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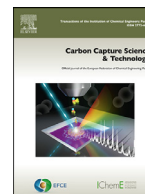
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Full Length Article

A technical and environmental comparison of novel silica PEI adsorbent-based and conventional MEA-based CO₂ capture technologies in the selected cement plant



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ABSTRACT

The cement industry accounts for almost 7 % of anthropogenic carbon dioxide emissions globally. Therefore, it is imperative to identify innovative solutions to mitigate carbon dioxide emissions from the cement industry.

This study aims to evaluate and compare the technical and environmental aspects of integrating two post-combustion carbon capture processes (CCS) into a cement plant: the conventional monoethanolamine (MEA)-based CCS process and the novel silica-alkoxyated polyethyleneimine (SPEI)-based CCS process. Three scenarios were considered: (i) a reference cement plant without CCS, (ii) the conventional MEA-based CCS system integrated into a cement plant and (iii) the novel SPEI-based CCS system integrated into a cement plant. The technical evaluation results showed that the regeneration energy requirements for the conventional MEA and novel SPEI-based CCS processes were 3.53 GJ/tonne CO₂ and 2.36 GJ/tonne CO₂, respectively, to achieve a capture rate of 90 %. However, the performance of MEA-based carbon capture processes can be improved by using advanced amine formulations that offer lower regeneration heat requirements at 3.3 GJ/tonne CO₂, although this is still higher than the SPEI-based carbon capture processes.

The novel SPEI-based CCS process showed superior environmental performance compared to the conventional MEA-based CCS process. The endpoint single score was conducted which showed that the SPEI-based CCS process had a lower impact on human health, ecosystems, and resources (7 %, 9 %, and 26 % lower, respectively) compared to the MEA-based CCS process.

1. Introduction

Certain human activities are causing an increase in greenhouse gas emissions (GHGs), which causes global warming and climate change. One of the major reasons for the increase in GHGs is industrialization (Jaffar et al., 2019; Avagyan, 2021). Several policies have been implemented to reduce GHGs to curb climate change (Commission, 2012). The main goal of these policies is to reduce carbon dioxide (CO₂) emissions to 40 % by 2030 and up to 95 % by 2050 in comparison to the 1990 level. According to International Environmental Agency (IEA), the cement industry is one of the main sources of anthropogenic CO₂ emissions that accounts for about 7 % of global emissions (IEA, 2020).

Despite higher CO₂ emissions from the process, the demand for cement-related goods and products are continuously increasing due to

the growth of industrial and economic sectors (Pacheco-Torgal et al., 2014; Alex et al., 2016). The United States Geological Survey has stated that cement production reached 4.1 tera kg in 2020 and is expected to increase to 6 tera kg by 2050 (USGS, 2021; Wojtach-Rychter et al., 2021). Additionally, according to the European Cement Association (CEMBUREAU), global cement production increased from 3.6 billion tons in 2012 to 4.17 billion tons in 2020 (Baeza et al., 2013; CEMBUREAU, 2021).

One of the main components of cement is clinker, which is produced by calcining limestone to produce calcium oxide (CaO) and then sintering of CaO with aluminosilicate and other raw materials. Almost 50 % of the total CO₂ emissions from cement plants are linked to the calcination of limestone, while 40 % of the emissions are produced by burning fuel in the rotary kiln and calciner. The electricity requirement accounts for

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almost 5 % of the emissions, and the remaining 5 % of CO₂ emissions are linked to transportation (Summerbell et al., 2017). It has been reported that each kilogram of cement generates approximately 0.5–0.7 kg of CO₂ emissions, depending on the production technology and clinker factor (USGS, 2021; Wojtacha-Rychter et al., 2021).

Strategies such as the usage of alternative fuels, improving electrical and thermal efficiency, substituting clinker with supplementary cementing materials, and carbon capture and storage (CCS) are being developed to reduce emissions from the cement industry (Galusnyak et al., 2022). The first three strategies can only partially eliminate the CO₂ emissions from the cement industry; therefore, the implementation of CCS provides a promising pathway to reduce CO₂ emissions from the cement industry (Xu et al., 2016). According to the IEA, it is expected that the implementation of CCS will contribute approximately 55 % to the reduction of GHG emissions (IEAGHG, 2008).

Currently, there are three carbon capture processes available: (i) pre-combustion CCS process, (ii) post-combustion CCS process, and (iii) oxy-fuel combustion process (Cormos et al., 2017). As previously mentioned, most of the CO₂ emissions in the cement industry are due to the calcination of limestone, therefore, pre-combustion CO₂ capture is not a viable approach (IEAGHG, 2008). In the case of the oxyfuel combustion process, modifications to the design of the rotary kiln and pre-calciner are necessary, making it difficult to retrofit existing cement plants. In contrast, the post-combustion CCS process is the most feasible option for the cement industry. It not only reduces emissions from the cement industry but, it can also be retrofitted to existing cement plants without requiring any design modifications (Hong, 2022; García-Gusano et al., 2015).

The most common method adopted for the post-combustion CCS process is through the chemical absorption process, with first-generation liquid amine-based solvents, such as monoethanolamine (MEA). MEA is mostly used due to its high capture rate and selectivity at low partial pressures. However, the integration of the MEA-based capture process has certain disadvantages, including high energy requirements for regeneration, the potential for equipment corrosion, and significant losses due to oxidative degradation and evaporation (Peu et al., 2023). Therefore, researchers are working towards developing second and third-generation solid sorbents for their application in the post-combustion CCS process.

Solid sorbent-based processes have several advantages over liquid amine-based processes, such as low regeneration energy requirements, reduced sorbent losses, and a lower potential for equipment corrosion. The solid sorbents can be either chemisorbents, including metal-organic frameworks, amine-functionalized mesoporous silica, and alkaline-based sorbents, or physisorbents, such as activated carbon and zeolites etc. (Bonenfant et al., 2008; Lu et al., 2008; Su et al., 2010; Belmabkhout and Sayari, 2009; Zhao et al., 2022; Khosravi et al., 2022). Compared with the other solid sorbents, various amine-based solid adsorbents have shown promising results because of their lower heat of adsorption and high dynamic sorption capacity (Kim et al., 2021). Among the amine-based sorbents, polyethyleneimine-based (PEI) solid sorbents have been widely developed. The sorbent based on PEI is essentially composed of a silica support material that has been impregnated with PEI and utilized in a circulation fluidized bed or bubbling fluidized bed reactor for CO₂ capture (Harlick et al., 2006; Sayari et al., 2016; Zhang et al., 2020, 2014b).

A number of life cycle assessment (LCA) studies have been performed to study the environmental impact. Stafford et al. (2016a) performed the LCA of a cement plant and investigated the use of waste in partial replacement of fossil fuels. They reported that the major contributor in all impact categories are the emissions from the rotary kiln. In another study, Stafford et al. (2016b) considered the Brazilian cement industry and reported that the emissions related to transportation have the maximum emissions, followed by burning fossil fuels in the kiln. Georgiopolou and Lyberatos (2018) investigated the influence of using refused derived fuel (RDF), waste tires, and biological

sludge as alternative fuels. They presented that the RDF fuel has a minimal environmental effect followed by waste tires, while the biological sludge-based fuel resulted in higher emissions. Furthermore, Cankaya and Pekey (2019) studied the influence of alternative fuels and raw materials. They showed that almost a 12 % reduction in total environmental impact can be achieved when alternative fuels and raw materials are employed.

