Journal of Cleaner Production 485 (2024) 144366

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Eco-efficiency assessment of carbon capture and hydrogen transition as decarbonisation strategies in alumina production

Javier Sáez-Guinoa, Inés Senante, Sara Pascual, Eva Llera-Sastresa *, Luis M. Romeo

School of Engineering and Architecture, University of Zaragoza, Campus Río Ebro, María de Luna 3, 50018, Zaragoza, Spain

ARTICLE INFO

Handling Editor: Yutao Wang

Keywords:
Alumina
Decarbonisation
Life cycle costing
Life cycle assessment
Eco-efficiency
Calcium looping

ABSTRACT

Studies seeking to thoroughly couple the environmental and economic performances of a process or product are becoming more prominent as the needs for decarbonisation grow. This study explores the use of Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) in the alumina extraction industry. Models for various thermal energy supply methods and CO_2 mitigation strategies were developed to obtain an inventory for analysis and the environmental impacts and life-cycle costs of the supply chain were evaluated. Finally, economic and ecological results were integrated using eco-efficiency indices and a sensitivity analysis of the most significant variables detected was conducted. The research indicated that the integration of a calcium-looping plant to capture post-combusted CO_2 could reduce 55.5% of the CO_2 equivalent emissions, while also obtaining a better economic performance due to the CO_2 avoided taxes. However, other environmental indicators had slightly more significant impacts because of the additional use of natural gas. The potential use of green hydrogen instead of natural gas could enable a 70.0% reduction in CO_2 equivalent emissions, as well as a reduction in all other environmental indicators studied, except for water consumption. However, transitioning to green hydrogen production could incur higher costs. This study introduced an eco-efficiency ratio index, indicating that CO_2 capture and storage proved to be the most eco-efficient scenario, regardless of economic fluctuations in CO_2 emission taxes. The substitution of natural gas with green hydrogen also emerged as a viable eco-efficient solution, provided electricity prices remain below 0.045€/kWh.

1. Introduction

Aluminium is one of the most demanded metals, with 67 million tonnes produced worldwide in 2021 (International Aluminium, 2022). Nevertheless, the elevated energy demand of its production was responsible for emitting around 275 million tonnes of direct CO_2 in that same year, which represents about 3% of the direct industrial emissions in the world (IEA, 2022). 98% of primary aluminium is produced from bauxite mineral (Schwarz, 2004). Bauxite is treated in an alumina refinery, where it is converted into Al_2O_3 through the Bayer Process. Subsequently, alumina is smelted following the Hall-Héroult process to obtain aluminium, which is finally cast to produce aluminium ingots. The cumulative energy demand of this whole process was estimated at 210 ± 10 MJ/kg of aluminium (Cushman-Roisin and Cremonini, 2021). This considerable energy demand comes mainly from two different stages of the aluminium production chain: approximately one-third comes from the thermal energy required to calcine the alumina during the Bayer process and the other two-thirds represent the electricity necessary to melt it during the Hall-Héroult process (International Aluminium Institute, 2023; Farjana et al., 2019).

Many studies agree that using renewable electricity during the Hall-Héroult process is the key to minimising environmental impacts and achieving carbon-neutral aluminium production. This deduction has been achieved either by studying the carbon footprint of the process (Saevarsdottir et al., 2020), analysing global carbon transfers and emission trades (Yi et al., 2022), or using emission factor methods (Wang et al., 2023). The proposed solutions involving thermal energy usage and waste production from alumina extraction are more diverse: Alternatives to the Bayer process, including the Pedersen process (Ma et al., 2022), where iron is first smelted and alumina subsequently extracted from the resultant slags, and coal fly ash acid digestion (Valeev et al., 2022; Yu et al., 2023), have been explored alongside enhancements and modifications related to the Bayer process (Suss et al., 2010; ZHOU et al., 2022). Unfortunately, most of these methods are hampered by the lack of economic viability (Zhou et al., 2023). Therefore, efforts are being made to find economical and environmentally suitable

* Corresponding author.
E-mail address: ellera@unizar.es (E. Llera-Sastresa).

https://doi.org/10.1016/j.jclepro.2024.144366
Received 22 December 2023; Received in revised form 21 November 2024; Accepted 30 November 2024
Available online 1 December 2024

solutions to this problem.

Concerns regarding Global Warming (GW) and Climate Change are continually increasing and appearing in the foreground of political and social agendas. The European Union Commission aims to drastically reduce its greenhouse gas emissions using a long-term strategy to become carbon neutral by 2050 (European Commission, 2018). Thus, new technologies and/or alternative energy sources must be explored in industries such as aluminium production to achieve this objective.

Many proposed solutions have focused on secondary aluminium recovery. The production of aluminium from mixed scraps, for instance, can emit approximately 95% fewer kilograms of CO_2 equivalent than traditional aluminium from bauxite ore (Adhikari et al., 2022). However, the extraction of alumina from aluminium dross can entail a 32.5% reduction in CO_2 equivalent emissions compared to extraction from bauxite (Zhu et al., 2020). However, only a few publications have focused on studies of primary alumina production, even though the projected aluminium demand forces the production of primary aluminium (European AluminiumCRU Group, 2020).

Therefore, it is of interest to conduct a primary alumina study that combines the economic and environmental aspects of alumina production. Life Cycle Costing (LCC) and Life Cycle Assessment (LCA) are two essential tools that separately evaluate the environmental and economic impacts of a product or system throughout its life cycle. (Ashby, 2024; Muralikrishna and Manickam, 2017). Although LCA is a well-stablished and standardised methodology (ISO, 2006a, 2006b), there are multiple guidelines and standards for performing an LCC study (Swarr et al., 2011; Toniolo et al., 2020), which usually differ in the scope of the study and the externalities considered. Hence, studies that perform combined LCA and LCC analyses present numerous differences in their methodologies and treatments of the results. In a review of multiple LCA and LCC combined studies (Miah et al., 2017), up to six different types of studies involving LCA and LCC were identified, and a hybridised framework that combined all of them to integrate both frameworks was presented.

Integrating environmental and economic studies is not exclusive of LCA and LCC studies. The World Business Council of Sustainable Development introduced the concept of eco-efficiency in the early 1990s. This term measures both economic and environmental performances, usually as a ratio between the created value and the environmental impact of a system (Čuček et al., 2015). Although the term has gained popularity over the years, methodologies and parameters have not been well established or defined (Koskela and Vehmas, 2012). Many different indicators exist for studying the eco-efficiency of a process depending on the goals and variables of each analysis (Peças et al., 2019). The definitions of the eco-efficiency ratio can vary from specific indicators (e.g. items sold, volume production, or employees, versus raw material exploitation, waste production, or land area used for the production) to global indicators such as LCA and LCC studies.

In the metal industry, performing both LCA and LCC has been proven to have many benefits in supply chains (Rebitzer, 2002), facilitating decision-making for process design comparisons and strategies. For instance, a recent comparison between two different hydrometallurgical routes to produce copper was carried out (Yang et al., 2022), highlighting the decompensation between environmental and economic development. In particular, regarding alumina industry, a life cycle environmental and economic assessment was performed (Zhu et al., 2020) comparing alumina recovery from secondary aluminium dross and alumina production through the Bayer process, concluding that both economic and environmental indicators were lower when recovering alumina from secondary aluminium dross.

However, these studies lack a deep integration of both LCA and LCC analyses, with practically two parallel analyses. Among the integrated studies, an LCA and LCC analysis of aluminium production was implemented (Luthin et al., 2021), with the goal of identifying dependencies between environmental and economic performance while focusing on the comparison between primary and secondary aluminium. Win-win situations, such as using domestic bauxite or renewable electricity, were detected in different scenarios that lacked an analysis of thermal energy consumption and its sources. Another environmental and economic assessment focused on aluminium production (Zhu et al., 2022) included a sensitivity analysis of different Bayer process configurations and electricity mixes for aluminium smelting. It was concluded that the Bayer process-hydropower-based method entailed the most significant reduction in environmental impacts and performed the best in achieving economic benefits. However, thermal energy usage has not been studied, despite being the variable with the most significant environmental impact from alumina refinement (Nunez and Jones, 2016).

Among the studies that analysed thermal energy consumption and explored pathways for decarbonisation (Peppas et al., 2023), carried out an LCA study of aluminium production, assessing the impacts of using an aqueous amine solvent to capture and store post-combustion carbon dioxide. The production of green hydrogen and its use instead of fossil fuels have also been studied, resulting in a scenario with the most negligible environmental impact. However, the economic implications were beyond the scope of this study. CO_2 absorption using amines is a well-implemented technology for mitigating CO_2 emissions, although it carries an important energy penalty (Anekwe et al., 2023). Other decarbonisation strategies, such as a calcium-looping plant, allow for more efficient energy recovery despite its implementation still being at a lower level (International Energy Agency, 2020).

Accordingly, the main goal of this study was to perform a combined LCA and LCC study, which included a sensitivity analysis of different thermal energy suppliers and possible carbon mitigation strategies. For this purpose, a study of primary alumina production through Bayer process is proposed, given its relevance and the significant usage of thermal energy. The analysis included the base-case scenario, in which natural gas is burned to meet energy demands, and three additional cases: the integration of a CO_2 capture plant using calcium looping and its later CO_2 storage and the substitution of natural gas by H_2 produced through electrolysis, with renewable and grid electricity. To achieve a complete integration of both the LCA and LCC studies, the hybrid framework guidelines found in the literature (Miah et al., 2017) were followed, and the environmental sustainability and economic benefits of the different proposed scenarios were displayed using eco-efficiency performance indices.

2. Materials and methods

The work consists of four different stages, as shown in Fig. 1, and is aligned with the principles of ISO 14040 and ISO 14044. The first stage defined the goals and scope of the study. This includes the system boundaries of both LCA and LCC analyses, the designation of the functional unit and the selection of the LCC perspective. The second stage is the analysis, which includes data gathering for the life cycle inventory. The third represents the calculation of LCC costs and the environmental impact assessment of the LCA. Finally, the last stage consists of the integration of the results of both analyses, the calculation of the eco-efficiency indices for every scenario, and the representation and interpretation of the results.

