



Can bioleaching of NIB magnets be an answer to the criticality of rare earths? An ex-ante Life Cycle Assessment and Material Flow Cost Accounting

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ARTICLE INFO

Handling Editor: Kathleen Aviso

Keywords:

Rare earth elements (REEs)
Ex-ante life cycle assessment (LCA)
Waste electrical and electronic equipment (WEEE)
Circular economy
Waste management
Critical raw materials (CRMs)

ABSTRACT

The instability of rare earth elements (REEs) supply chains due to, among others, geopolitical factors brought alternative sources of REEs under the spotlight. Waste from electrical and electronic equipment (WEEE) is considered one of such sources. WEEE recycling is seen as a way not only to mitigate the aforementioned REEs supply risk but also to benefit the environment and society caught currently in a precarious position. Within this context, bioleaching for REEs recovery is gaining attraction, considering that, so far, this process has mainly been used to recover other elements (e.g., Cu, Ni, Zn, Al, Au, Ag). Hitherto, a few lab-scale studies on Nd, Dy, and Pr bioleaching from NIB magnets were identified in the open literature, whereas only one study attempted to perform a simplified LCA analysis of the process. Ergo, this study aims at filling this knowledge gap. For this purpose, the Life Cycle Assessment (LCA) and Material Flow Cost Accounting (MFCA) were performed to assess the process' environmental and economic feasibility after scaling it up from a lab to a pilot scale. Moreover, a break-even analysis was performed to assess the competitiveness of the technology. As the bioleaching of NIB magnets is an emerging concept, this study aimed to identify future process optimisation and development directions. The process was divided into six stages (i.e., demagnetising, shredding, bacteria cultivation, bioleaching, REEs extraction, and oxidation), analysed individually and collectively. Electricity and oxalic acid consumption, together with investment costs, were identified as the main hotspots for future improvement.

1. Introduction

The resource recovery from waste electrical and electronic equipment (WEEE) has been in the spotlight for more than 20 years. Nevertheless, it recently became critical for ensuring the stability of supply chains threatened by geopolitical factors and inevitably increasing demand for critical raw materials (CRMs).

According to the European Commission, CRMs are the materials of economic importance which traditional supply methods (e.g., mining) are becoming increasingly arduous due to, among others, the aforementioned geopolitical or environmental aspects. Therefore, the EU updated a list of 30 CRMs (European Commission, 2020a), from which rare earth elements (REEs) are of particular interest due to their unique

chemical composition along with magnetic and fluorescent properties (Balaram, 2019).

Presently, China is the leading supplier of REEs worldwide (Arshi et al., 2018), and its monopoly has galvanised research on the REEs global market diversification, especially after tightening export quotas in 2011 (Binnemans et al., 2021; Guyonnet et al., 2015). This diversification shall occur in a twofold manner (Binnemans et al., 2021). The first direction stands for looking for other REEs deposits worldwide, for instance, in Madagascar, where the REE-absorbed clays were characterised as structurally analogue to that from China (Borst et al., 2020), or in Europe having substantial REEs resources (Goodenough et al., 2016) which could partially cover European demand for, for instance, Nd (Guyonnet et al., 2015). The second REEs market diversification

Abbreviations: CAPEX, Capital costs; CRM, Critical raw material; HDD, Hard disk drive; LCA, Life cycle assessment; LCI, Life cycle inventory; LCIA, Life cycle impact assessment; MFCA, Material flow cost accounting; NIB magnet, Neodymium magnet; OPEX, Operating costs; PBC, Printed circuit board; REE, Rare earth element; REO, Rare earth oxide; TRL, Technology readiness level; WEEE, Waste electrical and electronic equipment.

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<https://doi.org/10.1016/j.jclepro.2022.132672>

Received 30 October 2021; Received in revised form 31 May 2022; Accepted 10 June 2022

Available online 14 June 2022

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direction is developing additional sources of REEs, such as WEEE (Omodara et al., 2019).

In 2019, 38% of REEs were used for producing permanent magnets (Statista, 2022), including neodymium magnets (NIB magnets) consisting of predominantly (30 wt%) Nd but also Pr and Dy (Yang et al., 2017). NIB magnets are commonly used in consumer electronics (e.g., hard disk drivers (HDDs), loudspeakers), wind turbines, and electric vehicles motors (Lixandru et al., 2017). Therefore, due to their ubiquity, there has been growing research interest in recovering Nd, Pr, and Dy from waste NIB magnets (Frost et al., 2021), and this article follows this trend.

Several potential hydrometallurgical paths of NIB magnets recycling were developed. None of those, however, was commercialised due to high investment and operational costs (Arduin et al., 2020), even though hydrometallurgical recycling of NIB magnets combined with further reuse of the recovered Nd proved to be more economically and environmentally feasible than using virgin Nd (Karal et al., 2021). Although the conventional hydrometallurgical methods have achieved appreciable recovery of REEs (Karal et al., 2021; Tanaka et al., 2013), they require multiple steps to enhance leaching selectivity, expensive pre-treatment techniques and consumption of large amounts of chemicals. Therefore, recent research focused on sustainable hydrometallurgy by, for instance, lowering the process' energy demand or utilising acids with lower environmental impact. As an example (Önal et al., 2017), replaced H_2SO_4 with HNO_3 , thereby lowering the roasting temperature of NIB magnets from 800–1000 °C to 200 °C. In contrast (Reisdörfer et al., 2019), and (Gergoric et al., 2019) performed conventional chemical leaching of NIB magnets using organic acids obtained from agroindustrial waste. An overview of other recycling methods for NIB magnets can be found in Table A1 in Appendix A.

Another emerging research path on REEs recovery from NIB magnets is the metal ions mobilisation through their biological complexing and oxidation by microorganisms, called bioleaching or biohydrometallurgy (Isildar et al., 2019). Biohydrometallurgy is already an established technique used to extract metals from their ores, for instance, for about 10–20% of global Cu production (Auerbach et al., 2019). However, only a few studies on bioleaching from NIB magnets were performed hitherto (Dev et al., 2020). For this reason, there is still a need for fundamental research on the process to comprehend it and thus be able to control it fully (Isildar et al., 2019). Notwithstanding that, NIB magnets bioleaching is gaining attraction due to its higher selectivity towards chosen metals than chemical leaching (Isildar et al., 2019). Given this, the bioleaching of NIB magnets was selected as a subject of this study.