Gracia-Gusaon et al. (2015) studied the influence of integrating an MEA-based post-combustion CCS system with a cement plant. They found that the integration of the post-combustion CCS system showed improvements in global warming, ozone depletion, and abiotic depletion potential. Rolfe et al. (2018) investigated the effect of integrating a CCS system with a cement plant and compared the environmental impact of a calcium looping-based CCS system with oxyfuel combustion. They showed that the integration of a calcium looping carbon capture system had better environmental performance compared to oxyfuel combustion, and the global warming impact was reduced by 89 %. Also, An et al. (2019) compared a MEA-based carbon capture system with the oxyfuel combustion process. They showed that the integration of a CCS system could result in better performance in terms of global warming, eutrophication, acidification, and photochemical ozone formation.

Although considerable attention has been given to the SPEI-based CCS system for optimizing the carbon capture process at the lab scale and pilot scale, however, to the best of our knowledge, no work has been reported on the potential environmental benefits that can be gained through its integration with a cement plant. This research aims to perform a technical and environmental assessment of the novel SPEI-based CCS process integration with a cement plant and compare it with the conventional MEA-based CCS process integration. Various technical and environmental performance indicators are assessed when the two CCS technologies are integrated with a reference cement plant.

This study is based on real-time data obtained from an existing cement plant CEMEX, while the SPEI-based process is simulated based on pilot scale experimental results provided by our partners from the University of Nottingham, UK (Kim et al., 2021; Zhang et al., 2014a). A detailed technical and environmental assessment is conducted through simulation modeling and LCA methodology. This research has the potential to benefit those involved in the cement industry, policymakers, and academics by providing them with deep insights to assess the technical and environmental benefits of integrating CCS in the cement industry, enabling them to make informed decisions.

2. Carbon capture technologies

2.1. Case scenarios and descriptions

The influence of the integration of CCS technologies has been studied through simulation modeling and environmental assessment by considering the following scenarios.

Scenario 1: The reference cement plant without CCS.

Scenario 2: Integration of an MEA-based CCS system with the cement plant.

Scenario 3: Integration of SPEI-based CCS system with the cement plant.

In this study, the CCS process includes the compression and liquefaction of captured CO₂, while the storage scenario of CO₂ is not considered.

2.1.1. Scenario 1: the reference cement plant without CCS

Scenario 1 involves the cement plant without any CCS system, which is assumed as a benchmark. In the cement production process, the raw meal is processed to produce clinker, which is further grounded to produce a powder (cement) with specific physical and chemical properties. The processes involved in clinker production are shown in Fig. 1.

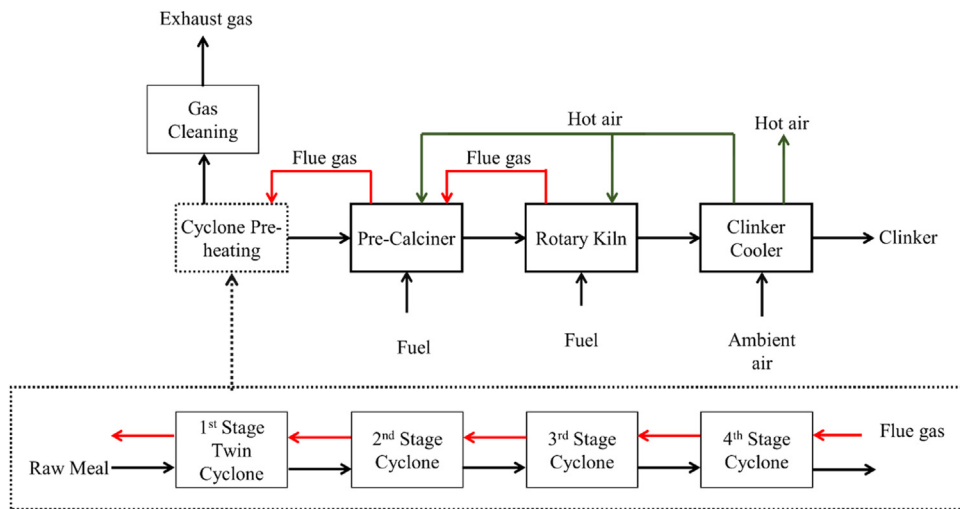


Fig. 1. The reference cement plant without CCS.

During clinker production, the flue gas and raw meal move in counter-current flow. The raw meal is fed into the cyclone preheater, where it is heated, and moisture is removed by the hot flue gas produced from the pre-calciner and rotary kiln. The number of cyclone preheaters employed is dependant on the moisture content of the feedstock. In the studied scenario, four cyclone preheaters are integrated into the cement plant. The raw meal achieves a temperature of 800 °C in the cyclone preheaters before being entered into the pre-calciner.

The reactions involved in the clinker production process are presented in Table 1 (Jaffar et al., 2023). In the pre-calciner, the calcination process takes place and CO₂ is removed from the limestone Eq. (1). After the calcination process, the calcined meal enters the rotary kiln, where sintering takes place at a high temperature of 1400–1500 °C. The reaction that occurs during the sintering process is given in Eqs. (2) to (6). The high temperature in the rotary kiln is achieved by burning coal in the kiln. After sintering in the kiln, the produced clinker undergoes a cooling process, where cold air is blown over the hot clinkers. The hot air is recovered and split into three portions. The secondary and tertiary air are used to heat the rotary kiln and pre-calciner, while the remaining hot air is used to heat the raw meal.

2.1.2. Scenario 2: integration of an MEA-based CCS system with the cement plant

In Scenario 2, the MEA-based CCS system is integrated with the cement plant. The MEA-based system is a single unit that captures CO₂ from the flue gas produced by the cement plant. Fig. 2 presents the schematic of integration of MEA based CCS into the cement plant. The flue gas is extracted directly from the first stage of the cyclone preheater

and cooled to remove the moisture. After the moisture is removed, the flue gas enters the absorption column, where 90 % of the CO₂ is removed using MEA solvent. The CO₂-rich MEA solvent is then directed to the desorption column, where the solvent is regenerated using steam and the CO₂ is released. The released CO₂ is then recovered and liquefied, while the regenerated MEA solvent is transferred back to the absorption column.

In the liquefaction process, the recovered CO₂ is compressed in a four-stages. The CO₂ is initially cooled to a temperature of 25 °C before entering each stage. The compressor operates at an adiabatic efficiency of 85 %. After each intercooler, flash separators are integrated to remove the highly purified liquid CO₂. Through this process, the CO₂ purity level reaches 96 %, which is considered ideal for CO₂ sequestration (Jaffar et al., 2023).