2.1. Goal and scope definition

The goal of this study is to conduct an integrated cost and environmental analysis of alumina production following the Bayer process, including renewable energy for H_2 usage or carbon capture. Thermal energy is the primary input of the process; thus, four alternative scenarios were studied to analyse the consequences of using different energy sources. The baseline case (S0) used the combustion of natural gas as the energy source, which is the most common practice in European alumina refineries. In the next one (S1), the possibility of integrating a CO_2 capture plant using Calcium Looping is discussed. Finally, for the two remaining scenarios, hydrogen produced in an electrolyser was used

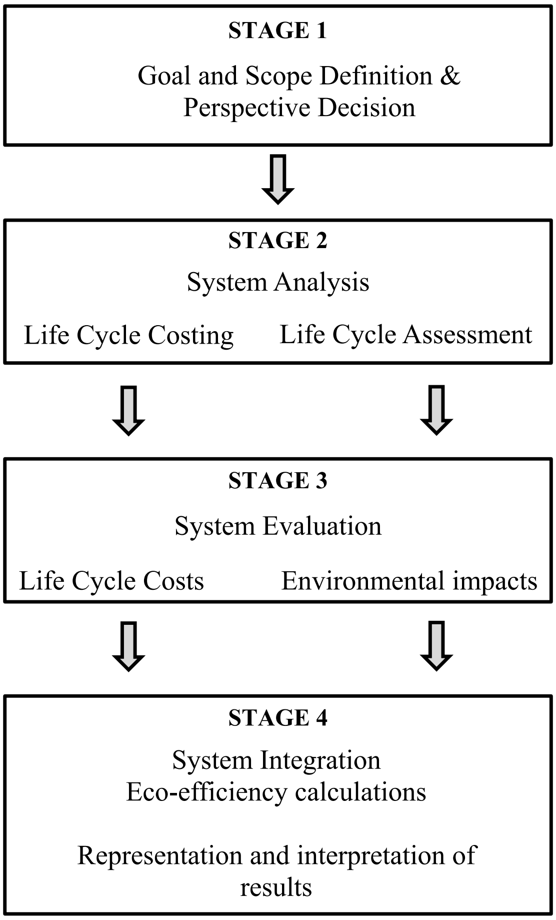

Fig. 1. Methodology followed during the study. Stages and actions.

as the energy source. Renewable electricity (S2) and the average European electricity mix (S3) were used for H_2 production. A summary of all four alternatives analysed in the study is shown in Table 1.

System boundaries were selected by applying a cradle-to-gate study from mineral extraction to alumina production. The main inputs and outputs are shown in Fig. 2. The system includes the extraction of bauxite rock and its transportation from the mine, which is assumed to be located in Guinea, to an alumina refinery in Europe. Bauxite pretreatment consisting mainly of grinding and desilicating the rock are performed. Subsequently, digestion occurs in NaOH liquor. Undissolved solids, known as red mud, are separated by filtration and disposed of in a landfill, whereas aluminium ions are precipitated in the form of aluminium hydroxide. Most of the NaOH liquor is recovered via evaporation, and the hydroxides are calcined to obtain smelter grade alumina.

A CO_2 capture plant was included in S1 to capture CO_2 from the flue gases generated during the aluminium hydroxide calcination stage. To accomplish this, gases containing mainly nitrogen, carbon dioxide, steam and oxygen are fed into a carbonator reactor, where CO_2 reacts with CaO to yield $CaCO_3$, and clean gases with minimal carbon dioxide content leave the system. $CaCO_3$ is then calcined in oxy-fuel combustion with natural gas, producing steam that is separated by condensation and pure CO_2. Most of the CaO is recycled into the carbonator, although some must be purged because of deactivation and replaced with fresh $CaCO_3$ (Abanades, 2002; Mastin et al., 2011). The main disadvantages of this technology are the use of additional natural gas to meet the calcination temperatures, the necessity of pure oxygen for oxy-combustion, and the unavoidable purging of the solid sorbent.

For S2 and S3, different sources of thermal energy were investigated. Among the suitable alternatives for achieving decarbonisation, hydrogen combustion for the production of heat has emerged as a promising option for high-temperature processes (Saint-Just and Etemad, 2016), even though its technological readiness level (TRL) is low (International Energy Agency, 2019). The use of hydrogen for high-temperature heating carries no CO_2 or NO_x emissions, making it appropriate for carbon-neutral applications as long as green hydrogen is used. The production of green hydrogen, even though it represents only 0.1% of current hydrogen production, stands at a high TRL and is rapidly increasing, with an expected global capacity growth from 2 GW in 2023 to 175 GW by the end of the decade (International Energy Agency, 2023). Therefore, an electrolyser that converts water into H_2 and O_2 was included and implemented as a source of thermal energy for scenarios S2 and S3.

Regarding the LCC definition and methodology, an investor perspective was adopted. Thus, a financial life cycle costing was performed, in which some binding externalities of the alumina industry, such as the costs of the red mud disposal or CO_2 emissions were also taken into consideration.

The functional unit of the study was decided to be time-referenced so that no discrepancies between the LCA and LCC were observed during the analysis, aiming to ease data gathering. One year of continuous production at an alumina refinery was selected as the basis for this study. The primary material and energy consumptions, waste, emissions and economic costs were based on this functional unit.

Table 1
Summary and designed names for each scenario.

Scenario	Thermal Energy Supply	CO_2 Mitigation/Prevention Strategy
S0	Natural Gas	–
S1	Natural Gas	CO_2 Capture Plant and Storage
S2	H_2	H_2 production using renewable electricity
S3	H_2	H_2 production using grid electricity

2.2. System analysis

The system analysis was divided into two parts: first, the data gathering used for the evaluation of both LCA and LCC and then the assumptions and boundaries considered to calculate economic costs using the Life Cycle Inventory collected.

2.2.1. Life cycle inventory

The primary energy and material consumptions were gathered after carrying out a thermodynamic simulation of the process using Aspen Plus V12.1 software (Aspen Process Economic Analyzer). Secondary data, such as electricity consumption and chemicals (NaOH, CaO) were retrieved from European Aluminium reports (European Aluminium Association, 2018). To perform the Bayer process modelling, a plant with the capacity to treat nearly 1.5 million tonnes of bauxite per year was considered. The composition of the bauxite was fixed, in %wt, as: 57.6% AlO(OH), 28.6% Fe_2O_3, 6.2% SiO_2, 4.5 TiO_2 and 3.1% CaO. The digestion of the rock was carried out at 280 °C and 3 atm, and the yield of the aluminium dissolution in the liquor was set at 97% (Donaldson, 2008; Donoghue et al., 2014). Afterwards, a liquid separation and a subsequent filtration were performed to recover as much liquor as possible. The undissolved rock, with a water content of 16.8%wt, was subjected to different washing steps before being ready for disposal. The liquid containing aluminium ions was cooled down to 80 °C and atmospheric pressure before entering the crystallizer, where $Al(OH)_3$ precipitates. Finally, a natural gas combustor with 5% excess air was modelled to calcine the hydroxides and obtain smelter-grade alumina. The rest of the liquor was then heated up to 105 °C to increase its OH^- concentration, and the water evaporated was recirculated into the precipitation stage (Ruys, 2019).

The sole energy input of the plant in cases S0 and S1 is natural gas. Its direct combustion heats the solid $Al(OH)_3$ to 1200 °C, and heat residues

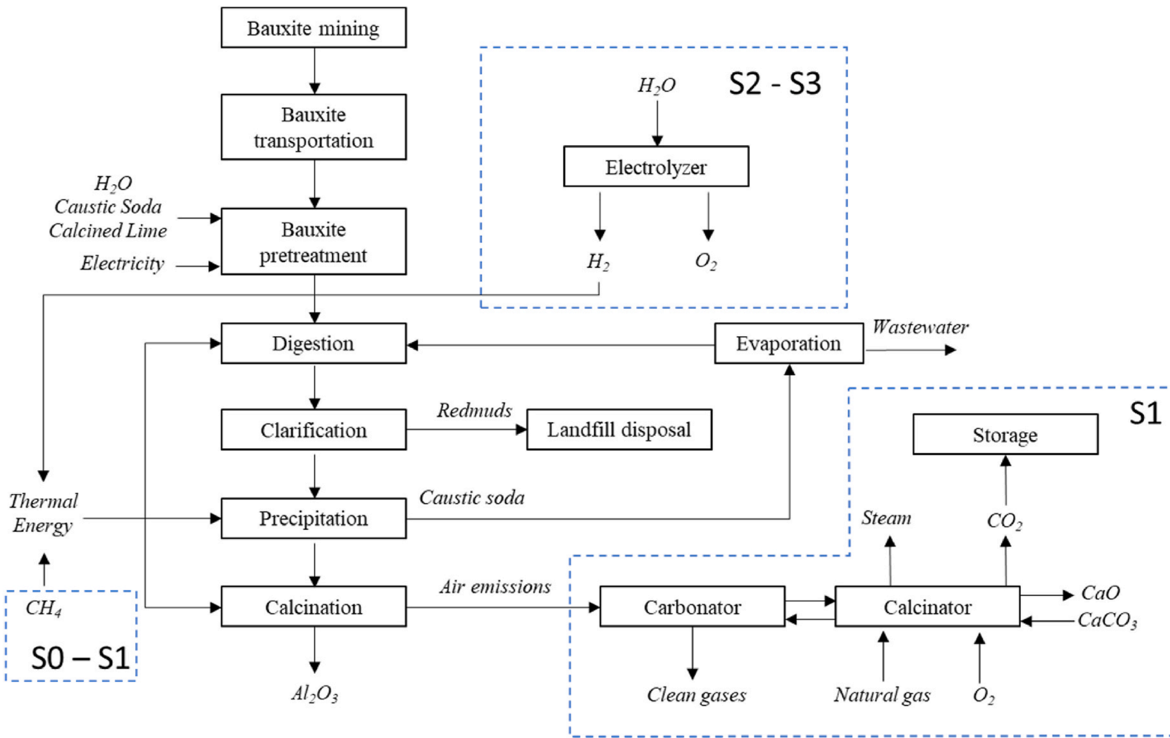

Fig. 2. System of the study. Boundaries and scope of each scenario.

from hot flue gases and alumina are exploited during flash evaporation and preheating at the digestion step, respectively. For scenarios S2 and S3, hydrogen combustion was modelled to calcine $Al(OH)_3$, and the remaining energy requirements (to preheat and evaporate the liquor) were included as electrical requirements.

