

Carbon Dioxide Capture in the Iron and Steel Industry: Thermodynamic Analysis, Process Simulation, and Life Cycle Assessment



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The iron and steel sector is one of the dominant drivers behind economic and social progress, but it is also very energy-intensive and hard-to-abate, making it a major cause of global warming. Improving energy efficiency, introducing hydrogen for direct reduction, and utilising CCS technologies are the three most viable options for reducing CO₂ emissions from steel mills. This investigation deals with a life cycle comparison of three different carbon capture processes, the inventory data of which have been obtained using process simulation based on rigorous phase and chemical equilibrium equations. In-silico models for the absorption of carbon dioxide employing MDEA, membranes, or sodium hydroxide to produce sodium bicarbonate have been developed and compared from a life cycle viewpoint. The research findings showed a variable amount of CO₂ removal in the three cases, where membranes achieved the best performance (95 % CO₂ removal). Since NaOH absorption produces a valuable by-product (sodium bicarbonate, which is commonly produced by Solvay process), the other two technologies were modified to integrate the utilisation of CO₂ for the synthesis of sodium bicarbonate with NaOH rather than transporting and storing the carbon dioxide. As a result, this production pathway for sodium bicarbonate generates lower environmental burdens than traditional Solvay process. The environmental performances of the alternatives are nearly equal, even though the environmental impacts associated with capturing the CO₂ and subsequently reacting with NaOH are always slightly higher than those involved with reacting directly during absorption. Among the evaluated alternatives, the direct conversion to sodium bicarbonate appears to be the most promising approach for converting CO₂ emissions in the steel sector.

Keywords:

carbon capture, steel, process modelling and simulation, life cycle assessment, sodium bicarbonate

Introduction

Global climate changes have been caused by the ongoing growth of the human population, the escalating demand for food and fuel, and the rise in new population requirements. The greatest factor contributing to rising temperatures is human activity, particularly the combustion of fossil fuels, which releases CO₂, methane, and other greenhouse gases into the atmosphere, including nitrous and sulphur oxides. The effects are clearly visible, from ice melting at Earth's poles and glaciers, rising of sea level, spreading of wildfires, more intense heat waves and storms, flooding, and longer periods of drought¹.

From the preindustrial era to 2017, according to the Intergovernmental Panel on Climate Change (IPCC), the average temperature rose between 0.8 °C and 1.2 °C, an increase of approximately 0.2 % per decade in the last 30 years². According to IPCC, global net anthropogenic CO₂ emissions must be decreased by 45 % from 2010 levels by 2030 in order to keep global warming at 1.5 °C or less, while net-zero CO₂ emissions are targeted for around 2050.

CO₂ is the greenhouse gas that is produced most intensively, and as a result, it has the greatest effect on climate change. The Publications Office of the European Union has stated that six countries are responsible for approximately 60 % of the global CO₂ emissions: China (29.30 %), United States (13.77 %), India (6.62 %), Russia (4.76 %), Japan

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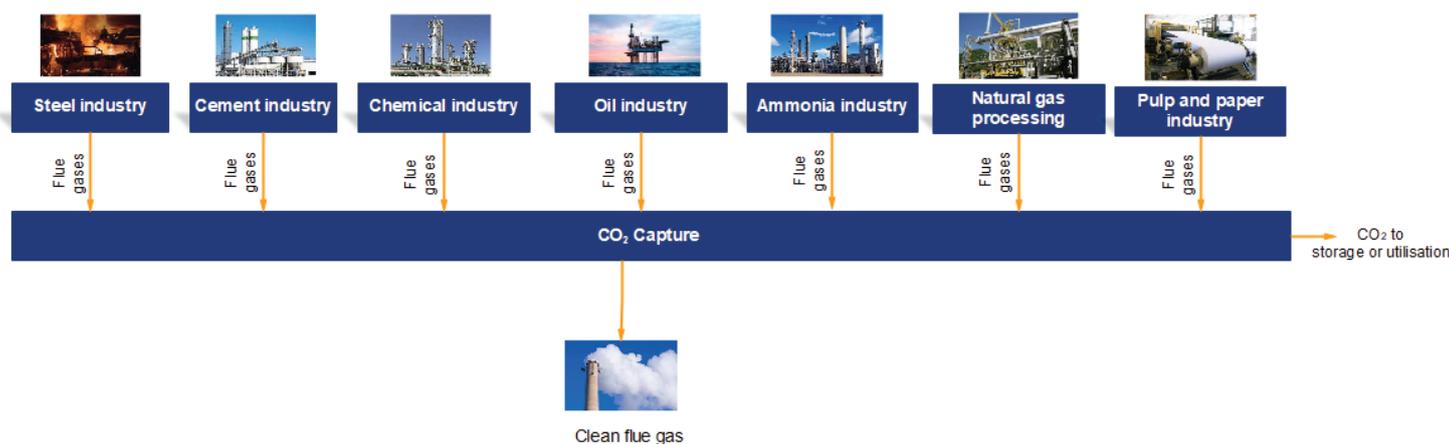


Fig. 1 – Overview of CO₂ capture from hard-to-abate sectors

(3.56 %), and Germany (2.15 %)³. The peak reduction in daily fossil CO₂ emissions due to the COVID-19 pandemic was approximately –17 %, which corresponds to the 2006 emission level. The population is beginning to recover from the pandemic and will once again consume more, so this sharp decline will be considerably less pronounced in the future⁴.

There are several ways to reduce global CO₂ emissions, which include enhancing energy efficiency, increasing renewable energy production, implementing a carbon tax, planting trees, conserving existing forests and grasslands, and capturing CO₂ from power plants and other sources⁵. Scientists, engineers, and governments have created a number of solutions to minimise the causes of climate change, such as replacing fossil fuels with green energy sources like wind and solar power. Other technologies are those integrating carbon capture and storage (CCS) or carbon capture utilisation and storage (CCUS) into currently used processes¹. For various applications, CCS has the potential to help the world achieve its climate change goals by enabling the use of fossil fuel-powered facilities while lowering CO₂ emissions⁶.

CCS entails trapping CO₂ before it is released into the atmosphere, transporting, and storing it for an extended length of time underground in huge geological formations, such as the underground reservoirs remaining after the extraction of oil or natural gas. The CCS can be an essential technology to reduce the CO₂ emitted by hard-to-abate sectors, like iron and steel, cement, oil refining or chemicals, ammonia production, natural gas processing, and pulp and paper industry (Fig. 1)⁷.

Due to the large CO₂ emissions around the world, the CO₂ from industrial sources has increased globally over the past decade⁸. As a consequence, the need for CCS implementation has also in-

creased. This is particularly true in Europe, where the European Commission has set ambitious reduction goals to achieve carbon neutrality by 2050⁹.

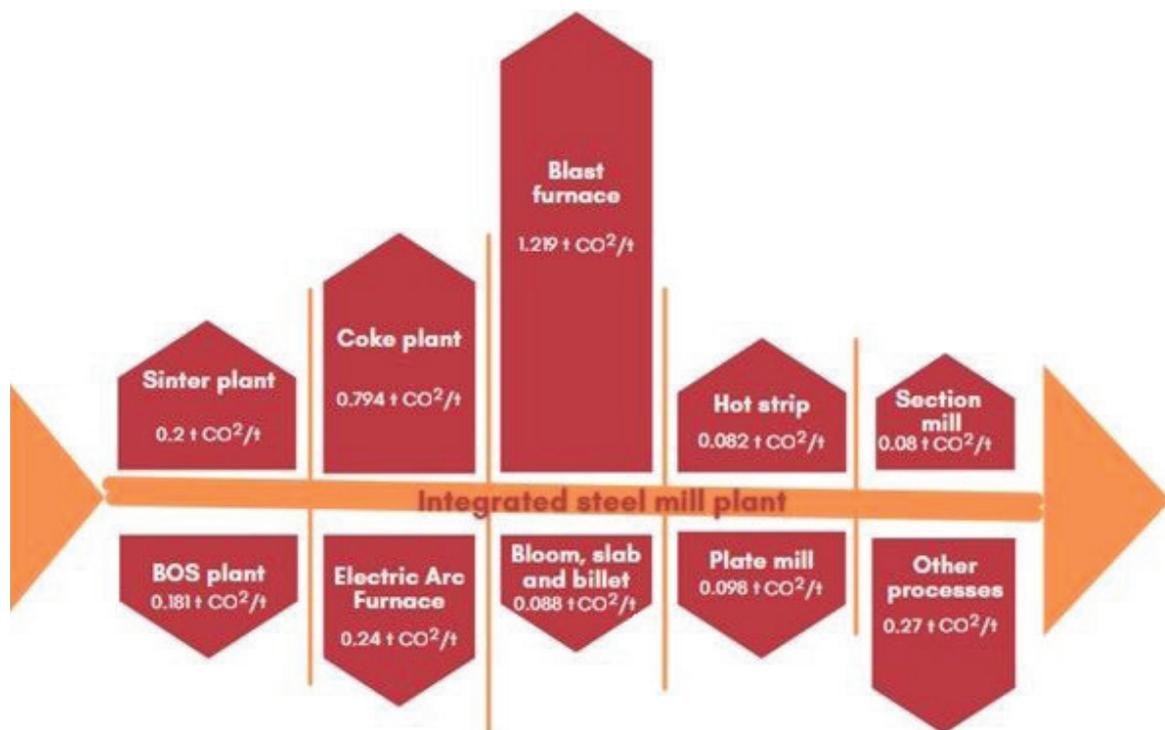
There are several technological pathways available for CO₂ capture, such as post-combustion, pre-combustion, oxy-fuel combustion, chemical looping combustion (CLC), and direct air capture (DAC) systems¹⁰. The advantages and disadvantages of these technologies are summarised in Table 1.

