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Recovery of Materials from Refrigerator: A Study Focused on Product Distribution, Recyclability and LCA Evaluation

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Abstract: This study outlines a recycling initiative conducted at Rekular GmbH, focusing on the recycling of 100 refrigerators. The recycling process employed a combination of manual dismantling, depollution, and mechanical processing techniques. Manual dismantling followed a predefined protocol to extract various materials, while the mechanical and physical processes involved shredding, zigzag, magnetic, and eddy current separation (ECS) to liberate and separate different materials. The resulting ferrous, non-ferrous and polymer product fractions were analyzed and categorized, providing valuable insights into the quality of interim products in the refrigerator recycling process. Simulations were then performed using FactSage^{TM} version 8.2 and HSC Chemistry 10 version 10.3.7.1 software to simulate the recovery of metals from the ferrous and non-ferrous fractions using pyro metallurgical and hydrometallurgical methods. An electric arc furnace (EAF) was utilized for iron (Fe), while a re-smelter process for aluminium (Al), and the black copper route was simulated for copper (Cu) recovery. The recovery rates including metallurgical, mechanical, and physical processes are as follows: Fe (78%), Al (68.4%), and Cu (52.4%). In contrast, the recovery rates through metallurgical processes are as follows: Al (99%), Fe (79%), and Cu (88%). This discrepancy is attributed to losses of these elements resulting from incomplete liberation in mechanical processing. Additionally, a product/centric approach was applied and the recycling index reached 76% for recovery the Al, Cu, and Fe metals in a refrigerator recycling process. Turning to the environmental impact evaluation within the life cycle assessment (LCA), the process unit with the highest emissions per refrigerator in the recycling process was the use of nitrogen during the shredding process, accounting for 3.7 kg CO_2 eq/refrigerator. Subsequently, the consumption of medium voltage electricity from the German grid during mechanical and physical separations contributed to 0.6 kg CO_2 eq/refrigerator. The EAF, and electrolytic refining stages in the metallurgical recovery process also had a notable impact, generating 10.7 kg CO_2 eq/refrigerator.

Keywords: metallurgy; processing; design for recycling; refrigerator recycling; recycling index; life cycle assessment; circular economies

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Citation: Paz, F.A.G.; Heibeck, M.; Parvez, A.M.; Torrubia, J.; van den Boogaart, K.G.; Raatz, S. Recovery of Materials from Refrigerator: A Study Focused on Product Distribution, Recyclability and LCA Evaluation. *Sustainability* **2024**, *16*, 1082. https://doi.org/10.3390/su16031082

Academic Editor: Giovanni De Feo

Received: 26 November 2023
Revised: 4 January 2024
Accepted: 10 January 2024
Published: 26 January 2024

1. Introduction

Electric and electronic equipment persist as a swiftly expanding category of waste within the European Union, exhibiting an annual growth rate of 2%, and it is estimated that less than 40% of waste electric and electronic equipment (WEEE) undergo the recycling process [1]: Annually, in Germany around three million domestic refrigeration appliances necessitate disposal [2]. This signifies that about 62,000 tons/year of a ferrous fraction, primarily consisting of Fe, and 1500 tons/year of a non-ferrous fraction, mainly composed of Al and Cu, require recovery. Moreover, it is important to mention that the environmental impact of products, to the extent of 80%, are established during the design phase [3]. Therefore, the circular economy (CE) concept aims to efficiently use resources by prolonging product life and transform waste into valuable materials. However, modern society's complex waste and consumer goods challenge CE viability. Advanced metallurgy, coupled with physical and mechanical separation is essential for successful CE technologies, addressing challenges in thermodynamics, technology, digitalization, and design for recycling (DfR). This integrated approach enhances resource efficiency in cities, aligning with the United Nations Sustainability Development Goals [4]. Moreover, characterizing the composition of materials in products such as refrigerators holds paramount importance in the realm of sustainable resource management and CE practices.

Within refrigerators, elaborate metal matrices incorporating various metallic elements such as Al, Fe, Cu, Cr, Ni, along with plastics such as polystyrene (PS), acrylonitrile butadiene styrene (ABS), and polyvinyl chloride (PVC) [5], creates a multi material structure (MMS) that poses unique challenges for recycling due to liberation and separation behavior [6]. Hence, understanding the precise composition and ensemble of refrigerators is crucial for devising efficient recycling strategies. This knowledge empowers the industry to recover materials including rare earths and critical raw materials, reducing reliance on primary production, which often carries substantial environmental burdens.

Furthermore, according to the 'Extended Producer Responsibility in Europe', products should be designed with a focus on good recyclability as well as reparability [7], thus by meticulously characterizing these materials, we can not only extract maximum value from discarded products but also avoid unnecessary waste. Therefore, contributing to a more sustainable and environmentally conscious future.

The comprehensive approach to recycling, involving manual dismantling, mechanical, physical, and metallurgical processes, exemplifies the commitment to a CE model. Through techniques such as shredding, zigzag separation, magnetic separation, ECS, pyro-hydro metallurgical processes valuable materials are reclaimed, diminishing the need for primary extraction [8]. It is imperative to integrate such practices into the LCA to accurately gauge their environmental impact.

Moreover, the extraction of materials such as steel alloys and the utilization of Polyurethane in the refrigerator's construction demonstrate the intricate knowledge required for efficient material recovery [9]. This meticulous approach not only minimizes waste but also promotes sustainable practices within the recycling industry, aligning with global efforts towards a greener and more environmentally conscious future. Furthermore, an in-depth evaluation of the recycling process, coupled with a comprehensive LCA encompassing both inventory and impact assessment, offers invaluable insights into the concept of DfR and the broader implementation of a CE.

DfR emphasizes the importance of considering recyclability at the initial stages of product development. By scrutinizing the composition of materials and their potential for recovery, designers can make informed choices that facilitate the eventual recycling of products [10]. This approach not only streamlines the recycling process but also reduces the environmental impact associated with resource extraction and manufacturing.

Additionally, a holistic LCA provides a comprehensive understanding of the environmental impacts associated with a product's entire life cycle. This evaluation enables us to quantify the benefits of a CE approach by considering the environmental burdens of each stage, from material selection to end-of-life treatment, we can identify opportunities for resource optimization and waste reduction [11]. This knowledge is pivotal in steering industries towards sustainable practices and driving innovation in materials and processes. Ultimately, the combination of a thorough recycling process evaluation and a robust LCA not only advances our understanding of sustainable product design but also paves the way for a more circular and environmentally conscious economy, where resources are conserved, waste is minimized, and the ecological impact of products is significantly reduced. The present study therefore aims to systematically identify and quantify the environmental impacts arising from the recycling and material recovery processes of refrigerators, through LCA approach with a specific focus on the integration of DfR principles within a CE framework.

2. Methodology

2.1. Manual Dismantling and Physical Processing of Refrigerators

This study outlines a recycling initiative conducted at Rekular GmbH, located in Baumholder, Germany, focusing on the recycling of 100 refrigerators, characterized as outlined in Table 1.

Table 1. Characteristics of refrigerators in the recycling process.

Refrigerator Type	Quantity	High (cm)	Wide (cm)	Weight (kg)
Small	31	185 ± 12	60 ± 5	70 ± 10
Wide	69	175 ± 4	90 ± 1	110 ± 20

The recycling process included two main techniques: manual dismantling and a mechanical procedure, where the material was transported on a conveyor belt with an energy consumption of 0.6 kWh/t. In the initial manual dismantling phase, electrical components such as printed circuit boards (PCBs), displays, cables, fans, glass shelves, and copper circuit-pipes were extracted using a hydraulic spreader-cutter and screwdrivers. Following this step, a depollution phase was conducted to remove the refrigeration oil and refrigerant. Lastly, a second manual dismantling process was employed to extract the compressor from the depolluted refrigerators. Throughout the process, the energy consumption was 1.03 kWh/t. The detailed breakdown of the materials removed during this process is presented in Table 2.

Table 2. Mass fraction of manually dismantled components and depollution in the refrigerator recycling process.

