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Environmental Impact of the Recycling of Ni-Co-Containing Saggars—A LCA Case Study in China

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Abstract: Scrapped saggars, used for the calcination of the cathode materials of lithium-ion batteries, contain large amounts of nickel, cobalt and manganese compounds, which have high economic value and significance to the ecological environment if recycled. This paper uses the life cycle assessment method to evaluate the environmental impact of the recycling process, compares its impact intensity with that of the direct disposal of Ni-Co-containing saggars and the production of corresponding products with alternative processes, and then compares the impact of each process. Sensitivity analysis of each material input and output item is carried out to find whether the input or output item that has a greater impact on the environment. The results show the following: (1) the environmental impact of the recovery of Ni-Co-containing saggars is much lower than that of equivalent products produced by alternative production methods, and the weighted person equivalent is only 14.5% of the alternative process; (2) from the perspective of processes, the crystallization and leaching processes demonstrate the greatest impact; and (3) among all input and output items, the sulfuric acid input in the reduction and leaching process, the potassium carbonate and steam input in the crystallization process, the potassium carbonate and potassium hydroxide input in the cascade separation process, and the ammonia input in the purification process are the items with the greatest environmental impact, accounting for 86.05% of the overall environmental impact sensitivity and becoming the focus of future process improvement.

Keywords: Ni-Co-containing saggars; eco-efficiency; life cycle assessment; recycling industry; metal recycling; environmental pollution; heavy metals; health hazards

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1. Introduction

China has put forward the goal of achieving its peak carbon emission in 2030 and carbon neutralization in 2060 (Li, 2021) [1]. The adjustment of the energy structure is the basic guarantee for achieving the carbon reduction goal. With the gradual promotion of new energy power generation, replacing the use of fossil energy power with batteries, such as in the automotive field, has become one of the key areas of energy conservation and emission reduction.

Due to their high energy density, excellent charging performance and long service life, lithium-ion batteries have become the mainstream electric vehicle power supply (Yang et al., 2014) [2]. Among them, lithium-ion batteries using nickel cobalt manganese ternary materials as cathode have the advantages of strong stability, moderate cost, etc., and the overall performance is superior to that of the previous single component materials, becoming the main development direction of lithium-ion batteries. Thus the demand for ternary cathode materials is growing rapidly. In the past few years, it has achieved an average annual growth rate of 20% (Li et al., 2015) [3]. With the rapid expansion of the electric vehicle industry, the consumption of related metals required for ternary cathode materials will continue to increase in the future. The world's lithium, nickel and cobalt metal resources are relatively scarce, with very limited reserves. With the rapid growth of demand, the reserves of these metals will have difficulty meeting market demand. Therefore, promoting the recovery and recycling of lithium-ion battery cathode materials is an inevitable requirement to ensure the development of the electric vehicle industry in the future. According to the new energy vehicle industry development plan 2020–2035 (Exposure Draft) [4] issued by the State Council of China, new energy vehicles will account for 25% of vehicle sales by 2025 and 40% by 2030. According to the estimation of Xing et al. (2019) [5], China's annual demand for battery cathode materials for new energy vehicles will be 6.6×10^4 tons of lithium, 4.6×10^5 tons of nickel and 5.4×10^4 tons of cobalt, equivalent to 330%, 47% and 113% of the national consumption in 2018, respectively. China's energy metal reserves are small. According to USGS data (2021) [6], China's lithium, nickel and cobalt mineral reserves (metals) are 1.5 million tons, 2.8 million tons and 80,000 tons, respectively. If China's domestic mineral reserves were to meet the battery demand at that time, it would only meet 22.7 years, 6.1 years and 1.5 years, respectively.

In the production process of lithium battery cathode materials, relevant materials need to be calcined in a ceramic saggar to form a cathode material consisting of $LiNi_xCo_yMn_{1-x-y}O_2$. During the calcination process, the battery cathode material reacts with the inner wall of the ceramic saggar to form complex compounds attached to the surface of the saggar (Zhai et al., 2018) [7]. The service life of ceramic saggar is short, and it will be scrapped after five to 20 uses. Scrapped ceramic saggars are often used as refractory materials or directly discarded, and a variety of rare metals attached to their surfaces cannot be recycled. In China alone, 80 million ceramic saggars are scrapped every year, including more than 20,000 tons of lithium, nickel, cobalt, manganese and other metals needed for the manufacture of lithium-ion battery cathode materials, resulting in a serious waste of resources (Li et al., 2016) [8]. At the same time, since the materials required for lithium-ion batteries will cause a great environmental impact in the process of mining, extraction and smelting, it will help to reduce the environmental impact by reducing the primary smelting through the recovery of related metals.

The Institute of Process Engineering of the Chinese Academy of Sciences, in conjunction with an enterprise producing ceramic saggars, has developed a high-efficiency separation and purification process of lithium/nickel/cobalt/manganese for the attachment of the inner surface of waste saggars by establishing a multimetal low-temperature reduction technology system. The process has passed the pilot test and has good economic prospects. However, the environmental impact of the whole life cycle of the process remains to be evaluated.

From the literature, research on nickel or cobalt metal recycling mainly focuses on the recycling technology of used battery cathode materials (Zeng et al., 2015; Shi, 2017; Wang, 2017; Yu, 2018; Povali et al., 2020) [9–13]. However, there are relatively few studies on the ecological impact of its recycling process. Temporelli et al. (2020), Richa et al. (2017) and Yang et al. (2020) [14–16] analyzed the ecological efficiency of lithium battery recycling and believed that lithium battery recycling could effectively reduce the environmental impact of its ecotoxicity and improve the efficiency of its ecological life cycle. Zackrisson et al. (2010), Unterreiner et al. (2016) and Raugei et al. (2019) [17–19] used life cycle assessment to compare the ecological impact of different solvents on automotive lithium batteries. Wu et al. (2019) [20] compared the ecological footprint of different types of renewable lithium batteries. Other studies have focused on the environmental impact of the metals involved in the initial smelting process (Ali et al., 2015; El Alfy et al., 2020) [21,22]. The relevant research on sagittal materials of battery cathode materials mainly focuses on the characteristics of sagittal materials during the firing process of battery cathode materials (Liu, 2015) [23]. It is worth noting that there are few studies on the recovery of nickel cobalt-containing saggars and their environmental impact.