The iron and steel sector is one of the most significant drivers of economic and social development, playing a vital role in meeting society's needs as it provides services in vast areas, such as buildings and infrastructure, mechanical and electrical equipment, transport systems, metal products, and domestic appliances. On the other hand, the steel industry is very energy-intensive, being one of the main contributors to global warming¹¹. The International Energy Agency (IEA) has identified the iron and steel (with 2.6 Gt CO₂/y), the cement (with 2.4 Gt CO₂/y), and the chemical industries (with 1.4 Gt CO₂/y) as the major CO₂ emitters, with a contribution of about 70 % of industry's direct CO₂ emissions¹². CO₂ comes from various sources in an integrated steel plant. The blast furnace, which is the centre of the steel-making process and requires over 70 % of the operation's total energy needs, is one of the key sources, followed by the coke plant. The CO₂ emissions coming from an average steel plant are reported in Fig. 2.

Globally, steel is produced following two main routes: blast furnace (BF)/basic oxygen furnace (BOF) also referred to as “integrated steelmaking”, and electric arc furnace (EAF) known as “mini-mill”¹⁴. The BF/BOF methods of producing steel are more frequently employed in countries with plentiful natural gas resources than in Europe¹⁵. According to literature data, the BF/BOF technology

Table 1 – Advantages and disadvantages of different CO₂ capture technologies

Capture technologies	Advantages	Disadvantages
Post-Combustion	easy to retrofit to existing plants	reduced efficiency if low CO ₂ partial pressure; high energy demand; high costs of operation and implementation
Pre-Combustion	improved efficiency if high CO ₂ partial pressure; fully developed technology; can be retrofitted to existing plants	temperature and efficiency issues; sorbent regeneration needed; high costs for sorption systems
Oxy-Fuel Combustion	very high CO ₂ concentrations enhances absorption efficiency; mature air separation technologies available; suitable for compact boiler and other equipment with reduced volume of flue gas	high efficiency; high costs of oxygen production; susceptible to corrosion problems
CLC	high purity CO ₂ can be obtained; low-cost carriers for O ₂	poorly developed technology; metal sulphides formation has to be prevented before processing
DAC	atmospheric CO ₂ is retained; high efficiency for CO ₂ separation	high costs due to low CO ₂ concentration in air

Fig. 2 – CO₂ emissions in the steel plant¹³

(Other processes contain pellet plant, pickling line, cold mill, annealing, hot dip metal coating, organic coating)

produces emissions between 1.6 and 2.2 tCO₂/t_{steel}, while the EAF route produces emissions between 0.6 and 0.9 tCO₂/t_{steel} when using scrap metal, and between 1.4 and 2 tCO₂/t_{steel} when starting with iron ore¹⁶. Although the EAF route has lower CO₂ emissions compared to the BF/BOF route, it should be emphasised that the two production routes are not comparable due to differences in feedstock, steel quality, and products. The BF/BOF route is the pre-

dominant steelmaking route, accounting for about 70 % of the world steel production¹⁷.

According to the World Steel Association, modern steel mills exhibit extraordinarily high rates of energy usage, and advanced technologies have resulted in optimum energy efficiency. However, there are currently no alternative manufacturing methods to replace unsustainable iron and steel-making processes¹⁸. As the integrated steelmaking

process depends significantly on carbon-based fuels and reductants, improved strategies for reducing CO₂ emissions are needed in the iron and steel sector. The most promising candidates taking into consideration the reduction of CO₂ emissions from steel plants include improving energy efficiency, introducing hydrogen for direct reduction¹⁹, and using CCS technologies²⁰. Numerous academic articles in the field, research projects on the topic, and pilot plants testing various CCS technologies demonstrate the scientific community's ongoing interest in finding effective answers to the problems of climate change.

Various research exists that specifically address the application of CCS in the steel sector according to the state-of-the-art. The majority of published studies view post-combustion capture configurations as the best option for decarbonising the steel sector. For instance, Arasto and co-authors²¹, studied CO₂ reduction possibilities from a steel mill by applying liquid absorption using monoethanolamine (MEA). The IEAGHG R&D Program conducted a thorough analysis of the financial implications of using CCS in an integrated steel production, examining two different capture scenarios: (i) a post-combustion absorption using MEA, and (ii) a BOF with

Table 2 – Projects on CCS applied to steel production

Project name	Project acronym	Project period	Project website	Entities involved	Technology involved
Ultra-Low CO ₂ steelmaking ²⁸	ULCOS	September 2004 – August 2010	www.ulcos.org/en	47 partners and 15 European countries	Direct production of steel by electrolysis; Hydrogen-based reduction using hydrogen from CO ₂ -lean technologies, etc.
CO ₂ Ultimate Reduction System for Cool Earth 50 ²⁹	COURSE50	Phase 1: 2008 – 2017 Phase 2: 2018 –	https://www.course50.com/en/	Nippon Steel; JFE Steel; KOBELCO Steel	Chemical absorption technology
SEWGS technology platform for cost effective CO ₂ reduction in the iron and steel industry ³⁰	STEPWISE	May 2015 – May 2019	www.stepwise.eu/	9 partners and 5 European countries	Sorption Enhanced Water Gas Shift (SEWGS) technology
Hydrogen meeting future needs of low carbon manufacturing value chains ³¹	H2FUTURE	January 2017 – June 2021	www.h2future-project.eu/	6 partners and 3 European countries	Proton Exchange Membrane (PEM) electrolyser technology
Turning waste from steel industry into a valuable low-cost feedstock for energy intensive industry ³²	RESLAG	September 2015 – February 2019	www.reslage.eu	19 partners and 8 countries	Thermal energy storage for concentrated solar power
Green steel for Europe ³³	Green Steel	January 2020 – June 2021	https://www.estep.eu/green-steel-for-europe	10 partners	Integration of carbon capture technologies; Direct or smelting reduction; Iron ore electrolysis
Innovative industrial transformation of the steel and chemical industries of Europe ³⁴	INITIATE	November 2020 – October 2025	https://www.initiate-project.eu/	11 partners and 5 European countries	Innovative ammonia production technology
Salzgitter Low CO ₂ Steelmaking ³⁵	SALCOS		https://salcos.salzgitter-ag.com/en	8 partners	Green hydrogen production to replace coal in the steelmaking process

top gas recycle and methyl diethanolamine (MDEA) as absorbing solvent²². Other studies investigated more advanced technologies, such as Pressure Swing Adsorption (PSA)²³, Vacuum Pressure Swing Adsorption (VPSA) together with Oxygen Blast Furnace (OBF) technology²⁴, Sorption Enhanced Water-Gas Shift (SEWGS)²⁵, calcium looping²⁶, or calcium looping combined with Ca-Fe looping using steel slag²⁷.

The major CCS projects applied to steel production are summarised in Table 2.

The current study focuses on three CO₂ capture systems used in the steel-making industry. The most developed and well-known processes use amines as absorption agents, and they can be employed as benchmarks for comparison with other novel, upcoming technologies. Membrane technologies represent another promising technology leading to carbon capture rates higher than 85 %. According to the author's best knowledge, the coupling of CO₂ capture utilising NaOH and steel manufacturing has yet to be explored. Since NaOH is even more plentiful, less expensive, and more well-known than MEA, and since its theoretical CO₂ absorption capability is larger than MEA's (1.39 t/tCO₂ vs. 0.9 t/tCO₂), using soda is a viable option³⁶.

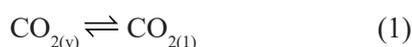
After the Introduction dealing with the presentation of the subject, the necessity and significance of the current research, the next section, entitled Materials and Methods, focuses on a detailed description of the methods and tools used in the investigation of the three cases under study, i.e., thermodynamic models, process modelling and simulation, and life cycle assessment (LCA). This section is followed by the results and discussion section, where the outcomes of the analysis are deeply analysed, and by some concluding remarks.

Materials and methods

This section presents the thermodynamic analysis of the systems, followed by the description of the process simulations, and the assumptions used for the LCA.

