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Environmental impacts and carbon footprint of high-value recycling of NdFeB scrap under life cycle assessment

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Abstract: The NdFeB scrap, as a representative solid waste of rare earths, possesses significant recyclable value. This study focused on NdFeB waste and investigated the environmental impacts of pyro- and hydro-metallurgical process (PH-M process) and its improved version, the pyro- and hydro-metallurgical improvement process (PH-Mi process). The results demonstrate that, although the PH-Mi process consumes higher amounts of energy, electricity, and chemicals compared to the PH-M process, it is more environmentally friendly and economically efficient (i.e., net profit increased by 34.12%). To quantify and compare the environmental performance of the two scenarios, life cycle assessment methodology was applied. It is concluded that the PH-Mi process is superior to the PH-M process for eutrophication potential (EP) and the total environmental impacts. In comparison with PH-Mi process, PH-M process exhibits a certain advantage in terms of carbon footprint due to increased consumption of electricity and chemicals after the technological upgrade.

Key words: NdFeB scrap; molten salt electrolysis waste; life cycle assessment; environmental impact; carbon footprint

1 Introduction

China is the largest producer of NdFeB magnets in the world and the production accounts for more than 90% of world production [1,2]. NdFeB magnets are extensively used in wind power generation, electronic equipment, and other industries for their comprehensive advantages such as high remanence, high coercivity and high magnetic energy [3]. After the equipment was

scrapped, leaving NdFeB scrap not only causes serious environmental pollution, but also leads to the waste of valuable resources. NdFeB scrap contains 30%–40% of rare earth elements (REEs), which can bring considerable financial returns [4–6]. In addition, REEs are important non-renewable strategic resources all over the world, and limited access to REEs resources has already led to the supply risk [7,8]. The recycling industry has seen a business opportunity in the surge of NdFeB scrap, mainly associated with the intrinsic value of the

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rare earth elements present in NdFeB scrap [9,10]. Comprehensive utilization of rare earth secondary resources is an effective way to save resources and prevent pollution. Increasing the recycling and utilization of NdFeB scrap can bring great social, environmental, and economic benefits.

The treatment methods for this waste include the pyrometallurgical process, hydrometallurgical process, electrolytic process, ionic liquid method, hydrolysis process, etc. TIAN et al [11] dissolved NdFeB waste with 6 mol/L HCl and added tartaric acid and hexamethylenetetramine (HMTA) chelating agent. The results showed that the purity of rare earth products recovered reached 96% and the recovery rate exceeded 90%. UDA [12] used carbon powder and excessive FeCl_2 as chlorination agents at 800 °C to effectively promote the chlorination reaction of waste Nd–Fe–B materials. VENKATESAN [13] found that Fe^{2+} in the solution was oxidized to the highest degree after 4 h at current of 1.2 A. The ionic liquid is a typical new NdFeB scrap recycling process. MATSUMOTO et al [14] studied the extraction recovery of Nd and Dy elements from NdFeB scrap by undiluted hydrophobic ionic liquid. However, there are three common problems in the comprehensive recycling industry of rare earth secondary resources. First of all, the processing process of NdFeB scrap is complicated and inefficient. It is necessary to study the efficient and clean technology of waste solid pretreatment or dissolution of rare earth. Secondly, the recycled product generally is flakes of rare earth oxide with the thickness of 3–5 μm , lacking the control of the purity and morphology. Finally, the wastewater containing rare earth and chloride produced in the recycling process is also a thorny problem.

With the development of the NdFeB magnet, NdFeB magnet products with high contents of Ce, Ho, Tb, and Dy appear [15]. The total amount of REEs and the proportion of REEs in the NdFeB recycled materials have undergone great changes, and the recovery process needs to be improved urgently. Besides, the improvement of the recovery process responds to the call of carbon peak and carbon neutrality [16]. An enterprise in China designed a comprehensive utilization process of NdFeB scrap through a technology upgrade, which is called the pyro- and hydro-metallurgical improvement process (PH-Mi process). In contrast

to conventional technology of the pyro- and hydro-metallurgical process (PH-M process), this technology shows a better data presentation in aspects of product and environmental impacts. The comprehensive utilization process of NdFeB scrap changes the kiln fuel from coal to natural gas, which makes the material nearly fully burned and improves the recycling rate of rare earth metal oxides from 90% to more than 95%. Meanwhile, the extracting stage increases from 488 to 618, and the varieties of rare earth metal oxides increase from 4 to 7 REEs after extraction. According to the operation practice of enterprises, the PH-Mi process is proven to be able to greatly reduce the generation of hazardous waste residue and effectively solve the environmental problems caused by wastewater containing rare earth, thus increasing the economic benefits of enterprises.

Life cycle assessment (LCA) is a valuable tool used to assess the environmental factors and potential impacts associated with products or processes, which is a systematic approach to assess and quantify the environmental benefits of the whole recycling process [17]. In recent years, numerous studies of LCA have been conducted about REEs recovery from the wastes [18,19]. KARAL et al [20] conducted LCA research on the feasibility and environmental performance of the hydrometallurgical Nd metal recycling. CHOWDHURY et al [21] presented an innovative technology with acid-free dissolution, which can reduce global warming impacts by up to 73% compared to the prevailing rare-earth oxide (REO) production routes in China. SCHULZE et al [22] reported a new process for the extraction of rare earths from NdFeB magnet scrap. The study demonstrated that 76% of the GWP 100 years impact is attributable to direct perfluorocarbon (PFC) emissions from the electrolysis process. BECCI et al [23] used a LCA to identify the most sustainable choice from two hydrometallurgical treatments (the first one with HCl and the second one with solid-state chlorination) and a pyrometallurgical technique. As reported in this research, the hydrometallurgical process shows the lowest environmental burden. WANG et al [24] conducted a LCA of three regeneration technology routes for sintered NdFeB magnets using actual industrial data. The LCA results confirm that the environmental impacts of the NdFeB magnets produced by

regeneration technology routes are lower than the virgin sintered NdFeB magnets. However, those of the evaluation of extraction, processing, and utilization is rarely performed on the environmental impacts in a systematic approach with NdFeB scrap enterprise. Accordingly, a LCA case study for REEs recovery from NdFeB scrap was conducted, wherein the potential environmental impact was quantified and the key factors to improve the processes in the technology upgrade scrap recycling were identified. In this study, the significance of the NdFeB scrap enterprise technology upgrade may be enlightening to other waste recycling enterprises.