The existing research mainly focuses on the environmental impact of the recycling process of lithium-ion batteries, while less attention is given to the recycling of the waste from the manufacturing process. There are three main research methods. In the early stage, a direct comparison method was used to assess the environmental impact by analyzing the energy directly consumed by the production line and the pollutants directly discharged. The second is the ecological footprint method, which calculates the carbon emissions or water consumption in the whole production process. The third method is life cycle assessment.

Life cycle assessment (LCA) is the process of assessing the environmental impact of products, processes or activities, from the collection of raw materials to the production, transportation, sales, use, reuse, maintenance, and final disposal in the environment of products. Therefore, LCA shows the whole life cycle of the subject. It first determined and quantified the energy and material consumption and environmental release in the whole life cycle, then assessed their impact on the environment, and finally determined and evaluated the opportunities to reduce these impacts (Santoyo-Castelazo, Azapagic, 2014) [24]. The LCA process is generally divided into four steps: definition of objectives and scope, inventory analysis, impact assessment and improvement assessment. This method can comprehensively quantify and assess the resource consumption, ecological pressure and human health impact of specific substances in the whole life cycle of their production and utilization and can further analyze the impact of changes in different raw materials or products on the ecological environment (ISO, 2006) [25]. Compared with other impact assessment methods that directly assess the production itself, it can more comprehensively assess the overall environmental impact of a specific product or process.

The purpose of this study is to achieve the following objectives: (1) quantitative analysis of the life cycle impact of the entire recycling process of nickel cobalt-containing waste saggar; (2) determination of the key areas, key links and key substances of environmental impact in the saggar recovery process through quantitative analysis s are; (3) propose possible technical improvement directions according to the analysis results. The innovative points of this study are: (1) the evaluation of life-cycle resources and environmental impact of the nickel-cobalt-containing saggar recycling production line and (2) the proposed improvement direction for the further optimization of the resource and environmental effects of the production line. The structure of the remaining part of this paper is as follows: the second part describes the methods and data sources used in this paper, the third part interprets the analysis results, and the fourth part gives corresponding conclusions and suggestions based on the analysis of the results and discusses the shortcomings and further research directions.

2. Methodology

2.1. LCA Goal and Scope Definition

The pilot test was jointly completed by the Institute of Process Engineering of the Chinese Academy of Sciences and a lithium-ion battery production company in Hunan. The company is one of the most important suppliers of cathode materials for lithium-ion batteries in China. The company has been committed to the recovery of cathode materials for a long time. The joint project extracts lithium, nickel, cobalt, manganese and other lithium-ion battery cathode materials from waste saggars, as well as alum, potassium sulfate and other byproducts, through physical separation, reduction and leaching, cascade separation, crystallization, and purification processes, with good economic benefits.

Physical separation is the first process. In this process, the inner surface of the waste saggar with a large amount of metal attached is first polished, and the ground powder is used in the reduction and leaching processes. The rest of the discarded saggar body will be crushed as refractory or raw material for ceramic production. After the reduction and leaching process, the powder on the inner surface of the saggar is soaked with strong acid to form an acid-leaching solution and enters the separation process. The remaining acid-leaching residue is further treated as waste. In the step separation process, the crude Ni-Co-Mn precursor is extracted from the acid-leaching solution, and the product is further purified. The byproducts produced, such as aluminum oxide, aluminum hydroxide and magnesium hydroxide, are sold directly, and the filtrate enters the crystallization process. In the crystallization process, the filtrate generates lithium carbonate and enters the purification process. The byproduct, potassium sulfate, is sold directly, and the remaining mother liquor returns to the cascade separation process. The crude Ni-Co-Mn precursor and lithium carbonate are purified and sold as the main products (Figure 1).