For scenario S1, in which a CO_2 capture plant was added, a thermodynamic simulation of a carbonator-calcinator cycle was implemented. To this end, the flue gases from S0 were sent to a calcium looping plant, where CO_2 was removed from the flue gases for use or storage. Calcium looping technology is made up of two main reactions: during the first one, carbonation, CaO is fed to the flue gases so that solid calcium carbonate can be produced at 650 °C; in the second one, calcination, natural gas is burned with pure oxygen while $CaCO_3$ decomposes at 900 °C, producing CaO that is recirculated back to carbonation, and CO_2. One of the drawbacks of this configuration is the sorption capacity decay of CaO in each cycle of carbonation and calcination (Grasa and Abanades, 2006). To overcome this and obtain a suitable conversion of the carbonation reaction, a fraction of CaO must be purged, and fresh CaCO3 must be introduced into the loop (Romeo et al., 2009).

The modelling of the carbon capture plant was modelled by considering the number of cycles of carbonation-calcination (*N*) of a limestone (CaO) particle as a reference. As the sorption capacity of the solid decays after each cycle, a typical limestone sorption capacity was used. Equation (1) shows the conversion of one particle of CaO (X_N) in the carbonator as a function of the number of cycles *N*.

$$X_N = \frac{1}{\frac{1}{1-X_r} + k \cdot N} + X_r \tag{1}$$

In this equation, k is the conversion decay constant, set as 0.52, and X_r is the residual activity of CaO particles, fixed as 0.075 (Grasa and Abanades, 2006) for the limestone used in this study. The average conversion achieved by the solid (X_{ave}) is the product of its number of cycles distribution (r_N) and particle conversions X_N, as shown in Equation (2) (Rodriguez et al., 2008).

$$X_{ave} = \sum_{N=1}^{N=\infty} r_N \cdot X_N \tag{2}$$

The variable r_N represents the percentage of CaO particles after a certain number of cycles and is calculated using the molar flow rate of CaO entering the carbonator (F_{CaO}) and the molar flow rate of $CaCO_3$ entering the calciner (F_0). The calculation of the variable r_N is shown using Equation (3).

$$r_N = \frac{\frac{F_0}{F_{CaO}}}{\left(1 + \frac{F_0}{F_{CaO}}\right)^N} \tag{3}$$

Thus, to achieve an admissible carbon capture efficiency (>90%) and minimise economic costs from energy and $CaCO_3$ consumption, a F_{CaO}/F_0 ratio of 5.5 was selected, and a purge of 2% of the CaO was effectuated on the calciner (Romeo et al., 2009).

Residual heats from "clean" gases after the carbonation reaction (at 650 °C) and from CO_2 and steam produced on the calcination (at 900 °C) were integrated into the Bayer process plant. Heat was used to supply energy during bauxite digestion. Thus, fossil fuel consumption by the Bayer plant decreased, and the raw output of CO_2 to be captured was reduced accordingly.

For scenarios S2 and S3, an alkaline electrolyser with an efficiency of 0.65 (Ozturk and Dincer, 2021) was also modelled. Gaseous H_2 was then utilized as fuel instead of natural gas, with the remaining energy requirements (which were unmet due to the significant reduction in combustion gases) being met directly by electricity, attributed to its suitability for relatively low temperatures (around 100-105 °C). The combustion of H_2 to obtain thermal energy was assumed to be complete and carried out with 5% air excess. To overcome heat transfer limitations of hydrogen combustion, a 3% penalty of energy demand was assumed with regards to natural gas scenarios (Demuynck et al., 2010).

The final foreground inventory of this study is presented in Table 2. Regarding other data to complete the Life Cycle Inventory, the electricity mix used for scenarios S0, S1 and S3 is shown in Table 3. In

Table 2
Foreground life cycle inventory.

	S0	S1	S2	S3
Inputs				
Bauxite (tonne)	$1.42\cdot10^6$	$1.42\cdot10^6$	$1.42\cdot10^6$	$1.42\cdot10^6$
Natural Gas (MJ)	$7.10\cdot10^9$	$7.54\cdot10^9$	–	–
Electricity (MWh)	$9.21\cdot10^4$	$9.21\cdot10^4$	$2.96\cdot10^6$	$2.96\cdot10^6$
NaOH (tonne)	$3.63\cdot10^4$	$3.63\cdot10^4$	$3.63\cdot10^4$	$3.63\cdot10^4$
CaO (tonne)	$2.41\cdot10^4$	–	$2.41\cdot10^4$	$2.41\cdot10^4$
$CaCO_3$ (tonne)	–	$7.10\cdot10^4$	–	–
Water (m^3)	$2.08\cdot10^6$	$2.08\cdot10^6$	$2.41\cdot10^6$	$2.41\cdot10^6$
Sea transport (t·km)	$6.54\cdot10^9$	$6.54\cdot10^9$	$6.54\cdot10^9$	$6.54\cdot10^9$
Outputs				
Alumina (tonne)	$6.53\cdot10^5$	$6.53\cdot10^5$	$6.53\cdot10^5$	$6.53\cdot10^5$
Bauxite residue (tonne)	$8.60\cdot10^5$	$8.60\cdot10^5$	$8.60\cdot10^5$	$8.60\cdot10^5$
Wastewater (m^3)	$1.70\cdot10^5$	$1.70\cdot10^5$	$1.70\cdot10^5$	$1.70\cdot10^5$
CaO (tonne)	–	$1.56\cdot10^4$	–	–
CO_2 emitted (tonne)	$3.91\cdot10^5$	$1.73\cdot10^4$	–	–
CO_2 stored (tonne)	–	$4.15\cdot10^5$	–	–
Oxygen (tonne)	–	–	$2.92\cdot10^5$	$2.92\cdot10^5$

Table 3
Disaggregated sources of electricity grid used in S0, S1 and S3.

Source Power	Composition (%)
Nuclear	23.5%
Wind Energy	20.8%
Coal	18.1%
Hydropower	14.7%
Cogeneration	10.5%
Combined Cycle	9.0%
Solar Energy	3.4%

contrast, the electricity mix for S2 based on renewable electricity is shown in Table 4. The CO_2 emissions and impact of CO_2 storage for scenario S1 were obtained from (Peppas et al., 2023). All data were collected mainly from the EcoInvent 3.7 database integrated in SimaPro software ("ecoinvent v3.7 – ecoinvent," 2020), using the Cut-Off Allocation option. In addition, the ELCD databases were used when no other data were found.

The inventory displayed in Table 2 shows that 10,877 MJ of energy are required per tonne of alumina produced in the base case of S0. Energy consumption is in agreement with the average industrial data provided by International Aluminium, which in 2019 accounted for 10,316 MJ for European refineries and 10,456 MJ for alumina refineries worldwide (International Aluminium, 2022). The slight difference in energy consumption may be due to the fact that this model is based on boehmite bauxite, while actual bauxite is usually a mix of gibbsite and boehmite mineral forms (Sáez-Guinoa et al., 2024). Considering that the energy consumption of primary aluminium production is approximately 74,900 MJ per tonne (International Aluminium, 2022), the alumina production stage is estimated to represent 29% of that energy (assuming that 2 kg of alumina is necessary for the production of 1 kg on aluminium ingot).

After the integration of the CO_2 capture plant, energy consumption of S1 stands at 11,547 MJ per tonne of alumina produced. This is one of the most essential features of the calcium looping technology and its implementation in the alumina industry. The high energy recovery of the capture plant could allow its integration with less than a 7% increase in energy consumption. Accordingly, the specific energy consumption amounts to 1.45 MJ per kg of CO_2 avoided.

Table 4
Disaggregated sources of electricity used in S2.

Source Power	Composition (%)
Wind Energy	53.4%
Hydropower	37.8%
Solar Energy	8.8%

The energy consumption of cases S2 and S3 is approximately 16,318 MJ per tonne of alumina, mainly because of the inefficiency of the electrolyser required to produce green hydrogen, which was set to 65%. In these cases, the specific energy consumption per kilogram of CO_2 avoided increased up to 8.51 MJ per kilogram of CO_2.

Across all four cases, producing one tonne of alumina required 2.17 tonnes of bauxite. The bauxite consumption can differ significantly owing to its variable composition. The average consumption by European refineries stands at 2.35 tonnes of bauxite per tonne of alumina (International Aluminium, 2022). However, water consumption increased for S2 and S3 from 3.18 m^3 to 3.69 m^3 per tonne of alumina produced, due to water electrolysis for H_2 production. Other relevant raw materials are limestone ($CaCO_3$) and pure oxygen for the CO_2 capture plant of S1, in which 0.11 tonnes of $CaCO_3$ and 0.24 tonnes of oxygen are needed per tonne of alumina.

2.2.2. *Life cycle cost*

Following the guidelines of the integrated life cycle cost and environmental assessment, the cost analysis of each case was implemented from an investor point of view, with the aim of carrying out a conventional financial Life Cycle Costing (fLCC) (Dyson, 2024). fLCC was divided into two different parts, according to the type of costs incurred: (i) the capital investment necessary for the construction and installation of the refinery plant, and (ii) the operational costs of producing alumina throughout a year. The operational costs were divided in three different categories: the costs of raw materials, the costs of energy and the costs of disposal/emission/management of the residues.