2 Problem statement and methodology

2.1 Problem statement

The source of NdFeB scrap is extremely complex, and the morphological characteristics are quite different. In this work, NdFeB scrap came from the whole life cycle of NdFeB magnet production until device failure. In the process of NdFeB magnet processing, a large amount of machining debris, rib stock, and sludge debris are generated, and there are also residual raw materials in the process of milling, pressing, sintering, and finished product testing. These wastes are the main sources of processing recycled materials, which account for 20%–50% of the amount of NdFeB scrap [25]. NdFeB waste also includes obsolete products that flow into the secondary utilization market after the products exhaust the service life. In those complex systems of NdFeB scrap, a few new treatment processes such as hydrogenation disproportionation desorption and recombination (HDDR) processing [26], bioleaching processing [27], and electrochemical extraction [28] are probably disabled. However, the traditional recycling process such as hydrometallurgical or pyrometallurgical process has problems of high pollution and high energy consumption. Driven by the requirements of China's environmental protection policies, the traditional recycling process urgently needs technical iteration.

PH-M process is a process suitable for industrial production developed by combining the advantages of pyro- and hydro-processes, which can realize high-value and green recycling of NdFeB scrap. However, the PH-M process mentioned in this study involves a few types of

NdFeB scrap and mainly depends on hard coal as the main energy source. Simultaneously, NdFeB scrap is enriched with high-value rare earth elements, and the amount of scrap gradually increases over the years, which prompts factories to upgrade the original recycling process. Molten salt electrolysis waste of rare earth is enriched with more REEs, and direct emission may also cause greater environmental pollution problems. The PH-Mi process, which utilizes NdFeB scrap and molten salt electrolysis waste as raw materials, effectively addresses the issue of the low secondary utilization rate of rare earth waste material that is currently prevalent. The improved process of PH-Mi provides a bridge for the after-mining extraction and separation unit when necessary, thus increasing the production capacity of the company.

Before the process modification, the PH-M process was economically efficient and eco-friendly, although the usage of traditional energy sources caused large pollutant emissions. Conversely, the modified PH-Mi process enhances the annual disposal capacity of wastes but leads to an elevation in the emission of partial pollutants. A trade-off is always present between pollutant emission and waste direct emission. The fossil energy and raw material in the two processes are diverse. Therefore, the superiority of these two processes cannot be judged entirely by the quantity of pollutants emitted. For better understanding of the comprehensive benefit of these two processes, in this work the environmental performance and sensitivity analyses were performed, and pollutant emissions in CO₂-based footprint were predicted. In addition, the techno-economic in different scenarios was evaluated based on cost and revenue.

2.2 Methodology

The analyses of environmental performance and sensitivity of “4000 t NdFeB scrap” as the functional unit were performed under the PH-M and PH-Mi processes. REEs content of NdFeB scrap can be up to 34.92 wt.%, meanwhile REEs content in molten salt electrolysis waste is 29.12 wt.%. The details of the chemical composition concerning the NdFeB scrap and molten salt electrolysis waste are shown in Table 1. When calculated in terms of REEs units, it is reasonable to consider that the PH-Mi process improves the effective mass yield to 12.5%, compared to the PH-M process. The

environmental impacts of recycling processes are quantified by defining system boundaries and building process models. The process simulations and data estimation are developed to further investigate the technical, economic, and environmental performance. Sensitivity analysis on the main influencing factors is conducted with different scenarios by adjusting the proportion. The projection of future trends on CO₂-based footprint is implemented.

Table 1 Chemical compositions of NdFeB scrap and molten salt electrolysis waste (wt.%)

Composition	NdFeB scrap	Molten salt electrolysis waste
REEs	34.92	29.12
FeO	56.84	14.35
SiO ₂	2.69	25.58
Al ₂ O ₃	NA	7.22
B	1.69	NA
Co	0.26	0.02
Others	3.18	23.71*

NA represents trace or none detected; * stand for containing 15.09% loss of combustion

2.2.1 Research scope

In this work, 4000 t NdFeB scrap was chosen as the functional unit in the PH-M process and the PH-Mi process. In particular, 600 t molten salt electrolysis waste of rare earth was selected as one of the accessories in the PH-Mi process. The processes of raw materials and energy production, waste disposal, and direct emissions of all stages of NdFeB scrap recycling processes are included in the system boundary. The system boundary for the PH-M process includes acidolysis, extraction separation, CoCO₃ purification, precipitation washing, and precipitation roasting. The system boundary for the PH-Mi process includes roasting, acidolysis, molten salt waste pretreatment, extraction separation, and precipitation roasting. Figure 1 illustrates the system boundaries of the PH-M and PH-Mi processes and the assumptions are as follows. The environmental impacts of the plant construction are not included in our work. Additionally, NdFeB scrap and molten salt electrolysis waste are waste products generated in the upstream production process, with no

consideration for environmental impacts. To simplify the calculation, several complex processes are integrated into one step. For example, the extracting stage is increased from 488 to 618 after process improvement, whereas just one unit is considered in the present study.

2.2.2 Life cycle inventory and data sources

Life cycle inventory (LCI) is the basis for examining the environmental impacts of processes [18,29]. In this study, over 1000 data according to all the processes defined in the system boundary have been gleaned from the typical enterprises of NdFeB scrap recycling in China, including resources, energy, materials, waste gas, waste liquid, and solid waste from each sub-process. There are two types of data involved in the research: foreground data and background data.

The foreground data pertain to the materials and energy directly incorporated into the production process. In this study, the foreground data are furnished by the pertinent representative enterprises engaged in NdFeB scrap recycling within Ganzhou city, China. Ganzhou is one of the origins of heavy rare earth in China and is one of the largest rare earth industry clusters in the world. A complete industrial chain has been developed around heavy rare earth including mining, smelting, separation, processing, and recycling, etc [30,31]. It represents a typical situation for NdFeB scrap recycling. The operational data are the foreground data of LCI closest to the ideal conditions, which are always available and reliable for some processes to be modelled [32]. These data encompass comprehensive records of all raw material input data for processes such as acidolysis, extraction separation and precipitation roasting, and the corresponding data with economic value. The background data contain the resource consumption and the environmental impacts of materials and energy in respective manufacturing processes. Environmental impact data including various feedstocks and emissions are obtained as background data from the GaBi database. Notably, the geographical scope is relevant to the accuracy of impact related to energy generation and other inputs, and the background data should select production country data of inputs [33]. Hence, the data are collected from GaBi database, and most of the primary background data are specific to China. Moreover, all other data not specifically cited in this