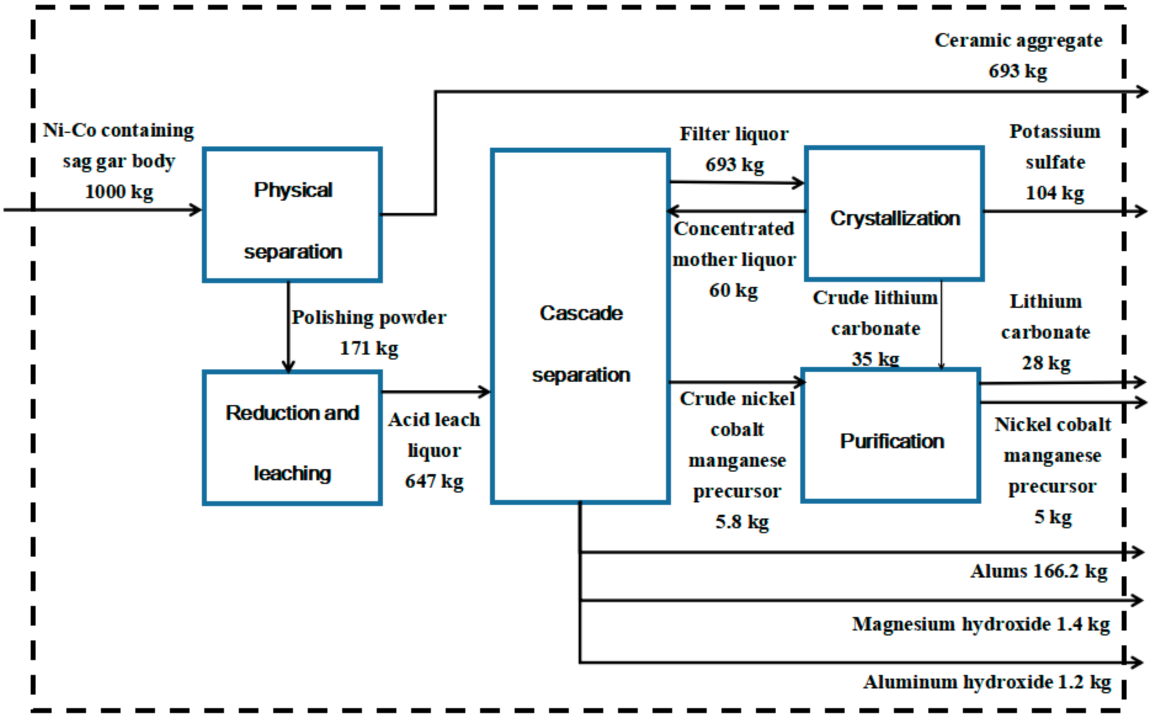

Figure 1. Definition of the LCA scope of the recovery of Ni-Co-containing saggars.

We selected 1000 kg nickel-cobalt-containing saggar as the functional unit to be studied. The material flow between different production processes is shown in Figure 1. This paper focuses on analyzing the environmental impact of the production process. Therefore, "cradle to gate" is selected as the boundary of the study area, including five steps of physical separation, reduction and leaching, cascade separation, crystallization and purification, that is, from the development of various raw materials to products, as well as the waste treatment process in the production process. However, the environmental impact of equipment and infrastructure construction is not considered. For ceramic aggregate, alum, potassium sulfate and other byproducts in the production process, economic distribution is adopted to distribute the impact on the environment in the production process. According to the calculation, the total distribution coefficient of the main products of nickel cobalt manganese precursor and lithium carbonate is 47.33%. When selecting the cutoff method, as the main raw material used in the process is scrapped saggars, it is considered that it does not bear the environmental load of primary production.

2.2. Life-Cycle Inventory

Using GaBi 9.0 ts software (Herrmann, Moltesen., 2015; Saynajoki et al., 2017; Emami et al., 2019) [26–28], the raw materials directly used were associated with the production process, and the emissions that needed to be treated were associated with the treatment process, thereby establishing a complete life cycle inventory. The material input and output of the inventory should be balanced. Supplementary Table S1 shows the material input and output tables of each process. All of the direct input-output data in the processes are supplied by the company, and indirect data, such as the full lifecycle environmental impact data of various raw materials, comes from the Gabi database.

After the Ni-Co-containing saggars were recovered, they entered the physical separation process. The surface metal of the saggars was polished with water to form a 30% water-containing powder. For each treatment of 1000 kg of waste saggars, 148 kg of water needed to be inputted to generate 171.4 kg of polishing powder (30% water content) during the reduction and leaching process, and the remaining ceramic aggregate was 977.8 kg.

The reduction and leaching process took the polishing powder from the physical separation process as the main material, and then concentrated sulfuric acid, reducing agent and water were added, generating an acidic leaching liquid that entered the cascade separation step and produced acidic leaching residue as solid waste. For each treatment of 1000 kg of saggars, 171.4 kg of polishing powder, 216 kg of concentrated sulfuric acid, 376 kg of water and 12 kg of other reducing agents were required to produce 647 kg of acid leachate and 128.4 kg of acid leaching residue (60% water content).

The cascade separation took the acid-leaching solution from the reduction leaching as the main raw material, and then potassium hydroxide, potassium sulfate, water, and other auxiliary materials were added to produce the crude Ni-Co-Mn precursor product, which entered the purification process and produced byproducts, such as magnesium hydroxide, aluminum hydroxide and alum, which were sold, while the remaining filtrate was further refined in the crystallization process. For processing 1000 kg of saggars, 647 kg of acid leachate, 20 kg of potassium sulfate, 55.6 kg of potassium hydroxide, 59.6 kg of saturated potassium sulfate solution, and 85.6 kg of water were needed to produce 5.8 kg of the crude Ni-Co-Mn precursor, 166.2 kg of alum, 1.4 kg of magnesium hydroxide, 1.2 kg of aluminum hydroxide, and 693.2 kg of filtrate.

The crystallization utilizes filtrate from cascade separation as the main material, adding potassium carbonate and water to generate crude lithium carbonate, which enters the purification process and generates potassium sulfate for sale. The remaining concentrated mother liquor returns to the step separation process, and the evaporated condensate is recycled. For each 1000 kg of waste sagger body, 693.2 kg of filtrate, 66 kg of potassium carbonate, 96.4 kg of water are required to produce 35.2 kg of crude lithium carbonate, 104 kg of potassium sulfate, 60 kg of concentrated mother liquor, and 656.8 kg of condensed water.

The purification process took the crude Ni-Co-Mn precursor (from the cascade separation step) and lithium carbonate (from the crystallization step) products as raw materials, and then sulfuric acid, carbon dioxide, ammonia and sodium hydroxide were added for purification to form the final products to sell. To process 1000 kg of waste saggars, 35.2 kg of crude lithium carbonate, 5.8 kg of crude Ni-Co-Mn precursor, 10 kg of concentrated sulfuric acid, 11 kg of carbon dioxide, 9 kg of ammonia (25%), 6 kg of sodium hydroxide, and 16 kg of pure water were required to produce 5 kg of battery-grade nickel-cobalt-manganese precursor and 28.2 kg of battery-grade lithium carbonate to sell. It also produced 1.8 kg of solid waste containing calcium and magnesium and 68 kg of wastewater.