A thorough literature review revealed a knowledge gap on the scale-up evaluation of REEs recovery via bioleaching. The identified studies were performed on a laboratory scale (Auerbach et al., 2019; Marra et al., 2018); however, only scaling up the process enables comparing its environmental performance with that of established technologies (Villares et al., 2017). Moreover, following (Marra et al., 2018), evaluating the scaled-up process based on the results of lab-scale studies allows guiding the further technological progress by providing information on possible directions of the process design improvements. This is why the environmental assessment of emerging technologies using LCA has recently gained attention (Moni et al., 2020), often referred to as ex-ante LCA (Bergerson et al., 2020). In this case, LCA enables the technology assessment with systematic rigour and long-term view before it becomes fully developed and well established commercially. It also allows the acceleration of technology's maturation towards environmentally preferable formulations.

In the area of WEEE treatment (Villares et al., 2016), performed an ex-ante LCA of a bioleaching process for the recovery of Cu from printed circuit boards (PCBs). The authors modelled the lab-scale bioleaching process using primary data from laboratory experiments and performed an attributional LCA of this system. Eventually, the system was scaled up to an industrial scale, and its potential environmental impacts were determined using different scenarios. Schulze et al. (2018) conducted an ex-ante LCA of an early-development process of REEs extraction from

NIB magnet scrap using molten salt electrolysis, thus identifying potential environmental hotspots and the potential impacts of this secondary REEs production route. Recently (Karal et al., 2021), conducted an ex-ante LCA of the hydrometallurgical recovery of Nd from NIB magnets to investigate possible environmental effects and cost of the process. System models were scaled up from a lab to a pilot scale, and the LCA results were compared to the conventional way of NIB magnets production. However, only one study on the LCA of NIB magnets bioleaching was identified in the open literature (Auerbach et al., 2017), but it did not consider the scaling up of the process.

Given the identified knowledge gap, an attributional ex-ante LCA of the scaled-up (from lab to pilot-scale) process of the REEs bioleaching from waste NIB magnets was performed in this study. Three REEs, namely Nd, Dy and Pr, were considered. Furthermore, Material Flow Cost Accounting (MFCA) was performed to assess potential saving spots within the process. Following (Schmidt, 2015) and (Nakano and Hirao, 2011), combining LCA with MFCA allows presenting environmental and economic facets of the technology and, consequently, enhances improvements in both of those aspects simultaneously. Such an interdisciplinary approach is also technically possible as both LCA and MFCA are based on similar material understanding and methodology (Rieckhof and Guenther, 2018).

To sum up, the study attempts to answer whether the NIB magnets bioleaching can be part of the answer for REEs criticality. Therefore, the study's main objective was to assess the environmental impacts of the REEs bioleaching from waste NIB magnets and to identify possible ways of technology improvement from ecological, monetary and physical points of view. To the best of the authors' knowledge, this is the first study on LCA and MFCA of a pilot-scale NIB magnets bioleaching presented in the open literature.

2. Material and methods

The maturity of the technology and market into which technology would be deployed is a critical defining factor of the emerging technology assessments (Bergerson et al., 2020). Some companies have already started developing technology for REEs recycling from NIB magnets, and several EU projects address this topic. Indeed, as recently reported in (CEWASTE Project Final Report, 2021), REEs recycling from NIB magnets is technically feasible, but the end-treatment technology readiness level (TRL) is probably below 9. Therefore, there is still space for further development in this area. Considering this and the knowledge gap on NIB magnets bioleaching, the authors claim that the TRL of the technology assessed herein is 4, given that the validation of integrated prototype in lab environment has been performed.

As far as market maturity is concerned, while WEEE treatment is a mature market, REEs recycling is not. In particular, NIB magnets are recycled at a level below 1% of the total amount produced globally (Lixandru et al., 2017) because of their complex composition. However, recycled REEs would compete with virgin REEs having a well-established market already. Consequently, the assessed technology would be deployed into an already existing mature REEs market. Therefore, following the classification reported by (Bergerson et al., 2020), the case within this study can be classified as an emerging technology in mature markets.

2.1. Scaling-up procedure

According to the framework described by (Tsoy et al., 2020), the technology scaling up was done following a three-step procedure: 1) projected technology scenario definition; 2) preparation of a projected LCA flowchart; and 3) projected data estimation, mainly performed through manual calculation. The data estimation step was performed in iteration with the second step.

The data on bioleaching used within this study is based on the lab-scale experiments, a simplified plant flow diagram, and a preliminary

LCA with cost assessment performed by (Auerbach et al., 2017, 2019). As only data from laboratory experiments is available, the five-step framework described by (Piccinno et al., 2016) was adopted for processes involving using chemicals as it helps to scale up chemical production processes for LCA studies. The framework suggests designing a simple plant flow diagram based on the lab protocol, which includes all the process steps with quantities of used chemicals. Subsequently, each process step is scaled up, including the linkage and consolidation of the in- and output data of all the involved process steps. All the obtained results are used in the concluding step to perform the LCA.

Following (Villares et al., 2016), the scaled-up scenario for the plausible bioleaching development targeted a medium scale as an extension of a waste management facility since a bioleaching plant could be incorporated on such facilities' sites than being implemented in a stand-alone large-scale facility. Therefore, in this study, the pilot-scale process was designed and based on the case of Stena Technoworld's WEEE recycling plant located in Sweden and described in (Lixandru et al., 2017). HDDs were assumed as the source of NIB magnets because: i) they are the largest NIB magnets application among household goods; ii) they are often readily accessible, considering general electronic scrap (Sprecher et al., 2014), and iii) because of the constant rapid turnover of computers (Walton et al., 2015). At the Stena's plant, approx. 2.5 t of HDDs are collected every month from a WEEE input of 1000 t/month. This corresponds to 27.5–102.5 kg of NIB magnets per month (Lixandru et al., 2017). This mass range was assumed further in the study as a minimum and maximum capacity of the bioleaching plant.

2.2. The case study of REEs recovery from waste NIB magnets using bioleaching: the scaled-up system

Fig. 1 shows a general flow chart of Nd, Dy, and Pr recovery from WEEE using bioleaching. LCA and MFCA analyses were performed for the processes within system boundaries.