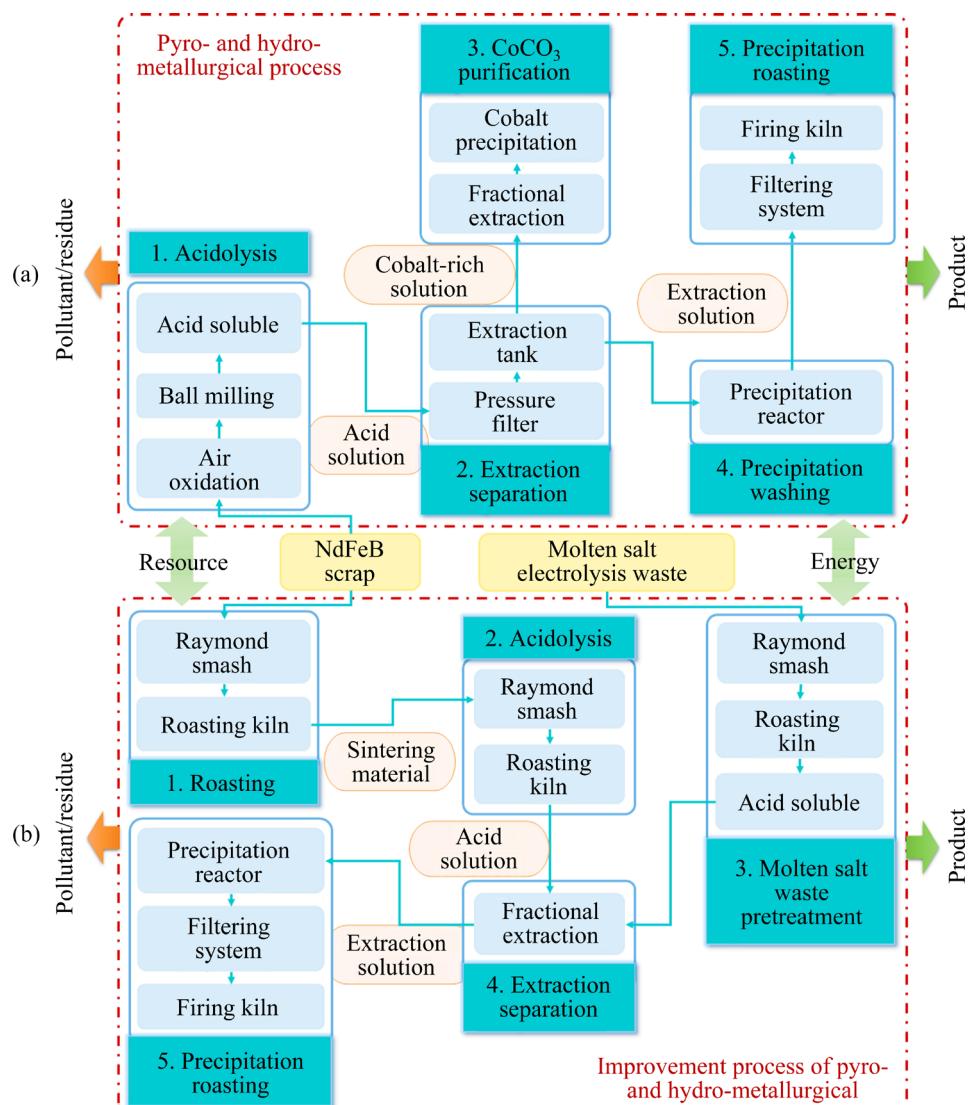


Fig. 1 System boundaries of PH-M (a) and PH-Mi (b) processes

paper are derived from GaBi modeling and calculations. Table 2 presents the LCI of the PH-M process and Table 3 presents the LCI of the PH-Mi process.

Table 2 shows that the direct emissions from the PH-M process include exhaust gases, wastewater, and solid waste. The exhaust gases mainly consist of roasting furnace flue gas, hard coal combustion flue gas, crushing dust, calcination kiln flue gas, and HCl waste gas produced during the acidolysis and precipitation processes. The wastewater mainly includes rare earth counter-extraction organic phase washing regeneration water, rare earth precipitation wastewater, rare earth precipitation washing water, acid mist purification tower wastewater, etc. The solid waste mainly includes acid-soluble slag, three-phase slag,

neutralization slag from wastewater treatment, and boiler ash slag.

The PH-Mi process in Table 3 displays a large input of raw materials and accessories, resulting in a substantial amount in direct emissions. The exhaust gases mainly consist of roasting and combustion flue gases, acid mist from acid dissolution and precipitation processes, and boiler flue gas. The main pollution units are roasting and acidolysis units, with particular attention needed for the environmental impacts of S, NO_x, and other substances. The wastewater mainly includes extractant residual wastewater, extraction washing acid water, extraction saponification wastewater, precipitation mother liquor, precipitation washing water, boiler wastewater, flue gas purification wastewater, and acid gas or waste gas

Table 2 LCI of treating 4000 t NdFeB scrap by PH-M process

Input/output	Matter	Consumption/t	Unit process
Input from nature	Water	14100	Water washing, heating
	Air	21845.70	Air combustion promoter (O ₂)
Input from processes	NdFeB scrap	4000	NdFeB scrap raw material
	Hard coal	720	Charcoal burning
	Electricity	3200*	Electricity machine running
	Sodium hydroxide	150	NaOH extraction
	Calcium hydroxide	1200	Ca(OH) ₂ (98%) neutralization
	Oxalic acid	84	Oxalic acid (98%) precipitation
	Sodium bicarbonate	16	NaHCO ₃ (98%) precipitation
	Hydrochloric acid	6030	HCl (31% in H ₂ O) acidolysis
	P507	10	P507 extraction
Output for products	Kerosene	20	Kerosene extraction
	Pr–Nd oxide	820.80	Pr–Nd oxide (>99%) product
	Dysprosium oxide	72.50	Dysprosium oxide (>99.9%) product
	Terbium chloride	293.70	Terbium chloride (>10% in H ₂ O) product
Direct emissions	Cobalt carbonate	28.30	CoCO ₃ (>60%) product
	Slag (acid-soluble)	3500	Solid waste
	Slag (three-phase)	50	Solid waste
	Slag (boiler ash)	191	Solid waste
	Hydrochloric acid	1.26	Exhaust to air
	Sulfur dioxide	3.97	Exhaust to air
	Flue gas	7.95	Exhaust to air
	Waste gas	22565.70	Exhaust to air (N ₂ >78%, CO ₂ >18%)
	Waste liquid	20640.50	Wastewater to water

* MW·h

purification wastewater. The main pollutants contain heavy metal elements such as Pb, Zn, Ni, Co, Cd, As, and a large amount of F and Cl elements. The main wastewater pollution units are the extraction separation and precipitation roasting units. The solid waste mainly includes waste acid-soluble slag, calcium fluoride precipitation slag, iron removal slag, roasting dust water spray sludge, dust collected during the crushing process, and pretreatment neutralization slag.