The data quality analysis and consistency analysis of the input data of all 5 production processes showed that the data used had high technical and time representativeness, and the location representativeness was slightly lower (Supplementary Table S2). Thus, the material input and output quality difference of each process was less than 0.5%, passing the consistency test.

The life cycle inventory assessment on the input-output table showed the resource consumption and waste discharge classification and total data of the whole production process. The corresponding data and analysis are provided in Section 3.1.

2.3. Life Cycle Impact Assessment Method

The ReCiPe 2016 indicator system (Huijbregts, Steinmann et al., 2017) [29] was selected in this study because it combines the advantages of CML and Ecoindicator at the same time. The use of 2 sets of evaluation indicators, midpoint and endpoint, can more comprehensively reflect the environmental impact characteristics of related processes. In addition, Recipe's parameter settings are more global, greatly increasing the applicability of this indicator in different regions. This indicator system adopts the following 16 types of midpoint indicators: global warming potential (GWP), ozone disposal potential (ODP), ionizing radiation potential (IRP), particulate matter formation potential (PMFP), ozone formation potential (OFP), acidification potential (AP), freshwater industrialization potential (FEP), human toxicity potential (HTP), terrestrial ecotoxicity potential (TETP), fresh water ecotoxicity potential (FETP), marine ecotoxicity potential (METP), marine eutrophication potential (ME), agricultural land occupation potential (LOP), water consumption potential (WCP), surplus ore potential (SOP), and fossil fuel potential (FFP). Through the weighted normalization of 16 categories of indicators, 3 types of end-point indicators are obtained. Finally, they are compared with the global environmental impact background values, further normalized and added to obtain the weighted equivalent, which is used to measure the overall environmental impact of the process (Figure 2).

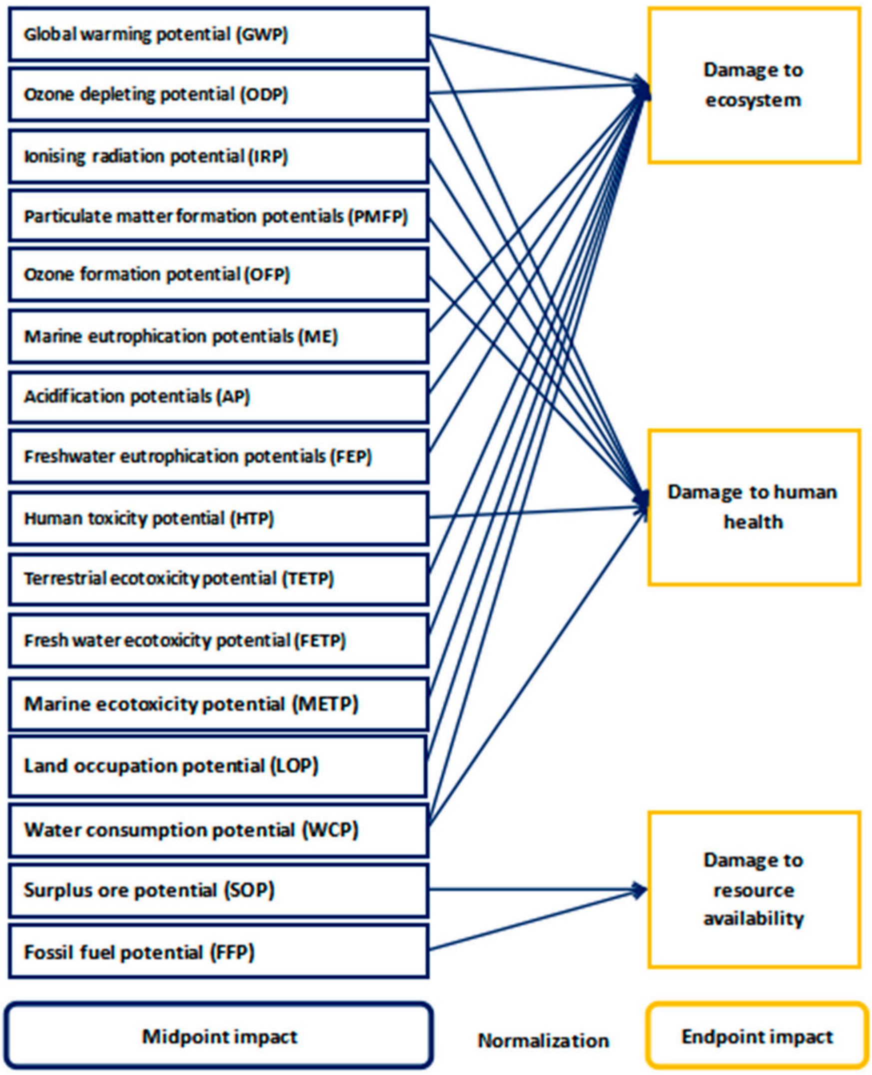

Figure 2. LCA-ReCiPe 2016 life cycle impact assessment process (according to Huijbregts, Steinmann et al. 2017 [29]).

3. Results

3.1. Life Cycle Inventory Assessment Results

As shown in Table 1, the life cycle energy consumption per 1000 kg of Ni-Co-containing saggars is 83.97 kg, and the material resource consumption is 1.41×10^5 kg. From the point of production processes, the largest energy consumption is during the reduction leaching process, accounting for 35.3% of the total energy consumption, followed by the crystallization and cascade separation steps, which have values of 27.36 kg and 16.79 kg, respectively. In terms of material resources, cascade separation is the most consumed. It consumes 8.09×10^4 kg per 1000 kg of Ni-Co-containing saggars, which is more than half of the total resource consumption, accounting for 57.2%, followed by the reduction and leaching and crystallization steps, which are 2.89×10^4 and 1.52×10^4 kg, respectively. In terms of emissions, the cascade separation step still has the highest value, with total emissions accounting for 57.3% of all emissions, followed by the reduction and leaching and crystallization steps; additionally, all subitems of emissions mostly come from cascade separation. Overall, from the perspective of material quality, cascade separation is the process with the greatest environmental impact, followed by the reduction and leaching, crystallization, purification and physical separation steps. This is not because there are too many direct input materials in the cascade separation process but because the materials used in the process are mostly expensive reagents, which require a large amount of materials to produce.