WEEE might contain toxic compounds (e.g., epoxy-coated substrate) for microorganisms used in the bioleaching process, resulting in inhibition of their activity and, consequently, decreased bioleaching efficiency (Isildar et al., 2019; Liang et al., 2010; Marra et al., 2018). For that reason, manual disassembling of WEEE containing NIB magnets is advisable. Moreover, shredding of WEEE without removing NIB magnets might result in significant material losses (Sprecher et al., 2014) and difficulties with the up-concentration of REEs (Lixandru et al., 2017; Yang et al., 2017). Accordingly, manual disassembling offers the highest environmental benefits due to significantly reducing the amount of NIB magnets wasted (Arshi et al., 2018; Sprecher et al., 2014). Also, the manual dismantling of HDDs benefits the recycling of their other components, such as printed circuit boards and aluminium (Sprecher et al., 2014; Talens Talens Peiró et al., 2020).

Given the above, the study case includes WEEE disassembling and manual sorting out NIB magnets, following the study of (Lixandru et al., 2017) based on the case of Stena Technoworld's WEEE recycling plant. In this recycling plant, WEEE are manually disassembled, and pieces containing NIB magnets (e.g., HDDs, loudspeakers from laptops and screens) can be separated. The average composition of the NIB magnet from HDDs is given in Table A2 in Appendix A.

Prior to the processing of NIB magnets, their demagnetising is

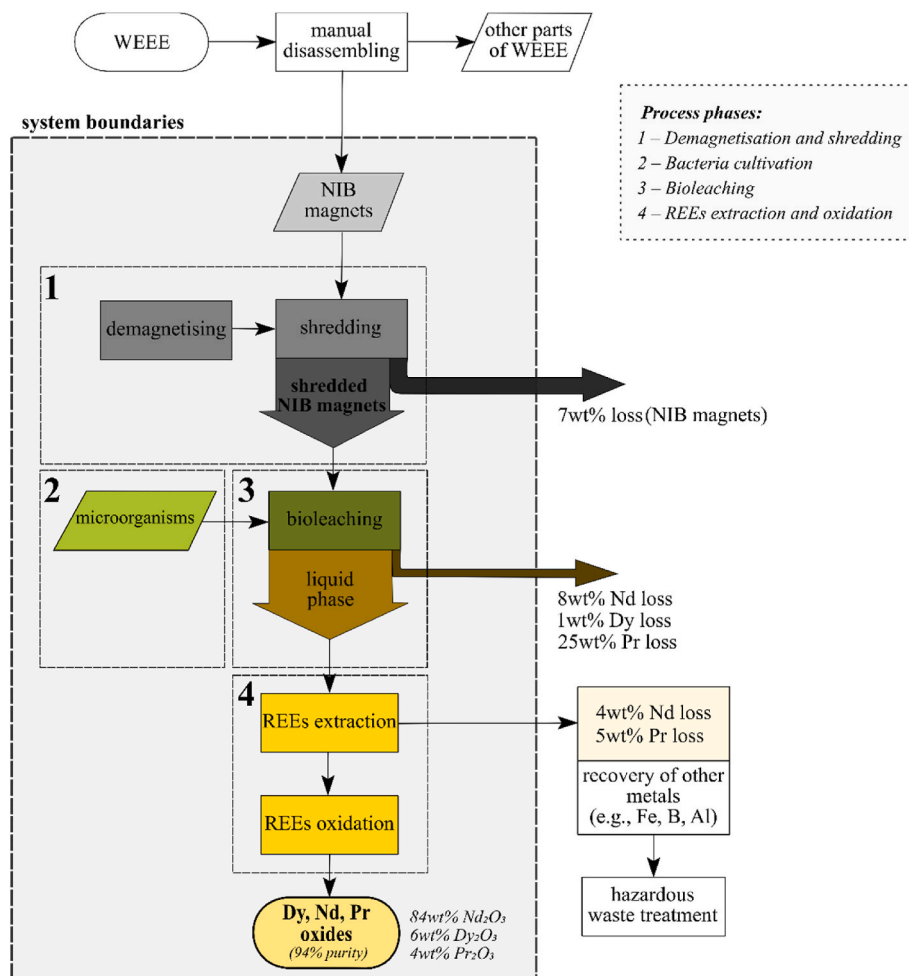


Fig. 1. A general flow chart of REEs recovery using the bioleaching process (the losses of Nd, Dy and Pr are given referring to their initial contents).

necessary for safety (Bahl et al., 2020) and practical reasons (i.e., enabling collecting of the powder after shredding) (München and Veit, 2017; Walton et al., 2015). There are several NIB magnets demagnetising ways, one of which is heating them to 350 °C in a furnace. Following (Karal et al., 2021), the energy consumption of the demagnetising process per 1 kg of the magnets is 0.075 kWh/kg, and such value was taken into account in this study. Afterwards, the demagnetised feedstock is shredded. The feedstock size reduction is beneficial for bioleaching efficiency (Auerbach et al., 2019); however, it results in a 7 wt% loss of the initial material mass (Karal et al., 2021), which was taken into account in this study as well.

As mentioned before, the current bioleaching analysis is based on the works of (Auerbach et al., 2017, 2019). Following (Auerbach et al., 2017, 2019; Marra et al., 2018), bioleaching occurs primarily due to microorganisms producing acids and only partially because of direct microorganisms-feedstock interactions. Therefore, according to (Auerbach et al., 2019), scaling up the process to 100 l is plausible. Additionally, to enhance the flexibility of the installation, it is advised to build a battery of reactors. These reactors would work simultaneously with the possibility of powering them on and off, depending on the capacity needed.

The assumed capacity of bioleaching plant varies between 27.4 and 102.5 kg of NIB magnets per month (Lixandru et al., 2017). To process this amount of waste, a set of 4 stirred reactors (one spare reactor), each with a volume of 100 l, is necessary, according to the process parameters presented in the study of (Auerbach et al., 2019). Furthermore, following (Auerbach et al., 2019), it was assumed that processing 7.8 kg of NIB magnets powder results in around 2 kg of Nd, Dy and Pr concentrate with a purity of 95% after extraction using oxalic acid (with mainly Fe impurities of approx. ~4.5%). *Leptospirillum ferrooxidans* (DSM 2705) were used for bioleaching (bioleaching mechanism - acidolysis (H₂SO₄) and redoxolysis (Fe³⁺)). Bacteria preparation lasted 13 days in (Auerbach et al., 2019); however, the authors claimed that this process might be shortened to 4 days when using shake batch reactors and increasing inoculum quantity. Therefore, in this study, an incubation period (the duration of the bacterial growth process) of 5 days was assumed (4 days with 1 day as a safety margin). The composition of the bacteria medium was assumed following (Auerbach et al., 2019) and guidelines from DSMZ (German Collection of Microorganisms and Cell Cultures). In the study of (Auerbach et al., 2019), the leaching process occurred primarily during the first few hours after loading the feedstock to the reactor; therefore, 1 day (including a safety margin) was assumed as the duration of the bioleaching process. Afterwards, REEs extraction from the liquid phase was necessary, using a concentrated oxalic acid and subsequent precipitation (Auerbach et al., 2019). The extraction yielded a mixture of REOs (rare earth oxides) with a purity of 94%. 1 day (including a safety margin) was assumed as the duration of the extraction step. Eventually, the extracted REEs were subjected to oxidation in a muffle furnace.