Material	Compressor	Electrical Parts, Display, Fans, etc.	Glass (Shelf)	Copper Circuit-Pipe	Refrigeration Oil and Refrigerant
(wt.-%)	9.20	2.2	1.2	0.20	0.20

In Figure 1, a block flow diagram delineates the entire recycling process in the studied refrigerator recycling plant. In the mechanical process, and as mentioned by [12], a shredding technique was employed to break down the MMS from the refrigerators into smaller fragments with an expected high liberation degree and energy consumption of 1.7 kWh/t. This process should facilitate favorable conditions for subsequent recycling processes such as metallurgical recovery processes [13]. Thereafter, a tube chain conveyor with an energy consumption of 1.91 kWh/t facilitates the transport of the shredded material to the zigzag separator which was utilized to segregate the lighter fraction from the shredded material, hence this separation step aims to isolate components with lower density, such as plastics and foams, from the heavier components [14], with and energy consumption of 3.67 kWh/t. Following the zigzag unit, a magnetic separator model Steinert MOR 95 156 MT 40 A (Cologne, Germany), was employed to separate the ferrous fraction from the non-ferrous fraction [15] with an energy consumption of 1.60 kWh/t. Furthermore, the non-ferrous fraction, which primarily consists of Al and Cu, was then subjected to an ECS model Steinert NES 100 220 E 50097 (Cologne, Germany). This separator utilizes eddy currents induced by a varying magnetic field to efficiently separate non-ferrous metals from other materials with an energy consumption of 0.76 kWh/t. This physical process plays a crucial role in the recovery of valuable non-ferrous metals, ensuring their separation and subsequent processing for recycling purposes [16]. The material composition of the intermediate materials streams in this process is presented in Section 3..

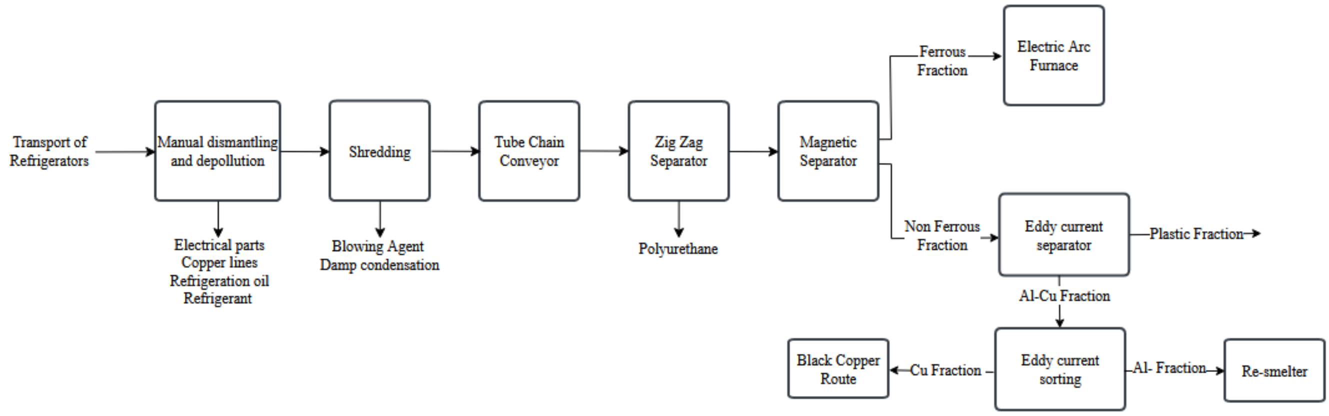

Figure 1. Block flow diagram of refrigerator recycling process.

Overall, the combination of manual dismantling and depollution, along with the mechanical process involving shredding, zigzag separation, magnetic separation, and ECS, enables the recovery of valuable materials from the refrigerators [17]. For the simulation of metal recovery, the composition of materials and alloys is presented in Section 3. However, it is crucial to emphasize that within the shredding process unit, Damp Condensation and Blowing Agent, constituting 3.7 wt.-% and 0.5% of the total input mass, respectively, were extracted from the refrigerators. The latter is routed to a Cryo-Condensation Plant. Furthermore, in the Zig Zag separator unit, Polyurethane, accounting for 21.7% of the total input mass, was also extracted from the refrigerators. It is noteworthy to emphasize that the primary regulatory reference cited by facility operators is the (TA Luft), denoting the technical instructions on air quality control.

The internal structure of the refrigerator is constructed using a specific type of steel known as S275JR/1.0044, characterized by properties outlined in [18]. This particular steel constitutes 70% of the total steel content in the refrigerator by weight. Regarding the housing or casing of the refrigerators, two distinct types of steel have been identified by characterization of metal alloys by X-ray fluorescence measurements. The first type, accounting for 26.1% by weight, corresponds to Steel type DCO5/1.0312, with properties specified by [19]. The second type, comprising 3.9% by weight, is fashioned from stainless steel, specifically an Austenitic grade denoted as 304 in the AISI classification. This composition aligns optimally with the requirements for utilization in refrigerator production, as indicated by reference [20]. In the case of the Al alloy composition, Al 3033 has been employed owing to its suitability for use in refrigerator production [21]. Regarding the magnet, a Neodymium-based one has been chosen due to its widely application in refrigerators production [22]. The composition of plastics has been determined based on the average proportions of various plastic types found in different studies about recycling of refrigerators [23–25]. Finally, the composition of PCB had been determined based on previous research's on metal recovery from WEEE [26].

2.1.1. Recovery of Steel-Electric Arc Furnace

Once the manual, mechanical and first physical separation was completed, the simulation for metal recovery was performed on the ferrous fractions, and it was conducted to explore the recovery of Fe using pyro metallurgical methods, specifically EAF technology [27]. As shown in Figure 2, all of the mechanical and physical aforementioned processes were transferred to HSC Chemistry 10 version 10.3.7.1 [28]. Furthermore, for the pyro metallurgical recovery simulation process, FactSage^{TM} version 8.2 [29] was applied. This software platform provides advanced computational tools for thermochemical calculations, allowing for accurate and efficient simulations of complex metallurgical processes.

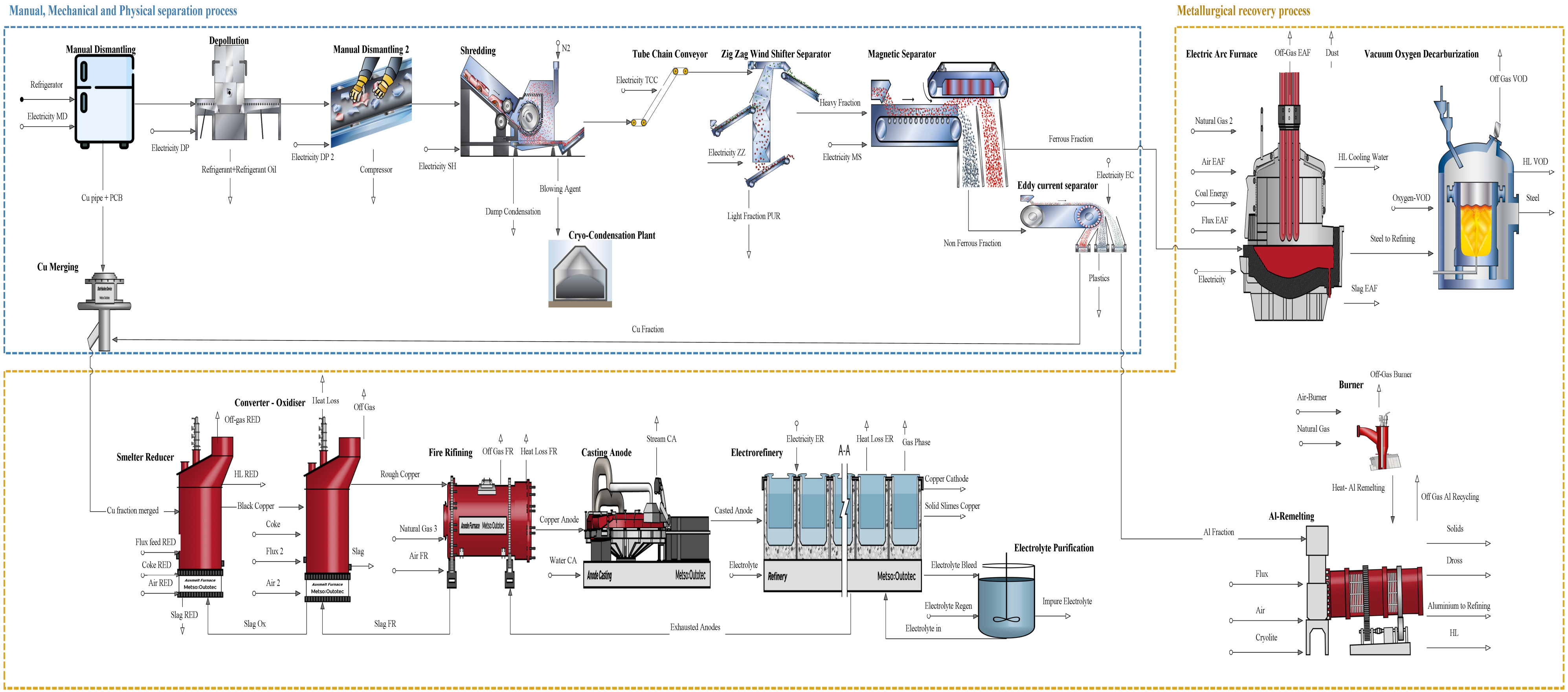

Figure 2. HSC Flowsheet of refrigerator recycling process and metallurgical recovery processes for iron, aluminium and copper2.2. Metallurgical recovery of metals from refrigerator recycling process.