Table 1. Life cycle resource depletion and emissions comparison of the processes of Ni-Co-containing saggar recovery.

Unit: kg	Total	Physical Separation	Reduction and Leaching	Cascade Separation	Crystallization	Purification
Total Flows	281,487.69	4914.43	51,754.89	161,663.66	31,028.75	21,125.98
Energy resources	83.97	1.18	29.66	16.79	27.36	8.98
Nonrenewable energy resources	83.97	1.18	29.66	16.79	27.36	8.98
Renewable energy resources	0.00	0.00	0.00	0.00	0.00	0.00
Material resources	141,317.35	2722.75	21,949.37	81,877.59	11,193.84	11,573.78
Nonrenewable resources	467.89	3.06	38.94	171.92	223.00	30.97
Renewable resources	141,849.23	2719.69	18,910.35	81,705.63	11,970.80	11,542.77
Emissions	141,860.19	2653.38	21,775.85	81,769.27	11,118.47	11,543.21
Deposited goods	336.30	2.92	88.02	109.67	120.81	14.88
Emissions to air	1569.12	48.14	263.72	674.41	434.28	148.56
Emissions to freshwater	131,349.77	2598.35	21,318.15	71,668.99	11,454.20	11,310.09
Emissions to seawater	605.00	3.97	105.97	316.20	109.18	69.68
Emissions to agricultural soil	0.00	0.00	0.00	0.00	0.00	0.00
Emissions to industrial soil	0.00	0.00	0.00	0.00	0.00	0.00

3.2. Life Cycle Impact Assessment Results

Using the ReCiPe 2016 characterization standard, the impact intensity of different processes on the ecological environment was analyzed from 16 indicators, such as climate change and land acidification. Table 2 shows that the recycling process of Ni-Co-containing saggars is superior to alternative production in all 16 indicators. The recycling of the Ni-Co-Mn precursor and lithium carbonate from Ni-Co-containing saggars through this process has obvious environmentally friendly advantages compared to direct production. For example, in terms of the greenhouse effect, the process is equivalent to only 16.1% of alternative production, the human toxicity effect is equivalent to only 8.9% of alternative production, and the metal consumption is equivalent to only 1.2% of alternative production.

Since there is little literature on the recovery of metal resources in Ni-Co-containing saggars, this article can only be compared with the literature on lithium battery manufacturing and secondary utilization. Compared with Yang (2020) [16] for the recycling and utilization of lithium batteries for vehicles, through unit conversion, the resource and environmental impact intensity per kilogram of battery mass is similar to the Ni-Co-containing saggar recovery process, and the Ni-Co-containing saggar recovery process is lower in the global warming field. While fossil consumption is slightly higher, the above analysis shows that the environmental impact of the Ni-Co-containing saggar recovery process is similar to that of lithium battery recycling. Compared with the Ni-Mn-Co (NMC) lithium battery manufacturing process of Temporelli (2020) [14] and Wang (2020) [30], after converting to the same quality, the environmental impact of the Ni-Co-containing saggar recovery process is significantly less than that of the direct production of lithium battery materials, which also confirms that the environmental impact of the process is less than that of alternative production.

Table 2. Comparison of the life cycle impact indicators for the recovery of Ni-Co-containing saggars.

Factor	Unit	Ni-Co-Containing Saggar Recovery	Alternative Production	Yang (2020) [16] LIB Secondary Use	Temporelli (2020) [14] NMC Production	Wang (2020) [30] NMC Production
GWP	kg CO_2 eq.	1.58×10^2	9.80×10^2	5.00×10^2	2.00×10^5	1.50×10^5
PMFP	kg PM2.5 eq.	3.19×10^{-1}	1.27×10^0			
FFP	MJ eq.	3.13×10^3	1.59×10^4	2.10×10^2		1.89×10^4
WCP	m^3	9.89×10^{-1}	6.67×10^0			
FETP	kg 1,4-DB eq.	2.43×10^{-2}	3.54×10^{-1}			2.10×10^4
FEP	kg P eq.	2.67×10^{-4}	1.94×10^{-3}	1.10×10^0	4.00×10^2	3.60×10^2
HTP	kg 1,4-DB eq.	6.70×10^2	7.48×10^3			6.00×10^5
IRP	kBq Co60 eq. to air	1.50×10^1	1.73×10^2			
LOP	Annual crop eq.·y	2.25×10^0	4.53×10^1			
METP	kg 1,4-DB eq.	2.14×10^2	5.49×10^3			1.80×10^4
ME	kg N eq.	8.52×10^{-3}	2.76×10^{-2}			1.80×10^2
SOP	kg Cu eq.	1.78×10^0	1.40×10^2	2.70×10^2		7.50×10^2
OFP	kg NO_x eq.	7.69×10^{-1}	8.99×10^0			
ODP	kg CFC-11 eq.	5.73×10^{-5}	3.71×10^{-4}		1.00×10^0	1.50×10^{-2}
AP	kg SO_2 eq.	1.10×10^0	4.22×10^0		2.00×10^3	3.00×10^3
TETP	kg 1,4-DB eq.	3.49×10^1	1.11×10^3			

From the perspective of each production process (Figure 3), reduction and leaching have the greatest impact in seven aspects: PMFP, FFP, FETP, HTP, METP, AP, TETP, and crystallization has the greatest impact in six aspects: GWP, WCP, ME, SOP, OFP, and ODP. Cascade separation has the greatest impact on three aspects: FEP, IRP, and LOP. Generally, reduction and leaching have the greatest impact in most areas. This is related to the need to use a large amount of sulfuric acid as a solvent in the reductive leaching process, and the environmental cost of sulfuric acid production is often higher.