2.3. The Life Cycle Assessment and the circularity approach

The LCA was performed following ISO 14040 and ISO 14044 standards (ISO 14040, 2006; ISO 14044, 2006). LCA was applied using the 4 phases of goal and scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA), and interpretation. An attributional LCA was performed to quantify the potential environmental impacts of recovering Nd, Dy, and Pr from spent NIB magnets and identify hotspots (i.e., the key processes or materials that have the most significant impact on the environment).

Following (Cinelli et al., 2014; Villares et al., 2017), three main problems can emerge in performing the ex-ante LCA, namely: i) difficulties in defining the goal and scope of the LCA at such an early stage; ii) uncertainty involving the process data due to its lack or poor quality, which results in dubious potential environmental impacts; iii) the establishment of an accurate confidence level in data interpretation. In

order to tackle these issues, an extensive literature review was performed, and the results of several studies were cross-checked to improve the quality of obtained data. Additionally, the study of the results' sensitivity to the variation of the system model and the two most relevant parameters was performed to enhance the correctness of data interpretation.

The software SimaPro® 8.5 was used for this work. LCA database was Ecoinvent v3.4, modified and licensed by SimaPro. The selected databases were both "Allocation at point of substitution (APOS)" and "Allocation, cut-off by classification". The sources used for life cycle inventory were summed up in Table 1, and a more detailed description was included in Appendix B.

LCIA was carried out at the midpoint level by applying CML-IA baseline version 3.05 characterisation factors included in the Ecoinvent v3.4 database. It consists of the following categories: Abiotic depletion, Abiotic depletion (fossil fuels), Global warming (GWP100a), Ozone layer depletion (ODP), Human toxicity, Fresh water aquatic ecotoxicity, Marine aquatic ecotoxicity, Terrestrial ecotoxicity, Photochemical oxidation, Acidification and Eutrophication.

The functional unit used (a reference basis) is the quantified function provided by the product system, namely the recovery of a mass of REEs concentrate between 6.78 and 25.27 kg per month (equal to the mass of REOs, which ranges between 7.9 and 29.4 kg per month) from WEEE treated at the plant gate.

The system boundary was from cradle to gate. The cradle is the end-of-life EEE (when it becomes WEEE) and enters the recycling plant. Extraction and emissions from previous life cycle phases were out of system boundaries, as well as impacts of waste collection (Fig. 1). The impacts of facility and infrastructure were not included in the system boundaries.

The geographical reference is Sweden. Thus, data for Sweden were applied when available in the database (e.g., energy mix). Otherwise, European regional data were used and finally, by default, any data available in the database. Where no information was available for specific chemicals, proxies were used.

Since no data were available for the treatment of the bioleaching solid residue, it was considered waste and cut off, following (Karal et al., 2021; Villares et al., 2016).

2.4. MFCA and break-even analysis

MFCA was applied, following the ISO 14051:2011 standard (ISO 14051, 2011). According to the standard, the objective of MFCA is to motivate and support the efforts of organizations to enhance both environmental and financial performance through improved material and energy use by linking physical and monetary data. This aligns with this study's goals.

Table 1

The data sources for LCI (foreground and background).

Treatment Stage	References
Disassembly Demagnetisation	Lixandru et al. (2017) (Karal et al., 2021) (electricity consumption); Ecoinvent (market for electricity)
Shredding	Primary data (power and capacity of the shredder); (Karal et al., 2021) (shredding efficiency); Ecoinvent (market for electricity)
Bacteria cultivation	Primary data (power consumed by the bioreactor; amount of chemicals); (Auerbach et al., 2017) (bioreactor capacity); Ecoinvent (market for electricity; market for part of chemicals used)
Bioleaching	Primary data (power and steam needed by the reactor); (Auerbach et al., 2017) (amount of magnet powder in input); Ecoinvent (market for electricity and steam)
REEs extraction and oxidation	Primary data (power needed by the reactor); (Auerbach et al., 2017) (amount of concentrate and oxides); Ecoinvent (market for electricity);

The same system boundaries (Fig. 1) and geographical reference (Sweden/Europe) as for the LCA were applied. Differently from the LCA, however, the time reference is one year. Five quantity centres were identified, i.e. demagnetisation, shredding, cultivation of bacteria, bioleaching, REEs extraction and oxidation. Operating costs (OPEX) for material, energy and labour were considered, as well as capital costs (CAPEX) of the machinery and its maintenance. What has to be noted, however, is that, following the MFCA methodology, CAPEX-related costs (i.e., depreciation and maintenance) together with labour costs are collectively referred to as the so-called 'system costs'. A straight line methodology was applied to calculate the yearly depreciation of machinery, assuming a 10-year product life and residual value equal to 10% of the initial price. Maintenance costs were assumed to be equal to 2% of the price for each piece of machinery. Labour costs were allocated to each quantity centre, considering the percentage of maintenance costs as an allocation factor. In the case of machinery used in more than one centre, the CAPEX and maintenance costs were allocated to each quantity centre, considering the process time. Eventually, the losses of REEs imputable to the process inefficiencies were also economically evaluated and allocated to costs due to material loss.

Potential incomes from selling the REOs obtained as the process output were also assessed. The obtained REOs are characterised by lower purity (94%) than these available currently on the market and are a mixture of Nd, Dy, and Pr. In NIB magnets production, the mixture of Nd and Pr is commonly used as an alloying agent as their separation is challenging due to their similar chemical properties (Sprecher et al., 2014); therefore, only the separation of Dy from the Nd and Pr mixture would likely be necessary before using the obtained REOs in the production of NIB magnets. However, considering the current immaturity of the recycled REOs market, it is hard to predict the reasonable price of recycled REOs; therefore, in this study, the current market prices of REOs (purity >99%) were used.