Table 4 presents the mass of elements and their respective recovery efficiencies for 1 t raw material, which is calculated based on Eq. (1). The waste materials are NdFeB scrap and NdFeB scrap

with molten salt electrolysis waste in the PH-M process and the PH-Mi process, respectively. It is noticeable that the recovery of the most elements has increased significantly after the process improvement, while the number of recovered elements increased by 60% and the total recycling efficiency rose to 27.09%. Particularly, the recovery efficiency is higher for Nd and Pr in the PH-Mi process from 8.88% and 8.68% to 12.23% and 11.95% respectively because of the process improvement. The lower recovery efficiency of Dy and Co in the PH-Mi process is primarily attributed to the low content of high-value elements in the molten salt electrolysis waste.

Table 3 LCI of treating 4000 t NdFeB scrap and 600 t molten salt electrolysis waste by PH-Mi process

Input/output	Matter	Consumption/t	Unit Process
Input from nature	Water	46870	Water washing, heating
	Air	34087	Air combustion promoter (O ₂)
	NdFeB scrap	4000	NdFeB scrap raw material
	Molten salt electrolysis waste	600	Molten salt electrolysis waste accessory
	Natural gas	1.8×10 ⁶ *	CH ₄ burning
	Electricity	4200**	Electricity machine running
	Hydrochloric acid	8600	HCl (31% in H ₂ O) acidolysis
	Liquid caustic soda	1500	NaOH (>30% and 50% in H ₂ O) extraction
Input from processes	Sodium hydroxide	90	NaOH extraction
	Oxalic acid	1430	Oxalic acid (98%) precipitation
	P507	10	P507 extraction
	Sodium bicarbonate	20	NaHCO ₃ (98%) precipitation
	Sodium chlorate	25	NaClO ₃ (98%) precipitation
	Kerosene	59	Kerosene extraction
	Quicklime	90	CaO (98%) neutralization
	Calcium hydroxide	1650	Ca(OH) ₂ (98%) neutralization
Output for products	Pr–Nd oxide	1300	Pr–Nd oxide (>99%) product
	Dysprosium oxide	34.7	Dysprosium oxide (>99.9%) product
	Gadolinium oxide	53.60	Gadolinium oxide (>99%) product
	Cerium oxide	32.30	Cerium oxide (>99%) product
	Holmium oxide	10	Holmium oxide (>99%) product
	Terbium chloride	312	Terbium chloride (>10% in H ₂ O) product
	Cobalt carbonate	29	CoCO ₃ (>60%) product
Direct emissions	Slag (acid-soluble)	3864	Solid waste
	Slag (calcium chloride)	143	Solid waste (60% in H ₂ O)
	Slag (boiler ash)	72.51	Solid waste
	Slag (iron removal)	723	Solid waste
	Hydrochloric acid	5.80	Exhaust to air
	Sulfur dioxide	0.36	Exhaust to air
	Flue gas	2.29	Exhaust to air
	Nitrogen dioxide	3.23	Exhaust to air
	Water, in the air	2460	Exhaust to air
	Waste gas	36989.20	Exhaust to air (N ₂ >78%, CO ₂ >18%)
	Waste liquid	54196	Wastewater to water

* m³; **MW·h

Table 4 Recycling efficiencies of different elements in two scenarios for 1 t waste

Element	PH-M		PH-Mi	
	Mass/kg	$\eta/\%$	Mass/kg	$\eta/\%$
Nd	88.82	8.88	122.32	12.23
Pr	86.79	8.68	119.52	11.95
Gd	–	–	10.11	1.01
Dy	15.79	1.58	6.57	0.66
Ce	–	–	5.72	0.57
Ho	–	–	1.90	0.19
Tb	3.13	0.31	2.89	0.29
Co	2.10	0.21	1.87	0.19
Total	196.63	19.66	270.91	27.09

$$\eta = \frac{\sum m_{i,\text{out}}}{\sum m_{i,\text{in}}} \times 100\% \quad (1)$$

where $m_{i,\text{out}}$ and $m_{i,\text{in}}$ represent the output quantity of each elemental product and the input quantity of the waste material, respectively.

2.2.3 Environmental impact assessment method

Life cycle impact assessment (LCIA) is an emphasis step of the LCA, which helps to associate each elementary flow or other intervention in LCI with environmental impacts categories. The common LCIA methods are CML, ReCiPe, and TRACI. CML is a problem-oriented approach that focuses on process characterization [34]. CML adopts a midpoint method using European or global average data to reduce the number of assumptions and the complexity, and can comprehensively assess the impact of products on the environment [35]. ReCiPe is a kind of method that combined the midpoint method, endpoint method, and end-point-based approach of Eco-indicator 99 methods [36]. TRACI is developed by the U.S. EPA to provide characterization factors for 10 impact categories, typically used for analysis related to U.S. geographic region [37].

The LCA is implemented by using GaBi. According to the characteristics of industrial production in China, the CML2001 method (2015 revision) is chosen since it is the most comprehensive to reflect the effect of assessment. The CML2001 method (2015 revision) divides the environmental impacts into 12 types. In this study, there are multiple types of environmental impacts due to the

input of various resources, energy, and materials in the process of NdFeB scrap recycling. Therefore, 8 indicators are selected for analysis in this work: abiotic depletion potential (ADP), acidification potential (AP), eutrophication potential (EP), freshwater aquatic ecotoxicity potential (FAETP), global warming potential (GWP), human toxicity potential (HTP), photochemical ozone creation potential (POCP), and terrestrial ecotoxicity potential (TETP). The impacts of other categories are negligible.

2.3 Mass balance and material flow

To further establish the reliability of the LCI, the mass balance of the PH-M and PH-Mi processes is studied, as shown in Figs. S1 and S2 in Supplementary Materials (SM), respectively. The original mass of NdFeB waste is 4000 t which includes 717.59 t of Nd and 416.25 t of Pr. The total inputs of Nd and Pr are 662.05 and 581.32 t in the PH-Mi process. However, the masses in products of Nd and Pr in the PH-M process are 355.27 and 347.14 t with a mass loss of 50.49% and 16.60%, respectively. Similarly, in the PH-Mi process, the masses of Nd and Pr in products are 562.69 and 549.81 t, with a mass loss of 9.54% and 5.42%, respectively. The mass loss may be attributed to measurement errors and chemical reaction loss. However, by comparing the mass loss results between scenarios PH-M and PH-Mi, it becomes evident that non-essential losses are significantly reduced following process improvements.