The initial capital investment was calculated following the methodology presented by Peters et al. (2003). The leading equipment was identified, sorted and sized based on the expressed data. The purchase costs were retrieved from the cost functions presented by Peters et al. (2003). These cost functions are exclusive to each type of equipment and depend primarily on the required capacity and construction material. Next, the obtained costs were updated using the chemical engineering plant cost index (CEPCI) from January 2023. For other equipment in which size or different specifications were not suitable, the Aspen Process Economic Analyzer (Aspen Tech, 2024) was used. In the case of scenario S1, the equipment costs of the carbonator, calciner and air separation unit were estimated as functions of the thermal energy output, thermal energy input and mass flow of oxygen, respectively (Lim et al., 2023). For scenarios S2 and S3, the capital costs were calculated as functions of the capacity required for the electrolyser to produce the necessary amount of H_2 (Thema et al., 2019). Once total purchase equipment costs were calculated, a factor between 2.69 and 3.60 depending on each case, was added to represent total direct costs (installation of equipment, electrical systems, piping, service facilities, buildings …) and another factor between 1.26 and 1.44 was used to estimate indirect costs (construction expenses, legal expenses, contingencies …) (Peters et al., 2003). A detailed list of the equipment included in the study and their disaggregated purchase, direct, and indirect costs can be found on Appendix A of the Supplementary Information.

All economic variables that affect the operational costs of the study (material inputs, energy inputs and treatment/disposal/emission costs of outputs) are summarised in Table 5. One critical factor is the economic cost of emitting CO_2, which is usually unsteady because of market fluctuations. In this case, it was assumed to be fixed at 85€/tonne of CO_2 emitted, similar to clearing prices of auctions in January 2022 (European Commission, 2023). Nonetheless, sensitivity analyses were performed to determine their importance. Similarly, two electricity prices were used during the study: electricity grid prices (for cases S0, S1 and S3) were set at 0.076€/MWh, the average European prices in first semester of 2021 (European Commission; Directorate-General for

Table 5
Economic variables used to calculate operational costs.

	Cost	Reference
Material Inputs		
Bauxite (€/tonne)	20	Qaidi et al. (2022)
CaO (€/tonne)	60	Romeo et al. (2008)
NaOH (€/tonne)	345	"Caustic Soda Prices | Historical and Current | Intratec.us," n.d.
$CaCO_3$ (€/tonne)	20	Chirone et al. (2022)
Water (€/tonne)	0.10	Barón et al. (2023)
Energy Inputs		
Natural Gas (€/kWh)	0.041	(Eurostat, 2022)
Grid electricity (€/kW	0.076	(European Commission, 2021)
Renewable electricity (€/kWh)	0.040	(European Commission, 2023)
Outputs		
Bauxite residue disposal (€/tonne)	9.13	Qaidi et al. (2022)
CO_2 emission rights (€/tonne)	85.00	European Commission (2023)
Wastewater (€/tonne)	0.20	Behnami et al. (2019)
CO_2 storage (€/tonne)	7.00	Romano et al. (2013)

Energy unit A4 Market Observatory for Energy, 2021), whereas renewable electricity for scenario S2 was estimated at 0.040€/MWh using actual power purchase agreements from renewable electricity production (European Commission, 2023). Other costs, such as manpower or contingencies were calculated as 18% of all operational costs, excluding CO_2 emission costs.

2.3. System evaluation

To assess the environmental impacts of each alternative configuration, the ReCiPe 2016 midpoint (H) method was used after SimaPro 9.0 was utilized to implement the Life Cycle Inventory. The ReCiPe methodology, selected because of its wide application, provides 18 midpoint indicator results. Among these, the primary focus was on GW, measured in kilograms of CO_2 equivalents emitted, as it is mainly influenced by thermal energy consumption. Other important indicators considered in the study were Water Consumption (WAC) (m^3), Land Use (LU) (m^2 of crop eq.), Fossil Resource Scarcity (FRS) (kg of oil eq.), Terrestrial Acidification (TA) (kg of SO_2 eq.) and Marine Eutrophication (ME) (kg of N eq.). The hierarchist perspective (H) was chosen as the impact assessment method due to its prevalence in scientific studies, with a time horizon of 100 years.

No normalisation or weighting among the different indicators was carried out, as they are not necessary steps according to ISO standards and can lead to potentially associated biases (Pizzol et al., 2017). Nonetheless, the impacts of different scenarios were normalised with reference to the most significant impact for each indicator to allow a fair comparison among configurations and complete integration of the analysis. Thus, normalisation was carried out among scenarios, but not among indicators, as is sometimes performed in LCA analysis.

For the life cycle cost calculations, capital investment costs of each process alternative were annualized using Equation (4), in which a capital recovery factor (*CRF*) is calculated.

$$CRF = \frac{i \cdot (1+i)^n}{(1+i)^{n-1}} \tag{4}$$

where n is the planned lifetime and i is the interest rate. The planned lifetime was set at 25 years, and the interest rate was assumed to be 8% (Haaf et al., 2020). Capital expenditures were multiplied by this CRF factor to obtain the annualized capital costs. The planned lifetime of the water electrolysis plant was set at 15 years (International Renewable Energy Agency, 2020). Hence, the capital recovery factor varied between the different scenarios, penalizing the investment of shorter lifetime plants. Operational costs were also estimated on a yearly basis, considering a continuous inflation rate of 2.5% per year (Samitha Weerakoon et al., 2024). A conservative value of the annual inflation rate was selected to prevent the effect of incurring high capital costs from being diluted by high inflation rates over 25 years. Considering all the economic factors, the life cycle annualized costs (LCAC) used for the analysis were calculated as shown in Equation (5).

$$LCAC = C_{CAPEX} \cdot CRF + \sum_{n=1}^{n=25} C_{OPEX} \cdot \frac{(1+r)^{n-1}}{n} \tag{5}$$

in which C_{CAPEX} are the capital investment costs, *CRF* is the capital recovery factor, C_{OPEX} is the sum of operational expenditures during one year, n is the number of years, and r is the inflation rate.

To calculate a suitable eco-efficiency index (EI), the added economic value created by the business activity was defined as the product system value (PSV), and it was estimated as the total possible revenues from marketable products of one year (RMP), minus the life cycle annualized costs. Thus, eco-efficiency indices were calculated according to Equation (6).

$$EI = \frac{PSV}{GWi} = \frac{RMP - LCAC}{GWi} \tag{6}$$

in which GWi is the environmental impact of the Global Warming indicator obtained during the life cycle assessment. RMP were also calculated using a 2.5% annual inflation rate, as shown in Equation (7). Revenues of 365€/tonne of alumina (Qaidi et al., 2022) and also 100€/tonne of oxygen (Parra and Patel, 2016) in scenarios S2 and S3 were considered for the analysis.

$$RMP = \sum_{n=1}^{n=25} F_{Al2O3} \cdot S_{Al2O3} \cdot \frac{(1+r)^{n-1}}{n} + \sum_{n=1}^{n=25} F_{O2} \cdot S_{O2} \cdot \frac{(1+r)^{n-1}}{n} \tag{7}$$

in which F_{Al2O3} and F_{O2} are the tonnes of alumina and oxygen produced respectively during one year of operation, and S_{Al2O3} and S_{O2} are the sale prices of alumina and oxygen in euros per tonne of product.

As observed in Equation (6), the Eco-efficiency index represents the ratio of the net economic profit (measured in € per year) versus the amount of CO_2 emitted during the same time frame. Its objective is to show the scenarios that get an adequate ratio between profits and environmental impacts and detect if any option that shows economic viability involves too significant environmental consequences. Given the uncertainty of most of the economic variables, a sensitivity analysis was conducted to confirm the eco-efficiency results. Two major variables with significant fluctuations, as the price of electricity and the cost of CO_2 allowances, were analysed in the range of 0–0.08€/kWh and 0–150€/tonne of CO_2 respectively, with the goal of evaluating their influence in the overall results and determining the potential breakeven points of market profitability and eco-efficiency.

3. Results and discussion

Table 6 shows the performance of each case for the six midpoint indicators assessed during the study period. Results of the base scenario indicate that 1.10 tonnes of CO_2 equivalent are emitted per tonne of

Table 6
Environmental impacts results assessed for each scenario (per ton of Al_2O_3).

	S0	S1	S2	S3
GW (tonne CO_2 eq.)	1.10	0.49	0.33	1.86
WAC (m^3)	5.68	5.70	6.01	9.21
LU (m^2a crop eq.)	3.76	3.81	2.83	3.91
FRS (kg oil eq.)	387	393	80.5	465
TA (kg SO_2 eq.)	3.79	3.78	3.34	13.0
ME (kg N eq.)	$4.14 \cdot 10^{-3}$	$4.20 \cdot 10^{-3}$	$2.13 \cdot 10^{-3}$	$1.91 \cdot 10^{-2}$

alumina produced when natural gas is used as fuel and no CO_2 mitigation strategies are implemented. This value is aligned with those found in literature, which range from 0.99 (Sáez-Guinoa et al., 2024) to 1.23 (Farjana et al., 2019) tonnes of CO_2 per tonne of alumina when the selected fuel of the study is natural gas. If other fossil fuels are considered, the GHG emissions indicator increases significantly, showing the dependence of the industry on the thermal energy supplies. An LCA estimated the GHG emissions at 1.33 tonnes per tonne of alumina when a mix of raw coal and natural gas was used (Peng et al., 2019). This indicator increased to 1.52 according to a study in which heavy fuel oil was used to produce alumina (Schmidt and Thrane, 2009) and to 1.65 when the average European electricity was proposed as source of thermal energy (Ma et al., 2022). On the other hand, the results of the scenario S1 show that the implementation of a CO_2 capture plant mitigates the GHG emissions, decreasing from 1.10 to 0.49 tonnes per tonne of alumina. Similarly to other studies, calcium-looping technology achieves a significant reduction in CO_2 emissions, but the other environmental indicators assessed increase slightly because of the increase in the energy consumption (Carbone et al., 2023).