2.1.3. Scenario 3: integration of an SPEI-based CCS system with the cement plant

In Scenario 3, the cement plant is integrated with the SPEI-based CCS system. The SPEI-based system uses a modular system and solid SPEI sorbents to capture CO₂ from the flue gas. Fig. 3 shows the schematic of integration of SPEI-based CCS into the cement plant. SPEI-based CCS system is integrated into modules due to limitations related to the maximum bed diameter required for fluidization. Our calculations have shown that four parallel units are required to capture CO₂ from the flue gas.

Similar to the MEA-based CCS system, the flue gas is extracted directly from the first stage of cyclone preheaters and cooled to remove moisture. After 90 % of moisture removal, the flue gas is directed to the bubbling bed adsorber reactor, where it passes through the solid SPEI sorbent. The solid SPEI sorbent can capture up to 90 % of CO₂ from the flue gas. The CO₂-rich SPEI sorbent is then transferred to the bubbling bed desorber, where the CO₂ is released, and the sorbent is regenerated. The recovered CO₂ is liquefied using the same methodology as for the

Table 1
Calcination and clinker formation reactions.

Reactions	Eqs.
Calcination Reaction (Pre-calciner step)	
$\text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2 \Delta H = +178.2 \text{ KJ/mol}$	Eq. (1)
Clinker Formation Reaction (Rotary kiln step)	
$2\text{CaO} + \text{SiO}_2 \rightarrow \text{Ca}_2\text{SiO}_4 \Delta H = -126.4 \text{ KJ/mol}$	Eq. (2)
Dicalcium silicate (C ₂ S)	
$3\text{CaO} + \text{SiO}_2 \rightarrow \text{Ca}_3\text{SiO}_5 \Delta H = -113.0 \text{ KJ/mol}$	Eq. (3)
Tricalcium silicate (C ₃ S)	
$3\text{CaO} + \text{Al}_2\text{O}_3 \rightarrow \text{Ca}_3\text{Al}_2\text{O}_6 \Delta H = -7.3 \text{ KJ/mol}$	Eq. (4)
Tricalcium aluminate (C ₃ A)	
$\text{Al}_2\text{O}_3 + 4\text{SiO}_2 + \text{H}_2\text{O} \rightarrow \text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2 \Delta H = -80.7 \text{ KJ/mol}$	Eq. (5)
Pyrophyllite (AS ₄ H)	
$\text{Al}_2\text{O}_3 + 2\text{SiO}_2 + \text{H}_2\text{O} \rightarrow \text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 \Delta H = -7.3 \text{ KJ/mol}$	Eq. (6)
Kaolinite (AS ₂ H ₂)	

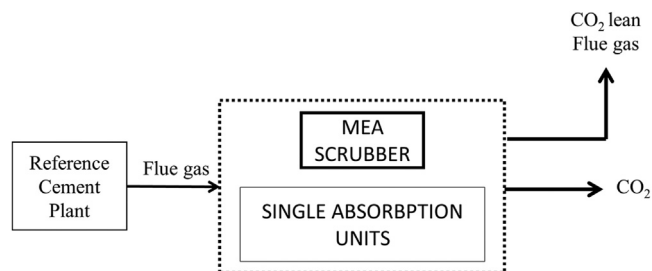


Fig. 2. Schematic diagram of an MEA-based CCS system with the cement plant.

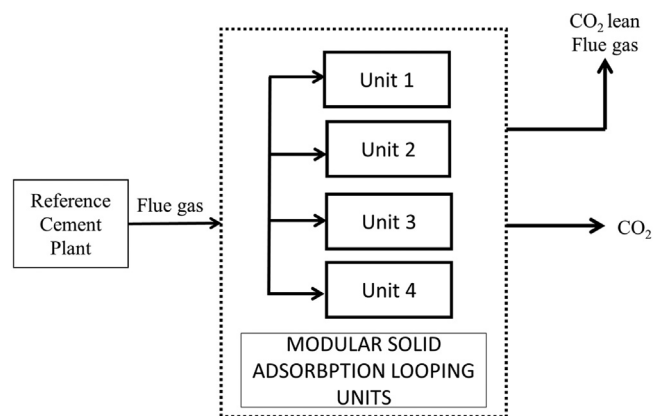


Fig. 3. Schematic diagram of an SPEI-based CCS system with the cement plant.

MEA-based CCS system, and the regenerated SPEI sorbent is transferred back to the bubbling bed adsorber reactor.

3. Modelling and boundary conditions

3.1. Feedstock composition

The raw meal used for the clinker formation is composed of limestone, iron, clay, and sand. The percentage of limestone is 77.7 wt. %, iron is 1.4 wt. %, clay is 18.6 wt. % and sand is 2.3 wt. %. The moisture content percentage in limestone is 15.2 wt. %, in iron is 5.3 wt. %, in clay is 14.0 wt. %, and in sand is 22.1 wt. %.

RDF, waste tires, and coal are used as fuel for heating pre-calciner, while only coal is used as fuel in the rotary kiln to achieve the desired temperature. The detailed ultimate analysis on a received basis is presented in Table 2.

3.2. Indirect heat consumption

Indirect heat consumption is defined as the ratio of net primary energy consumption to the efficiency of a reference cement plant. Indirect heat consumption refers to the net primary energy consumed in relation to the net electricity consumption produced by the reference power plant. Therefore, to calculate the indirect heat consumption, the reference power plant needs to be defined. For the current study, it is assumed that electricity is provided by the national grid, which has an electricity production efficiency of 62 % and an average CO₂ emissions rate of 274 kg CO₂/MWh (BP, 2022).

In the case of CCS integration, it is assumed that heat for sorbent regeneration is provided by a natural gas boiler with an efficiency of 90 % and a CO₂ emission rate of 56.1 kg of CO₂/GJ of natural gas (Voldsund et al., 2019).

Table 2
Ultimate analysis of fuels.

	Coal	RDF	Tyres
Hydrogen (Wt.%)	5.0	7.2	8
Carbon (Wt.%)	65.7	53.0	52.1
Sulphur (Wt.%)	0.5	0.3	2
Oxygen (Wt.%)	11.8	0	8
Nitrogen (Wt.%)	1.7	1.4	0
Water (Wt.%)	2	23.7	15
Ash (Wt.%)	13.3	14.4	0
LHV (MJ/kg fuel)	26.0	18.8	25.9

3.3. The reference cement plant operating conditions

We developed a simulation model for clinker production with a capacity of 133.4 tonnes/hr, based on real-time data provided by our industrial partner. Fig. 4 presents the main operating parameters of the reference cement plant. The raw meal is heated up to 880 °C using flue gas, with the first, second, third, and fourth stage cyclone preheaters having temperatures of 370, 563, 737, and 880 °C, respectively. The simulation model assumes ideal solid-gas separation during the preheating stage, and the flue gas leaving the preheaters enters the raw meal mill at 325 °C. Additionally, a portion of flue at 325 °C is utilized in the coal mill.

The clinker cooler uses ambient air, which splits into three preheating streams. The secondary air is fed to the rotary kiln at 973 °C, while the tertiary air is added to the pre-calciner at 1000 °C. A small portion of this air stream, at 213 °C, is utilized to heat the raw meal to the desired temperature at the inlet.