Data were gathered through market analyses (e.g., discussions with experts, company offers, official electricity prices). The exact data sources can be found in Table A9 in Appendix A.

According to (Bergerson et al., 2020), a system-wide analysis incorporating some market aspects can be helpful despite the uncertainty. Break-even analysis could help to define thresholds of performance required for the technologies to be competitive.

Moreover, the sensitivity of the analysed system to the amount of NIB magnets treated was studied. In this additional scenario, the treatment of NIB magnets from loudspeakers, laptops and PC/TV screens was considered. It is estimated that approx. 28.5–70 kg of the NIB magnets could be recycled from loudspeakers in laptops and PC/TV screens collected in Stena's recycling plant (Lixandru et al., 2017). Thus, these amounts and that of the NIB magnets from HDDs equal 56–172.5 kg per month. This means that to process the feedstock, a set of 6 reactors (100 l each) is necessary in this case. Then, the mass of recovered REEs concentrate ranges between 13.8 and 42.5 kg per month (equal to a mass of REOs between 16.1 and 49.5 kg per month). Eventually, the variation of costs and incomes in the scenario was analysed.

3. Results and discussion

The results of performed LCA and MFCA, together with their discussion, are presented herein. Additionally, the concluding discussion placing the study's result in a broader context is included at the end of the section.

3.1. LCA results

Firstly, the differences in the results imputable to the choice of the system model (i.e., cut-off by classification or APOS) were assessed. As shown in Tables A3–5, the use of the APOS system model causes a slight increase in all the impact categories, except for *Acidification*, while the impact category which increases the most is ODP. All the process phases

would display only slight variations in each impact category (always between –3.61% and +3.05%). Thus, the cut-off by classification system model was adopted in the remainder of the analysis.

The environmental impacts of each process phase (i.e., demagnetisation and shredding, cultivation of bacteria, bioleaching, REEs extraction and oxidation) are shown in Fig. 2. An exact data can be found in Table A6 in Appendix A. The allocation of impacts to the analysed REEs can be performed using two criteria: mass and economic values. Considering the mass of the extracted elements, 89% of impacts are imputable to Nd, 7% to Dy and the remainder 4% to Pr. On the other hand, considering the prices (reported in Table A9 in Appendix A), 14.9% of impacts are imputable to Nd, 70.3% to Dy and 14.8% to Pr.

Two process phases, namely *Cultivation of bacteria* and *REEs extraction and oxidation*, dominate in all analysed environmental impact categories. The *Cultivation of bacteria* impact exceeds 68% for 3 categories: ODP, Freshwater aquatic ecotoxicity, and Terrestrial ecotoxicity. This is likely related to producing chemicals used as a bacteria medium (e.g., hydrochloric, boric, sulphuric acids, or ammonia), which results in, among others, electricity and water consumption. On the other hand, the *REEs extraction and oxidation* impact is the highest (around 45%) for Abiotic depletion (fossil fuels) and Acidification. This can be primarily linked to citric acid production (commonly done via submerged fermentation) - more precisely to the feedstock and energy consumption (steam, natural gas and electricity) during the process (Parsons et al., 2019; Wang et al., 2020).

The results of the preliminary LCA described in (Auerbach et al., 2017) showed that bioleaching accounted for 74–75% of the impacts in various impact categories, while cultivation of bacteria accounted for 25–26% by adopting the ReCiPe characterization method. The environmental impacts in these two process steps are predominantly due to the electricity demand of the bioreactor. Despite the different characterisation methods, the results confirm that the impacts of the bioleaching become lower after the scale-up. However, electricity consumption still plays a significant role: the analysed environmental burdens can be predominantly linked to the electricity consumption and the production process of citric acid (oxalic acid surrogate) (Fig. 3). An exact data can be found in Table A7 in Appendix A. However, it has to be taken into account that the impact of citric acid production can also be linked to electricity consumption (Parsons et al., 2019; Wang et al., 2020), but the process data is aggregated in Ecoinvent. Therefore, it is not possible to determine the exact contribution of individual parameters (e.g., electricity or natural gas consumption, steam production) to the total environmental burden of the process. Nevertheless, part of the citric acid's contributions to the overall environmental impact could be linked to electricity consumption, thereby increasing the values for electricity shown in Fig. 3.

Given the above, it can be concluded that electricity consumption is a primary parameter influencing the environmental performance of the analysed bioleaching process. As already reported by (Auerbach et al., 2017), the environmental impacts of the two significant process steps (i.e., bacteria cultivation, REEs extraction) are predominantly due to the electricity demand, which includes: stirring, temperature control, system control, adjustment of the pH value (pumps), supply air and exhaust air control, as well as cleaning and sterilisation after the end of the process. Similarly (Karal et al., 2021), identified electricity as one of the main parameters highly contributing to the environmental burdens of hydrometallurgical REEs recovery from NIB magnets.

Additionally, a sensitivity analysis was performed regarding the change in electricity and oxalic acid consumption. The results are shown in Fig. 4, and the exact values are shown in Table A8 in Appendix A. The results confirm the previous observations from Fig. 3 that electricity consumption has a significantly stronger influence on all analysed environmental impact categories than oxalic acid.