The simulation for the recovery of Fe from the ferrous fraction, which comprises around 3.55 tons, was performed using an EAF. Table 3 provides a detailed breakdown of the elemental composition of the ferrous fraction, encompassing metals, oxides, and plastics.

Table 3. Composition (in wt.-%) of the ferrous product from refrigerator recycling.

Fe	C	Cr	Ni	Mo	Mn	Si	S
94.7	0.2	0.7	0.3	0.03	1.2	0.04	0.03
P	**N**	**Cu**	**Al**	**B**	**Nd**	**Nb**	**Dy**
0.03	0.01	0.4	0.1	0.01	0.16	0.005	0.01
Sn	**Pb**	**Au**	**Zn**	**Ag**	**Pd**	**Ta**	**Co**
5.00×10^{-4}	4.70×10^{-4}	1.00×10^{-5}	1.20×10^{-4}	2.00×10^{-5}	3.00×10^{-6}	3.00×10^{-6}	3.00×10^{-6}
SiO_2	**MgO**	**Al_2O_3**	**CaO**	**K_2O**	**Na_2O**	**TiO_2**	**C_2F_4**
0.12	3.00×10^{-4}	9.00×10^{-4}	2.00×10^{-4}	2.00×10^{-4}	3.00×10^{-4}	5.00×10^{-4}	3.00×10^{-4}
C_2H_6	**C_2H_3Cl**	**C_2H_4**	**C_3H_6**	**C_3H_6O**	**C_8H_8**		
0.0033	0.10	0.10	0.10	0.10	1.63		

The simulation involved subjecting the ferrous fraction to a temperature of 1600 °C [30]. Additionally, chemical energy is introduced by oxygen in the air feed, oxides present in fluxing agents, and carbonaceous materials such as coal and natural gas, thus facilitating the smelting and recovery of Fe. This pyro metallurgical approach offers an efficient and environmentally friendly method for extracting Fe from the ferrous fraction [31]. The calculation of the model output was simulated in FactSage^{TM} version 8.2 [29], and as illustrated in Figure 3, the EAF and vacuum oxygen decarburization (VOD) was simulated in HSC Chemistry 10 version 10.3.7.1 software, following the methodology as previously demonstrated by other studies [11].

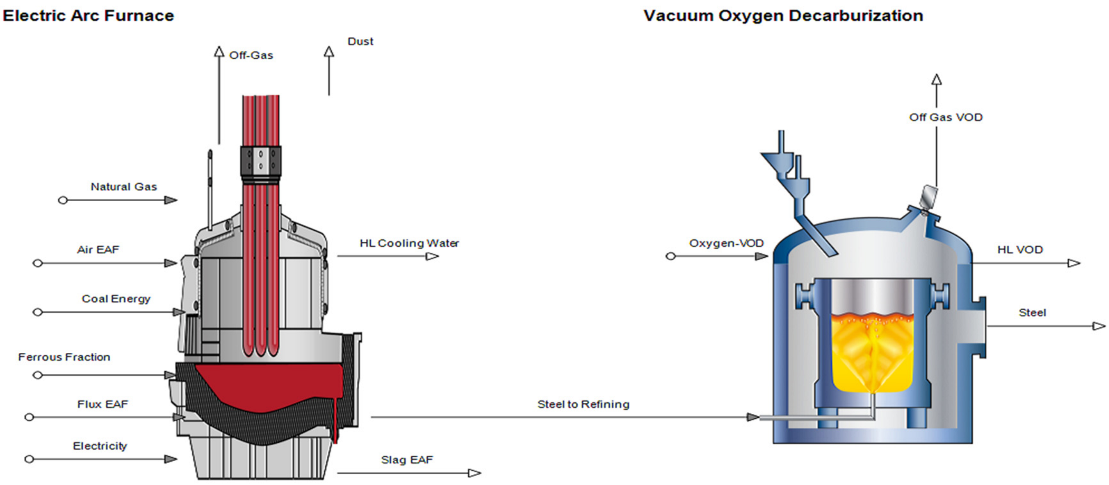

Figure 3. In- and output flows of Fe recovery units in HSC simulation.

For FactSage^{TM} calculations, the following phases were allowed: (Fact PS), for the gas phase, (FToxid) for the slag phase, and the molten metal phase, which is Fe-based (Fsteel). Throughout the operation, the ferrous fraction was loaded into the EAF and subjected to 391 kWh/t of electrical energy, which generates an electric arc and produces intense heat, melting the scrap [32]. During the melting process, impurities were mainly eliminated in oxidic forms within the gas and slag phases, thus, the chemistry of the molten metal was altered to produce the desired recovery rate. Moreover, to aid in this process, CaO was added as a fluxing agent [33]. Notably, electrical energy was the primary source of energy, accounting for more than half of the energy required in the EAF as mentioned by [31]. During the EAF process, byproducts such as off-gases, dust, and slags were generated along with the molten metal, which was then sent for further refining in a VOD process. VOD unit was simulated to further improve the quality of the finished product by blowing O_2, and lowering the carbon content in Cr-rich melts with C values below 1 in wt.-%, thus removing other impurities after the metal had been melted and refined [34].

2.1.2. Recovery of Aluminum—Remelting

Compared to primary production, recycling Al saves approximately 95% of the energy needed [35]. In the present case study, the collection of Al scrap (66.30 kg) is obtained after ECS, and its elemental composition is delineated in Table 4.

Table 4. Aluminum Scrap composition (wt.-%).

Cu	**Al**	**Mn**	**Zn**	**Si**	**Mg**	**Fe**	**B**
0.85	98.59	0.053	0.027	0.026	0.026	0.017	2.80×10^{-4}
Nd	**Nb**	**Dy**	**Sn**	**Pb**	**Ni**	**Au**	**Ag**
0.0074	2.34×10^{-4}	2.80×10^{-4}	0.0013	0.0013	4.45×10^{-4}	1.78×10^{-5}	4.45×10^{-5}
Pd	**Ta**	**Co**	**SiO_2**	**MgO**	**Al_2O_3**	**CaO**	**K_2O**
8.90×10^{-6}	8.90×10^{-6}	8.90×10^{-6}	0.022	8.90×10^{-4}	0.0026	4.45×10^{-4}	4.45×10^{-4}
Na_2O	**TiO_2**	**C_2F_4**	**C_2H_6**	**C_2H_3Cl**	**C_2H_4**	**C_3H_6**	**C_3H_6O**
8.90×10^{-4}	0.0013	8.90×10^{-4}	0.0093	0.017	0.017	0.019	0.017
C_8H_8							
0.27							

The re-melting process for Al recycling was simulated using HSC Chemistry [28] as shown in Figure 4. Furthermore, the FactSage^{TM} version 8.2 software with the following phases: (Fact PS), for the gas phase, (FToxid) for the slag phase, (FT salt) for the inorganic salt database, and the molten metal phase which is Al based (FTlite) [29] has been used for the Al recovery thermodynamic simulation.