Thermodynamic analysis: Chemical and phase equilibria

CO₂ solubility in aqueous amine-NaOH solutions is determined by both its physical solubility and the chemical equilibrium for the aqueous phase reactions among CO₂, water, and solvent. Physical solubility is the equilibrium between gaseous CO₂ molecules and CO₂ molecules in the aqueous solutions:



It can be expressed by Henry's law:

$$P \cdot y_{\text{CO}_2} \cdot \hat{\phi}_{\text{CO}_2}^V = x_{\text{CO}_2} \cdot H_{\text{CO}_2} \cdot \gamma_{\text{CO}_2}^* \quad (2)$$

where P is the system pressure, y_{CO_2} is the mole fraction of CO₂ in the vapour phase, $\hat{\phi}_{\text{CO}_2}^V$ is the CO₂ fugacity coefficient in the vapour phase, H_{CO_2} is the Henry's law constant of CO₂ in the mixed solvent of water and solvent, x_{CO_2} is the equilibrium CO₂ mole fraction in the liquid phase, and $\gamma_{\text{CO}_2}^*$ is the asymmetric activity coefficient of CO₂ in the mixture of water and solvent.

Henry's constant in the mixed solvent can be calculated from those in the pure solvents³⁷:

$$\ln\left(\frac{H_i}{\gamma_i^\infty}\right) = \sum x_A \ln\left(\frac{H_{iA}}{\gamma_{iA}^\infty}\right) \quad (3)$$

where H_i is Henry's constant of supercritical component i in the mixed solvent, H_{iA} is Henry's constant of supercritical component i in pure solvent A , γ_i^∞ is the infinite dilution activity coefficient of supercritical component i in the mixed solvent, γ_{iA}^∞ is the infinite dilution activity coefficient of supercritical component i in pure solvent A , and x_A is the mole fraction of solvent A .

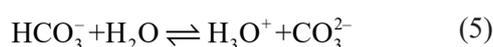
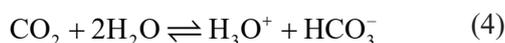
The ELECTrolyte Non-Random Two-Liquid (ELECNRTL) thermodynamic model was selected to evaluate the properties of the aqueous systems considered. ELECNRTL was developed to account for the non-ideality of a single salt that completely dissociates in water, and it has been widely adopted and validated for CO₂ absorption/stripping applications using both MEA and hot potassium carbonate (HPC) solvents³⁸.

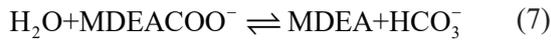
The vapour phase fugacity coefficients are calculated using the Peng Robinson (PR) equation of state. Water, hydroxides, and carbonates are treated as pure components at the reference state, which is the system pressure and temperature, whereas the reference state for CO₂ is infinite dilution.

For all the absorption/stripping columns, a rigorous rate-based modelling approach, accounting for mass-transfer and appropriate sizing to ensure a correct fluid-dynamic regime, was employed.

The molecule-molecule non-randomness parameters, H₂O-salt pairs non-randomness parameters, and the CO₂-salt pairs non-randomness parameters were fixed, and temperature-dependent energy parameters were modelled as described by Hesse *et al.*³⁹ All the parameters for the ELECNRTL model were taken from the Aspen Plus data bank.

The chemical reactions occurring when carbon dioxide (CO₂) is absorbed in an aqueous MDEA solution are summarised as follows:





The equilibrium constants of the reaction were calculated from the reference-state Gibbs free energies of the participating components:

$$-RT \ln K_j = \Delta G_j^0(T) \quad (9)$$

where K_j is the equilibrium constant of reaction j , $\Delta G_j^0(T)$ is the reference-state Gibbs free energy change for reaction j at temperature T , R is the universal gas constant, and T is the system temperature.

For the aqueous phase reactions, the reference states chosen are pure liquid for the solvents (water and MDEA/NaOH), and aqueous phase infinite dilution for the solutes (ionic and molecular).

The Gibbs free energy of solvents is calculated from that of ideal gas and the departure function:

$$G_s(T) = G_s^{ig}(T) + \Delta G_{js}^{ig \rightarrow l}(T) \quad (10)$$

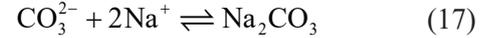
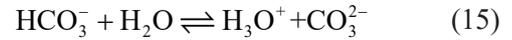
where $G_s(T)$ is Gibbs free energy of solvent s at temperature T , $G_s^{ig}(T)$ is the ideal gas Gibbs free energy of solvent s at temperature T , and $\Delta G_{js}^{ig \rightarrow l}(T)$ is Gibbs free energy departure from ideal gas to liquid at temperature T .

The Gibbs free energy of an ideal gas is calculated from the Gibbs free energy of formation of an ideal gas at 298.15 K, the enthalpy of formation of an ideal gas at 298.15 K, and the ideal gas heat capacity.

$$G_s^{ig}(T) = \Delta_f H_{j,298.15}^{ig} + \int_{298.15}^T C_{P,s}^{ig} dT - T \cdot \left(\frac{\Delta_f H_{j,298.15}^{ig} - \Delta_f G_{j,298.15}^{ig}}{298.15} + \int_{298.15}^T \frac{C_{P,s}^{ig}}{T} dT \right) \quad (11)$$

where $G_s^{ig}(T)$ is the ideal gas Gibbs free energy of solvent s at temperature T , $\Delta_f G_{j,298.15}^{ig}$ is the ideal gas Gibbs free energy of formation of solvent s at 298.15 K, $\Delta_f H_{j,298.15}^{ig}$ is the ideal gas enthalpy of formation of solvent s at 298.15 K, and $C_{P,s}^{ig}$ is the ideal gas heat capacity of solvent s . The reference-state properties, $\Delta_f G_{j,298.15}^{ig}$ and $\Delta_f H_{j,298.15}^{ig}$, and the ideal gas heat capacities were taken from Aspen Plus v12.1 data banks⁴⁰. For water, the Gibbs free energy departure function was obtained from the ASME steam tables. For MDEA and NaOH, the departure function was calculated from the ELECNRTL model included in Aspen Plus v12.1⁴⁰.

The chemical reactions taking place during usage of NaOH to convert CO_2 into sodium bicarbonate (NaHCO_3) are described as follows:



The kinetic parameters and equilibrium constants were retrieved from the literature⁴¹.

To model CO_2 compression and pipeline transport, the PR Equation of State (EoS) with Boston–Mathias modifications (PR-BM) was selected. Barbera *et al.*³⁸ have shown that the PR EoS is able to describe the behaviour of pure CO_2 as well as of binary/ternary mixtures comprising most typical impurities, such as nitrogen (N_2) and oxygen (O_2), provided that suitable binary interaction parameters (k_{ij} s) are employed. The validity of the Aspen Plus PR-BM model in the near supercritical region against experimental data for pure CO_2 , and for binary mixtures of gases containing CO_2 was also verified³⁸.

It is commonly acknowledged that, to minimise the energy costs, CO_2 should be transported at a pressure above the critical value P_c (73.8 bar), in order to have low viscosity and high density. The presence of impurities causes the mixture properties to be different from those of pure CO_2 . In particular, a phase envelope can be identified depending on the composition, so that sufficiently high pressures should be kept along the pipeline to avoid the formation of a 2-phase system. In this work, it was verified that no retrograde condensation phenomena occur under the conditions investigated.

Process modelling and simulation

The *in-silico* approach involves a wide panorama of modelling techniques, which allow to gain life cycle inventories of various products, from materials⁴², to chemicals⁴³, or processes⁴⁴. A description of the characteristics of the process simulations performed with the aim of gaining material and energy balances are reported within this section.

Description of Case 1: CO_2 capture using gas-liquid absorption (MDEA solvent)

The CO_2 removal using MDEA as a solvent is presented in Fig. 3.