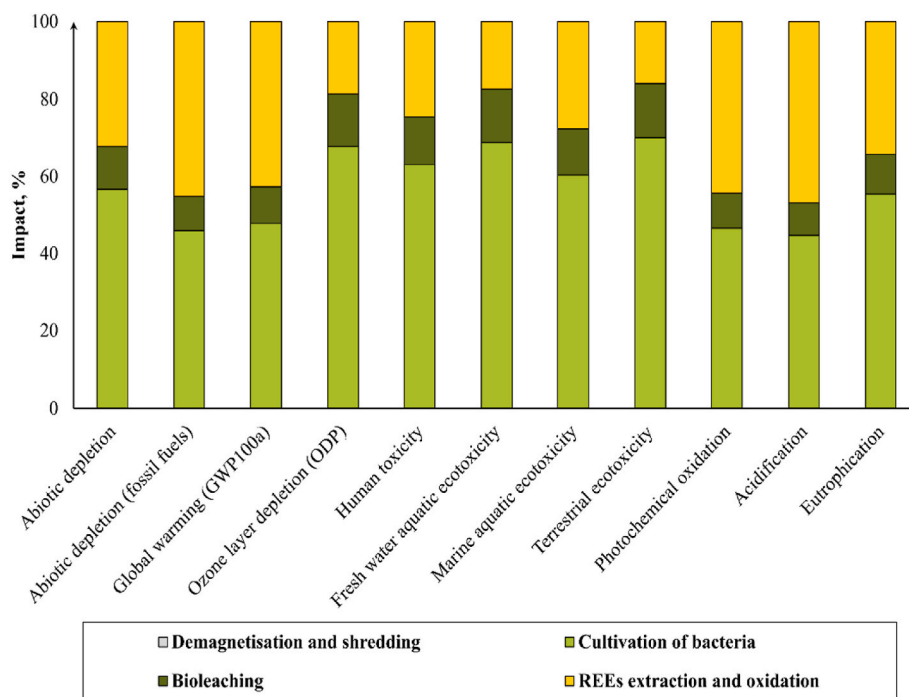


Fig. 2. The process' phases' contribution to various environmental impact categories, cut-off by classification system model.

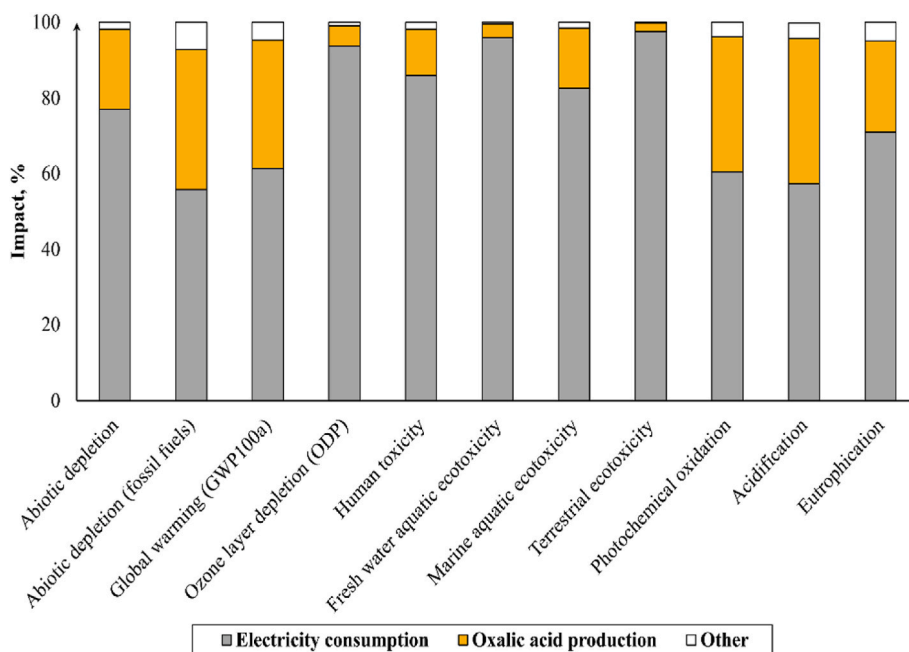


Fig. 3. The contribution of electricity and oxalic acid to the process' environmental burdens.

3.2. MFCA and break-even analysis results

The results of the MFCA are shown in Fig. 5. The exact values are shown in Table A10 in Appendix A. In general, material costs dominate among the analysed costs (74% of the total costs), followed by system costs (20% of the total costs). As the materials (i.e., bacteria medium and oxalic acid) are used in the *Bacteria cultivation* and *REEs extraction and oxidation*, these processes are responsible for most of the costs (52% and 44%, respectively). Moreover, the increased share of *Bacteria cultivation* in the total costs is related to using bioleaching reactors within this process. The costs of those reactors are linked to the depreciation costs,

which constitute around 60% of the total system costs.

As far as REEs losses are concerned, an almost equal amount of them is imputable to *Bioleaching* and *Shredding* (each around 42% of the total material loss costs), while the remaining 15% is allocated to *REEs extraction and oxidation*. What has to be noted is that reducing the REEs loss might result in a considerable increase in the process income by 17%.

To sum up, the analysis points out that the process is not profitable at this stage, mainly because of the high costs of materials used and the necessary equipment. Indeed, the results show a negative cost of 202 € per kg of REOs. Therefore, optimising the bacteria medium composition

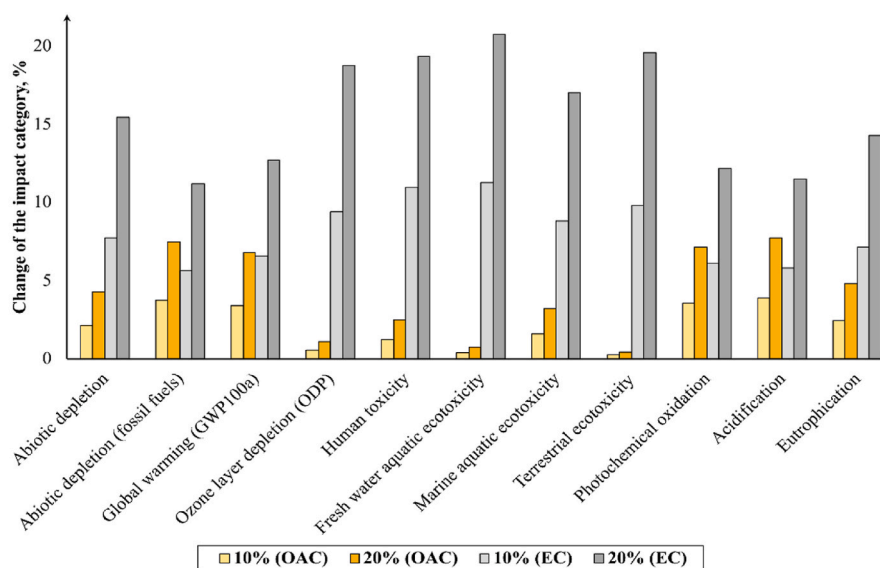


Fig. 4. The sensitivity analysis results for a 10% and 20% increase in oxalic acid* (OAC) and electricity consumption (EC); * - citric acid as an oxalic acid surrogate.

and decreasing the investment costs shall be the first step on the path to enhancing the installation's economic feasibility.