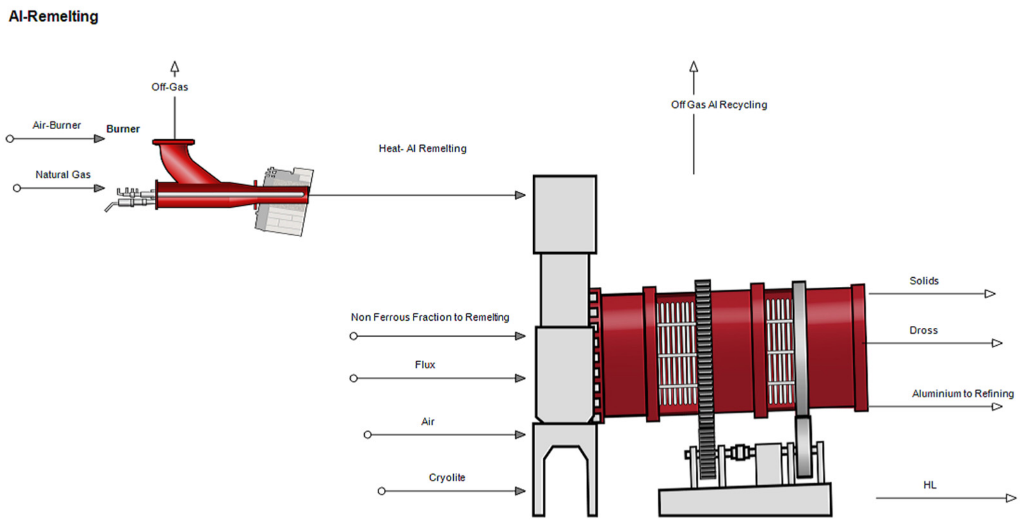

Figure 4. HSC simulation of aluminum recovery from aluminum scrap.

The simulation investigated the influence of a burner as the primary heat source and the incorporation of fluxing agents, including NaCl, KCl, and Cryolite (Na_3AlF_6) to lower the melting point of alumina, hence the amount was determined based on its proportion relative to the overall fluxing agent composition, thus a calculated percentage of 5% was implemented in the simulation [36]. For the burner combustion, natural gas was added, and the temperature was set at 800 °C to facilitate the melting of the aluminum scrap [37]. Moreover, as it was also mentioned by [38], the simulation generated several outputs, including off gases, a solid phase, dross, and molten aluminum which needs to be further refined according to specific industry requirements [39].

The solid phase analysis revealed the presence of impurities and non-metallic materials, while the dross composition comprised non-reduced metallic elements and oxide compounds. Efficient dross management is crucial for maximizing Al recovery and minimizing material losses [40].

2.1.3. Recovery of Copper

Copper scrap coming from the mechanical and physical separator process, is indicated in Table 5. This scrap material consists of a non-ferrous composition predominantly comprising Cu, which has undergone separation via an ECS, additionally; copper tubes and PCB coming from manual dismantling are also considered in the feed composition.

Table 5. Copper scrap composition (wt.-%).

Cu	Al	Mn	Zn	Si	Mg	Fe	B
64.32	7.68	1.34	0.83	0.67	0.67	1.60	7.04×10^{-5}
Nd	**Nb**	**Dy**	**Sn**	**Pb**	**Ni**	**Au**	**Ag**
0.19	5.86×10^{-5}	7.03×10^{-5}	0.61	0.61	0.20	8.15×10^{-5}	0.02
Pd	**Ta**	**Co**	**SiO_2**	**MgO**	**Al_2O_3**	**CaO**	**K_2O**
4.0×10^{-5}	4.07×10^{-5}	4.07×10^{-5}	3.44	0.41	1.22	0.20	0.20
Na_2O	**TiO_2**	**C_2F_4**	**C_2H_6**	**C_2H_3Cl**	**C_2H_4**	**C_3H_6**	**C_3H_6O**
0.41	0.61	0.41	4.28	0.84	0.43	1.45	0.43
C_8H_8							
6.86							

As can be shown in Figure 5, and as mentioned by [11], the proposed methodology entails a sequence of operations, following a path known as the black copper route.

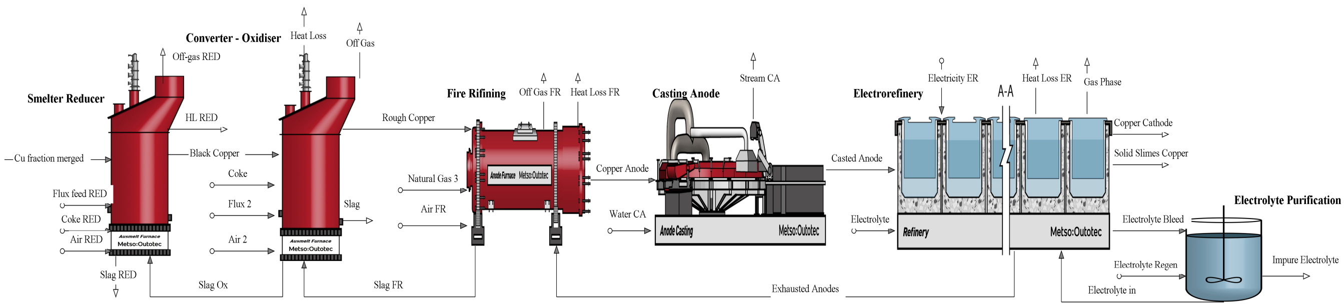

Figure 5. HSC simulation of copper recovery process.

This route commences at the smelter-reducer, followed by the converter-oxidizer stage. Subsequently, the material undergoes fire-refining, casting, and ultimately concludes with electro refining, with the aim of achieving a copper cathode with a Cu purity level of 99.99% [41]. The Cu recycling process was simulated using HSC Chemistry [28] and FactSageTM version 8.2 software with the following phases: (Fact PS), for the gas phase, (FToxid-SLAGA) for the slag phase, and the molten metal phase which is Cu based (FScopp-Liqu). In the smelter, plastics within the scrap serve as an alternative source of energy, replacing coke. Additionally, metals such as Al, Mn, and Mg will not be reclaimed, resulting in their conversion into oxides in the slag phase [26]. This stage involves subjecting the Copper scrap material to high temperatures (1300 °C), under a pO_2 of 10^{-8} atm and enriched oxygen, also with the presence of fluxing agents such as FeO, CaO, SiO_2, The purpose is to generate a Cu content in black copper up to 80 wt.-% as well as 1 wt.-% of Cu_2O in slags and minor metals in the off gas phases [30]. Moreover, the converter, follows the smelter and focuses on the oxidation of impurities present in the black copper at (1300 °C), under a pO_2 of 10^{-5} atm, to obtain rough copper or so called blister copper with a Cu content of 96–97 wt.-% [42]. The converter utilizes coke as a reductive agent, in conjunction with fluxing materials such as FeO, CaO, and SiO_2, to facilitate slag formation [41]. During fire refining, impurities in the blister copper, including Fe, Sn, Pb, and other minor elements, are segregated into the slag phase with the aid of fluxing agents through high-temperature smelting at (1200 °C), under a pO_2 of 10^{-6} atm. This meticulous procedure substantially augments purity Cu levels, reaching between 98.5 and 99.8 wt.-% [43]. In the casting stage, the molten copper is carefully poured into molds to form anodes [44]. Finally, Electrolytic refining process employs an electrolytic cell, typically consisting of an electrolyte solution at (64 °C), containing 170–200 g/L H_2SO_4, 40–50 g/L Cu and two electrodes: the anode (made of the cast copper anodes) and the cathode. The objective of electrolytic refining is to further purify the Cu obtained from previous stages and achieve an extremely high level of Cu purity, often reaching 99.99%, it also separates valuable impurities such as Au, Ag and Pd to recover them as by products [45].

2.2. Life Cycle Assessment

LCA serves as a method for comprehensively analyzing the environmental aspects and potential impacts of a product or service, considering its entire lifecycle, spanning from creation to disposal [46]. According to the [47] standard, the LCA process encompasses four key stages: establishing the goal and scope, compiling a Life Cycle Inventory (LCI), as well as a life cycle impact assessment (LCIA), and interpreting the findings. In-depth information regarding these stages is available in [48,49]. For the LCA, OpenLCA 1.10.0 software is coupled with Ecoinvent 3.8 database.

2.2.1. Goal and Scope

The goal of this comprehensive LCA is to quantitatively evaluate the environmental impacts associated with the recycling of 100 refrigerators at Rekular GmbH. The function of the recycling process is to recover Al, Cu and Fe from refrigerators. Hence, the functional unit will be evaluated as the amount of kg of Al, Cu, or Fe per Refrigerator within the recycling process. This aspect holds significant importance in the LCA study, as the results will be presented in relation to the chosen functional unit [50]. As illustrated in Figure 6, the boundary for the LCA was established employing a Gate-to-Gate approach.