The disaggregated contributions to CO_2 total emissions for each scenario are shown in Fig. 3. As expected, natural gas combustion was the main contributor, contributing nearly 55% of equivalent CO_2 emissions of base case S0, followed by natural gas extraction (13%) and the use of chemicals (12%). Accordingly, the S1 emissions decreased in a similar proportion, as most of the post-combustion CO_2 was captured and stored underground. The rest of the factors (bauxite extraction and transportation, use of chemicals and natural gas extraction) remained unchanged. The same reason explains the more significant decrease in S2 emissions. Avoiding the extraction, transport and combustion of natural gas minimises the CO_2 equivalent emissions in scenario S2, in which renewable electricity production has a minimal impact. However, the substitution of natural gas for grid electricity does not compensate for the emissions, as the equivalent emissions due to electricity production are even higher than the use of natural gas to produce heat.

One of the most noticeable results is the S3 scenario, which exceeds the performance of the other alternatives for any other indicator. However, the use of renewable energy for producing hydrogen results in a reduction in all indicators (except for water consumption) when compared to natural gas processes. While the addition of a CO_2 capture plant and storage can result in a reduction of 55.4% CO_2 equivalent emissions, the use of green hydrogen instead of natural gas can decrease that amount to 70.0%.

The results show that although CO_2 capture and storage is a valid alternative to mitigate GW, as explained by the avoided CO_2 emissions, the S1 results do not improve any other indicator with regard to the base S0 scenario, as opposed to the use of green hydrogen in S2. The increase in the use of natural gas of S1 impacted water consumption, land use, fossil resource scarcity and marine eutrophication, regardless of the post-combustion storage of CO_2. Thus, it can be concluded that replacing natural gas with green hydrogen is a more effective solution for minimising the generic environmental impacts of alumina production.

Table 7 lists the economic performances obtained during the LCC analysis. As expected, capital costs of scenario S0 were the lowest. Building up a single alumina refining plant of this capacity (0.65 million tonnes per year) would cost around 205.65 M€. The addition of a calcium-looping plant implies a 75% increase in the initial expenditure, with 359.77 M€. However, the purchase of an electrolyser in S2 and S3 is a significant disadvantage of this technology, representing more capital expenses than the refinery itself. The calculated capacity of the electrolyser to meet the thermal energy demands of the alumina refinery amounts to 253 MW. Therefore, the estimated capital costs for the hydrogen plant reach 589.48 M€. Even though the global installed capacity of water electrolysis plants is projected to be 170–365 GW by 2030 (International Energy Agency, 2023), and hydrogen plants with capacities of hundreds of megawatts are projected in the medium term (RWE AG, 2024), the technical and infrastructure challenges, yet to be addressed, rule out the actual implementation of the technology in a scenario such as the one proposed in this study, in which 100% of the fossil fuel consumption is avoided. Despite this, the progressive improved efficiency of electrolysers and the potential reduction of capital costs in the near future make these type of analyses of interest to researchers and industry professionals.

However, operational costs show interesting results: the avoided taxes for emitting CO_2 make S1 an alternative with minimal operational costs. Despite increasing natural gas consumption and the material necessities of the CO_2 capture plant, annual operational costs represent 171.09 M€, while conventional alumina production represent 197.29 M€ per year. A similar pattern is observed in studies found in literature. Strojny et al. studied the integration of calcium looping into a natural gas combined cycle power plant, resulting in economic profitability of the calcium-looping system from the 4th year assuming an avoided cost of 170€ per tonne of CO_2 (Strojny et al., 2023). Barón et al. also studied the integration of calcium-looping technology into the glassmaking industry. Results indicated the profitability of the technology assuming a

Table 7
Costs obtained for each scenario.

	S0	S1	S2	S3
Capital Costs, C_{CAPEX} (M€)	205.65	359.77	589.48	589.48
Operational Costs, C_{OPEX} (M€/year)	197.29	171.09	200.05	322.30
Life Cycle Annualized Costs (M€/year)	311.35	287.72	346.42	524.70

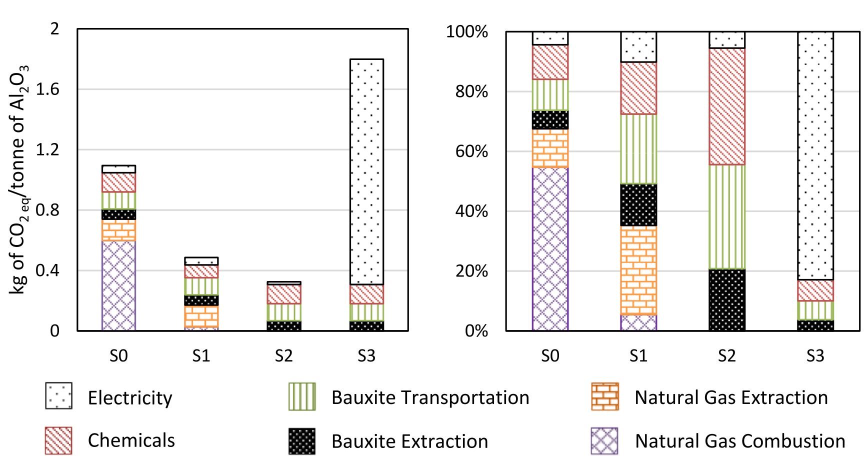

Fig. 3. Results of Global Warming indicator assessed. Net CO_2 equivalent emissions (left) and proportional contributions of most remarkable factors (right).

20 years life-time of the plant and carbon emission rights of more than 200€ per tonne of CO_2 (Barón et al., 2023). On the other hand, the significant influence of electricity prices on the operational costs of using green hydrogen is also observable in the different results for S2 (with an electricity cost of 0.04€/MWh) and S3 (0.076€/MWh). While the operational costs of S2 are comparable to S0 and S1, standing at 200.05 M€, operational costs of S3 come totally out of a viability range, being 322.30 M€ per year.

To provide better insight into the economic performances, the breakdown of the annual costs for each scenario is shown in Fig. 4. Annualising the initial disbursement costs reveals the limited impact of a mild increase in capital expenditure on the total costs and, thus, on the global performance of alumina production. Energy costs using either fossil fuels or electricity represent the main economic variables of the industry. For S0 and S1, the costs of natural gas are 37.4 and 40.5% respectively, whereas the electricity costs for S2 and S3 correspond to 45.9 and 58.5% of the total life cycle costs, respectively.

The results of the six environmental indicators and the LCC results were normalised to the most prominent Figure (S3 for all cases), and its graphical representation is shown in Fig. 5. Scenario S2 has the most negligible environmental impact, especially for indicators such as GW, Land Use and Fossil Resource Scarcity. In other indicators, such as Water Consumption, Terrestrial Acidification or even LCC results, the differences between S0, S1 and S2 were small, hampering a good comparison among them. Focusing only on the consequences of GW and the economic performance of each alternative, Fig. 6 shows the PSV results of the four different scenarios studied (Y-axis) represented in opposition to their respective CO_2 equivalent emissions (X-axis). To facilitate visual comprehension, the image includes range limits of market profitability and environmental impact tolerability. The market profitability is fixed as the range of positive values of PSV variable. Admissible environmental impacts are more arduous and subjective. In this case, a 33% reduction in the Global Warming indicator with respect to base case S0 was estimated as a tolerable performance. With these assumptions, the graphic shows how S3 configuration excessively surpasses both market and environmental limits because of the vast electricity costs and impacts associated with electricity production. Configurations S1 and S2 remained inside the range limits in terms of both economic costs and global warming impact, indicating the advantages of the potential implementation of this technology.

The same data were used to calculate the eco-efficiency indices, which are suggested as a method for comparing profitability and sustainability simultaneously. Their results are given in Table 8. Scenario S1 was the most eco-efficient scenario. A negative value of S3 index indicates the impossibility of obtaining commercial profit from the

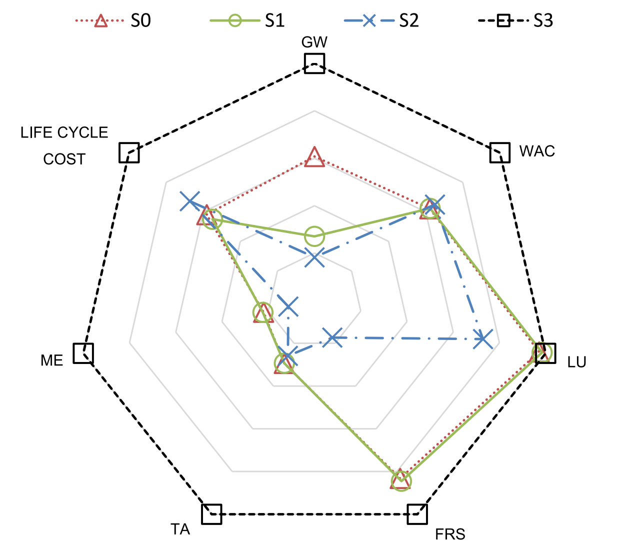

Fig. 5. Representation of all normalised LCA indicators, together with LCC results.

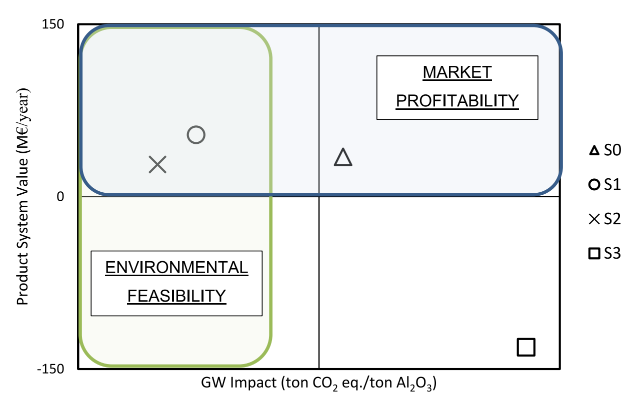

Fig. 6. Representation of defined eco-efficiency index: GW Impact vs Product System Value.