3.4. MEA-based CCS system operating conditions

In the MEA-based CCS system, it is assumed that 30 wt. % of the MEA solvent is used for the scrubbing process. The absorption column is operated at 40 °C with a CO₂ capture rate of 90 %. For a desorption column, a temperature of 150 °C is assumed with a CO₂ lean lime loading of 0.24 mol CO₂/mol MEA (Antzaras et al., 2023; Jaffar et al., 2023). The sorbent/CO₂ ratio is critical and significantly affects the CO₂ capture rate and regeneration energy. For the current study, a sorbent to CO₂ molar ratio of 2 is assumed, and the purge rate is assumed at 3.2 kg sorbent/tonne of CO₂ (Roussanaly et al., 2017; Atsonios et al., 2015; Abu-Zahra et al., 2007; Rezazadeh et al., 2016). The process flow diagram of the MEA-based CCS system is shown in Fig. 5.

3.5. SPEI-based CCS system operating conditions

In the SPEI-based CCS system, it is assumed that the sorbent used in the capture process is composed of 53 % silica and 47 % polyethyleneimine. The adsorption temperature in the bubbling bed is assumed to be 50 °C. Similar to the MEA-based CCS system, the CO₂ capture rate is assumed to be at 90 %. The bubbling bed desorption reactor temperature is assumed to be between 110 and 120 °C. The sorbent to CO₂ mass ratio is assumed to be 10 and the purge rate is assumed to be at 0.1 %. The working capacity of the SPEI-based CCS process is assumed to be 1.25 mmol of CO₂/g of SPEI (Kim et al., 2021; Zhang et al., 2014a, 2016). The process flow diagram of the SPEI-based CCS system is shown in Fig. 6.

4. Methodology

4.1. Technical analysis methodology

All three case scenarios were modeled and simulated using the ECLIPSE software to ensure that the comparisons are consistent and accurate. ECLIPSE, is a sophisticated suite of C-language programs, designed to facilitate an in-depth technical and economic analysis of both current and prospective fuel conversion and power generation systems. ECLIPSE stands out for its remarkable flexibility in approach and the seamless integration of technical and economic elements within the analysis. It is equipped with comprehensive databases encompassing chemical properties, utilities, and capital costing all of which are user-modifiable, thereby enhancing its applicability and utility in diverse scenarios (Williams and McMullan, 1996).

As shown in Fig. 7, technical and environmental analysis is carried out in four logical steps. In the first step, process flow diagrams made up of modules and streams are developed. In the second step, enthalpy calculations for each stream are performed that are used to calculate the mass and energy balance after defining the stream inputs and technical

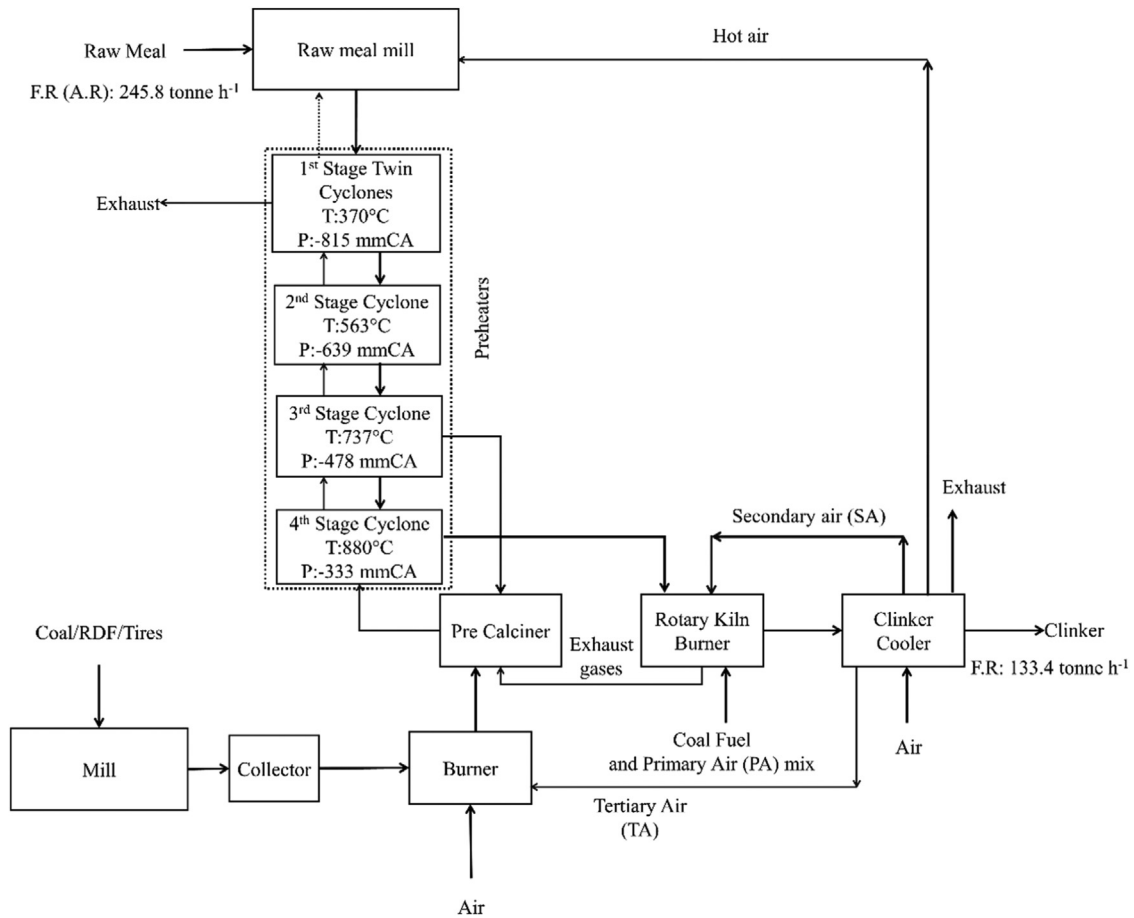
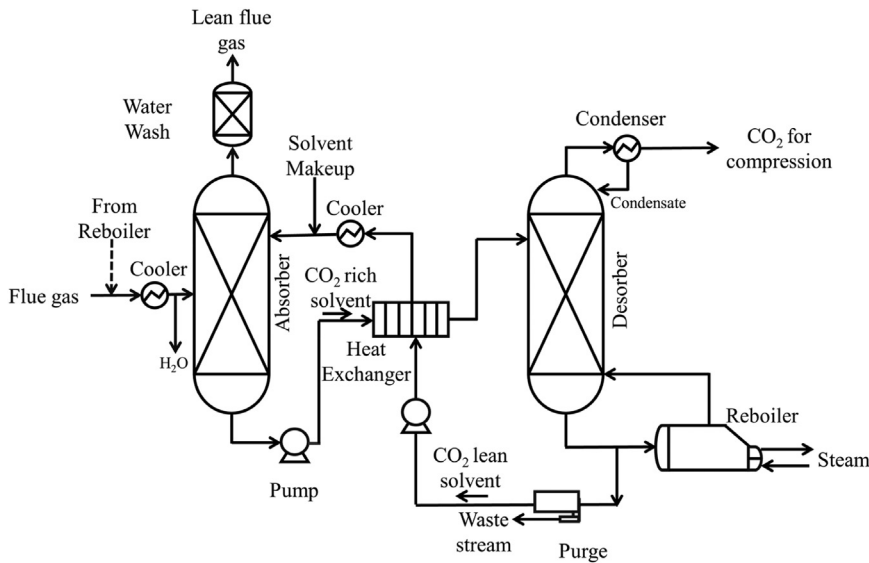


Fig. 4. Main operating parameters of the reference cement plant.