A stream containing CO_2 , O_2 , N_2 , Ar, and vapour H_2O resulted from a steel plant producing 576 t h^{-1} of steel is considered as input to the CO_2 capture process⁴⁵, with equipment sizing calculated accordingly. Captured carbon dioxide was the main

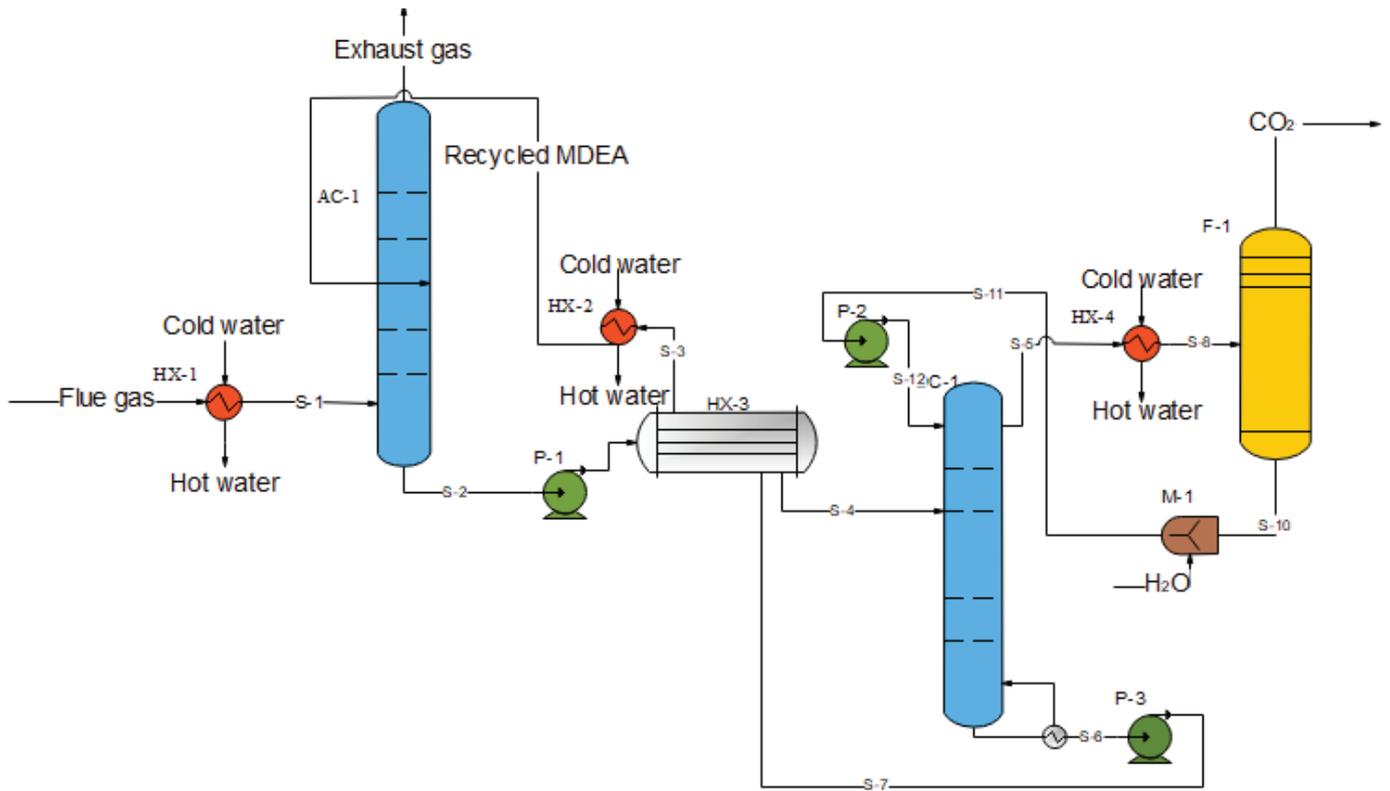


Fig. 3 – Schematic representation of CO₂ capture using MDEA

product of this process, while the by-product represented by an exhaust gas stream containing mainly water vapour, O₂, N₂, Ar, and CO₂ traces. Electrolyte thermodynamic model (E-NRTL) was used for Case 1 simulation, as implemented within ChemCAD. The absorption-desorption cycle of CO₂ takes place within absorption column (AC-1) and desorption column (DC-1) using a MDEA solution (50 wt.%). The regeneration of the MDEA solution occurs after the desorption column, when is cooled to 40 °C using two heat exchangers (HX-3 and HX-2) and recycled into AC-1. The stream coming from the steel mill is also cooled down, up to 40 °C, using the HX-1 unit. After the absorption of CO₂ in AC-1, the remaining gases (i.e., clean/exhaust flue gases) are released into the atmosphere. It is assumed that AC-1 has 11 stages and the inlet temperature is 40 °C. The CO₂-rich solution obtained from the bottom of AC-1 is transported to the desorption column using a pump (P-1) with an efficiency of 85 %. The DC-1 column has 10 stages, the inlet temperature being 105 °C. The CO₂ stream is released from the top of DC-1 column along with low quantities of water vapour. This carbon dioxide-water mixture is sent to the HX-4 heat exchanger, followed by a gas-liquid separator (F-1). CO₂ is obtained as a top product, while water is mixed with a make-up stream and recycled back to the desorption column (DC-1).

Description of Case 2: CO₂ capture using polymeric membranes

CO₂ capture using polymeric membranes was also investigated as a valuable technology. The simulation consists of two-stage co-current spiral wound membranes connected in series, as shown in Fig. 4. The co-current membrane configuration was chosen because it achieves the best separation performance⁴⁶. The permeances for the main compounds are summarised in Table 3, where the input and output streams are reported. The CO₂ capture plant's input stream is the same as in Case 1⁴⁵, but the ChemCAD thermodynamic model Ideal Vapour Pressure was used for this simulation. Cleaned CO₂ is the main product of the plant, with the by-product of a gas stream containing O₂, N₂, Ar, H₂O, and CO₂ traces. The gas stream coming from the steel plant is cooled to 50 °C using a heat exchanger (HX-1), and transported to the spiral wound membrane (MEMB-1) using a compressor (C-1). A percentage of 30 % of the permeate stream is recycled back to the first membrane as sweep, using a splitter (DIV-1). The gas mixture is cooled using three heat exchangers (HX-3, HX-4 and HX-5) with an output temperature of 35 °C. The first permeate stream reaches the MEMB-2 membrane, and it is assumed that 5 % of the permeate stream from the second membrane stage is recycled. Permeate-2 flow is further compressed in C-5, C-6, and C-7 compressors

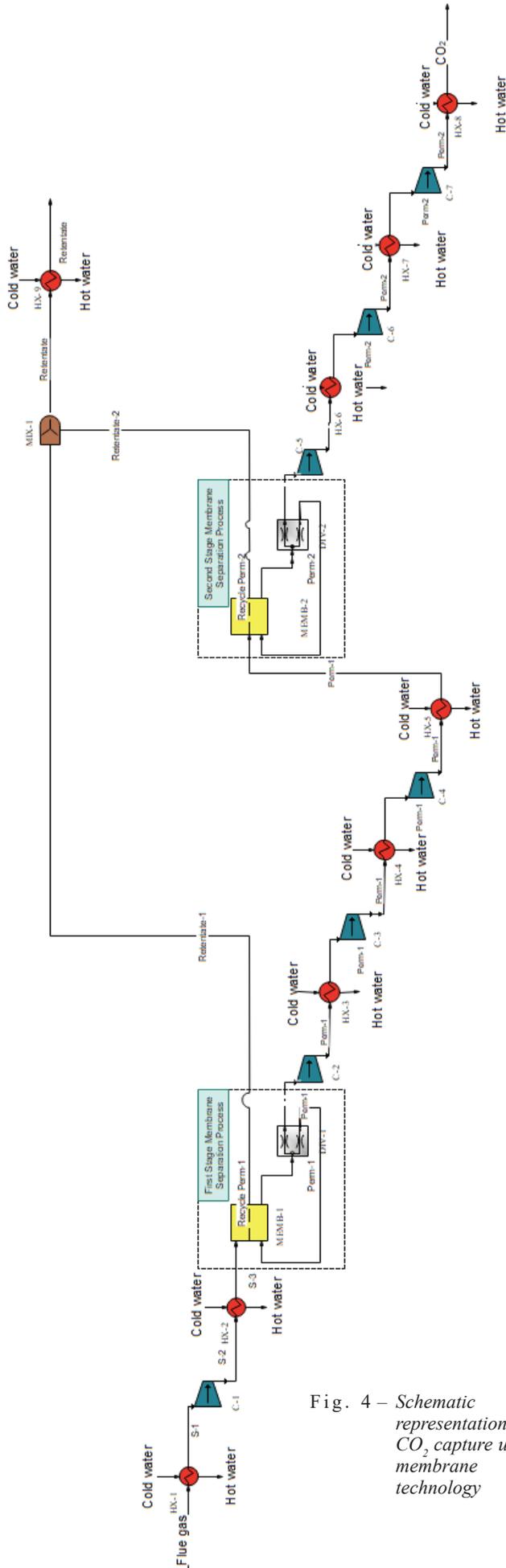


Fig. 4 – Schematic representation of CO₂ capture using membrane technology

Table 3 – Membranes main assumptions⁴⁶

System	Assumptions	Values
	CO ₂ permeance	1
	N ₂ permeance	5 · 10 ⁻³ m ³ (STP)
	O ₂ permeance	2 · 10 ⁻² bar h m ²
	H ₂ O permeance	2.25 · 10 ⁻⁹
Membranes for CO ₂ separation	1 st membrane area	22,800 m ²
	2 nd membrane area	10,000
	Pressure ratio for 1 st membrane	100
	Pressure ratio for 2 nd membrane	80

(with 90 % efficiency) and is cooled using three coolers (HX-6, HX-7 and HX-8) with an output temperature of 60 °C. The retentate streams from both membranes are combined using a mixer, and cooled to 68 °C using an additional heat-exchanger (HX-9). The main assumptions adopted for the process modelling are reported in Table 3.