Additionally, the impact of the mass of magnets processed on the process' economic feasibility was investigated. Fig. 6 presents the MFCA results for the second scenario with increased installation capacity (172.5 kg/month). The exact values are shown in Table A11 in Appendix A.

In this case, the cost structure is similar to the base scenario, as the material and system costs represent 79% and 17% of the total costs, respectively. Compared to the base scenario, with a 70% increase in the installation's capacity, system and material costs increase by 33% and 70%, respectively. It is related to the fact that two additional bioleaching reactors are taken into account in the second scenario, which considerably increases the depreciation and, consequently, system costs. In the case of material costs, they depend linearly on reactors' volume. Thus, an increase in the processed magnets' mass, number of reactors and process cycles needed to process magnets causes an increase in material costs. Simultaneously, income from selling the REOs increases by 64% but the costs related to material loss increase by 68%. The lower increase rate of income than that of costs related to material loss points out that the process's inefficiencies are another critical factor, after material costs and CAPEX, for the economic feasibility of the process. This means that increasing the installation's capacity does not significantly improve its economic performance (−188 € per kg of REO) when keeping the same technological configuration of the process. Therefore, the process redesign might be necessary when scaling up, for instance, by increasing bioreactors volume or optimising bacteria medium composition, thereby decreasing reactors number or reducing the amount of chemicals used within the process.

3.3. Concluding discussion

This study's relevance lies in the significant supply risk value for Dy, Nd, and Pr, as it is the highest of all the materials evaluated in the CRM list, with a 100% EU import reliance and a high concentration of supply. For these REEs, end-users outside of China will remain vulnerable to China's dominance of the global REEs value chain (mining, oxides, metals, alloys and magnets) in the foreseeable future. Moreover, the rising annual demand for Nd and Dy will significantly exceed global production by 2030, and, after the depletion of historically accumulated reserves, shortages are foreseen in case additional sources of supply are not developed (European Commission, 2020b).

Given the above, the study attempted to evaluate the potential of bioleaching of NIB magnets as the answer for REEs criticality, particularly emphasising its environmental and economic performance. Due to the ex-ante exploratory nature of the study, its aim was not to provide an exact assessment of environmental and economic impacts but to identify and analyse critical factors to guide further process developments and provide the knowledge to improve decision-making.

The highest toll on the environment is taken by electric energy consumption (Figs. 3 and 4), followed by oxalic acid consumption. However, from an economic point of view, energy consumption plays a minor role in process costs, whereas oxalic acid consumption constitutes around 50% of the total material costs for the process. As the process does not seem profitable at this stage of its development, recirculating chemicals (i.e., mainly oxalic acid) could improve the process's environmental and economic performance, as highlighted by (Karal, 2019) as well. Moreover, reusing water used within the process and designing a continuous process (instead of an energy-intensive batch process) could also be considered. Furthermore, reducing material (REEs) loss along the process, with a particular emphasis on magnet shredding, could increase the process's income by up to 20%. Last but not least, the bioleaching process could be coupled with additional processing of other HDD parts, like, for instance, printed circuit boards (PCBs), which, following (Talens Talens Peiró et al., 2020) is profitable considering the cost of PCBs disassembling vs the value of recovered Ag, Au, and Pd.

Moreover, the scaling-up of the process itself suffers several challenges that must be noted. These are, for instance, efficient aeration and temperature control or agglomeration and compaction of waste particles in a large-scale setup during bioleaching (Natarajan et al., 2015), resulting in the pulp density increase and, consequently, bioleaching efficiency decrease (Isildar et al., 2019).

What has to be also highlighted is that certain data gaps exist, especially regarding the recovery of other leached metals and hazardous wastewater treatment (Karal, 2019). As an example, there is a potential to recover Fe (which content in NIB magnets is around 668 g/kg according to (Auerbach et al., 2019)) and utilise it further (Kumari et al., 2021), thereby improving process economic feasibility.

Last but not least, it has to be noted that market trends of both NIB magnets and REEs are very variable. It is expected that the global demand for permanent REEs magnets will increase to 13.2% (worth \$41.41 billion) by 2022 (Research and Markets Dublin, 2016). On the other side, the market prices of Nd, Pr and Dy are very volatile since they are governed both by market forces and political aspects. For example,