Fig. 5. Process flow diagram of MEA-based CCS system.



characteristics of particular modules. The data gathered during the simulation step serves as the baseline for identifying crucial components. In the third step, all the energy inputs and outputs are analyzed. After completing the mass and energy balance, the environmental assessment is conducted.

4.2. Environmental assessment methodology

LCA methodology is used to assess the environmental effects of goods or services throughout the course of their whole lifetime i.e.,

from cradle to grave. However, the cradle-to-gate concept is the most widely applied. The structures and machinery used in manufacturing, are frequently left out of research, but, in a few cases, where accounting LCAs are studied; these should be included. In this study, we utilized the LCA software SimaPro. The ecoinvent built-in database and the ReCipe 2016 method is applied for the life cycle impact assessment (LCIA).

The ISO 14040 series is available specifically for LCA (ISO, 2006). A LCA is divided into four sections: the goal and scope, the life cycle inventory (LCI), LCIA, and the interpretation.

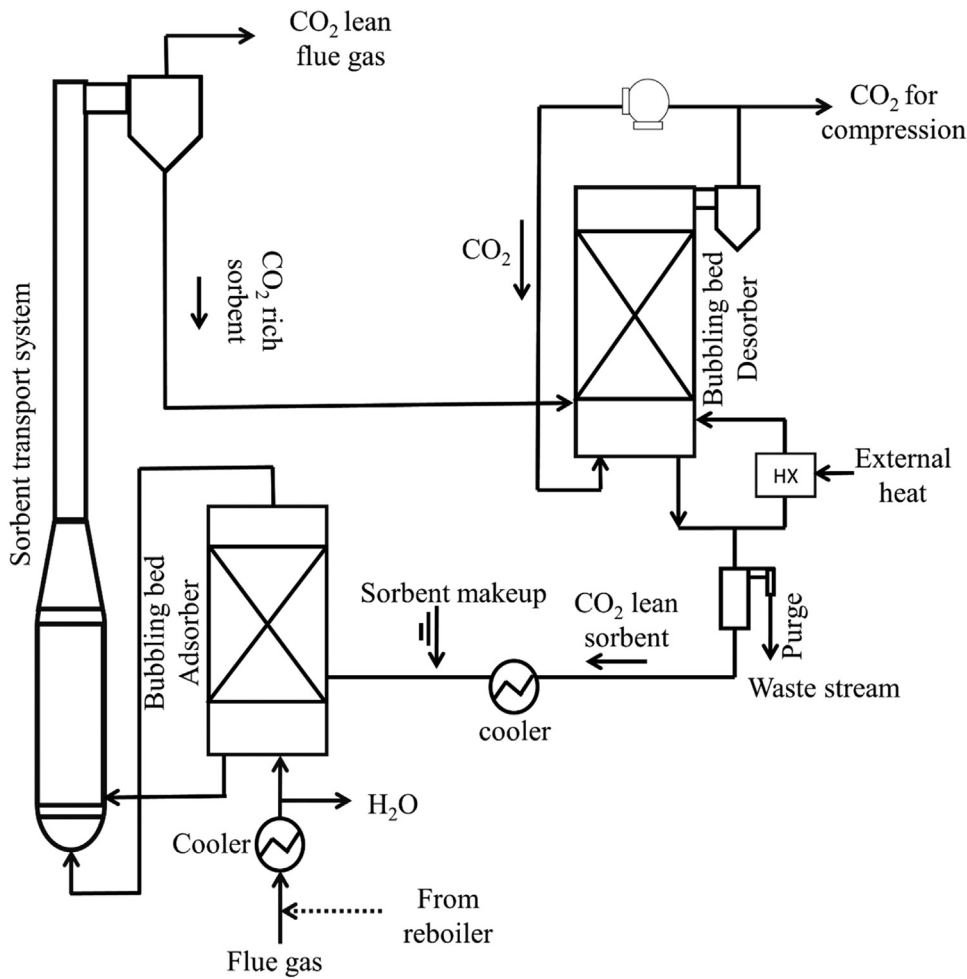


Fig. 6. Process flow diagram of SPEI-based CCS system.

4.2.1. Goal and scope

The goal of this research is to evaluate the environmental impact of the clinker production process by integrating the CCS system. The study compares the three case scenarios outlined in Section 2.1, to determine if the integration of CCS technologies can reduce environmental impact compared to a cement plant without CCS. Fig. 8 presents the system boundaries for life cycle assessment. The functional unit considered in this study is 1 kg of clinker production, which is consistent with similar research (Stafford et al., 2016a, 2016b).

Allocation is used to determine the share of environmental impact when multiple products are formed simultaneously. According to the ISO 14044 guidelines, the allocation should be minimized as much as possible. Baumann and Tillman (2004) and Weidema (2014) suggested splitting the unit processes or expanding the product system. In the SimaPro software the function of “Avoided Product” is applied to expand the system and the impact of avoided products are subtracted from the total impact of the system. However, in the case of RDF, a complication arises, because in the SimaPro software waste is not possible to model

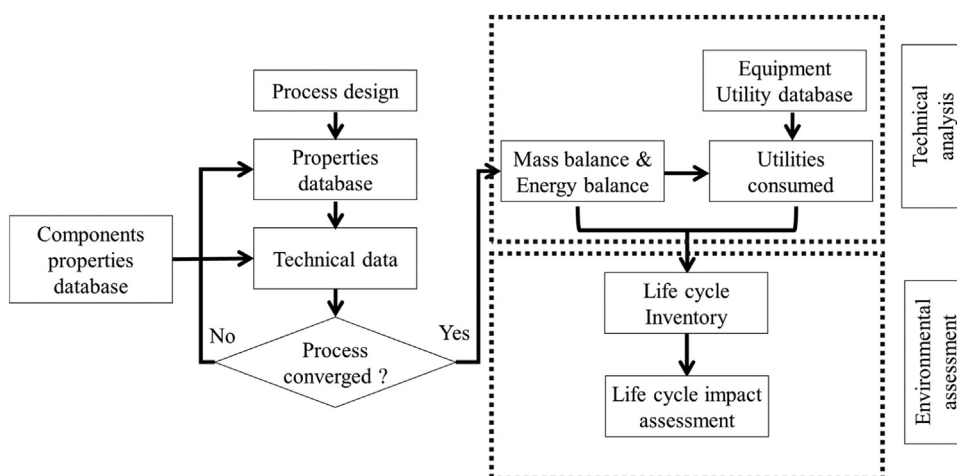


Fig. 7. Logical steps involved in process simulation and modeling for technical analysis and environmental assessment.