2.2.2. Life-Cycle Inventory

In conducting the LCI, the Ecoinvent 3.8 database was utilized. Herein, Reagents, Energy, Emissions, and Waste were categorized as technosphere and elementary flows, while 100 refrigerators constituted an intermediate flow, and Cu, Al, and Fe represented the product flow. The phase involves gathering and evaluating the LCI. According to [47], this process entails compiling and quantifying input and output data within the defined system boundary, accounting for the movement of materials, energy, waste, and resources. The data is specific to the functional unit, encompassing the energy and materials required for it, as well as the resulting emissions and waste generation. Additionally, allocation is crucial to exclude co-production and by-products from the system [48]. It is important to mention that the treatment for the blowing agent in the Cryo-Condensation plant has not been integrated into the LCI for the LCA.

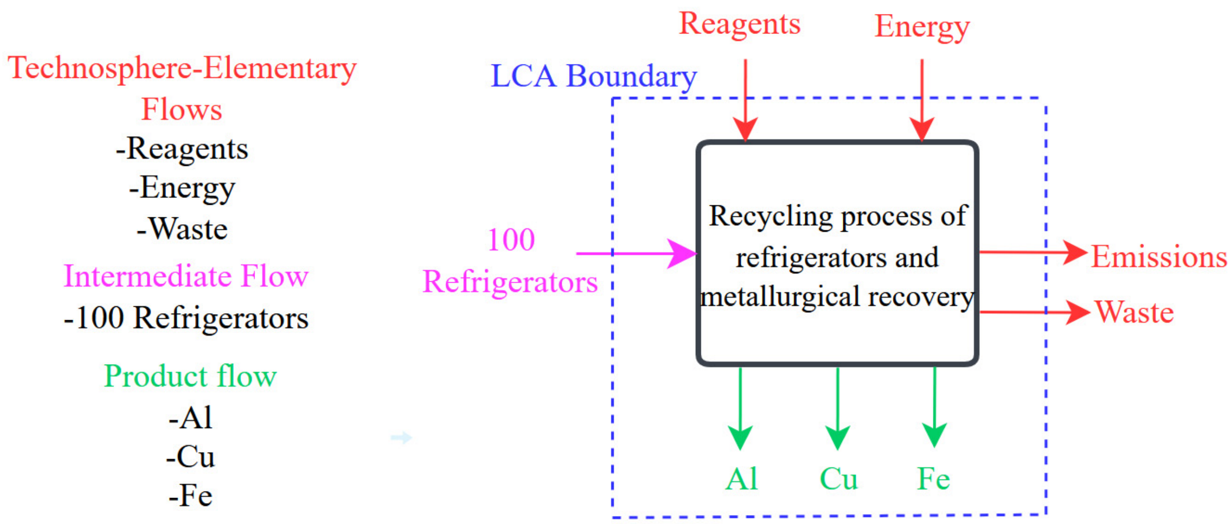

Figure 6. LCA Boundary.

2.2.3. Life-Cycle Impact Assessment

LCIA involves considering factors such as resource consumption, emissions, and waste generation. The initial step involves selecting impact categories, indicators, and characterization models. The subsequent stage involves categorizing individual flows based on the impact categories they contribute to. Finally, characterization factors are applied to these categorized elementary flows, yielding quantifiable values for comparison [46]. The midpoint approach scrutinizes effects occurring in the middle of the causality chain. The ReCiPe method is used in LCIA calculations, assigning impact scores to various emissions through characterization factors. These factors can be determined using two primary methods: at the midpoint or endpoint levels. ReCiPe provides calculations for both—18 midpoint indicators and 3 endpoint indicators [51].

2.2.4. Interpretation of the Results

In accordance with [47], the final phase encompasses interpretation. Here, the results of both LCI and LCIA are summarized and deliberated, considering the defined goal, scope, limitations, and sensitivity analysis within the refrigerator recycling process, and recovery of materials. The obtained insights hold valuable utility for a range of applications, including well-informed decision-making and DfR endeavors [46].

2.3. Design for Recycling

DfR, a pivotal strategy within the broader framework of Eco-design, embodies a holistic approach to product development, since it encompasses a diverse range of technical considerations, all aimed at optimizing end of life products recovery. This achievement could be facilitated by the creation of industrial ecological systems, guided by the use of advanced simulations [52]. These simulations play a crucial role in assessing the ecological footprint of a product, thereby informing the selection of materials based on criteria such as reusability, reparability, remanufacturability, and recyclability [53]. Hence, and as mentioned by [52], adopting a product/centric approach to recycling, we delve into a comprehensive methodology, since it goes beyond mere disassembly and physical recycling (separation), forging a vital link to metallurgical processes and other final treatment procedures. It is in this phase that materials find their way back into the resource cycle, ensuring a more sustainable utilization. As stated by [54,55], the adoption of a Recycling Index emerges as a pivotal metric, since it indicates the recycling performance of both products and their constituent materials. This index is derived by dividing the recovery rate of individual elements by the total sum, thereby providing a nuanced assessment of their sustainability credentials. This, in turn, propels the imperative for a product-centric recycling framework. Moreover, the quantification of recyclates is imperative, as it enables the utilization of thermodynamic and physical properties within simulation models calibrated with industry-specific data [6].

For a clearer quantitative understanding, the Recycling Index (RI) introduced by [55] has been employed, where the overall RI is derived from the weighted average of the individual recycling rates. This collective metric serves as the foundation for the comprehensive assessment of recycling efficiency.

3. Results and Discussions

3.1. Manual and Physical Separation

As depicted in Figure 7, the process involves a sequence of manual, mechanical, and physical separations, yielding distinct fractions. Notably, a light fraction, comprising 2100 kg of Polyurethane, which represents 21.7 wt.-% of the total refrigerator, is obtained from the Zig Zag separator. Subsequent to ECS, 2190 kg of plastic is extracted from the process, achieving a commendable recovery rate of 96%. The remaining 4% constitutes unrecovered plastic, which remains unliberated within the ferrous and non-ferrous fractions. Within the ferrous fraction, 3.2% of the unliberated plastic fraction is present in the 3555 kg total mass, while the remaining 0.8% persists unliberated within the 2358 kg of non-ferrous fraction.

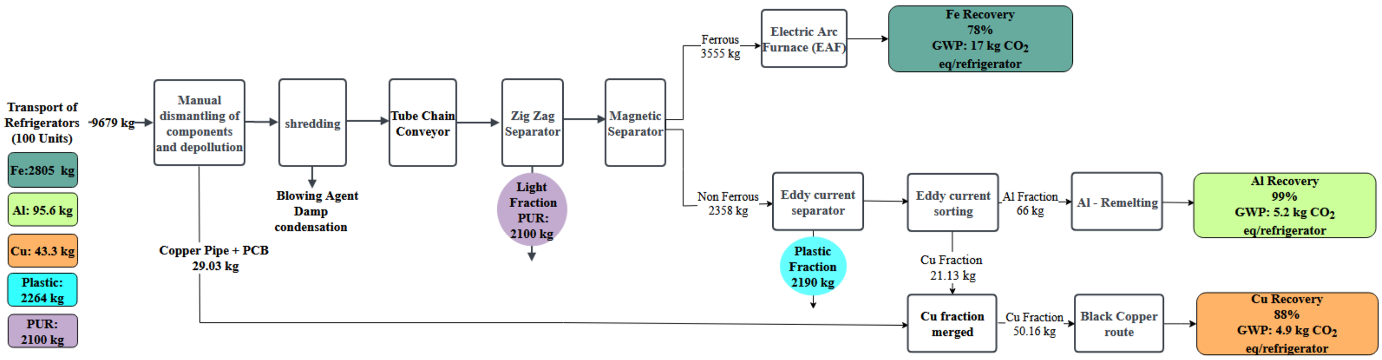

Figure 7. Distribution of materials after mechanical, physical, and metallurgical processes.

After manual dismantling, and as shown in Table 6, the mechanical and physical processes dismantle the refrigerators, resulting in fragments comprising ferrous and non-ferrous metals, magnets, plastics, glass, wood, and PCBs. Hence, downcycling principles manifest in the process units within the refrigerator recycling process. In this context, it is paramount to prioritize robust health and safety protocols to mitigate potential risks and safeguard the well-being of workers engaged in the recycling activity. A thorough risk assessment is essential to address possible hazards, including ergonomic strains, exposure to hazardous materials, the presence of sharp objects during dismantling, and the risk of injuries associated with heavy machinery or manual handling of materials. In the context of metallurgical recovery of metals, involving the use of reagents and high temperatures, additional risks must be considered. These include the safe handling of chemical reagents and the exposure to elevated temperatures, adding complexity to risk management and necessitating appropriate preventive measures. Adequate training, protective equipment, and adherence to established safety procedures are crucial elements in ensuring a secure working environment during the recycling process.