Figure 2 presents the Sankey diagrams which illustrate the material flow in two types of NdFeB scrap recycling processes, showing the flow direction of the main raw materials and accessories in the production process of Pr–Nd oxide products. With the comparison of PH-M process in Fig. 2(a) and PH-Mi process in Fig. 2(b), it is found that with the increase in the addition of main raw materials and accessories, the yield of Pr–Nd oxide increases by 58.4% of the pre-process improvement. Besides, with the dosage of oxalic acid surged by 1602%, the increase in raw materials and accessories is 15% and 78.22%, respectively. The application of oxalate precipitation method in the recycling of NdFeB scrap can efficiently separate REEs and Fe element. Therefore, the dosage of oxalic acid significantly increases in the PH-Mi process.

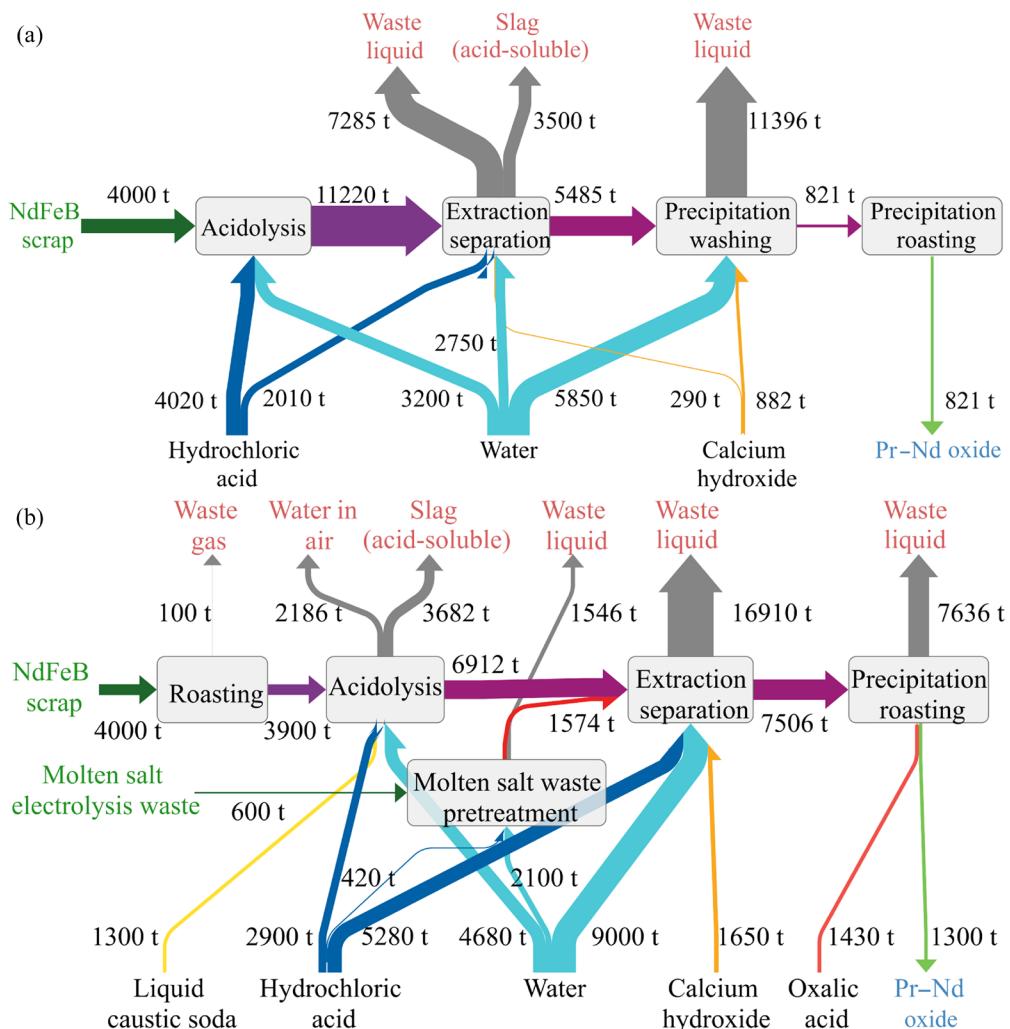


Fig. 2 Sankey diagrams of material flow with PH-M (a) and PH-Mi (b) processes (Green and blue fonts represent raw materials and Pr-Nd oxide, respectively, black font represents accessories, and red font represents three wastes generated during the recycling in NdFeB scrap)

3 Results

3.1 Characterization results

The contribution of various scenarios during resources recovery of waste magnet to the diverse environmental life cycle impact is shown in Fig. 3. The details of the environmental impacts shown in Fig. 3 are presented in Tables S1 and S2 in SM. In the PH-M process, the acidolysis unit consumes 50.4% coal, 33.3% water and 66.6% hydrochloric acid of the whole process in Fig. S1 in SM, thus generating significant environmental impacts, especially noteworthy for ADP, AP, EP, and GWP. Regarding ADP, the characterization value is 2.77×10^8 MJ, which represents 43% of the total. The energy in this scenario mainly originates from

the hot steam generated in the industrial boiler room and the boiler flue gas collected during the production process of the factory. Since coal is the main fuel for the boiler, it becomes the dominant influencing factor for ADP of the NdFeB waste recovery. AP indicator reflects the acidification potential by quantifying SO_2 . The source of 2.68×10^4 kg SO_2 in the acidolysis unit is mainly acid mist, including a substantial amount of SO_2 -containing acid mist from the combustion of sulfide in coal and acid mist from the evaporation of hydrochloric acid. The EP was 5.94×10^7 kg PO_4^{3-} , which mainly originated from the acidolysis unit (70.0%) and the precipitation roasting unit (30%). The eutrophic material probably originates from coal and NdFeB scrap, where the input from these two units enriches the phosphorus element. For

GWP, the characterization value in the acidolysis unit and extraction separation unit was 2.65×10^7 and 1.82×10^7 kg CO₂, respectively. CO₂ emissions are mainly from energy use and the higher energy consumption in the PH-M process is primarily from coal and heat utilization of industry. The CoCO₃ purification unit is invested with a multitude of organic compounds, leading to a sudden increase in the environmental impact factors of FAETP, HTP, and TETP.