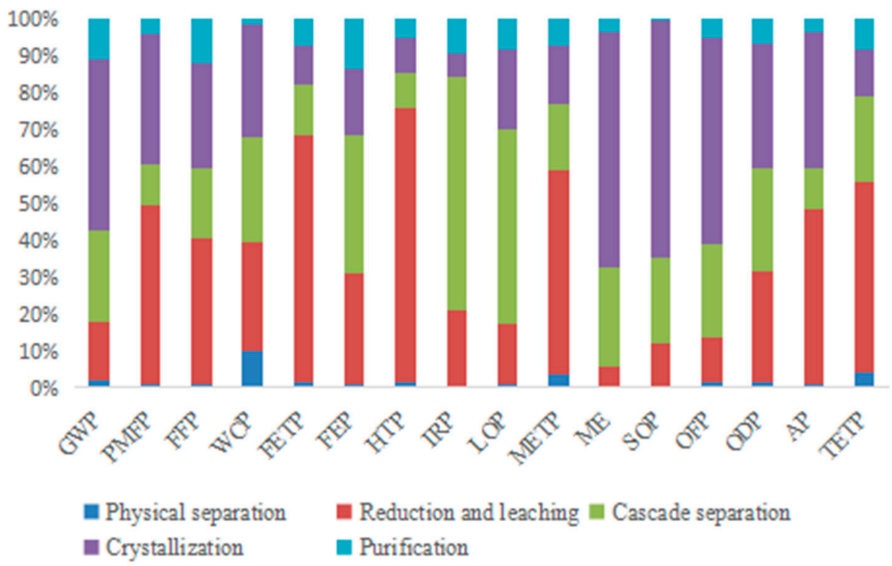

Figure 3. Comparison of the midpoint indicators of the various production processes for recovery of Ni-Co-containing saggars.

Through normalization, the endpoint value of the impact on the ecological environment is obtained. The weighted human equivalent of the Ni-Co-containing saggar recovery is 101.88, while the weighted human equivalent of the alternative process is 700.6. The overall environmental impact of the Ni-Co-containing saggar recovery process is only equivalent to 14.5% of the alternative process; thus, the advantage in environmental protection is obvious. The area with the greatest environmental impact of the process is the ecosystem (Figure 4), with a weighted human equivalent of 45.08; the second is the human health impact, with a weighted human equivalent of 42.64; and the impact of resource availability is the smallest, indicating that the process has indeed played a role in resource conservation. In further subdivisions (Figure 5), the GWP is the most influential field, accounting for 32% of the total impact, indicating that the energy structure dominated by fossil energy is the most important environmental influencing factor of the process. In the future, consideration should be given to increasing the proportion of clean energy use. The influence of FFP, which is ranked third, is mutually confirmed by this fact. The second place is OFP, which indicates that the process has certain photochemical pollution. From the point of view of the processes, OFP is mainly concentrated in the crystallization process, which means that the atmospheric emissions generated during the crystallization process still need to be strictly controlled. In addition, the impact of AP is also more than 10%, which is mainly concentrated in the reductive and leaching process and shows that the acid mist generated during the acid leaching step needs to be further reduced.

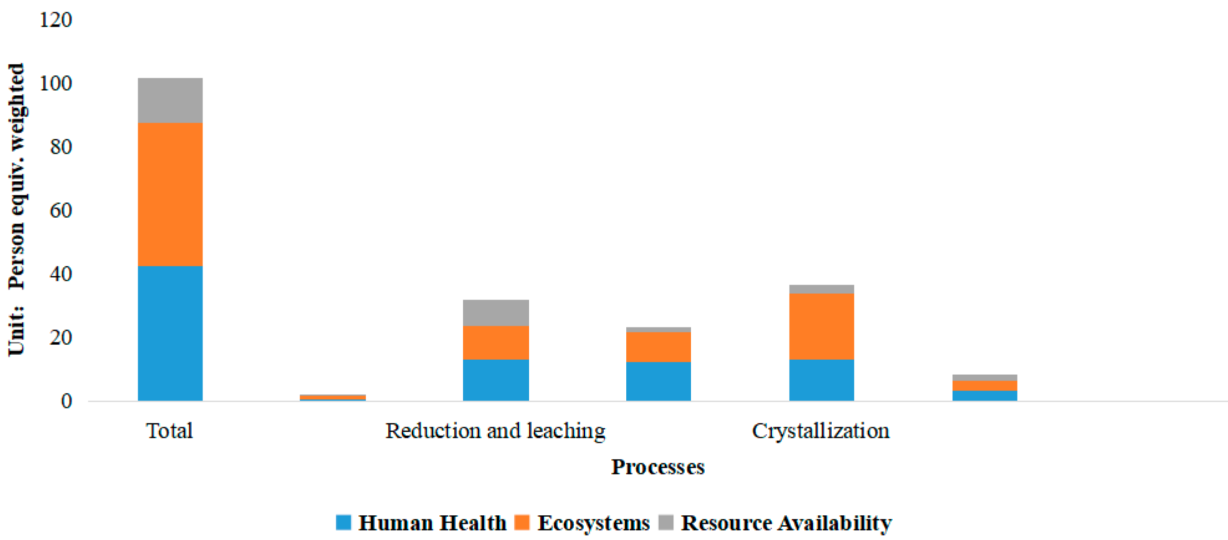

Figure 4. Comparison of the life cycle impact indicators of the various production processes for the recovery of Ni-Co-containing saggars.