Steel represents 35.9 wt.-% of the total mass of the refrigerator. Moreover the internal structure primarily utilizes S275JR/1.0044 steel, constituting 70% of its total steel content. The housing incorporates two distinct steel alloys: DCO5/1.0312; 26 wt.-% and AISI 304 Austenitic stainless steel 40 wt.-%. Moreover, Al 3033 alloy, representing 1 wt.-% of the total refrigerator mass, is chosen for its suitability in refrigerator production. A Neodymium-based magnet, accounting for 0.4 wt.-% of the total refrigerator mass, is selected for its common application in refrigerator production. Additionally, the plastic composition, constituting 23.4 wt.-% of the total refrigerator mass, is determined based on average proportions from various recycling studies, while the PCB composition is derived from prior research on metal recovery from WEEE. The wt.-% of the PCB are defined as the 0.1% of total mass of refrigerators. The elemental composition of these materials is presented in Table 7.

Table 6. Material obtained after mechanical and physical process.

Shredding (kg)						
Steel	**Cu**	**Al**	**Magnet**	**Plastics**	**Glass and Wood**	**PCB**
3474.76	19.36	96.79	38.72	2264.89	9.68	9.68
Zigzag Separator (kg)						
Steel	**Cu**	**Al**	**Magnet**	**Plastics**	**Glass and Wood**	**PCB**
3474.76	19.36	96.79	38.72	2264.89	9.68	9.68
Ferrous Fraction after Magnetic Separator (kg)						
Steel	**Cu**	**Al**	**Magnet**	**Plastics**	**Glass and Wood**	**PCB**
3457.39	1.28	1.84	17.65	72.48	4.21	0.55
Non Ferrous Fraction to ECS (kg)						
Steel	**Cu**	**Al**	**Plastics**	**Magnet**	**Glass and Wood**	**PCB**
17.37	18.08	94.95	2192.41	21.06	5.47	9.13

Table 7. Elemental composition of materials and alloys (wt.-%) present in refrigerator.

Stainless Steel (wt.-%)							
Fe	C	Cr	Ni	Mo	Mn	Si	S
70.5	0.07	17.5	8	0.8	2	1.0	0.015
P	N						
0.045	0.11						
Steel 1.0312 (wt.-%)							
Fe	C	P	S	Mn			
99.54	0.06	0.025	0.025	0.35			
Steel 1.0044 (wt.-%)							
Fe	C	Mn	P	S	N	Cu	
97.658	0.21	1.50	0.035	0.035	0.012	0.55	
Al 3003 (wt.-%)							
Al	Mn	Cu	Zn	Si	Mg		
97.0	1.0	0.5	0.5	0.5	0.5		
Magnet (wt.-%)							
Al	Fe	B	Nd	Nb	Dy		
0.4	64.2	1.2	32.0	1.0	1.2		
Plastic (wt.-%)							
PS	ABS	PVC	PP	PE	PC	POM	PMMA
60	10	5	5	5	5	5	5

Table 7. *Cont.*

Waste PCB (wt.-%)							
Cu	Fe	Al	Sn	Pb	Ni	Zn	Ag
22	6	4	3	3	1	0.8	0.1
Au	Pd	Ta	Co	C_3H_6	C_2F_4	C_2H_6	C_2H_3Cl
0.04	0.02	0.02	0.02	5	2	10	2
$C_{10}H_{20}O_2$	$C_{15}H_{16}O_2$	$C_{12}H_{22}O_4$	SiO_2	MgO	Al_2O_3	CaO	K_2O
5	5	1	15	2	6	1	1
Na_2O	TiO_2						
2	3						

3.2. Metallurgial Process

Within the pyrometallurgical process, the steel product composition after the EAF route is listed in Table 8. Further refinement is necessary to meet the requirements for its intended application. The electricity consumed in this process amounts to 1727 kWh. The recovery rate of iron (Fe) is 79%, resulting in direct CO_2 emissions of 0.7 ton CO_2 per ton of steel. However, the recovery rate from refrigerator is 78%. Additionally, 711 kg of oxides within the slag phase comprising FeO, Cr_2O_3, MnO, as well as 890 kg of gases containing Co, CO_2, N_2, and O_2, have been generated.

Table 8. Steel product composition after Vacuum Oxygen Decarburization.

Steel Composition	Fe	Cr	Ni	Mo	Mn	S	Cu	O_2
(wt.-%)	99.3	0.5	0.04	0.03	0.06	0.01	0.05	0.02

Within the pyrometallurgical process, the aluminum obtained (as shown in Table 9) from the process serves as the primary target metal for recycling, finding applications across various sectors of the aluminum industry. The recovery of aluminum from the non-ferrous fraction is 99%, resulting in direct CO_2 emissions of 0.13 ton CO_2 per ton of aluminum. However, the recovery rate from refrigerators is 68.4% due to the high unliberated fraction of aluminum present in the plastic fraction 24.8 wt.-% and the ferrous fraction 1.9 wt.-%.

Table 9. Composition of aluminum product after Re-smelting.

Aluminum to Refining	Al	Fe	Cu	Zn	Mg
(wt.-%)	99.9	0.001	0.001	0.0001	0.0001

For copper recovery as depicted in Table 10, the copper content in the copper cathode acquired through the electrolytic refining following the black copper route surpasses 99.99 wt.-%, with a recovery rate of 52.4% in the refrigerator recycling process, and 88% in the metallurgical recovery process. This substantial loss could be attributed to the significant fraction of copper utilized in Steel 1.004 (0.55%), which is consequently lost during the Iron recovery process. Finally, for the Black Copper route recovery process, the electricity consumption is 8.35 kWh.

Table 10. Composition of copper cathode.

Copper Cathode	Cu	Fe	Pb	Ni
(wt.-%)	99.99	0.0003	0.001	0.002

Furthermore, as shown in Table 11 the total energy consumption from the manual, mechanical, and physical separation process is 96 kWh, and 1723 kg of nitrogen is used in the shredding process to maintain an inert atmosphere for the Blowing Agent extraction. The total electrical energy required for the metallurgical recovery process is distributed as follows: EAF: 1727 kWh, and Black Copper Route: 8.35 kWh.

Table 11. Electricity consumption of processing steps in refrigerator recycling and metallurgical recovery.

Process	Electricity Consumption (kWh)
Conveyor Belt	6
Manual dismantling and depollution	10
Shredding	14.2
Tube Chain Conveyor	16
Zig Zag Separator	30
Magnetic Separator	13.4
Electric Arc Furnace	1727
Eddy Current Separator	6.4
Electrolytic refining	8.35

3.3. Recycling Index

The recycling and recovery rates achieved through mechanical, physical, and metallurgical processing in the refrigerator recycling process are meticulously documented in Table 12. The recovery rates for Al, Cu, Cr, Fe, and Mo are reported to be above 50%. However, the valuable metal Sn, which is present in oxidized form in the slag phase, is also indicated as a loss. Additionally, precious metals such as Au, Ag, and Pd are reported as losses (around 0.0008 wt.-%) in the copper anode slimes during the electrolytic refining process. Furthermore, rare earths such as Dy and Nd, present in magnets, are also considered losses (around 0.35 wt.-%), existing as oxides in the slag form after the EAF process. This product/centric approach provides a detailed breakdown, allowing for a thorough examination of the process method's efficacy in reclaiming valuable materials. Additionally, the overarching recycling rate label depicted, offers a visual representation that succinctly encapsulates the overall success of the recycling endeavor.

Table 12. Recycling rate and recovered weights for the recycling process.

Element	Weight in Refrigerator (g)	Recovery Weight (g)	Recovery Rate
Ag	0.19	0	0.00%
Al	955.90	653.8	68.40%
Au	0.07	0	0.00%
B	4.64	0	0.00%
Co	0.03	0	0.00%
Cr	235.96	133.3	56.51%
Cu	566.43	297.3	52.49%
Dy	4.64	0	0.00%
Fe	33,949.82	26,483.9	78.01%
Mg	3.54	0.00065	0.02%
Mn	432.14	16	3.70%
Mo	12.17	8.	65.71%

Table 12. *Cont.*

Element	Weight in Refrigerator (g)	Recovery Weight (g)	Recovery Rate
Nb	3.87	0	0.00%
Nd	123.89	0	0.00%
Ni	108.95	10	9.80%
P	11.41	0	0.00%
Pb	3.24	0.0028	0.09%
Pd	0.02	0	0.00%
Si	18.76	0	0.00%
Sn	5.80	0	0.00%
Ta	0.03	0	0.00%
Zn	5.09	0	0.00%
	Total		75.74%

3.4. Life Cycle Assessment

In the context of the LCIA pertaining to the recovery of Al, Fe, Cu, the chosen impact category is global warming potential (GWP). This selection is predicated on the potential emissions associated with the high-energy demands inherent to the refrigerator recycling process. The total GWP contribution for the recovery process for Al, Cu, and Fe is listed in Table 13.