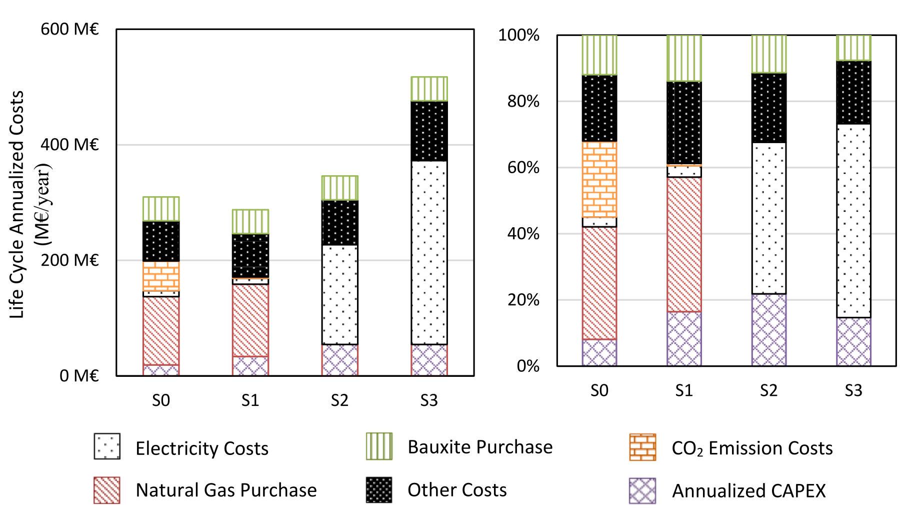

Fig. 4. Results of life cycle annualized costs. Total costs (left) and proportional contributions of most remarkable factors (right).

Table 8
Eco-efficiency indices of each scenario.

	S0	S1	S2	S3
EI (€/tonne CO_2 eq.)	31.46	109.94	84.72	−72.32

process. The positive value of the S2 index, on the other side, shows its potential profitability despite the high initial disbursement costs. The commercialisation of O_2 produced during water electrolysis helps also to make the process viable and even more eco-efficient than the base case S0, which is penalised by its high CO_2 emissions.

To check the influence of electricity prices and carbon emission taxes globally and verify the results, a sensitivity analysis was performed on these two variables. The results are presented as a 3D graph in Fig. 7a. To facilitate visual understanding, the front view of the X-axis (electricity prices) is also represented in Fig. 7b, showing the eco-efficiency indices for scenarios S0, S1 and S2 in the range of 0–150€/tonne of CO_2 of taxable emissions. To facilitate comprehension, the S3 results are not included in the graph because its environmental impact was found to be out of a tolerable range and its economic performance overlapped with that of S2. In Figs. 7a and b, electricity significantly influenced the eco-efficiency of S2. If electricity prices remained below 0.036€/kWh, S2 would represent the most eco-efficient process. However, if these prices increased to 0.045€/kWh, this configuration would no longer be profitable, and eco-efficiency would become negative. Another interesting conclusion is that case S1 always shows higher eco-efficiency than scenario S0, regardless of the penalty of the carbon taxes that are carried along. This does not mean that S1 would maintain better economic performance even if no CO_2 emission taxes were implemented; however, from an environmental perspective, profitability would be higher, as more value would be created per tonne of CO_2 emitted.

The overall results of the study show significant implications for the different stakeholders of the alumina industry. The sensitivity analysis indicates the favourable effects of the implementation of the emission rights system as a means of promoting decarbonisation. However, additional strategies are required by policymakers, such as tax incentives for investment in low-carbon technologies or a strong support to researchers and innovation sectors. Governments must also develop a reliable energy policy that guarantees the availability of renewable energies. The study indicates also the need for industrial practitioners to embrace innovation technologies, given the environmental and economic impact of the current state of the industry. Electrification and CO_2 capture technologies show the potential to reduce energy costs and increase competitiveness, despite the initial increase in costs because of the need of new technologies and infrastructure.

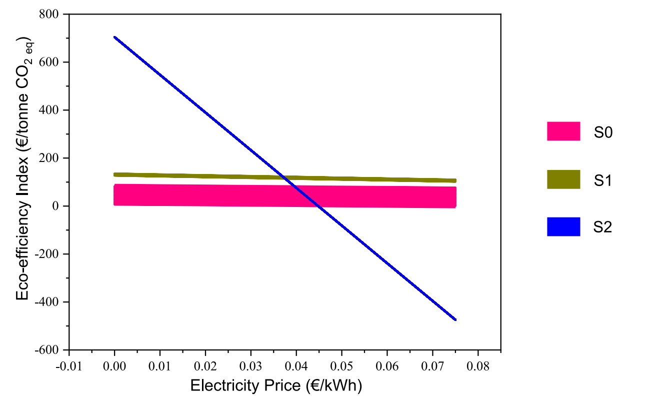

Fig. 7b. Sensitivity analysis. Front view: range variation of eco-efficiency index (Y-axis) according to changes on electricity price (X-axis).

4. Conclusions

In this study, four different sources of thermal energy and CO_2 mitigation strategies were considered for modelling an alumina refinery plant. Subsequently, integrated LCA and LCC analyses were performed for each of the four cases. This study aimed to quantify the environmental impacts of each scenario, determine if those cases could meet the economic performance requirements to be implemented in the alumina industry in the future, and comparing them with the current alumina production process.

Following the ReCiPe evaluation method, the environmental impact of the base case of an alumina refinery using natural gas was estimated at 1.10 tonnes of CO_2 per tonne of alumina produced. Results indicated that the integration of a calcium looping plant to capture and store post-combustion CO_2 could decrease the equivalent CO_2 emissions down to 0.49 tonnes of CO_2 per tonne of alumina. Replacing natural gas with

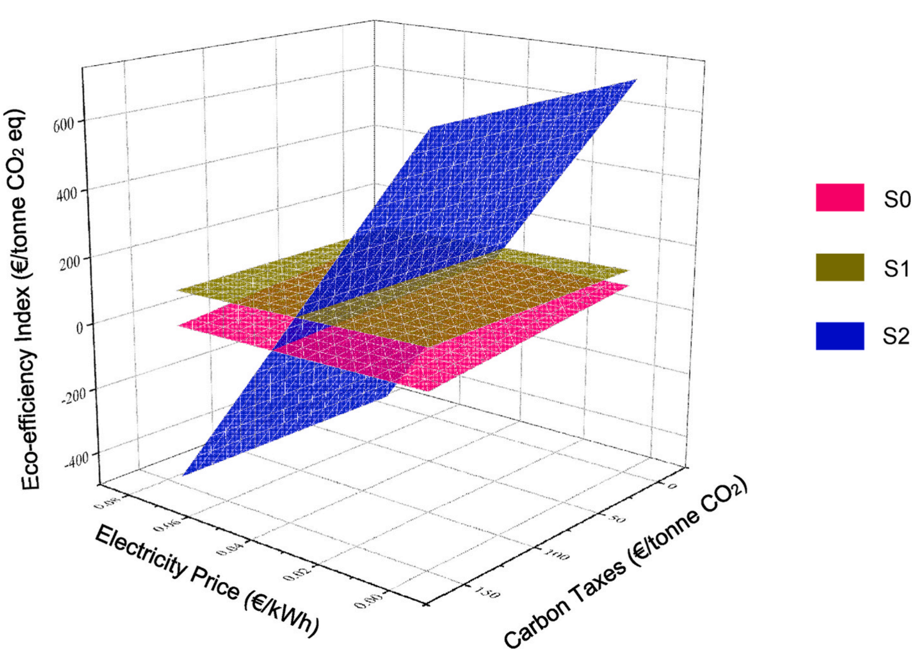

Fig. 7a. Sensitivity analysis. Variation of eco-efficiency index (Z axis) according to changes on electricity price (Y axis) and CO_2 emission taxes (X axis).

green hydrogen showed greater potential environmental benefits, reducing this indicator to 0.33. Regarding the economic performance from the LCC perspective, adding a CO_2 capture plant resulted in lower economic costs than in the single refinery case. Despite the more significant initial investments, the life cycle annualized costs when integrating the carbon capture plant were estimated at 287.72 M€ per year. In comparison, the base case costs were 311.35 M€, explained by costs of CO_2 emissions. Green hydrogen production instead of importing natural gas, however, implied higher costs, which were strongly affected by the electricity price and varied from 346.42 to 524.70 M€ per year.

To obtain a common overview of both economic and environmental performances, an eco-efficiency ratio index was defined and calculated for each case, showing the CO_2 capture and storage as the most eco-efficient case, regardless of the fluctuation of the economic penalty of CO_2 emissions. Substituting natural gas with green hydrogen was also a good eco-efficient solution as long as the electricity prices were kept under 0.045€/kWh.

Despite some limitations of the study, such as the purchase estimations, the modelling differences with a real industrial plant and the economic and technological unpredictability, these findings can serve as a valuable resource for various stakeholders in the aluminium industry seeking to enhance their production process in an environmentally friendly manner. This study emphasizes the necessity for industry professionals to adopt innovative technologies due to their environmental and economic significance. While electrification and CO_2 capture technologies may initially raise costs due to new infrastructure requirements, they have the potential to lower energy expenses and enhance competitiveness in the long run. Additionally, the decarbonisation of the alumina industry must be accompanied by policymakers, who need to adopt additional measures, such as offering tax incentives for low-carbon technology investments and supporting research and innovation, especially to ensure access to renewable energy sources. Finally, this study underlines the importance of adequately integrating environmental and economic analyses to assess sustainability and highlights the implications of thermal energy consumption in the alumina industry.

CRediT authorship contribution statement

Javier Sáez-Guinoa: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Conceptualization. **Inés Senante:** Resources, Investigation. **Sara Pascual:** Resources, Investigation. **Eva Llera-Sastresa:** Writing – review & editing, Supervision, Conceptualization. **Luis M. Romeo:** Writing – review & editing, Supervision.

Declaration of competing interest

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

Acknowledgements

This work was carried out in the frame of the AlSiCal project, funded by the European Union's Horizon 2020 research and innovation programme under grant agreement N. 820911. The work described in this paper is also partially supported by the Government of Aragon (Research Group DGA T46_23R: Energía y CO_2).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2024.144366.