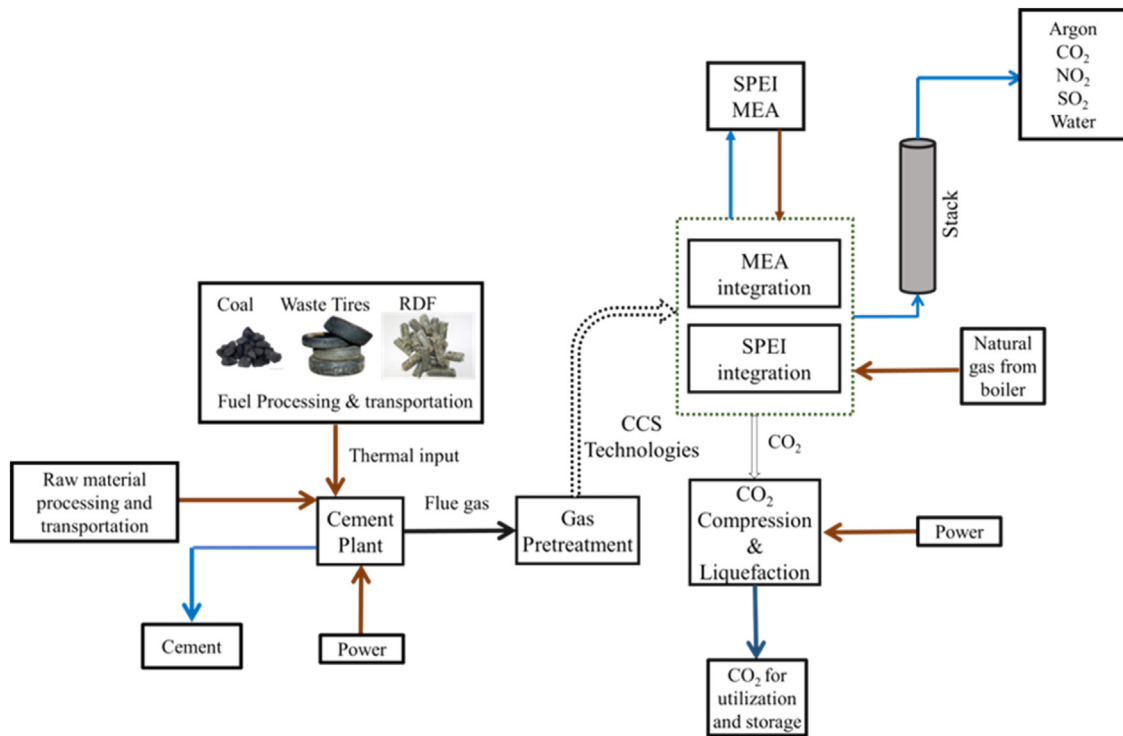


Fig. 8. System boundaries for LCA.

waste as an input product. Some literature avoided the issue because of the same input. For example, Dong et al. (2018) compared various waste-to-energy technologies using a functional unit of 1 tonne of municipal solid waste (MSW) received at the plant. To avoid the issue, they reported that the benefit of disposing of MSW in landfills would be the same for all technologies. Chen et al. (2010) explored different allocation methods for recycling waste into different products in the cement industry. They reported that if recycled material is used, there is no need to consider the extraction and production of the virgin base material, and it can be considered an avoided product. However, there is still a need to consider the environmental impact of the material recovery process. Similarly, according to European Waste Framework, the waste used to produce RDF has no use and should be disposed of. Therefore, in this study, the benefits of avoiding landfills disposal of RDF and waste tires are captured (Sauve and Van Acker, 2020).

Both midpoint and endpoint indicators were evaluated. The midpoint environmental indicator includes 18 categories, and the endpoint indicators include 3 categories (Brilhuis-Meijer, 2014). LCIA employ the characterization factor to quantify the environmental damages for

each impact category. However, comparing impacts is challenging since each impact characterization approach uses a different unit of measurement. Therefore, the findings from the characterization stage were divided by a reference situation for each impact in the normalization step and therefore make the result interpretation simpler (Baumann and Tillman, 2004).

In this study, background processes are modeled using the built-in database of SimaPro software where possible. It is considered that the pre-dried hard coal and the raw material are received therefore the drying processes are modeled using the ECLIPSE process simulator to obtain the utility inputs and flow rates. The RDF and waste tire processing are not included in the SimaPro software, therefore, LCA for these processes was developed by consulting the literature, and supply companies.

The process of RDF and tyres preparation and transportation are presented in the Fig. 9 and Fig. 10, respectively. Tables 3 and 4 present the assumptions considered for the preparation and transport of RDF and waste tires respectively (Dong et al., 2021; Abougilil et al., 2017; Merlin and Vogt, 2020; Kløverpris, 2010; Pavlovic et al., 2019; Rolfe et al., 2022). It is assumed that RDF and waste tires receive envi-

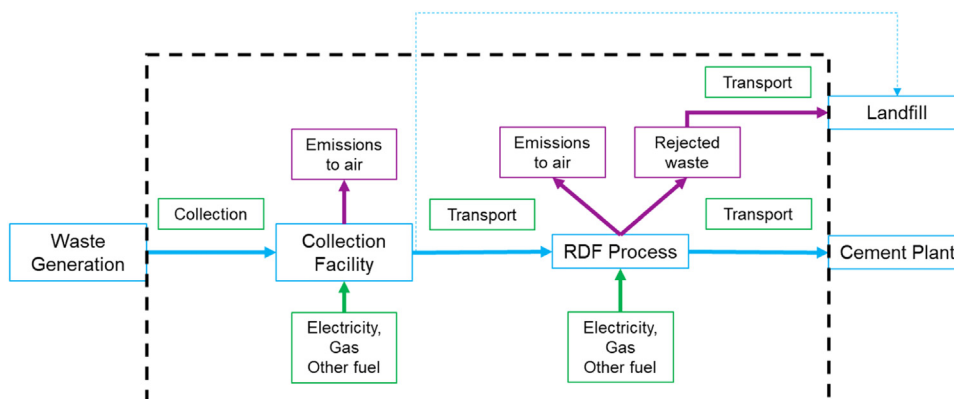


Fig. 9. Schematic of RDF preparation and transport.

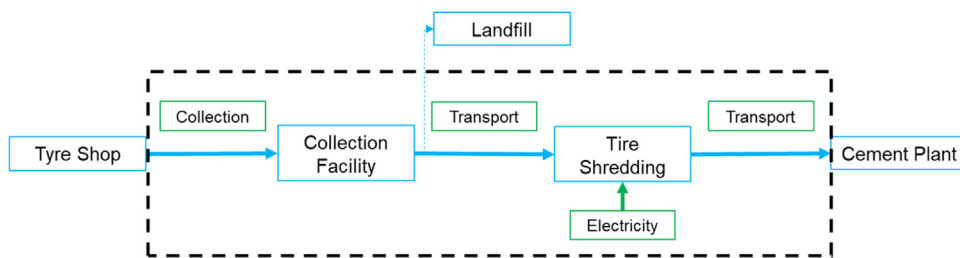


Fig. 10. Schematic of tyres preparation and transport.

Table 3
Assumptions for the preparation and transportation of RDF.