Description of Case 3: CO₂ capture using gas-liquid absorption (NaOH)

The inlet stream flow (23,353 kg h⁻¹), composition (CO₂, O₂, N₂, Ar, and steam) and conditions (1500 °C, 1 bar) are equivalent to those used in Cases 1 and 2⁴⁵. The main product is sodium bicarbonate (NaHCO₃), while the undesired by-products are sodium carbonate (Na₂CO₃), and exhaust gas stream containing mainly water vapour, O₂, N₂, Ar, and CO₂ traces. The reactions occurring in the reacting environment are described within the thermodynamics analysis section. The absorption and conversion of CO₂ takes place within two absorption columns in series (AC-1 and AC-2) working at different pH using a NaOH solution (10 wt.%). According to Shim *et al.*⁴⁷, when pH is less than 7, carbonic acid is the most abundant species, and as soon as the pH rises above 10, the carbonate ion takes over. In between, with nearly a complete conversion at pH equivalent to 8.5, the bicarbonate ion is energetically preferred. The aqueous solution at the bottom of AC-1 must therefore have a pH as close as possible to 8.5 pH in order to produce a pure sodium bicarbonate crystal after the crystallisation process. The inlet stream is cooled to 650 °C in HX-1 before flowing into a bubble reactor, where the reaction starts to occur. The bubble reactor serves as the bottom equilibrium stage of the absorbing column AC-1, where the aqueous solution

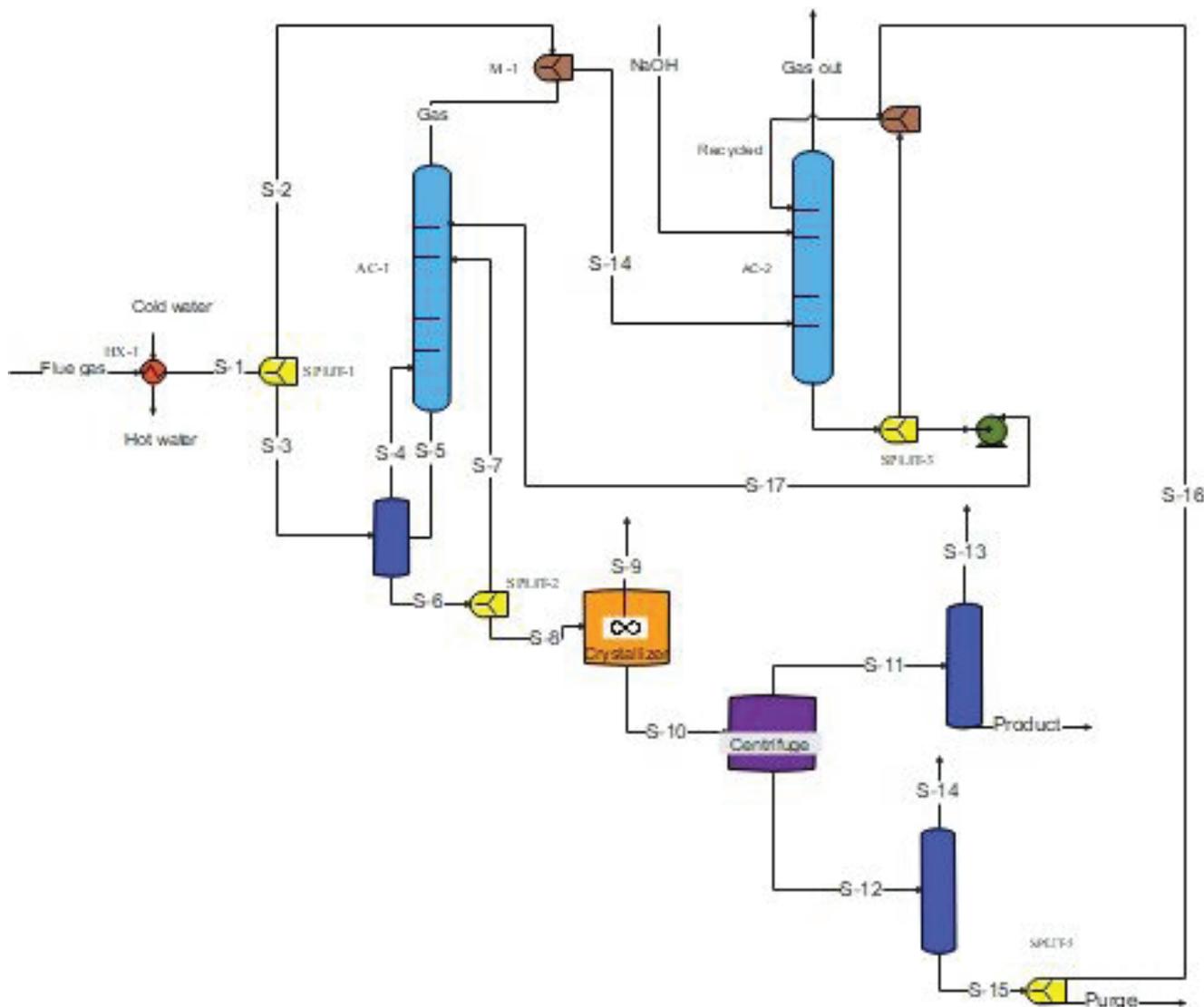


Fig. 5 – Schematic representation of CO_2 capture using NaOH

of sodium carbonate coming from AC-2 flows from the top. After the first absorption, the flue gas with a residual amount of CO_2 flows into AC-2's bottom, where the stream of sodium hydroxide solution is introduced at the top. Thereafter, CO_2 -free outlet gas stream is discharged into the atmosphere. The bicarbonate-rich solution is cooled from $80\text{ }^\circ\text{C}$ to $25\text{ }^\circ\text{C}$ to promote crystallisation, reducing the solubility of sodium bicarbonate. The precipitate is then separated through centrifugation, followed by a drying step. To remove the excess water present, the mother liquor is subsequently put through a flash separation step under vacuum. During evaporation, a fraction of carbonate and bicarbonate ions reacts to produce CO_2 following reactions (14) and (15). Since the battery of columns had almost completely absorbed the inlet CO_2 , the carbon dioxide emissions produced at this stage are the largest of the entire capture process. Nevertheless, the overall process capture efficiency is above 90 %. In order

to avoid losing reactants, the concentrated sodium/carbonate/bicarbonate ion solution is recycled within AC-2, which lowers the amount of soda needed for the total absorption of CO_2 .

Life cycle assessment

LCA is a methodology to comprehensively evaluate potential impacts along the life cycle of products and services throughout their life cycle. Standard LCA methodology^{48,49} follows four steps: (i) goal and scope definition, (ii) life cycle inventory (LCI), (iii) life cycle impact assessment (LCIA), and (iv) interpretation. The LCA should take into account every stage of the product's life cycle, including the extraction of raw materials, manufacturing of the product, its distribution, use, and end of life, which typically includes recycling, landfilling, or incineration. A schematic representation of LCA steps is given in Fig. 6.

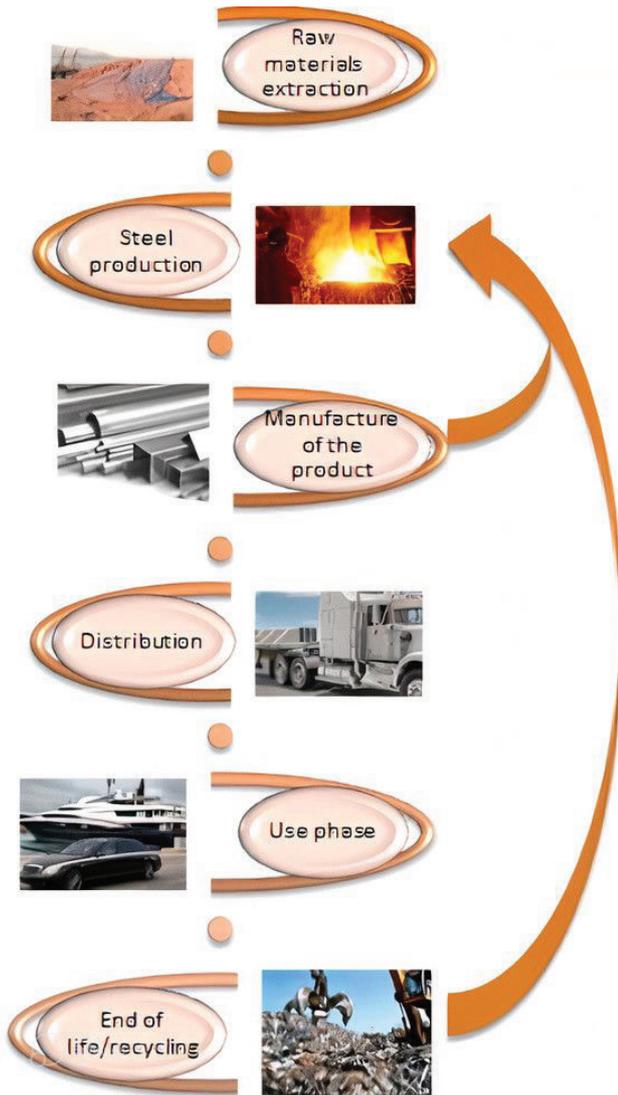


Fig. 6 – LCA steps for steel

The goal of the present study was to compare the environmental impacts of steel production coupled with various CO₂ capture technologies (using MDEA, membranes, and NaOH). The LCA results should identify the technology with the lowest impact on the environment. In the current investigation, one tonne of steel was used as the functional unit.