Data availability

Data will be made available on request.

References

Abanades, J.C., 2002. The maximum capture efficiency of CO2 using a carbonation/calcination cycle of CaO/CaCO3. Chem. Eng. J. 90, 303–306. https://doi.org/10.1016/S1385-8947(02)00126-2.
Adhikari, B., Lister, T.E., Reddy, R.G., 2022. Techno-economic and life cycle assessment of aluminum electrorefining from mixed scraps using ionic liquid. Sustain. Prod. Consum. 33, 932–941. https://doi.org/10.1016/j.spc.2022.08.020.
Anekwe, I.M.S., Tetteh, E.K., Akpasi, S., Atuman, S.J., Armah, E.K., Isa, Y.M., 2023. Carbon dioxide capture and sequestration technologies – current perspective, challenges and prospects. Green Sustain. Proc. Chem. Environ. Eng. Sci. Carbon Dioxide Capt. Util. 481–516. https://doi.org/10.1016/B978-0-323-99429-3.00034-5.
Ashby, M.F., 2024. Life cycle costing. LCC Mater. Sustain. Dev. 149–181. https://doi.org/10.1016/B978-0-323-98361-7.00007-5.
Aspen Plus, 2024. Leading Process Simulation Software | AspenTech [WWW Document], n.d. URL. https://www.aspentech.com/en/products/engineering/aspen-plus. (Accessed 14 February 2024).
Aspen Process Economic Analyzer [WWW Document], n.d. URL https://www.aspentech.com/en/products/engineering/aspen-process-economic-analyzer (accessed February.14.2024).
Barón, C., Perpiñán, J., Bailera, M., Peña, B., 2023. Techno-economic assessment of glassmaking decarbonization through integration of calcium looping carbon capture and power-to-gas technologies. Sustain. Prod. Consum. 41, 121–133. https://doi.org/10.1016/J.SPC.2023.07.029.
Behnami, A., Zoroufchi Benis, K., Shakerkhatibi, M., Fatehifar, E., Derafshi, S., Chavoshbashi, M.M., 2019. Integrating data reconciliation into material flow cost accounting: the case of a petrochemical wastewater treatment plant. J. Clean. Prod. 218, 616–628. https://doi.org/10.1016/j.jclepro.2019.01.218.
Carbone, C., Ferrario, D., Lanzini, A., Verda, V., Agostini, A., Stendardo, S., 2023. Calcium looping in the steel industry: GHG emissions and energy demand. Int. J. Greenh. Gas Control 125, 103893. https://doi.org/10.1016/j.ijggc.2023.103893.
Caustic Soda Prices | Historical and Current | Intratec.us [WWW Document], n.d. URL https://www.intratec.us/chemical-markets/caustic-soda-price (accessed September.23.2024).
Chirone, R., Paulillo, A., Coppola, A., Scala, F., 2022. Carbon capture and utilization via calcium looping, sorption enhanced methanation and green hydrogen: a techno-economic analysis and life cycle assessment study. Fuel 328, 125255. https://doi.org/10.1016/J.FUEL.2022.125255.
Čuček, L., Klemeš, J.J., Kravanja, Z., 2015. Overview of environmental footprints. Assess. Meas. Environ. Impact Sustain. 131–193. https://doi.org/10.1016/B978-0-12-799968-5.00005-1.
Cushman-Roisin, B., Cremonini, B.T., 2021. Materials. Data, stat. Useful numbers. Environ. Sustain. 1–16. https://doi.org/10.1016/B978-0-12-822958-3.00012-1.
Demuynck, J., De Paepe, M., Sierens, R., Verhelst, S., 2010. Heat transfer comparison between methane and hydrogen in a spark ignited engine. In: 18th World Hydrogen Energy Conference 2010, WHEC 2010, Proceedings. Essen, Germany, pp. 149–156.
Donaldson, D., 2008. Light Met. https://doi.org/10.1201/9781420011906.sec3.
Donoghue, A.M., Frisch, N., Olney, D., 2014. Bauxite mining and alumina refining: process description and occupational health risks. J. Occup. Environ. Med. 56, S12. https://doi.org/10.1097/JOM.0000000000000001.
Dyson, B., 2024. Integration of life cycle and life cycle sustainability assessments into decision analytic approaches for sustainable technologies. Encycl. Sustain. Technol. 643–655. https://doi.org/10.1016/B978-0-323-90386-8.00057-7.
ecoinvent v3.7 – ecoinvent [WWW Document], 2020. URL https://ecoinvent.org/the-ecoinvent-database/data-releases/ecoinvent-3-7/(accessed 7.May.2022).
European Aluminium Association, 2018. Environmental Profile Report.
European Aluminium, CRU Group, 2020. A vision for strategic, low carbon and competitive aluminium. In: Vision 2050: Vision 2050: European Aluminium's Contribution to the EU's Mid-Century Low-Carbon Roadmap.
European Commission, 2018. A Clean Planet for all: a European strategic long-term vision for a prosperous, modern, competitive and climate neutral economy. Brussels.
European Commission, 2023. Report from the Commission to the European Parliament and the Council on the Functioning of the European Carbon Market in 2022 Pursuant to Articles 10(5) and 21(2) of Directive 2003/87/EC.
European Commission; Directorate-General for Energy unit A4 Market Observatory for Energy, 2021. Quarterly Report on European Electricity Markets.
Eurostat - European Commission, 2022.). Gas Prices for Household Consumers - Bi-annual Data (From 2007 Onwards.
Farjana, S.H., Huda, N., Mahmud, M.A.P., 2019. Impacts of aluminum production: a cradle to gate investigation using life-cycle assessment. Sci. Total Environ. 663, 958–970. https://doi.org/10.1016/j.scitotenv.2019.01.400.
Grasa, G.S., Abanades, J.C., 2006. CO2 capture capacity of CaO in long series of carbonation/calcination cycles. Ind. Eng. Chem. Res. 45, 8846–8851. https://doi.org/10.1021/ie0606946.
Haaf, M., Anantharaman, R., Roussanaly, S., Ströhle, J., Epple, B., 2020. CO2 capture from waste-to-energy plants: techno-economic assessment of novel integration concepts of calcium looping technology. Resour. Conserv. Recycl. 162, 104973. https://doi.org/10.1016/J.RESCONREC.2020.104973.

IEA, 2022. IEA (2022), Aluminium. IEA. Paris License: CC BY 4.0 [WWW Document]. URL. https://www.iea.org/reports/aluminium. (Accessed 15 May 2023).

International Aluminium, 2022. Life Cycle Inventory Data and Environmental Metrics for the Primary Aluminium Industry.

International Aluminium, 2023. Primary Aluminium Production [WWW Document]. URL. https://international-aluminium.org/statistics/primary-aluminium-production/. (Accessed 25 June 2024).

International Energy Agency, 2019. The Future of Hydrogen: seizing today's opportunities. Future Hydrogen. https://doi.org/10.1787/1e0514c4-en.

International Energy Agency, 2020. Energy technology perspectives 2020. Energy Technol. Perspect 2020. https://doi.org/10.1787/ab43a9a5-en.

International Energy Agency, 2023. Global hydrogen review 2023. Global Hydrogen Rev. 2023. https://doi.org/10.1787/a15b8442-en.

International Renewable Energy Agency, 2020. Green Hydrogen Cost Reduction: Scaling Up Electrolysers To Meet The 1.5°C Climate Goal H.

ISO, 2006a. ISO 14040:2006 Environmental Management - Life Cycle Assessment - Principles and Framework.

ISO, 2006b. ISO 14044:2006 Environmental Management - Life Cycle Assessment- Requirements and Guidelines.

Koskela, M., Vehmas, J., 2012. Defining eco-efficiency: a case study on the Finnish forest industry. Bus. Strat. Environ. 21, 546–566. https://doi.org/10.1002/BSE.741.

Lim, L.H., Tan, P., Chan, W.P., Veksha, A., Lim, T.T., Lisak, G., Liu, W., 2023. A techno-economic assessment of the reutilisation of municipal solid waste incineration ash for CO2 capture from incineration flue gases by calcium looping. Chem. Eng. J. 464, 142567. https://doi.org/10.1016/J.CEJ.2023.142567.

Luthin, A., Backes, J.G., Traverso, M., 2021. A framework to identify environmental-economic trade-offs by combining life cycle assessment and life cycle costing – a case study of aluminium production. J. Clean. Prod. 321, 128902. https://doi.org/10.1016/J.JCLEPRO.2021.128902.

Ma, Y., Preveniou, A., Kladis, A., Pettersen, J.B., 2022. Circular economy and life cycle assessment of alumina production: simulation-based comparison of Pedersen and Bayer processes. J. Clean. Prod. 366, 132807. https://doi.org/10.1016/J.JCLEPRO.2022.132807.

Mastin, J., Aranda, A., Meyer, J., 2011. New synthesis method for CaO-based synthetic sorbents with enhanced properties for high-temperature CO2 -capture. Energy Proc. 4, 1184–1191. https://doi.org/10.1016/J.EGYPRO.2011.01.172.

Miah, J.H., Koh, S.C.L., Stone, D., 2017. A hybridised framework combining integrated methods for environmental Life Cycle Assessment and Life Cycle Costing. J. Clean. Prod. 168, 846–866. https://doi.org/10.1016/j.jclepro.2017.08.187.

Muralikrishna, I.V., Manickam, V., 2017. Environmental management life cycle assessment. Environ. Manag. 57–75. https://doi.org/10.1016/B978-0-12-811989-1.00005-1.

Nunez, P., Jones, S., 2016. Cradle to gate: life cycle impact of primary aluminium production. Int. J. Life Cycle Assess. 21, 1594–1604. https://doi.org/10.1007/s11367-015-1003-7.

Ozturk, M., Dincer, I., 2021. A comprehensive review on power-to-gas with hydrogen options for cleaner applications. Int. J. Hydrogen Energy 46, 31511–31522. https://doi.org/10.1016/J.IJHYDENE.2021.07.066.