Table 13. Contributions to GWP in refrigerator recycling process.

Element	Al	Cu	Fe
kg CO_2 eq/refrigerator	5.21	4.97	17.01

In relation to the process unit contributors for GWP, Figure 8 provides a comprehensive visual representation of the specific contributions made by each process unit. This evaluation takes into careful consideration the intricacies of manual dismantling, as well as the mechanical, physical, and metallurgical processes employed throughout the entirety of the refrigerator recycling procedure.

The total emissions in (kg CO_2 eq) incurred for the production of 0.28 kg of copper cathode, corresponding to one refrigerator, amount to 4.97 kg CO_2 eq/refrigerator. The primary contributor to this burden is the utilization of nitrogen in the shredding process, accounting for 3.71 kg CO_2 eq/refrigerator. This is necessitated to maintain an inert environment for the treatment of the blowing agent derived from polyurethane. Subsequently, the second most significant contributor is the consumption of medium voltage electricity from the German grid, amounting to 0.609 kg CO_2 eq/refrigerator. This electricity is employed in various stages, including manual dismantling, mechanical and physical separation processes, as well as in the electrolytic refining stage of the black copper route process. Furthermore, the use of enriched oxygen in both the smelter and oxidizer units within the black copper route process yields emissions of 0.277 kg CO_2 eq/refrigerator. Finally, the smelter and reducer units generate off gases, contributing 0.250 kg CO_2 eq/refrigerator and 0.139 kg CO_2 eq/refrigerator respectively to the overall emissions inventory.

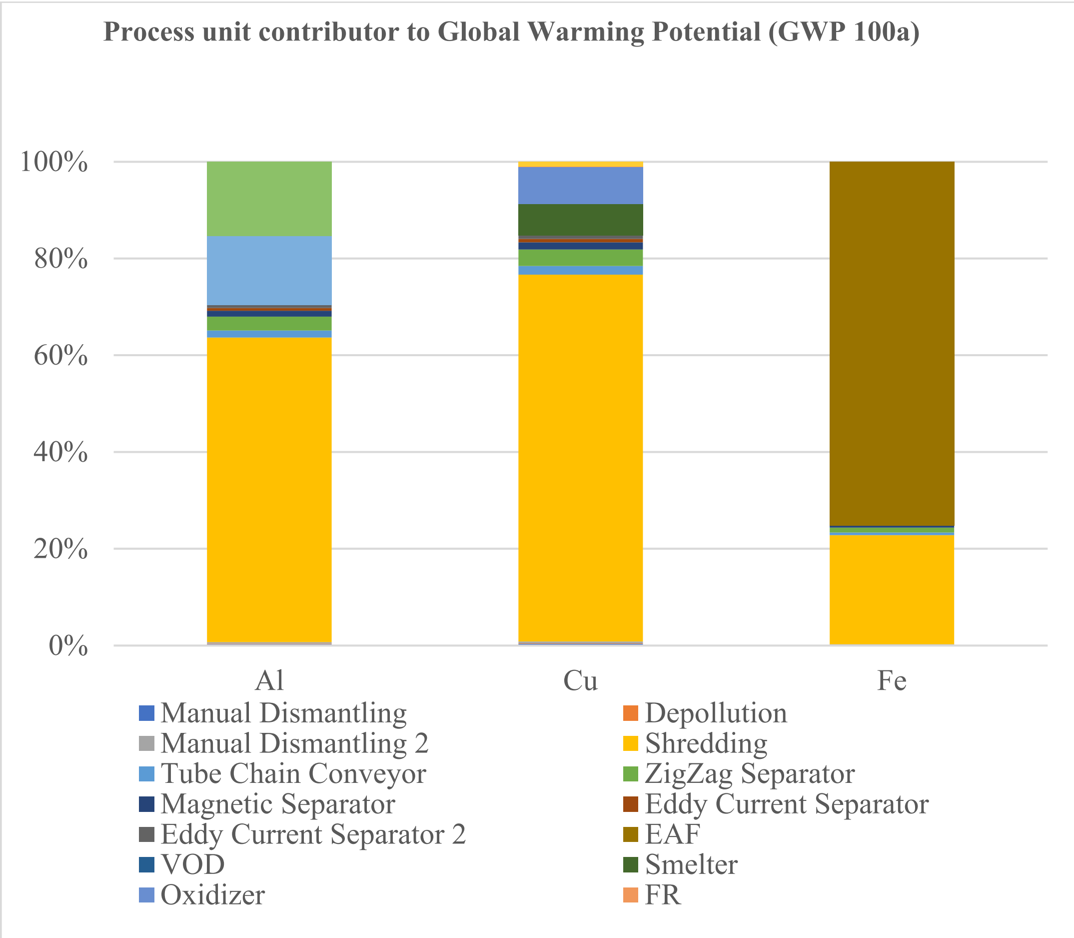

Figure 8. Process unit contributions to GWP in Al, Cu, and Fe recovery.

For iron recovery, the total emissions amount to 17.01 kg CO_2 eq/refrigerator. This value is attributed to the recovery of 26.67 kg of steel from one refrigerator. The most significant contributor to these emissions is the utilization of medium voltage energy sourced from the German grid in the EAF, accounting for 10.17 kg CO_2 eq/refrigerator. Nevertheless, this technosphere flow is prominently consumed across all manual, mechanical, physical, and metallurgical processes. Additionally, the use of nitrogen in the shredding unit contributed 3.76 kg CO_2 eq/refrigerator, while the use of fluxing agents in the pyro metallurgical units represents 2.231 kg CO_2 eq/refrigerator. The employment of coke as a reducing agent contributes 0.43 kg CO_2 eq/refrigerator, and lastly, the emissions from EAF off gases amount was 0.42 kg CO_2 eq/refrigerator. Moreover, in the context of the LCIA for aluminum recovery, the total emissions amount was 5.21 kg CO_2/refrigerator. This value corresponds to the recovery of 0.65 kg of aluminum from one refrigerator. The primary contributor to these emissions is the use of nitrogen in the shredding process, accounting for 3.76 kg CO_2 eq/refrigerator. Following, the consumption of medium voltage electricity from the German grid represented 0.533 kg CO_2 eq/refrigerator.

Additionally, the burner responsible for providing heat in the re-smelting process generates emissions of 0.44 kg CO_2 eq/refrigerator, attributed to the release of off gases after the combustion process, where natural gas serves as an energy source, resulting in 0.43 kg CO_2 eq/refrigerator. Furthermore, the use of salt as a fluxing agent in the re-smelting unit contributed to 0.027 kg CO_2 eq/refrigerator. This detailed analysis provides a comprehensive understanding of the specific emissions sources and their respective magnitudes within the assessed metal recovery process. It is apparent that the use of nitrogen in the shredding process, and the use of electricity constitutes a significant portion of the overall environmental impact in this context. Additionally, as mention by [56] assessing more LCIA methods could enhance the understanding of the life cycle assessment (LCA) in the refrigerator recycling process. Although other studies may adopt similar approaches regarding mechanical and physical processes, [8,57,58], the present study introduces a distinctive recycling index and LCA approach, taking into account the mechanical, physical, and metallurgical processing involved in the refrigerator recycling process.

