of belt filter)
Electricity consumption for the whole beneficiation is calculated in table 7.

**Table 7.** Energy consumption calculated on the basis of the mass balance of circuit for Covas.

| Step of beneficiation processes | Unit energy consumption [kWh/Mg] | Input material [kg] | Calculated energy consumption [kWh] |
|--------------------------------|----------------------------------|---------------------|-------------------------------------|
| Size reduction + Classification| 15                               | 1000                | 15                                  |
| Gravity Separation             | 0.5                              | 1000                | 0.5                                 |
| Magnetic separation            | 2                                | 184                 | 0.368                               |
| Filtration - dewatering        | 0.5                              | 184                 | 0.092                               |

Emissions to air are neglected in the modelling, because information on emissions from the pilot Covas beneficiation operation is not available.

After beneficiation Covas residues are useful for land reclamation and can be treated as a soil substrate. The best option for the residues treatment is their return to the disposal facilities, where they take from and subsequent reclamation for nature rehabilitation or forestry. Due to high acidity the reclamation of the Covas beneficiation residues should include their neutralization. Additionally fertilization should be applied with organic amendments sewage sludge or compost [12]. The calculated values used for land reclamation are: 5 kg of Ca(OH)$_2$ per ton of tailings and organic amendments, N dose: 150 kg per ha of reclaimed area.

5. Results of modelling - Life Cycle Impact Assessment and discussion

5.1. Results of modelling the REE concentrate production from New Kankberg tailings

The normalization step of REE concentrate production from New Kankerg tailings is shown in figure 3. The significant impact categories are: Urban land occupation, Natural land transformation, Fossil depletion. Within the category Urban land occupation positive and negative impacts are almost equal. The positive impact on the environment (negative value) results from avoiding the gold tailings (1000 kg of input material) disposal. On the other hand, tailings after REE beneficiation (990 kg), are
disposed at the tailing pond designed for gold tailings. Tailings quantity and quality is almost unchanged by beneficiation process. The fossil depletion category indicator is dependent on the consumption of fossil fuels (crude oil for petroleum production, natural gas and hard coal) for electricity production and fuel for transport.

The most significant unit processes are direct operations on tailings: both input material and final residues. Avoided disposal of gold tailings, which are taken for REE beneficiation, generate positive impact on the environment. On the other hand landfilling of residues after beneficiation process, generate negative impact. The others significant unit processes are reagents used for flotation and electricity consumption.

The highest environmental impact is caused by the disposal of the final residues after beneficiation. This unit process contributes significantly to Urban land occupation (midpoint category) and to Ecosystem (endpoint category). Nevertheless, avoiding disposal of initial gold tailings and using them as input material for REE concentrate production balances the negative impact for Ecosystem. Another significant contributors observed in endpoint categories are reagents for flotation, especially fatty acid.

5.2. Results of modelling the REE concentrate production from Covas tailings

The normalization step of REE concentrate production from Covas tailings is shown in figure 4. The significant impact categories are: Fossil depletion, Climate change Human Health, Climate change ecosystem, Urban land occupation. High value of Fossil depletion category indicator results from electricity production from fossil fuels (coal, natural gas, crude oil) as well as fuel for transport purposes. The high values in Climate change categories are caused mainly by carbon dioxide emissions from the production of electricity from fossil fuels and lime production for acidity reduction in land reclamation. Within the category Urban land occupation positive and negative impacts are almost equal. The positive impact on the environment (value below zero) results from avoided impact, that is taking from the environmental the old mine tungsten tailings. The negative impact (value above zero) is caused by disposal residues after REE beneficiation process.

![Figure 3. Normalisation step of the production of REE concentrate from New Kankberg tailings.](image-url)
The highest environmental impact is caused by the electricity consumption. This unit process contributes significantly to Climate change categories, Fossil depletion (midpoint categories) and to all endpoint categories (Human Health, Ecosystems and Resources). Another significant unit processes are: disposal of 850 kg of residues after beneficiation (negative impact on the environment) and avoided disposal of tungsten old mine tailings (positive impact). These unit processes contribute mostly to Urban land occupation category and to Natural land transformation (midpoint categories), and to Ecosystem (endpoint category).

Figure 4. Normalisation step of the production of REE concentrate from Covas tailings.

6. Conclusions
The goal of this study is to identify and assess the environmental aspects associated with REE concentrate production from New Kankberg tailings and production from Covas tailings, with the Life Cycle Assessment methodology.

The system boundaries for REE concentrate production from New Kankberg tailings include transport of gold tailings to REE processing facility, 3 stages of beneficiation, then residues deposition and land reclamation. The system boundaries for REE concentrate production from Covas include excavation and transport of old tailings to processing facility, 4 stages of beneficiation, then residues deposition and land reclamation.

The main difference between New Kankberg and Covas models is the fact that the input material for New Kankberg beneficiation (gold tailings) is obtained directly from the gold production facility and sent for further processing. While for Covas the old mining wastes, which represents 30 years underground mining activity focused in tungsten, are excavated from a disposal facility and then processed.

For the REE concentrate production from New Kankberg tailings the highest environmental impact is caused by the disposal of final residues after beneficiation. Avoiding of disposal the initial gold tailings and using them as input material for REE concentrate production balances the negative impact in the Ecosystem endpoint category. The significant midpoint impact categories are: Urban land occupation, Natural land transformation, Fossil depletion. Significant contributors observed in almost
all midpoint impact categories and in all endpoint categories are reagents used for flotation, especially fatty acid.

For the REE concentrate production from the old Covas tailings the highest environmental impact is caused by the electricity consumption. Another significant unit processes are: disposal of residues after beneficiation (negative impact) and avoided disposal of tungsten old mine tailings (positive impact). The significant midpoint impact categories are: Fossil depletion, Climate change Human Health, Climate change ecosystem, Urban land occupation.

It must be noted, that the ENVIREEE project has not been completed yet and not all steps of REE recovery have been fully developed. This article shows results of screening LCA for beneficiation processes for REE recovery from secondary sources. As the ENVIREEE project further develops, more detailed LCA studies will be performed, including other steps of REE recovery: leaching and separation.

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