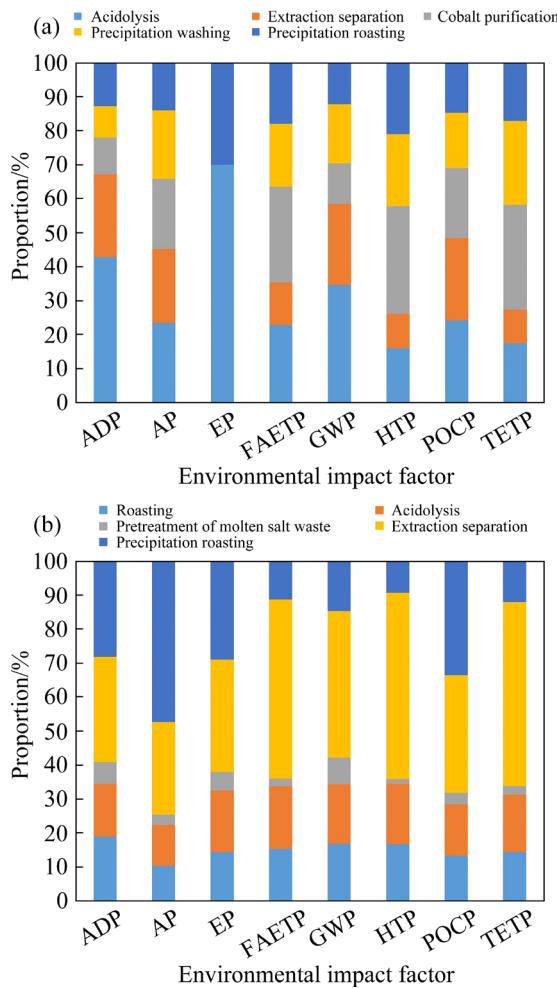


Fig. 3 Environmental life cycle impact of NdFeB scrap recycling with PH-M (a) and PH-Mi (b) processes

In Fig. 3(b), the most significant change compared to Fig. 3(a) is EP, which decreases to 2.95×10^4 kg PO₄³⁻ of the total. It is due to the replacement of hard coal with natural gas in the PH-Mi process. In addition, the process is modified to consume a remarkable number of organic compounds in the extraction separation unit. Therefore, the contribution of FAETP, HTP, and TETP by DCB equivalent is more than 50% in the PH-Mi process, 2.05×10^5 , 6.29×10^6 and

1.82×10^5 kg DCB (Table S2 in SM), respectively. AP and POCP occupy 47% and 34% of the total due to the input of more natural gas in the precipitation roasting unit (Fig. S2 in SM), which contains elements of S and P.

To further investigate the environmental impacts of both recycling technologies, in this work the CML2001 method (2015 version) is selected to obtain a single integrated environmental impact values by weighting and normalizing the calculation based on the characterization results in Fig. 4. The x-axis shows the five units of recycling high-value elements and the y-axis is the normalized value of each environmental category, which is denoted as global equivalent weighted incl biogenic carbon.

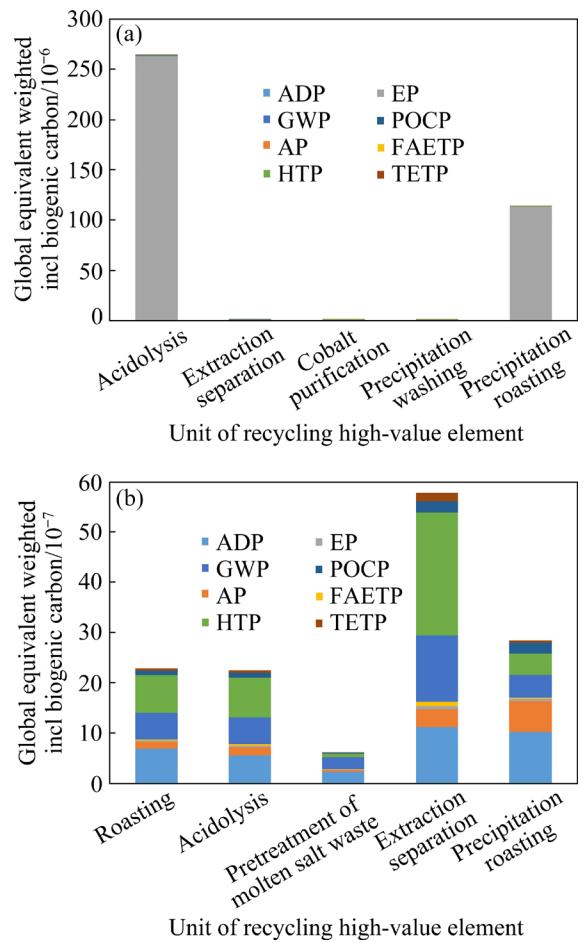


Fig. 4 Normalization results of producing high-value elements from NdFeB scrap with PH-M (a) and PH-Mi (b) processes

The normalized LCIA results corroborate the characterization value mentioned above. For the PH-M process, the total environmental impacts of high-value elements recycling are 3.83×10^{-4} . It is

remarkable that for EP in Fig. 4(a), the total environmental impact of the PH-M process is 3.76×10^{-4} with 69.94% of the acidolysis unit and 30.05% of the precipitation roasting unit. Therefore, other indicators such as ADP, AP, GWP, and HTP have a minor environmental burden, less than 2% of the total. The total environmental impact of the PH-Mi process is sharply reduced to 1.38×10^{-5} with the adoption of natural gas as the energy source (Fig. 4(b)), which is 96.41% compared to the period before the technological improvement. In particular, the environmental impact of EP declines to 1.86×10^{-7} , while other indicators generate an increase of more than 50%. HTP contributes more than 30% to the total environmental impact, which is probably caused by heavy metal elements such as Cu, Cd, and Ni in the waste of NdFeB and molten salt electrolysis. When emitted into air or directly emitted into water and soil, the heavy metals cause serious metal pollution and endanger human health. The unit of extraction separation has the most significant environmental impact on the PH-Mi process in Fig. 4(b). It is probably due to the dramatic increase in electricity consumption and the massive addition of chemical reagents, such as organic compounds, acids, and neutralizers.

3.2 Sensitivity analysis results

The sensitivity analysis is implemented to identify the key factors which dominate the environmental impacts. It is an approach that serves to illustrate and analyze the variability of potential environmental burden [38]. Sensitivity analysis is performed with the input value of key processes as the independent variable and the change value of key categories as the dependent variable. After analysis, sensitivity combined with characterization results identifies four impact factors: electricity,

hard coal, hydrochloric acid, and oxalic acid. For each factor, the 10% reduction in consumption is analyzed, and the results of the sensitivity analysis are shown in Tables S3 and S4 in SM. The results demonstrate that, compared to baseline data, the contribution of hydrochloric acid and oxalic acid to environmental impacts in each scenario is relatively feeble. Notably, fossil fuels and electricity are selected as the main influencing factors for detailed analysis, and the radar plot is shown in Fig. 5.