The weighted human equivalents of each process (Figure 4) are 1.65 for physical separation, 31.94 for reduction and leaching, 23.29 for cascade separation, 36.66 for crystallization, and 8.42 for purification. The most important part of the environmental impact is crystallization, followed by reduction and leaching, which account for 36.0 and 31.4% of the total environmental impact, respectively, and show the highest need for improvement.

Indicators: global warming potential (GWP), ozone disposal potential (ODP), ionizing radiation potential (IRP), particulate matter formation potential (PMFP), ozone formation potential (HOFP), ecosystem ozone formation potential (EOFP), acidification potential (AP), freshwater industrialization potential (FEP), human toxicity potential (HTP), terrestrial ecotoxicity potential (TETP), fresh water ecotoxicity potential (FETP), marine ecotoxicity potential (METP), marine eutrophication potential (ME), agricultural land occupation potential (LOP), and water consumption potential (WCP).

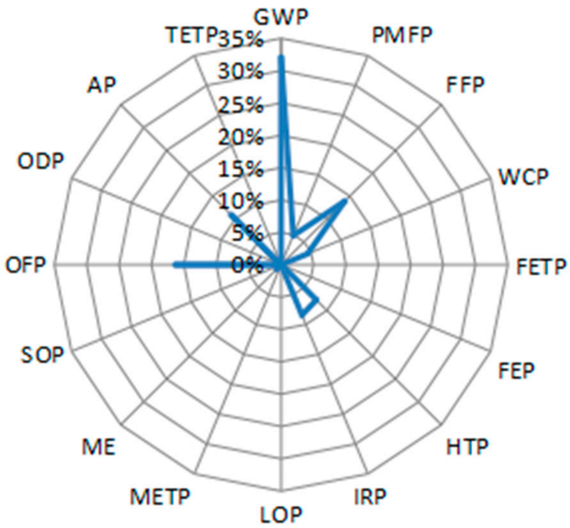

Figure 5. Subdivided endpoint indicators for the recovery of Ni-Co-containing saggars.

3.3. Uncertainty Analysis

Monte Carlo analysis was used to assess the uncertainty of the LCA results (Garcia Sanchez and Guereca, 2019) [31]. This method measures the uncertainty of the observation value by calculating the average value, standard deviation (SD) and coefficient of variation (CV) of the observation value. The confidence interval is 95%; that is, if the CV value of the observation value is less than 5%, it indicates that the observation value has a high degree of certainty.

The analysis results (Table 3) show that the CVs of the input-output quality analysis results are 5.89×10^{-10} and 5.20×10^{-10}, respectively. In the midpoint data, the largest CV is the CV of ODP (2.06%), while the CV of the endpoint data is 4.08×10^{-7}. Both of them are far less than 5%, which shows that the calculation results have high reliability.

Table 3. Monte Carlo analysis of the Ni-Co-containing saggar recovery results.

	Unit	Mean	SD	CV (%)
Mass-Input	kg	2.99×10^{5}	1.76×10^{-6}	5.89×10^{-10}
Mass-Output	kg	3.00×10^{5}	1.56×10^{-6}	5.20×10^{-10}
Midpoint-GWP	kg CO_2 eq.	3.33×10^{2}	1.03×10^{-6}	3.09×10^{-7}
Midpoint-PMFP	kg PM2.5 eq.	6.74×10^{-1}	1.82×10^{-6}	2.70×10^{-4}
Midpoint-FFP	MJ eq.	1.55×10^{2}	1.84×10^{-6}	1.19×10^{-6}
Midpoint-WCP	m^3	2.94×10^{2}	0.00×10^{0}	0.00×10^{0}
Midpoint-FETP	kg 1,4 DB eq.	5.13×10^{-2}	1.77×10^{-6}	3.45×10^{-3}
Midpoint-FEP	kg P eq.	5.63×10^{-4}	2.52×10^{-6}	4.48×10^{-1}
Midpoint-HTP	kg 1,4-DB eq.	5.82×10^{0}	4.60×10^{-7}	7.90×10^{-6}
Midpoint-IRP	kBq Co60 eq. to air	3.17×10^{1}	1.58×10^{-6}	4.98×10^{-6}
Midpoint-LOP	Annual crop eq.·y	4.75×10^{0}	5.64×10^{-7}	1.19×10^{-5}
Midpoint-METP	kg 1,4-DB eq.	4.51×10^{2}	0.00×10^{0}	0.00×10^{0}
Midpoint-ME	kg N eq.	1.80×10^{-2}	1.35×10^{-6}	7.50×10^{-3}
Midpoint-SOP	kg Cu eq.	3.77×10^{0}	2.49×10^{-6}	6.60×10^{-5}
Midpoint-OFP	kg NO_x eq.	8.17×10^{-1}	1.64×10^{-6}	2.01×10^{-4}
Midpoint-ODP	kg CFC-11 eq.	1.21×10^{-4}	2.49×10^{-6}	2.06×10^{0}
Midpoint-AP	kg SO_2 eq.	2.33×10^{0}	1.54×10^{-6}	6.61×10^{-5}
Midpoint-TETP	kg 1,4-DB eq.	7.37×10^{1}	7.75×10^{-7}	1.05×10^{-6}
Endpoint	Weighted person equivalents	4.50×10^{2}	2.16×10^{-6}	4.80×10^{-7}

3.4. Sensitivity Analysis Results

A −10% change sensitivity analysis was conducted with all 22 raw material and emission variables. There were 12 items with an environmental impact of approximately

0.1% on the entire production process (Table 4), of which six items were greater than 0.5%, thereby showing a greater impact. The ranks are the sulfuric acid input in the reduction and leaching process, the potassium carbonate and steam input in the crystallization process, the potassium carbonate and potassium hydroxide input in the cascade separation process, and the ammonia input in the purification process. These six environmental impact changes account for 86.05% of the total impact on the recovery of Ni-Co-containing saggars. Among them, the input-output item with the greatest environmental impact is the sulfuric acid input in the reduction leaching process. If the acidic agent can be used more accurately, the environmental impact of the entire production process can be effectively reduced. The second is the input of potassium carbonate and potassium hydroxide in the crystallization and cascade separation process. Since this input needs to be excessive, the degree of excess needs to be controlled to reduce environmental effects. As the main energy source for the crystallization process, steam is also an important source of environmental impact. Therefore, it is also important to improve the heat utilization efficiency of the crystallization process. Finally, the use of ammonia in the purification process needs to be further optimized.