The system boundary is shown in Fig. 7, including: (i) upstream processes: steel raw material acquisition, O₂ production process, MDEA, membrane, and NaOH production, (ii) core processes: steel production, CO₂ capture, CO₂ transport and storage (for Cases 1 and 2).

The main LCA assumptions for MDEA production, membrane production, CO₂ transport and storage, as well as the electricity production, are reported by Luca and Petrescu⁵⁰. For NaOH production, it was assumed that sodium hydroxide is obtained through sodium chloride electrolysis. The most common processes for NaCl electrolysis are based on different separation technologies, such as membrane, amalgam and diaphragm-based separations. The dataset used in the present LCA study (GaBi) is based on the diaphragm route, where sodium chloride solution is led through the anode chamber through the diaphragm into the cathode chamber. The overall energy consumption is higher compared to the mercury cell process⁵¹. Since the absorption using NaOH provide a valuable by-product other than steel, the system expansion approach was employed to include the additional functions related to the co-product⁴⁹. In fact, synthesising sodium bicarbonate by CO₂ recovery allows for the production of less sodium bicarbonate using the

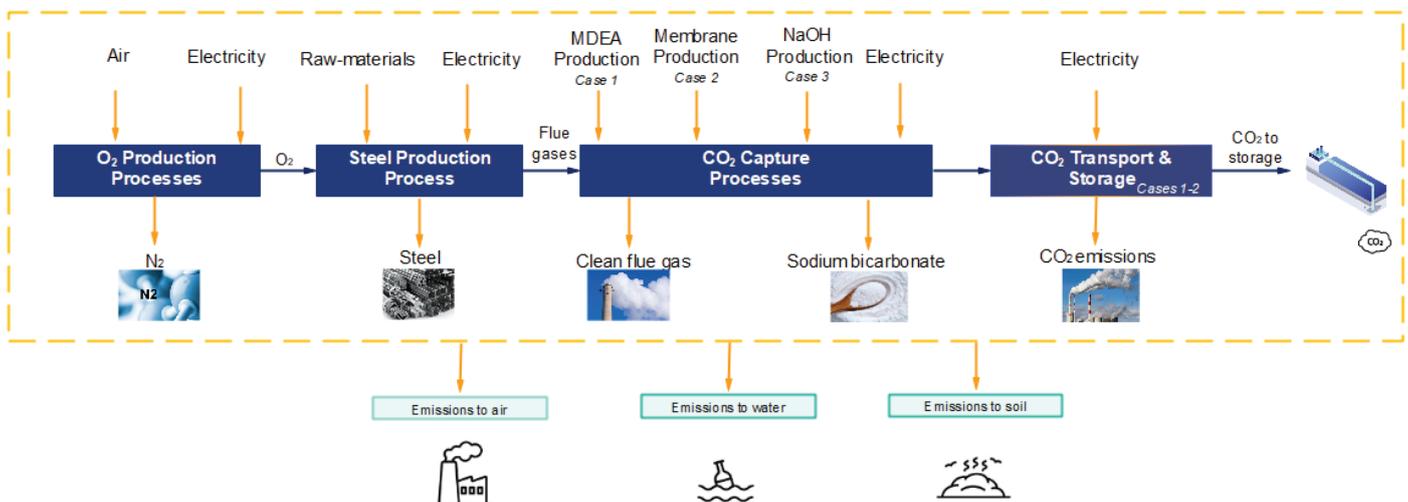


Fig. 7 – LCA boundary conditions

conventional Solvay process. In the Solvay process, ammonia is absorbed into sodium chloride brine, and the ammoniated brine reacts with carbon dioxide leading to NaHCO_3 and ammonium chloride (NH_4Cl). The carbon dioxide is introduced into the carbonation step. Ammonia is re-obtained from the mother liquid by treatment with quick lime according to the following chemical reaction:



Ammonia is reintroduced into the production process after being regenerated through a process that produces an additional amount of carbon dioxide. The by-product, calcium chloride, is disposed of into sea water with the effluent stream⁵¹.

The present LCA study follows a cradle-to-gate perspective, meaning that all the life cycle stages, from the raw-material extraction/production to the production of one tonne of steel were included, as shown in Table 4.

The LCI data used in the current research were obtained from process simulations performed using Aspen Plus and ChemCAD. Secondary data were gathered from the scientific literature or within GaBi database, while using the GaBi software v10 for calculations. Table 5 presents the main LCI data.

LCIA was carried out applying the ReCiPe Midpoint (H) 2016 method to calculate midpoint impact categories relevant for the cases under investigation. The impact categories considered are schematically presented in Fig. 8. ReCiPe was chosen since is one of the most recent and updated impact assessment methods available to LCA practitioners.

Table 4 – Sub-processes considered in the LCA

Processes considered	Case 1	Case 2	Case 3
Steel making	Yes	Yes	Yes
O ₂ production using ASU	Yes	Yes	Yes
MDEA production & transportation	Yes	No	No
Membrane production & transportation	No	Yes	No
NaOH production & transportation	No	No	Yes
CO ₂ capture using MDEA	Yes	No	No
CO ₂ capture using membranes	No	Yes	No
CO ₂ capture using NaOH	No	No	Yes
CO ₂ transport and storage	Yes	Yes	No
MDEA degradation	Yes	No	No
NaHCO ₃ production	No	No	Yes

Table 5 – LCI for Cases 1 – 3 for the production of 1 ton of steel

Inputs	Units	Case 1 (CCS with MDEA)	Case 2 (CCS with membrane)	Case 3 (CCS with NaOH)
To ASU				
Air	kg/t _{steel}	318.86	318.86	318.86
To steel plant				
Purchased scrap	kg/t _{steel}	116.92	116.92	116.92
Own scrap	kg/t _{steel}	73.07	73.07	73.07
Oxygen	kg/t _{steel}	73.70	73.70	73.70
Nitrogen	kg/t _{steel}	0.38	0.38	0.38
Ar	kg/t _{steel}	0.77	0.77	0.77
Lime	kg/t _{steel}	64.83	64.83	64.83
Iron ore	kg/t _{steel}	5.08	5.08	5.08
Dolomite	kg/t _{steel}	11.02	11.02	11.02
Desulph. hot metal	kg/t _{steel}	900.87	900.87	900.87
To CO₂ capture units				
Flue gas	kg/t _{steel}	37.99	37.99	37.99
MDEA	kg/t _{steel}	0.05	–	–
PSf	kg/t _{steel}	–	0.17	–
PVAm	kg/t _{steel}	–	1.15·10 ⁻⁵	–
NaOH	kg/t _{steel}	–	–	9.89
Outputs				
From ASU				
N ₂	kg/t _{steel}	240.95	240.95	240.95
O ₂	kg/t _{steel}	73.67	73.67	73.67
Ar	kg/t _{steel}	4.04	4.04	4.04
Water	kg/t _{steel}	0.18	0.18	0.18
From membrane				
O ₂ stream	kg/t _{steel}	–	96.58	–
Retentate 1	kg/t _{steel}	–	637.37	–
Retentate 2	kg/t _{steel}	–	232.20	–
From steelmaking plant				
Steel	kg/t _{steel}	1	1	1
Slag	kg/t _{steel}	114.23	114.23	114.23
Sludge	kg/t _{steel}	39.13	39.13	39.13
From CO₂ capture units				
CO ₂ emissions	kg/t _{steel}	0.018	0.019	–
CO ₂ stored	kg/t _{steel}	10.63	11.12	–
NaHCO ₃	kg/t _{steel}	–	–	20.05