In the PH-M scenario, hard coal contributes significantly to EP (85.26%), ADP (45.91%) and GWP (35.91%). Electricity is the highest contributor to HTP (10.10%). In the PH-Mi scenario, the highest variation in extraction separation unit at 10% electrical change is observed to be attributed to HTP (8.66%), followed by TETP (6.75%) and FAETP (6.71%). The environmental impacts of electricity are primarily caused by the burning of fossil fuels, as thermal power generation accounts for over 70% of China's electricity production. Meanwhile, ADP shows a maximal variation with 10% decrease in natural gas, and the environmental impacts of other factors are minimal to be negligible. To verify the positive influence of natural gas on reducing environmental impacts, hard coal in the PH-M scenario is replaced with a thermal equivalent of natural gas to investigate the environmental impacts (Table S5 in SM). As shown in Table S5 in SM, with the other conditions unchanged, the proportions of EP, GWP, and HTP decrease to 7.65%, 85.52%, and 85.34%, respectively, when the energy source is modified. It is exciting to observe that the total environmental impact is reduced to 9.18%. In summary, natural gas is a more environmentally friendly energy source than hard coal.

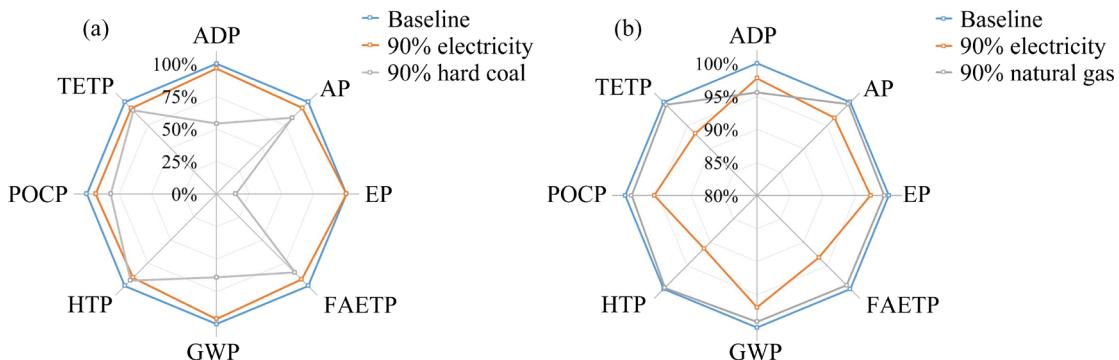


Fig. 5 Sensitivity analysis results of main contributors in PH-M (a) and PH-Mi (b) processes

3.3 Carbon footprint

As a quantified indicator for carbon emission analysis and assessment, carbon footprint becomes an important consideration for low-carbon economies [39,40]. In this study, LCA is implemented to quantify the carbon footprint of high-value element recovery in waste. Figure 6 shows the calculated carbon emissions in the CO_2 form only. Herein, material extraction is the emissions of feedstock materials in the generation process, and direct emission is the direct emissions of the recycling process.

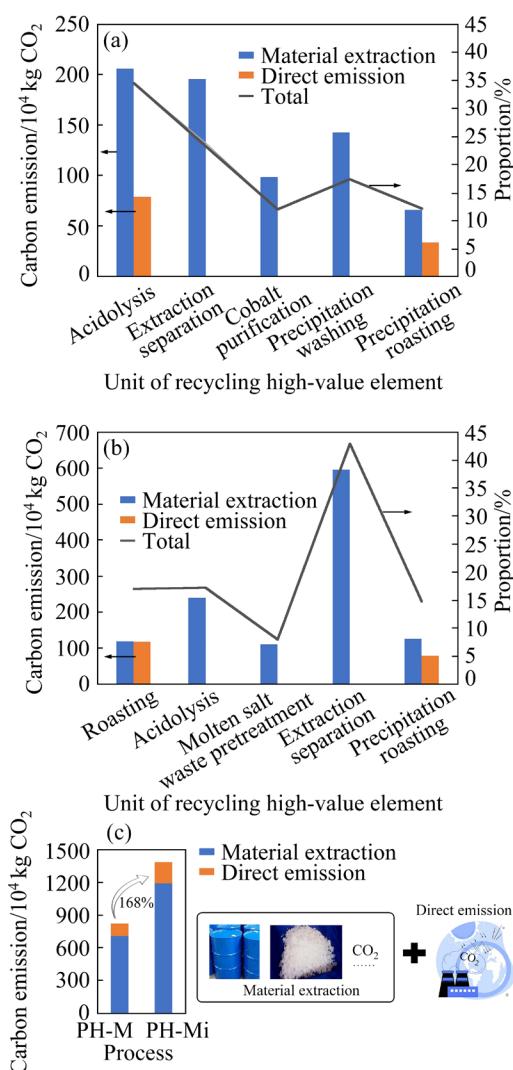


Fig. 6 Carbon footprint in PH-M (a) and PH-Mi (b) processes, and comparison of total carbon emissions in two scenarios (c)

Figure 6(a) demonstrates that the calculated annual carbon footprint of NdFeB scrap recovery using the PH-M process is $8.22 \times 10^6 \text{ kg CO}_2$, which is mainly derived from acidolysis unit (34.62%) and

extraction separation unit (23.84%). The material extraction stages have the most significant impact on greenhouse gas (GHG) emissions, representing 13.75% of total carbon emissions. The annual carbon footprint of the PH-Mi process to recycle NdFeB scrap and molten salt electrolysis waste is $1.38 \times 10^7 \text{ kg CO}_2$ in Fig. 6(b). In the PH-Mi scenario, the direct emission percentage declines to 14% of total carbon emissions. In particular, the extraction separation of 43% is an essential contributing unit, which is probably attributed to the increased consumption of chemicals resulting from the extraction process upgrading from grade 488 to grade 618. Figure 6(c) visually demonstrates the carbon footprint of both scenarios, with the PH-Mi process increasing carbon emissions by 68% over the PH-M process. There is an abnormal increase in carbon emissions due to the addition of electricity, calcium hydroxide, and hydrochloric acid. The carbon footprint assesses CO_2 emissions over the product life cycle, and the results indicate consistency with the GWP indicator in the characterization results. In the future, the PH-Mi process will consider further reducing carbon emissions through the following methods such as utilization of green chemicals, waste heat recovery, the clean energy alternative to electricity (from hard coal), and process optimization.