Collection (tonnes per kilometre)	29.4
Electricity (kWh)	154.4
Gas (m ³)	37.09
Transport (tonnes per kilometre)	95.61
CO ₂ (kg)	68,60166
RDF (tonnes)	1

Table 4
Assumptions for the preparation and transportation of tyres.

Collection (tonnes per kilometre)	30
Transport (tonnes per kilometre)	80
Electricity (MJ)	4.5
Shredded Tyres (tonnes)	1

ronmental credit for not being disposed of in landfills, but the environmental impact of waste collection, sorting, and transportation is considered. The average distance from waste generation to the collection facility, from the collection facility to the RDF processing site, and from the RDF processing site to the landfill site was estimated using literature and in agreement with the manufacturer. Similarly, in the case of waste tires, the transport distance from the tire shop to the collection site and from the collection site to the landfill site was estimated using literature. Additionally, the transport distance of hard coal and raw material was estimated using literature. It is assumed that the waste collection vehi-

cle was a 21 mt waste collection lorry. While, for the freight transport of hard coal, RDF, and waste tires the vehicle is assumed to be 32 mt freight transport lorry.

In the case of RDF, the waste is sorted at a municipal solid waste facility (MSF). The electricity and gas are utilized in the collection facility and RDF during the RDF processing stage. The electricity data and natural gas is taken from the built-in SimaPro database. After MSF, the waste is normally sent to the final stream (reuse, recycle, landfills). According to EU legislation, waste should be recycled or reused before being disposed of in landfills (energy recovery in this case). Therefore, it is assumed that the waste utilized in the RDF process is destined for landfills. The waste diverted from landfills and waste rejected from RDF processing are sent back to the landfills. Therefore, except for transportation, the process is considered neutral.

The LCA for this project is cradle-to-gate, which includes material extraction, fuel synthesis and drying, transport, clinker production process, and clinker production process integrated with CCS technologies. The main limitation of this project is the assumption of unknown elements that are outside the control of this study.

5. Results

5.1. Technical analysis results

ECLIPSE process simulation software was used to perform the technical analysis of all three case scenarios. The key technical analysis results are given in Table 5.

Table 5
Technical analysis results.

Main process data	Reference cement plant	MEA integration	SPEI integration
Raw meal input (tonne/hr, as received)		208.9	
RDF input (tonne/hr, as received)		14.0	
Tyres input (tonne/hr, as received)		1.4	
Coal input (tonne/hr, as received)		7.0	
Total thermal input (MWth)		133.4	
Clinker production (tonne/hr)		133.4	
Raw meal/clinker ratio (dry basis)		1.57	
Auxiliary power consumption (MWe)	16.3	17.2	16.8
CO ₂ purification/compression power (MWe)	n/a	13.8	13.0
Total power consumption (MWe)	16.3	31.0	29.8
Specific power consumption (MWh/tonne Clinker)	0.12	0.23	0.22
Specific direct heat required (GJ/tonne Clinker)	3.60		
Specific indirect heat required (GJ/tonne Clinker)	0.71	1.30	1.35
Specific heat requirement from natural gas boiler (GJ/tonne Clinker)	-	4.40	2.76
Equivalent specific heat requirement (GJ/tonne Clinker)	4.31	9.35	7.66
CO ₂ captured (tonne CO ₂ /hr)	n/a	134.5	126.5
CO ₂ emitted on-site (tonne CO ₂ /hr)	122.5	15.0	14.1
CO ₂ capture rate of the plant (%)	-	90	90
Specific direct CO ₂ emissions (kg CO ₂ /tonne Clinker)	919	92	92
Specific indirect CO ₂ emissions (kg CO ₂ /tonne Clinker)	34	64	61
CO ₂ emissions from natural gas boiler (kg CO ₂ /tonne Clinker)	-	20	16
Equivalent specific CO ₂ emissions (kg CO ₂ /tonne Clinker)	952	176	169
Equivalent CO ₂ emissions avoided (kg CO ₂ /tonne Clinker)	-	777	783
SPECCA (GJ/tonne CO ₂)	-	6.49	4.30

Table 6
Life cycle inventory of (i) reference cement plant (ii) MEA integration with the cement plant (iii) SPEI integration with the cement plant.

Inputs			Wastes & Emissions			Products		
Element	Value	Unit	Element	Value	Unit	Element	Value	Unit
Reference cement plant								
Limestone	1.5349	kg	Argon	0.0199	kg	Clinker	1	kg
Coal	0.0522	kg	Water	0.4130	kg			
Tyres	0.0101	kg	CO ₂	0.9176	kg			
RDF	0.1048	kg	NO ₂	0.0001	kg			
Electricity	0.44	MJ	SO ₂	0.0001	kg			
MEA integration								
Limestone	1.5349	kg	Argon	0.0199	kg	Clinker	1	kg
Coal	0.0522	kg	Water	0.8952	kg	CO ₂	1.0085	kg
Tyres	0.0101	kg	CO ₂	0.1133	kg			
RDF	0.1048	kg	NO ₂	0.0001	kg			
MEA	0.0027	kg	SO ₂	0.0001	kg			
Electricity	0.8502	MJ	MEA	0.0037	kg			
Natural gas	0.1066	m ³						
SPEI integration								
Limestone	1.5349	kg	Argon	0.0199	kg	Clinker	1	kg
Coal	0.0522	kg	Water	0.4130	kg	CO ₂	0.9483	kg
Tyres	0.0101	kg	CO ₂	0.1055	kg			
RDF	0.1048	kg	NO ₂	0.0001	kg			
SPEI	0.010	kg	SO ₂	0.0001	kg			
Electricity	0.8067	MJ	SPEI	0.0011	kg			
Natural gas	0.0711	m ³						

5.1.1. Technical performance of the reference cement plant

Based on the real-time data provided by our industrial partner CE-MEX, the modeling of the reference cement plant has been performed with a clinker production capacity of 133.4 tonnes/h. The specific heat required is 3.60 GJ/tonne of clinker produced, with almost 60 % of the thermal energy linked to the calcination of limestone in the pre-calciner. The total power consumption in the reference cement plant is 16.3 MWe, with a specific power consumption of 0.12 MWh/tonnes of clinker produced. The results show that the total CO₂ production from the cement plant is 122.5 tonnes/hr, with the highest CO₂ emissions linked to the pre-calciner. If the indirect CO₂ emissions from electricity are considered, then the equivalent CO₂ emissions are estimated to be 0.952 tonne CO₂/tonne clinker.

5.1.2. Technical performance of the integration of an MEA-based CCS process with the cement plant

The integration of an MEA-based CCS with a cement plant can remove 90 % of the CO₂ from the flue gas. To ensure a fair comparison, the raw meal input and clinker production capacity have been kept constant. However, MEA integration requires more electricity. Considering the base case cement plant, MEA-based carbon capture unit, and compression unit, the electricity consumption increased to 31.0 MWe, with almost 45 % of the total electricity consumption occurring in the CO₂ compression stage. In addition to the electricity consumption requirement, additional heat is necessary to regenerate the solvent in the desorber. In this study, it is assumed that the required thermal energy for solvent regeneration is provided by a natural gas boiler. The total thermal energy required from the natural gas boiler is 162.8 MWth, with a specific heat requirement of 4.4 GJ/tonne of clinker. The specific CO₂ emissions avoided (including direct, indirect, and natural gas CO₂ emissions) and specific primary energy consumption for CO₂ avoided (SPECCA) relative to the reference cement plant are 0.78 tonnes CO₂/tonne of clinker and 6.5 GJ/tonne of clinker, respectively.