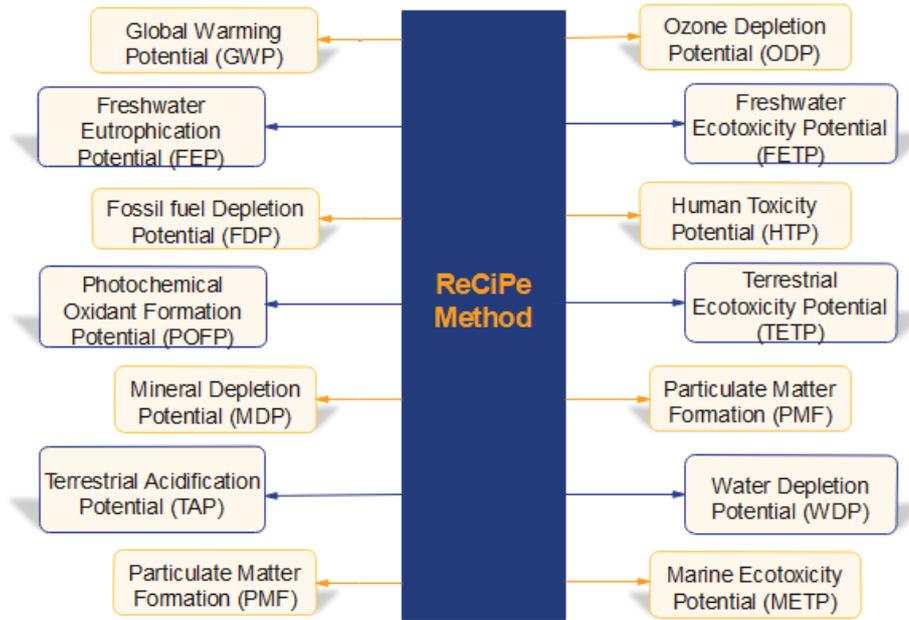


Fig. 8 – Impact categories according to ReCiPe method

Results and discussions

The primary inputs and outputs from process modelling and simulation for the three scenarios under consideration are summarised in Table 6, and were validated using data from the scientific literature for MDEA⁵², membranes⁴⁶, and sodium hydroxide⁴⁷.

It can be noticed that, starting from the same mass flow-rate (i.e., 23,352.82 kg h⁻¹) and composition of flue gas, a variable amount of CO₂ was removed in the three cases. From the initial flow-rate of 6,825.51 kgCO₂ h⁻¹, a flow-rate of 6,140.71 kgCO₂ h⁻¹ was removed for Case 1 (capture efficiency of 90 %), 6,490.64 kgCO₂ h⁻¹ was removed in Case 2 (capture efficiency of 95 %), while in

Table 6 – Material balance of the CO₂ capture plants using MDEA (Case 1), membranes (Case 2), and NaOH (Case 3)

Stream	Unit of measure	Case 1			Case 2			Case 3					
		Input	Output		Input	Output		Input	Output				
		Flue gas	Exhaust gas	Carbon dioxide	Flue gas	Retentate	Carbon dioxide	Flue gas	NaOH	Gasout	Product	S-14	Purge
Temperature	°C	1,500	40	30	1,500	60	60	1,500	25	77.5	25	81	
Pressure	bar	1	1	3.9	1	1.95	1	1	1.1	1	1	0.36	
Vapor fraction	–	1	1	1	1	1	1	1	0	1	0	1	
Components mass flow													
H ₂ O		210.96	636.84	27.56	210.96	210.96	–	210.96	51,300	5,418.06	–	45,829.32	306.53
Ar		264.05	264.05	–	264.05	264.05	–	264.05	–	264.04	–	0.01	–
N ₂	kg h ⁻¹	13,271.91	11,816.86	3.64	13,271.91	13,203.72	68.19	13,271.91	–	11,820.33	–	0.24	–
CO ₂		6,825.51	–	6,140.71	6,825.51	334.87	6,490.64	6,825.51	–	13.54	–	645.05	–
O ₂		2,780.39	2,778.79	1.59	2,780.39	2,629.39	151	2,780.39	–	2,780.28	–	0.10	–
NaOH _(l)		–	–	–	–	–	–	–	5,700	–	–	–	196.80
CO ₃ ²⁻ _(l)		–	–	–	–	–	–	–	–	–	–	–	143.05
HCO ₃ ⁻ _(l)		–	–	–	–	–	–	–	–	–	–	–	9.31
NaHCO _{3(s)}		–	–	–	–	–	–	–	–	–	11,558.43	–	–
Total mass flow	kg h ⁻¹	23,352.82	15,596.54	6,173.50	23,352.82	16,642.99	6,709.83	23,352.82	57,000	20,296.26	11,558.43	46,474.71	572.04

Case 3, a mass flow of 6,166.92 kgCO₂ h⁻¹ was converted to NaHCO₃ (capture efficiency of 90 %), leading to 11558.43 kg h⁻¹ of NaHCO₃.

The process simulations results were used to calculate the LCIA outcomes for the cases under investigation, which are reported in Table 7.

The impacts of Cases 1 and 2 are fairly similar, with Case 1's impacts related to MDEA adoption being a little less severe. The environmental performance for the CO₂ capture using NaOH are very high compared to the other two capture technologies investigated (gas-liquid absorption using MDEA and membrane CO₂ capture). The system expansion method resulted in negative impacts for

various environmental categories, where the impacts of producing sodium bicarbonate via the Solvay process were deducted from the impacts of producing steel with CCS. The impacts associated with the Solvay process are, in fact, often substantially larger than those produced using NaOH to convert CO₂ to sodium bicarbonate, indicating that there is a net advantage from utilising the second pathway, as shown in Table 8. For instance, in the case of the Global Warming Potential (GWP) indicator, the distribution of various sub-processes to the total GWP is illustrated in Fig. 9. It can be noticed that for Case 3, the production of NaOH via electrolysis accounted for around 89 % of the total GWP value

Table 7 – LCIA results according to ReCiPe impact assessment method

	Units of measure	Case 0 (no CCS)	Case 1 (CCS with MDEA)	Case 2 (CCS with membrane)	Case 3 (CCS with NaOH)
GWP	kg CO ₂ eq./t _{steel}	12.30	1.79	1.42	-5.79
FEP·10 ⁶	kg P eq./t _{steel}	8.09	9.01	9.63	27.90
FDP	kg oil eq./t _{steel}	0.18	0.23	0.36	-1.97
FETP·10 ⁴	kg 1,4-DB eq./t _{steel}	8.55	9.45	9.63	12.82
HTP·10 ²	kg 1,4-DB eq./t _{steel}	16.60	16.80	17.4	32.00
MDP	kg Fe eq./t _{steel}	3.32	3.33	3.33	3.36
POFP·10 ³	kg NMVOC/t _{steel}	2.18	2.35	2.84	-41.10
TETP·10 ⁵	kg 1,4-DB eq./t _{steel}	4.71	4.94	5.25	24.71
WDP·10 ³	m ³ /t _{steel}	33.60	36.7	40.00	-30.00
TAP·10 ⁴	kg SO ₂ eq./t _{steel}	20.60	67.00	52.50	-858.00
METP·10 ⁴	kg N eq./t _{steel}	7.58	9.81	9.55	-211.00
PMF·10 ³	kg PM10 eq./t _{steel}	6.27	6.90	6.47	-21.64

Table 8 – LCIA results for Case 3, with and without system expansion

	Units of measure	Case 3 without system expansion	NaHCO ₃ production via Solvay	Case 3 with system expansion
GWP	kg CO ₂ eq./t _{steel}	14.32	20.11	-5.79
FEP·10 ⁶	kg P eq./t _{steel}	50.39	22.50	27.90
FDP	kg oil eq./t _{steel}	4.24	6.21	-1.97
FETP·10 ⁴	kg 1,4-DB eq./t _{steel}	31.10	18.28	12.82
HTP·10 ²	kg 1,4-DB eq./t _{steel}	56.90	24.90	32.00
MDP	kg Fe eq./t _{steel}	3.41	0.05	3.36
POFP·10 ³	kg NMVOC/t _{steel}	28.80	69.90	-41.10
TETP·10 ⁵	kg 1,4-DB eq./t _{steel}	53.40	28.69	24.71
WDP·10 ³	m ³ /t _{steel}	224.00	254.00	-30.00
TAP·10 ⁴	kg SO ₂ eq./t _{steel}	262.00	1120.00	-858.00
METP·10 ⁴	kg N eq./t _{steel}	116.00	327.00	-211.00
PMF·10 ³	kg PM10 eq./t _{steel}	9.16	30.80	-21.64