Parra, D., Patel, M.K., 2016. Techno-economic implications of the electrolyser technology and size for power-to-gas systems. Int. J. Hydrogen Energy 41, 3748–3761. https://doi.org/10.1016/J.IJHYDENE.2015.12.160.

Peças, P., Götze, U., Bravo, R., Richter, F., Ribeiro, I., 2019. Methodology for selection and application of eco-efficiency indicators fostering decision-making and communication at product level—the case of molds for injection molding. Adv. Appl. Manuf. Eng. 1–52. https://doi.org/10.1016/B978-0-08-102414-0.00001-X.

Peng, T., Ou, X., Yan, X., Wang, G., 2019. Life-cycle analysis of energy consumption and GHG emissions of aluminium production in China. Energy Proc. 158, 3937–3943. https://doi.org/10.1016/j.egypro.2019.01.849.

Peppas, A., Politi, C., Kottaridis, S., Taxiarchou, M., 2023. LCA analysis decarbonisation potential of aluminium primary production by applying hydrogen and CCUS technologies. Hydrogen 4, 338–356. https://doi.org/10.3390/hydrogen4020024.

Peters, M.S., Timmerhaus, K.D., West, R.E., 2003. Plant Design and Economics for Chemical Engineers, fifth ed.

Pizzol, M., Laurent, A., Sala, S., Weidema, B., Verones, F., Koffler, C., 2017. Normalisation and weighting in life cycle assessment: quo vadis? Int. J. Life Cycle Assess. 22, 853–866. https://doi.org/10.1007/s11367-016-1199-1.

Qaidi, S.M.A., Tayeh, B.A., Isleem, H.F., de Azevedo, A.R.G., Ahmed, H.U., Emad, W., 2022. Sustainable utilization of red mud waste (bauxite residue) and slag for the production of geopolymer composites: a review. Case Stud. Constr. Mater. 16, e00994. https://doi.org/10.1016/J.CSCM.2022.E00994.

Rebitzer, G., 2002. Integrating life cycle costing and life cycle assessment for managing costs and environmental impacts in supply chains. Cost Manag. Supply Chain. 127–146. https://doi.org/10.1007/978-3-662-11377-6_8.

Rodriguez, N., Alonso, M., Grasa, G., Abanades, J.C., 2008. Heat requirements in a calciner of CaCO3 integrated in a CO2 capture system using CaO. Chem. Eng. J. 138, 148–154. https://doi.org/10.1016/J.CEJ.2007.06.005.

Romano, M.C., Spinelli, M., Campanari, S., Consonni, S., Cinti, G., Marchi, M., Borgarello, E., 2013. The calcium looping process for low CO2 emission cement and power. Energy Proc. 37, 7091–7099. https://doi.org/10.1016/J.EGYPRO.2013.06.645.

Romeo, L.M., Abanades, J.C., Escosa, J.M., Paño, J., Giménez, A., Sánchez-Biezma, A., Ballesteros, J.C., 2008. Oxyfuel carbonation/calcination cycle for low cost CO2 capture in existing power plants. Energy Convers. Manag. 49, 2809–2814. https://doi.org/10.1016/J.ENCONMAN.2008.03.022.

Romeo, L.M., Lara, Y., Lisbona, P., Escosa, J.M., 2009. Optimizing make-up flow in a CO2 capture system using CaO. Chem. Eng. J. 147, 252–258. https://doi.org/10.1016/j.cej.2008.07.010.

Ruys, A., 2019. Bauxite: the principal aluminum ore. In: Alumina Ceramics. Elsevier, pp. 39–47. https://doi.org/10.1016/b978-0-08-102442-3.00002-6.

Rwe, A.G., 2024. RWE Annual Report 2023.

Saevarsdottir, G., Kvande, H., Welch, B.J., 2020. Aluminum production in the times of climate change: the global challenge to reduce the carbon footprint and prevent carbon leakage. Jom 72, 296–308. https://doi.org/10.1007/s11837-019-03918-6.

Sáez-Guinoa, J., García-Franco, E., Llera-Sastresa, E., Romeo, L.M., 2024. The effects of energy consumption of alumina production in the environmental impacts using life cycle assessment. Int. J. Life Cycle Assess. 29, 380–393. https://doi.org/10.1007/S11367-023-02257-8.

Saint-Just, J., Etemad, S., 2016. Catalytic combustion of hydrogen for heat production. Compend. Hydrog. Energy Hydrog. Energy Convers. 3 (3), 263–287. https://doi.org/10.1016/B978-1-78242-363-8.00010-4.

Samitha Weerakoon, A.H., Kleszcz, S., Assadi, M., 2024. Exogenous-spatial scale techno-economic methodology for energy application assessment based on levelized cost of electricity: a case study in Norway. Energy Convers. Manag. X 22, 100560. https://doi.org/10.1016/J.ECMX.2024.100560.

Schmidt, J.H., Thrane, M., 2009. Life cycle assessment of aluminium production in new Alcoa smelter in Greenland [WWW Document]. Life cycle Assess. Alum. Prod. new Alcoa smelter Greenl. URL. https://lca-net.com/publications/show/life-cycle-assessment-aluminium-production-new-alcoa-smelter-greenland/. (Accessed 25 September 2020).

Schwarz, H.-G., 2004. Aluminum production and energy. In: Encyclopedia of Energy, pp. 81–95. https://doi.org/10.1016/b0-12-176480-x/00372-7.

Strojny, M., Gładysz, P., Hanak, D.P., Nowak, W., 2023. Comparative analysis of CO2 capture technologies using amine absorption and calcium looping integrated with natural gas combined cycle power plant. Energy 284, 128599. https://doi.org/10.1016/J.ENERGY.2023.128599.

Suss, A., Fedyaev, A., Kuznetzova, N., Damaskin, A., Kuvyrkina, A., Panov, A., Paromova, I., Lukyanov, I., 2010. Technology solutions to increase alumina recovery from aluminogoethitic bauxites. In: Johnson, J.A. (Ed.), Light Metals, pp. 53–56.

Swarr, T.E., Hunkeler, D., Klöpffer, W., Pesonen, H.L., Ciroth, A., Brent, A.C., Pagan, R., 2011. Environmental life-cycle costing: a code of practice. Int. J. Life Cycle Assess. 16, 389–391. https://doi.org/10.1007/s11367-011-0287-5.

Thema, M., Bauer, F., Sterner, M., 2019. Power-to-Gas: electrolysis and methanation status review. Renew. Sustain. Energy Rev. 112, 775–787. https://doi.org/10.1016/j.rser.2019.06.030.

Toniolo, S., Tosato, R.C., Gambaro, F., Ren, J., 2020. Life cycle thinking tools: life cycle assessment, life cycle costing and social life cycle assessment. Life Cycle Sustain. Assess. Decis. Methodol. Case Stud. 39–56. https://doi.org/10.1016/B978-0-12-818355-7.00003-8.

Valeev, D., Bobylev, P., Osokin, N., Zolotova, I., Rodionov, I., Salazar-Concha, C., Verichev, K., 2022. A review of the alumina production from coal fly ash, with a focus in Russia. J. Clean. Prod. 363, 132360. https://doi.org/10.1016/J.JCLEPRO.2022.132360.

Wang, Qiang, Huang, P., Wang, Qinmeng, Guo, X., 2023. Greenhouse gas emissions and future development trends of primary aluminum in China. J. Clean. Prod. 403, 136828. https://doi.org/10.1016/j.jclepro.2023.136828.

Yang, Zhaoyue, Yang, Zhendong, Yang, S., Liu, Ziliang, Liu, Zhenghua, Liu, Y., Drewniak, L., Jiang, C., Li, Q., Li, W., Yin, H., 2022. Life cycle assessment and cost analysis for copper hydrometallurgy industry in China. J. Environ. Manag. 309, 114689. https://doi.org/10.1016/J.JENVMAN.2022.114689.

Yi, X., Lu, Y., He, G., Li, H., Chen, C., Cui, H., 2022. Global carbon transfer and emissions of aluminum production and consumption. J. Clean. Prod. 362, 132513. https://doi.org/10.1016/j.jclepro.2022.132513.

Yu, W., Rao, Z., Yuan, H., Wei, P., Nyarko-Appiah, J.E., Jiang, W., 2023. An efficient and environmental friendly strategy for alumina extraction and Fe–Si alloys production from coal fly ash by combining vacuum thermal reduction, alkali dissolving, and magnetic separation. J. Clean. Prod. 408, 137129. https://doi.org/10.1016/J.JCLEPRO.2023.137129.

Zhou, G. tao, Wang, Y. lin, Qi, T. gui, Zhou, Q. Sheng, Liu, G. hua, Peng, Z. hong, Li, X. bin, 2022. Enhanced conversion mechanism of Al-goethite in gibbsitic bauxite under reductive Bayer digestion process. Trans. Nonferrous Metals Soc. China 32, 3077–3087. https://doi.org/10.1016/S1003-6326(22)66004-7.

Zhou, G.T., Wang, Y.L., Qi, T.G., Zhou, Q.S., Liu, G.H., Peng, Z.H., Li, X. Bin, 2023. Toward sustainable green alumina production: a critical review on process discharge reduction from gibbsitic bauxite and large-scale applications of red mud. J. Environ. Chem. Eng. 11, 109433. https://doi.org/10.1016/J.JECE.2023.109433.

Zhu, X., Jin, Q., Ye, Z., 2020. Life cycle environmental and economic assessment of alumina recovery from secondary aluminum dross in China. J. Clean. Prod. 277, 123291. https://doi.org/10.1016/J.JCLEPRO.2020.123291.

Zhu, S., Gao, C., Song, K., Chen, M., Wu, F., Li, X., 2022. An assessment of environmental impacts and economic benefits of multiple aluminum production methods. J. Clean. Prod. 370, 133523. https://doi.org/10.1016/J.JCLEPRO.2022.133523.