3.5. Design for Recycling

In the context of DfR, a thorough analysis of refrigerators recycling process reveals critical observations:

- **Polyurethane Insulation and Shredding Process**: The utilization of nitrogen in the mechanical process, particularly during shredding, holds great significance. Its role is pivotal in establishing an inert environment, a requirement stemming from the blowing agent found in polyurethane (PU). Given that PU constitutes a significant 21.7 wt.-% of the refrigerator, strict adherence to DfR principles is imperative. As demonstrated in Figure 9, by avoiding the use of PU, ergo, no N_2 in the shredding process, we can achieve a remarkable reduction in GWP—specifically, a 72% reduction for Al recovery, 74% for Cu recovery, and 22% for Fe recovery. This underscores the important role of material selection in mitigating environmental impacts throughout the recycling process. This presents an opening to investigate alternative insulation materials. These materials should aim for an equilibrium between thermal efficiency and recyclability.
- **Energy Source for Recycling Process**: The reliance on non-renewable energy sources, particularly electricity from the German grid, contributes significantly to the environmental burden 11.3 kg CO_2 eq/refrigerator. Transitioning to renewable energy sources or implementing energy-efficient technologies for a better liberation rate of MMS in the recycling process aligns with DfR principles and could substantially reduce environmental impacts. As demonstrated in Figure 9, if a cleaner electricity grid is employed, exemplified by the one in Norway [59], the GWP will be reduced to 5.03 CO_2 eq/refrigerator. Moreover, considering this in conjunction with abstaining from PU usage, it significantly contributes to a reduction in GWP. Specifically, this proposed approach leads to a GWP reduction of 82% in Al recovery, 86% in Cu recovery, and 80% in Fe recovery. This underscores the substantial environmental benefits attainable through a combination of sustainable energy sourcing and judicious material selection.
- **Material Complexity and Recyclability:** The use of eight different types of plastics in refrigerator construction presents a challenge for effective recovery and recycling. Simplifying material types, or incorporating standardized and easily separable plastics, aligns with DfR objectives. Additionally, efforts to liberate the 4% of plastic that remains unrecovered could enhance overall recyclability. A beneficial approach might involve developing a more comprehensive understand of physics-based models to delineate the mechanical recycling process, particularly in processes such as shredding for the liberation of MMS. One noteworthy example is the methodology proposed by [60], which employs finite element simulation to model the shredding process. Moreover, as illustrated in Figure 10 [54], the Recycling Index of 76% suggests that the material loss may be attributed to factors such as unliberated behavior already mentioned or material complexity during the alloy selection process for the intended product purpose.
- **Reuse, Repair, and Remanufacture Potential:** Considering design elements that facilitate component disassembly and replacement could extend the product's lifespan, reducing the demand for new materials and minimizing environmental burdens. As mentioned by [61], WEEE constitutes complex waste streams due to the technical challenges involved in its treatment. In the case of refrigerators, considering design elements that facilitate the disassembly and recovery of valuable components, such

as compressors during manual dismantling, can enhance the potential for repair and remanufacturing.

- **Steel Alloy Selection and Metallurgical Considerations**: Choices influenced by aesthetic considerations, such as the use of different steel alloys, should be scrutinized. The three different steel alloys, comprising 35.9 wt.-% of the refrigerator, contains valuable metals like such as Cr and Ni. The selection of alloys that facilitate easier separation and recovery during pyro metallurgical processes aligns with DfR objectives, preventing the loss of valuable metals within the slag phase in oxide form.

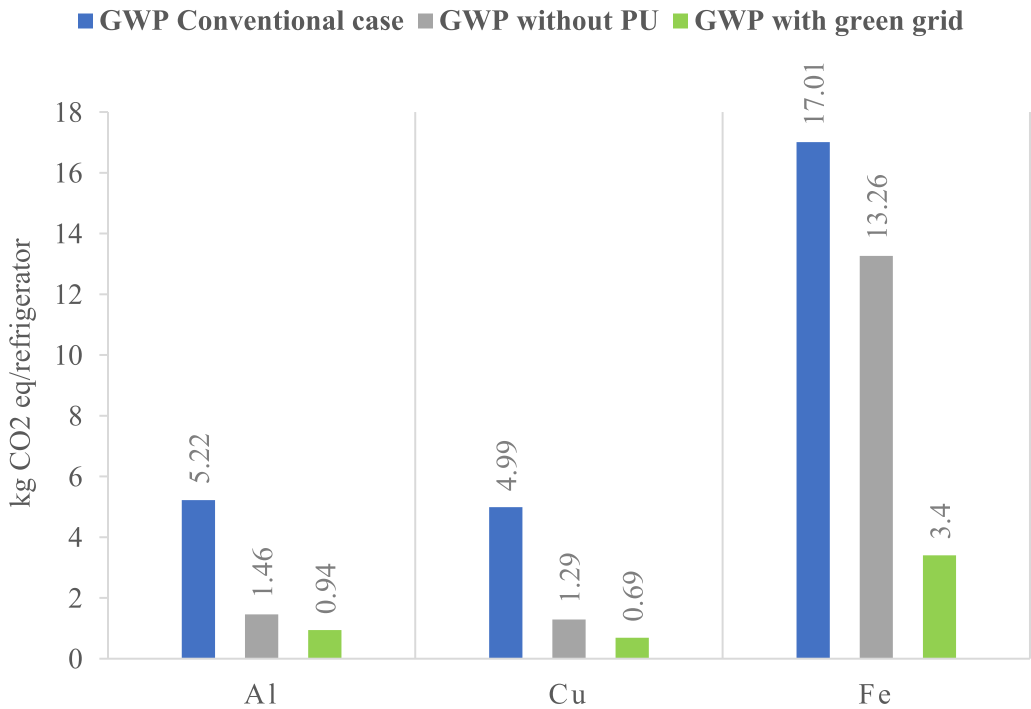

Figure 9. Global warming potential (GWP 100 a) reduction considering electricity source and material selection.

A+++	90–100%	
A++	80–90%	
A+	70–80%	76%
A	60–70%	
B	50–60%	
C	40–50%	
D	30–40%	
E	20–30%	
F	10–20%	
G	0–10%	

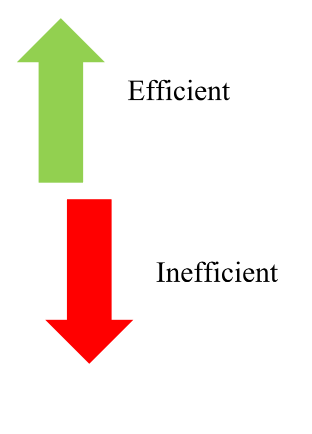

Figure 10. Recycling Index for refrigerator recycling process according to [55].

4. Conclusions

The process unit that contributes the most to the GWP is shredding, primarily due to the significant environmental burden incurred by the use of Nitrogen. Al and Cu exhibit recovery rates slightly above 50%, while Fe shows a recovery of about 78%. However, certain precious metals and rare earths are reported as losses after the pyro and hydrometallurgical recovery processes. Furthermore, it concludes that applying DfR principles to refrigerator design necessitates a multi-dimensional approach. This includes considerations for material selection, ease of disassembly, energy sourcing, and component reusability. By prioritizing recyclability, minimizing material complexity, and optimizing energy inputs, designers can create appliances that are more environmentally sustainable throughout their entire lifecycle. Additionally, strategic material selection and alloy choices can further enhance the recoverability of valuable resources, reinforcing the circularity of the product's life cycle.

The recycling process outlined in this study shows potential applicability in other countries, especially developing nations, contingent upon considerations such as infrastructure, technological capabilities, and regulatory frameworks. To enhance its international versatility, future work could explore optimization strategies in mechanical processing to address losses and improve overall metal recovery rates. Moreover, the recycling process aligns with sustainable development pillars, contributing to environmental preservation by diverting refrigerators from landfills and reducing raw material extraction. Economically, the recovery of metals demonstrates resource efficiency, and socially, the process hints at job creation and skill development opportunities.

Author Contributions: Conceptualization, F.A.G.P. and A.M.P.; methodology, F.A.G.P.; software, F.A.G.P.; formal analysis, F.A.G.P.; investigation, M.H., F.A.G.P. and J.T.; writing—original draft preparation, F.A.G.P.; writing—review and editing, A.M.P., K.G.v.d.B. and S.R.; visualization, F.A.G.P. and A.M.P.; supervision, A.M.P.; project administration, K.G.v.d.B.; funding acquisition, S.R. All authors have read and agreed to the published version of the manuscript.

Funding: The project "Circular by Design" is funded within the funding measure "Resource-efficient Circular Economy—Innovative Product Cycles (ReziProK)". "ReziProK" is part of the research concept "Resourceefficient Circular Economy" of the Federal Ministry of Education and Research (BMBF) and supports projects that develop business models, design concepts or digital technologies for closed product cycles. Funding reference number: 033R244. Project duration: 1 July 2019–30 June 2022.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data are contained within the article.

Acknowledgments: I express my sincere gratitude to Rekular GmbH for providing the invaluable opportunity of understanding the refrigerator recycling process. This first-hand experience has profoundly shaped the content of this paper.

Conflicts of Interest: The authors declare no conflicts of interest.

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