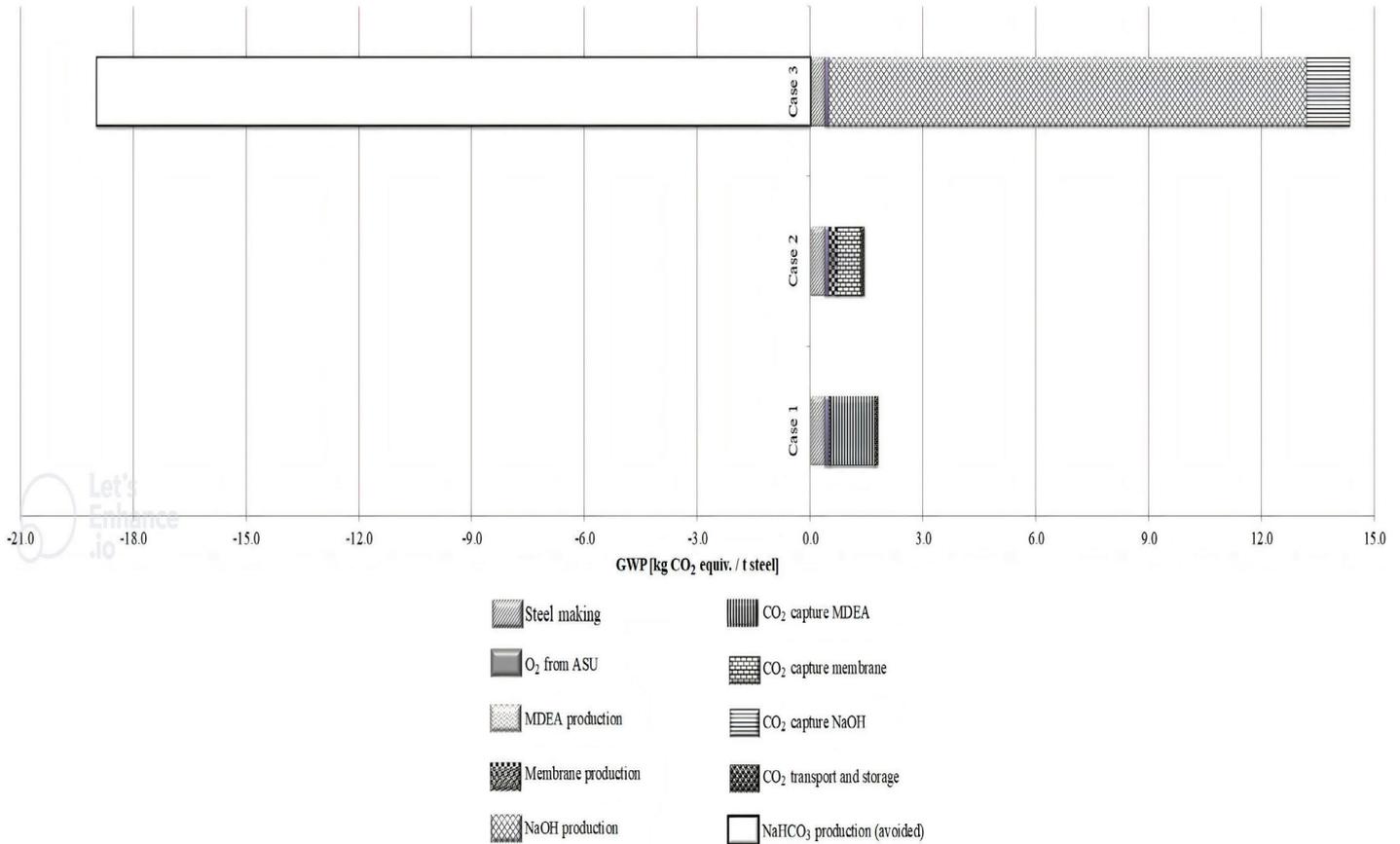


Fig. 9 – GWP distribution for Case 1, Case 2, and Case 3

Table 9 – LCIA results for Case 1*, 2*, and 3

	Units of measure	Case 1*	Case 2*	Case 3
GWP	kg CO ₂ eq./t _{steel}	-5.11	-5.17	-5.79
FEP·10 ⁶	kg P eq./t _{steel}	28.13	32.20	27.90
FDP	kg oil eq./t _{steel}	-1.77	-1.72	-1.97
FETP·10 ⁴	kg 1,4-DB eq./t _{steel}	13.35	14.32	12.82
HTP·10 ²	kg 1,4-DB eq./t _{steel}	32.40	33.80	32.00
MDP	kg Fe eq./t _{steel}	3.36	3.37	3.36
POFP·10 ³	kg NMVOC/t _{steel}	-38.60	-40.00	-41.10
TETP·10 ⁵	kg 1,4-DB eq./t _{steel}	25.23	26.51	24.71
WDP·10 ³	m ³ /t _{steel}	-22.00	47.00	-30.00
TAP·10 ⁴	kg SO ₂ eq./t _{steel}	-788.00	-847.00	-858.00
METP·10 ⁴	kg N eq./t _{steel}	-198.00	-315.00	-211.00
PMF·10 ³	kg PM10 eq./t _{steel}	-20.06	-21.28	-21.64

(i.e., 12.70 kg CO₂ eq./t_{steel} from the total 14.32 kg CO₂ eq./t_{steel}), which is counterbalanced by the Solvay process that does not employ NaOH. However, there are still several impact categories, i.e., Freshwater Eutrophication Potential (FEP), Freshwater Ecotoxicity Potential (FETP), Human Toxicity

Potential (HTP) and Terrestrial Ecotoxicity Potential (TETP), which display higher impacts for the NaOH absorption.

Due to the unfairness of the comparison of carbon capture alternatives, Case 1 and Case 2 were amended to include the use of CO₂ for the synthesis

of sodium bicarbonate with NaOH rather than transporting and storing the carbon dioxide. The environmental indicators changed as a result of this new hypothesis, and Table 9 summarises the new indicators that relate to Cases 1*, 2*, and 3, applying the system expansion approach to each carbon capture technology. The quantities of NaHCO₃ obtained in Case 1* and Case 2* were 20.34 kg h⁻¹ and 21.24 kg h⁻¹, respectively, for each tonne of steel produced. A different amount of NaHCO₃ results from a variable quantity of CO₂ captured using MDEA and membranes.

The results in Table 9 can be analysed to draw the following conclusions: (i) the environmental indicators have very similar values, meaning that the environmental performances are roughly equivalent; and (ii) the environmental burdens associated with capturing CO₂ and then reacting with NaOH are always slightly higher than those associated with reacting directly during absorption. The only exception is related to the water depletion potential (WDP), which highlighted a higher usage of water when following the membrane pathway.

Conclusions

A comparison among the life cycle emissions of three different carbon capture pathways was carried out in order to identify the most promising technology for the emissions reduction in the iron and steel sector. After a thermodynamic description of the chemical equilibria and the reaction kinetics involved, several in-silico models for the absorption of carbon dioxide employing MDEA, membranes, or sodium hydroxide to produce sodium bicarbonate were developed. Process simulations allowed for obtaining the material and energy balances required for the generation of the life cycle inventories of each alternative option. The results demonstrated that the direct conversion of CO₂ to sodium bicarbonate, which creates a useful by-product while avoiding the emissions associated with its traditional synthesis via the Solvay process, was the most efficient technique for reducing emissions in the steel industry. The environmental performance of Cases 1 and 2 significantly improved when the CO₂ captured by them was converted to sodium bicarbonate, approaching the scores of Case 3, which was still the most environmentally friendly. As a result, since carbon capture continues to pose significant challenges for industrial-scale applications, the use of a safe, simple-to-implement, and low-cost technique such as NaOH absorption, may be beneficial in advancing this industry towards a more sustainable development.

Abbreviations list

AC	– adsorption column
ASME	– <i>American Society of Mechanical Engineers</i>
ASU	– <i>air separation unit</i>
BF	– blast furnace
BOF	– basic oxygen furnace
BOS	– basic oxygen steelmaking
C	– compressor
CCS	– carbon capture and storage
CCUS	– carbon capture utilisation and storage
CLC	– chemical looping combustion
COVID-19	– coronavirus disease 2019
DAC	– direct air capture
DB	– dichlorobenzene
DC	– desorption column
DIV	– divider
EAF	– electric arc furnace
E-NRTL	– electrolyte thermodynamic model
ELECNRTL	– ELECTrolyte Non-Random Two-Liquid
EoS	– equation of state
eq.	– equivalents
F	– flash (gas-liquid separator)
FDP	– fossil depletion potential
FEP	– freshwater eutrophication potential
FETP	– freshwater ecotoxicity potential
Gt	– giga tonne
GWP	– global warming potential
HPC	– hot potassium carbonate
HTP	– human toxicity potential
HX	– heat exchanger
IEA	– International Energy Agency
IEAGHG	– International Energy Agency Greenhouse Gas Research & Development Programme
IPCC	– Intergovernmental Panel on Climate Change
l	– liquid
LCA	– life cycle assessment
LCI	– life cycle inventory
LCIA	– life cycle impact assessment
MEMB	– membrane
METP	– marine ecotoxicity potential
MDEA	– methyl diethanol amine
MDP	– metal depletion potential
MEA	– monoethanolamine
MIX	– mixer
NMVOC	– non-methane volatile organic compounds

OBF	– oxygen blast furnace
P	– pump
Perm	– permeate
POFP	– photochemical oxidant formation potential
PMF	– particulate matter formation
PR	– Peng Robinson
PR-BM	– Boston-Mathias modifications
PSA	– pressure swing adsorption
PSf	– polysulfone
PVAm	– polyvinyl amine
s	– solid
S	– stream
SEWGS	– sorption enhanced water-gas shift
Split	– splitter
STP	– standard temperature and pressure
TAP	– terrestrial acidification potential
TETP	– terrestrial ecotoxicity potential
VPSA	– vacuum pressure swing adsorption
WDP	– water depletion potential
y	– year

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