3.4 Cost and profit analysis results

The costs and benefits of two high-value recycling processes for wastes are estimated without considering the disposal costs and personnel salaries for solid wastes such as acid-soluble slag and iron removal slag. The main expenses of recycling process include chemical reagents and energy. The income is calculated based on the products and byproducts to obtain the final profits. Table 5 summarizes the mass and revenue in two scenarios of NdFeB scrap recycling per year, considering net profit without equipment cost, depreciation, and operation expense. The data of total cost and net profit are derived from Tables S6 and S7 in SM.

The total production of commodities applied to the PH-M process is 951 t. Especially, Pr–Nd oxide and terbium chloride contribute over 85% of total revenue. After improving disposal technology, the mass of commodity is increased to 1490.8 t and the

Table 5 Mass and revenue of high-value element recycling processes of NdFeB scrap

Category	PH-M		PH-Mi		Unit price*/ (RMB¥·t ⁻¹)
	Mass/(t·a ⁻¹)	Contribution/%	Mass/(t·a ⁻¹)	Contribution/%	
Pr–Nd oxide	820.8	54.47	1300	66.39	1039700
Gadolinium oxide	—	—	53.6	1.67	635000
Dysprosium oxide	72.5	13.75	34.7	5.07	2972400
Cerium oxide	—	—	32.3	0.02	10000
Holmium oxide	—	—	10	0.92	1883000
Terbium chloride	29.4	31.15	31.2	25.44	16601700
Cobalt carbonate	28.3	0.63	29	0.49	347400
Total revenue	951	100	1490.8	100	—

Category	Value/(RMB¥·a ⁻¹)		Growth rate/%
	PH-M	PH-Mi	
Total cost	410906950	485643690	18.18
Total revenue	1566806160	2035988920	29.95
Net profit	1155899210	1550345230	34.12

* Unit price was from the website of www.cbcie.com

profit margin is boosted from 281.30% to 319.25%. It is remarkable that with nearly a 18.18% increase in feedstock costs, the PH-Mi process achieves a 34.12% net profit growth rate. Of all the products, Pr–Nd oxide and terbium chloride, as the most expensive products per unit, account for more than 90% of the total revenue. Overall, the PH-Mi process implements not only an improvement in annual treatment capacity through the technology upgrade but also an enhancement in net profit.

4 Discussion

From the findings, both strategies have their unique characteristics in terms of carbon emissions and economic viability. Against the backdrop of the Beautiful China initiative, the industrial cost reduction, and efficiency improvement, the PH-Mi process demonstrates clear advantages, albeit with a certain gap from existing carbon reduction targets. The PH-Mi process emerges as the optimal choice for enterprises considering this delicate balance. In the future, the PH-Mi process will need to further reduce carbon emissions from four methods to achieve regional carbon emission goals.

Firstly, the utilization of green chemicals is a better way to make improvements. By optimizing the use of chemicals and selecting environmentally friendly, low-carbon emission treating agents and

solvents, the carbon emissions of chemicals in the acidolysis unit and extraction separation unit can be reduced, for example, the substitution of industrial kerosene with kerosene produced from carbon dioxide or green methanol. This can significantly reduce greenhouse gas emissions, positively impacting carbon reduction. Secondly, establishing a rational recycling system and constructing an energy reuse process to recover waste heat are essential. Developing waste heat recovery systems to utilize the heat generated from the roasting, acidolysis, and extraction separation units for other production processes or energy provision can enhance energy utilization efficiency and reduce reliance on external energy sources. Additionally, clean energy will be used to replace the electricity produced by thermal power generation. Enterprises may consider deploying clean energy sources, such as solar power and wind energy, to meet the energy demands of the precipitation washing unit in the PH-Mi process. Lastly, further reduction of carbon emissions can be achieved through process optimization by minimizing the use of energy and raw materials.

5 Conclusions

(1) The phosphorus rich in hard coal contributes to the anomalous increase of EP in the

PH-M scenario. After the normalization of EP, the proportion of EP in the total environmental impact is observed to be over 99.9%, and the results of sensitivity analysis corroborate the negative environmental impacts of hard coal.

(2) The total environmental impact of the PH-Mi process is sharply reduced to 1.38×10^{-5} , which is 96.41% compared to that of the PH-M scenario. However, the carbon footprint shows that the PH-Mi process has a 68% increase in carbon emissions compared to the PH-M process.

(3) In terms of economic benefit, the importance of technology upgrades is adequately reflected. For the PH-Mi process, the net profit is improved to 34.12% of the original process with a 17% increase in cost. Among all commodities produced by two scenarios of recycling, Pr–Nd oxide and terbium chloride have the highest profit contribution.

(4) The environmental impacts, economic and social benefits of the PH-Mi process are substantially higher than those of the PH-M process, whereas there is no advantage regarding carbon emissions. Although the improvement of waste material recycling processes has indeed increased material and energy input to a certain extent, the social and economic values of process improvement should not be denied. In the context of the global energy crisis and environmental degradation conditions, it is necessary for governments and businesses to enhance their strategic understanding in the future, intensify efforts to overcome key technologies for resource conservation and recycling, organize the development of resource conservation and substitution technologies with significant promotional value, and create a resource-saving society.

CRediT authorship contribution statement

Jun-feng WANG: Conceptualization, Visualization, Writing – Original draft; **Lu-jing LIU:** Formal analysis, Writing – Review & editing, Supervision; **Ming YANG:** Methodology, Software; **Yang WANG:** Validation, software; **Yi-fan GU:** Resources, Software; **Ying-yan HU:** Investigation, Supervision, Funding acquisition; **Liang-fang LIAO:** Data curation.

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.

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Supplementary Materials

Supplementary Materials in this paper can be found at: http://tnmsc.csu.edu.cn/download/24-p1713-2023-1011-Supplementary_Materials.pdf.

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生命周期评价下钕铁硼废料高值回收的环境影响及碳足迹

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摘要: 钕铁硼废料作为典型稀土固废, 具有较高的经济价值。本文作者以钕铁硼废料为对象, 研究了火法-湿法冶金工艺((PH-M)及其改进方案(火法-湿法冶金改进工艺(PH-Mi))的环境影响。研究结果表明, 尽管 PH-Mi 消耗了更多的能源、电力和化学品, 但 PH-Mi 表现出良好的环境效益与经济效益(净利润增长了 34.12%)。采用生命周期评价方法量化比较两种方案的环境影响。PH-Mi 在富营养化潜力(EP)和总体环境影响方面优于 PH-M, 而 PH-M 在碳足迹方面优于 PH-Mi 的原因在于技术升级后消耗了更多的电力与化学品。

关键词: 钕铁硼废料; 熔融盐电解废料; 生命周期评价; 环境影响; 碳足迹

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