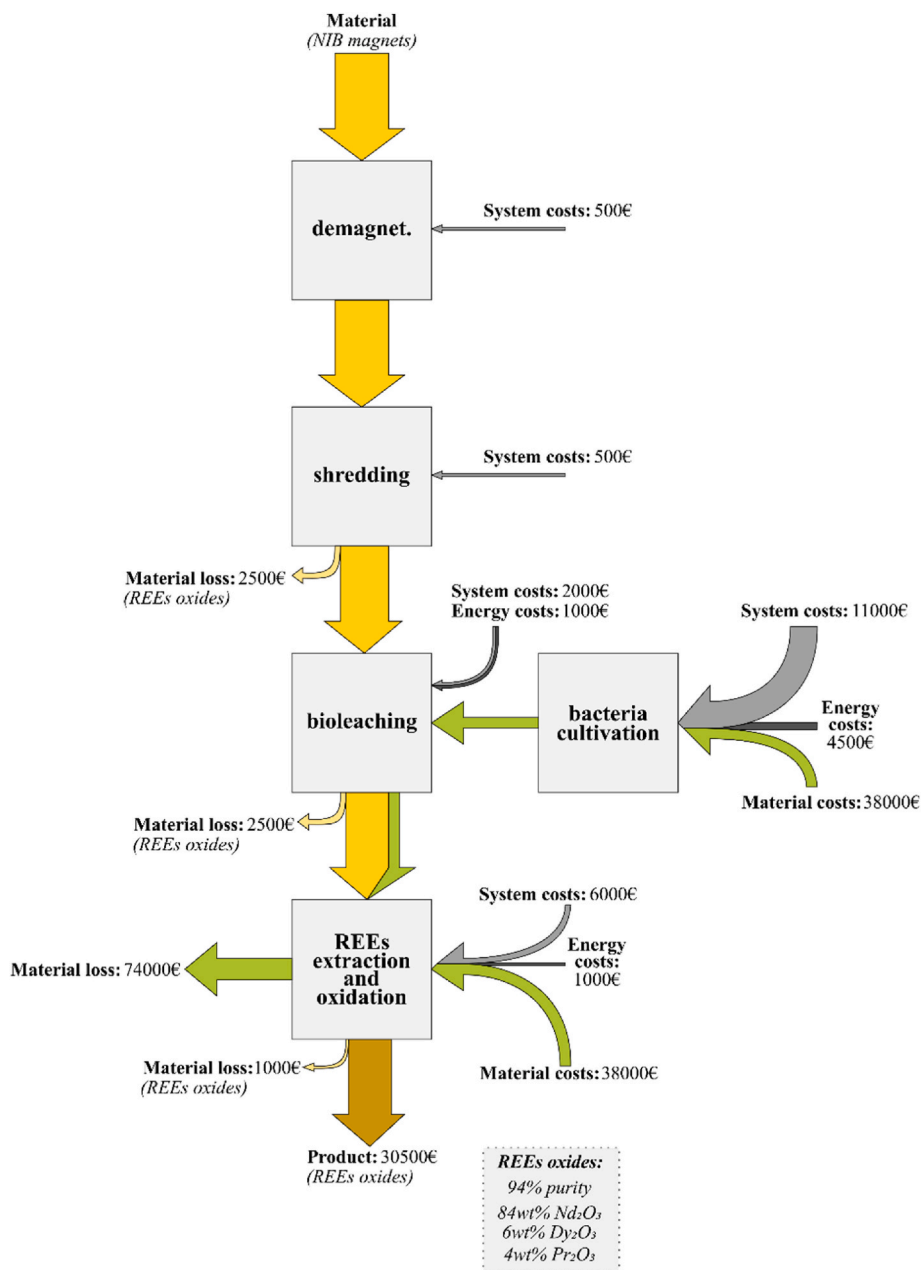


Fig. 5. The MFCA cost allocation for max. capacity (102.5 kg/month of NIB magnets processed); values are rounded to 500 €

in 2010, the export of REEs decreased by 40%, thereby increasing their price up to 1500%. In 2011, however, the export increased again, resulting in REEs prices decreasing to a level below that from 2010 (Auerbach et al., 2019).

To sum up, the potential of NIB magnets bioleaching is predominantly related to meeting the global metal demand for REEs (Natarajan et al., 2015). However, at this early stage of technology development, numerous improvements and optimisation still need to be implemented to enhance process environmental and economic feasibility. Besides, as the CAPEX of such installation is substantial, state subsidies might be necessary to trigger this technology development. This is in agreement with the findings of (Binnemans et al., 2021) that the government intervention in securing a sustainable REEs supply, for example, by setting a minimum recycled content or even through more direct intervention in the market, needs to be discussed.

4. Conclusions

The focus of this study is the evaluation of the environmental and economic feasibility of Nd, Dy, and Pr bioleaching from waste NIB magnets in a circular economy approach; the benefits of recycling other HDDs components, such as PCBs and Al, were not considered. Moreover, due to the early stage of technology development, the LCA and MFCA results cannot be considered definitive. Nevertheless, they serve as valuable preliminary information to assess potential optimisation ways to enhance the process performance and further support policymakers and decision-makers in developing this technological process. Overall, the study’s main findings can be summarised in two points.

Firstly, the LCA shows that electricity consumption should be particularly addressed as it takes the highest toll on the environment. However, the second identified parameter having a considerable ecological impact is oxalic acid production. Additionally, the MFCA pointed out the significance of oxalic acid costs. Therefore, to further

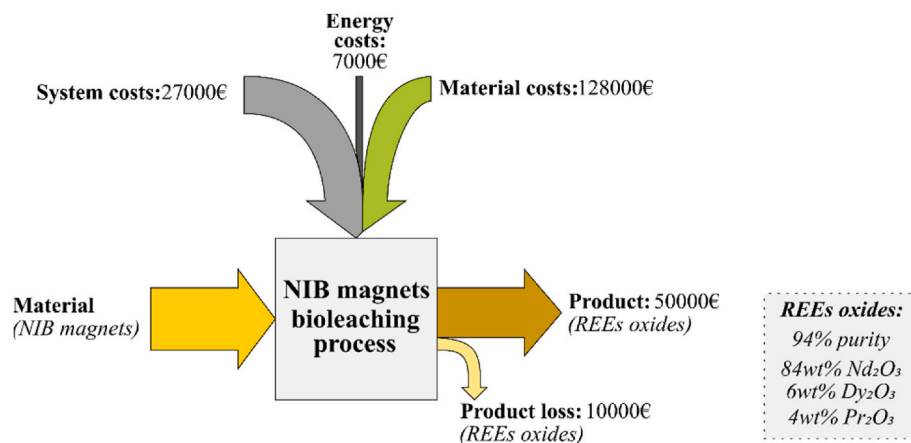


Fig. 6. The general MFCA scheme for the second analysed scenario; values are rounded to 500 €

improve the environmental performance of the process while simultaneously enhancing its economic feasibility, the oxalic acid amount used shall be reduced, for example, by its reuse or recirculation.

Secondly, the considerable share of the total process costs is also related to the cost of the bioreactor itself. Therefore, it is highly possible that state subsidies would be necessary to sustain process operation. Moreover, in the case of scaling up, process reconfiguration might be required because, for instance, increasing bioreactors volume might decrease the investment costs for the installation.

As NIB magnets bioleaching is an emerging technology, further investigations and development are required. Therefore, there are numerous possible directions for future work. However, two main directions can be indicated as to be considered for further investigations: I – further research on the process to validate the concept on a pilot scale and, subsequently, to assess the technology performance with a higher level of confidence; II – further research on recycling of remaining part of the NIB magnets (e.g., Fe) and of other HDD parts.

CRedit authorship contribution statement

Chiara Magrini: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Writing – review & editing.
Katarzyna Jagodzińska: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

The authors would like to thank R. Auerbach (Fraunhofer Research Institution for Materials Recycling and Resource Strategies IWKS, Alzenau, Germany), Engin Karal and Anna Banach-Wiśniewska (Regional Center of Water and Sewage Management, Tychy, Poland) for fruitful correspondence. Moreover, the authors would like to acknowledge the help and relevant feedback from the organisers, participants, and our colleague Mariana Gazire Lemos from the course "Circular Economy and Raw Material Competence for Sustainable Production" by EIT RawMaterials Academy.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2022.132672>.

[org/10.1016/j.jclepro.2022.132672](https://doi.org/10.1016/j.jclepro.2022.132672).

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