Table 4. Sensitivity analysis of the changes in the environmental impacts of major items.

No.	Process	Material Flow	Environmental Impact (%)
1	Reduction and leaching	Sulfuric acid	3.04
2	Crystallization	Potassium carbonate	2.03
3	Crystallization	Steam	1.22
4	Cascade separation	Potassium hydroxide	1.19
5	Cascade separation	Potassium carbonate	0.62
6	Purification	Ammonia water	0.52
7	Crystallization	Electricity	0.34
8	Purification	Carbon dioxide	0.21
9	Physical separation	Electricity	0.17
10	Purification	Sulfuric acid	0.14
11	Purification	Sodium hydroxide	0.13
12	Reduction and leaching	Electricity	0.10

4. Conclusions and Discussion

Based on the above analysis, we draw the following conclusions. The life cycle environmental impact of the recovery of waste Ni-Co-containing saggars by the Institute of Process Engineering of the Chinese Academy of Sciences is much lower than that of equivalent products produced by alternative production methods, and the weighted human equivalent is only 14.5% of the alternative process. Thus, this process is highly competitive in the ecological economy and has good application prospects. China produces approximately 5×10^5 tons of Ni-Co-containing saggars each year. For every ton of saggars processed, a profit of approximately 2100 RMB can be generated. If the entire process is used, a profit of 1.1×10^9 RMB can be generated. Additionally, it can reduce greenhouse gas emissions by 4.1×10^5 tons carbon dioxide eq., decrease fossil energy consumption by 1.5×10^5 tons oil eq., decrease metal consumption by 7×10^5 tons, and decrease the acidification effect by 1×10^3 tons SO_2 eq.

Out of all the indicators, carbon emissions are the indicators that have the greatest impact on the entire life cycle of Ni-Co saggar recycling, and the energy structure dominated by fossil energy is the most important environmental impact factor for this process. Second, there is a certain photochemical pollution effect in the crystallization process, which is related to exhaust gas emission during the crystallization process. In addition, the extensive use of acid in the reduction and leaching process increases the potential impact of acidification. In the future, the influence of the abovementioned areas should be controlled with emphasis.

Out of all the processes, crystallization has the greatest environmental impact, followed by the reduction and leaching process. This is because the crystallization process requires more energy and a large amount of the supersaturated sodium salt solution, while the reduction and leaching process is the reason that the raw material processing volume is large and more acidic liquid is required for input. Sensitivity analysis shows that in the total of 22 input and output items, the input of sulfuric acid in the reduction and leaching process, the input of potassium carbonate and steam in the crystallization process, the input of potassium carbonate and potassium hydroxide in the cascade separation process, and the input of ammonia in the purification process are the items with the greatest environmental impact and account for 86.05% of the overall environmental impact sensitivity, which is the focus of future process improvements.

Based on the above conclusions, we make the following recommendations:

1. Improve the energy utilization structure and reduce carbon emissions. Increase the use of clean energy and increase the efficiency of heat energy utilization in the crystallization process;
2. Improve the accuracy of the use of key raw materials, including sulfuric acid, potassium carbonate, potassium hydroxide, and ammonia. In the reductive leaching process, sulfuric acid is used more accurately. In the crystallization and cascade separation process, the degree of supersaturation of potassium carbonate and potassium hydroxide input is strictly controlled. Control the input of ammonia in the purification process. In addition, new materials that are more environmentally friendly should be explored;
3. Strengthen the air emission in the two processes of reduction and leaching and crystallization. Acid mist leakage in the reduction and leaching process should be reduced, the waste gas generated in the crystallization process should be treated, or a more environmentally friendly crystallization method should be used.

This paper used the life cycle assessment method to analyze the overall environmental impact of the recovery of Ni-Co-containing saggars, compared it with the environmental impact of alternative production methods, and evaluated the environmental impact of the recovery process. Furthermore, the final resource consumption and pollutant discharge of each production step in the recovery and utilization of Ni-Co-containing saggars were evaluated, and the environmental impact of each process in different fields was analyzed. The environmental benefits of Ni-Co-containing saggar recovery were far superior to alternative process production, and it was determined that crystallization and reduction and leaching were the two processes with the greatest environmental impact. Finally, through sensitivity analysis, the raw materials or emission items that should be prioritized for improvement were identified. This conclusion has high guiding significance for the improvement of the production process.

The study has some flaws due to the lack of progress in testing. First, due to defects in technical analysis, this paper fails to further analyze the process mechanisms of various major raw material consumption and waste discharge. In addition, this paper is limited to the analysis of the environmental impact of the production line and proposes directions for improvement. Therefore, suggestions for improvement are proposed to provide a clear process route. These problems will become an important direction for our further research.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su15097442/s1, Table S1: Material inventory when recovering Ni-Co-containing saggars; Table S2: Data quality assessment of the life cycle data of Ni-Co-containing saggar recovery. Table S3: Monte Carlo analysis of the Ni-Co-containing saggar recovery results.

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