5.1.3. Technical performance of the integration of SPEI-based CCS process with the cement plant

In the scenario where the SPEI-based CCS unit is integrated with the reference cement plant, 90 % of CO₂ can be removed from the flue

gas. The raw meal input and clinker production capacity of the reference case cement plant remain the same. The electricity consumption in the SPEI integration is 29.8 MWe, which is lower compared to the MEA integration, with almost 44 % of the total electricity required for the compression. This is due to the lower amount of CO₂ required to be captured because of lower thermal energy requirement. The total regeneration heat required for the sorbent regeneration is 102.4 MWth, corresponding to 2.36 GJ/tonne CO₂, which is almost 33 % lower compared to the MEA integration. The specific CO₂ emissions avoided (including direct, indirect, and natural gas CO₂ emissions) and SPECCA relative to the reference cement plant are 0.78 tonnes CO₂/tonne of clinker and 4.3 GJ/tonne of clinker, respectively.

5.2. Environmental analysis results

The LCI results obtained from the technical analysis are presented in Table 6. To ensure accuracy across the studied scenarios, the functional unit is assumed to be 1 kg of clinker produced. As the SimaPro software does not support flowrates, therefore, the LCI data timescale is set to one second.

5.2.1. Midpoint indicators

Table 7 and Fig. 11 present the LCIA characterization results of the midpoint indicators. As each impact category has a different set of units, they cannot be directly compared. However, the impact category scores for each scenario can be useful for comparison purposes. The base case cement plant exhibits the lowest scores in all the midpoint impact assessment indicators, except for global warming, which is highest in the MEA integration followed by SPEI integration. Meanwhile, the MEA integration results in the highest scores in nearly all the midpoint impact categories, except for global warming, which is almost 64 % lower than the reference cement plant. While, in the case of SPEI integration the global warming impact is 67 % lower than reference cement plant. The higher score of the global warming indicator in the reference cement plant is linked to the higher CO₂ emissions into the atmosphere from the process, while the higher impact scores of the other midpoint indicators in the case of CCS integration are linked to the utilization of a higher amount of natural resources such as electricity and natural gas. The SPEI

Table 7
Impact Assessment. ReCiPe 2016 Midpoint (H) V1.07 / World (2010) H.

Impact category	Unit	Reference cement plant	MEA integration	SPEI integration
Global warming	kg CO ₂ eq	1.01274	0.36221	0.33678
Stratospheric ozone depletion	kg CFC-11 eq	0.00000	0.00000	0.00000
Ionizing radiation	kBq Co-60 eq	0.03044	0.05408	0.05233
Ozone formation, Human health	kg NOx eq	0.00044	0.00061	0.00057
Fine particulate matter formation	kg PM2.5 eq	0.00019	0.00031	0.00029
Ozone formation, Terrestrial ecosystems	kg NOx eq	0.00045	0.00062	0.00058
Terrestrial acidification	kg SO ₂ eq	0.00049	0.00083	0.00076
Freshwater eutrophication	kg P eq	0.00013	0.00018	0.00017
Marine eutrophication	kg N eq	0.00001	0.00001	0.00001
Terrestrial ecotoxicity	kg 1,4-DCB	0.14905	1.33993	0.22298
Freshwater ecotoxicity	kg 1,4-DCB	0.00481	0.00758	0.00704
Marine ecotoxicity	kg 1,4-DCB	0.00655	0.01011	0.00959
Human carcinogenic toxicity	kg 1,4-DCB	0.00924	0.01419	0.01335
Human non-carcinogenic toxicity	kg 1,4-DCB	0.19079	0.26233	0.25397
Land use	m ² a crop eq	0.00354	0.00638	0.00583
Mineral resource scarcity	kg Cu eq	0.00014	0.00028	0.00025
Fossil resource scarcity	kg oil eq	0.05425	0.17274	0.13660
Water consumption	m ³	0.00121	0.00212	0.00205

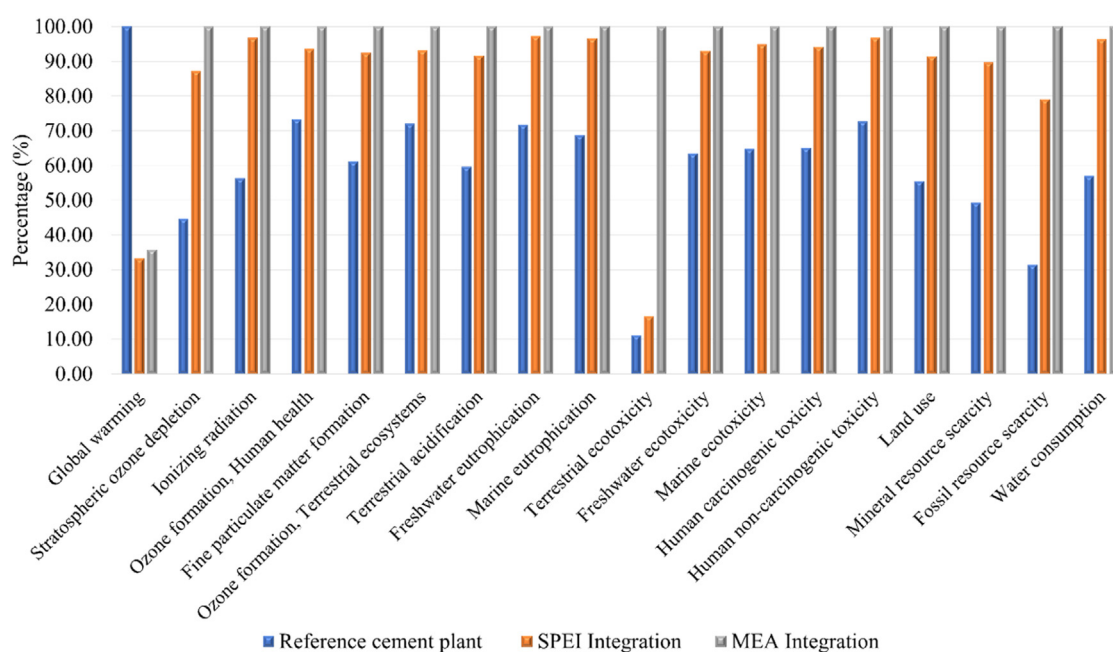


Fig. 11. Comparing the reference cement plant, SPEI-based CCS technology integrated with the cement plant and MEA-based CCS technology integrated with the cement plant. Method: ReCiPe 2016 Midpoint (H) V1.07 / World (2010) H.

integration leads to better performance and shows lower impact scores for all the categories when compared with the MEA integration. In some indicators such as stratospheric ozone depletion, terrestrial ecotoxicity, mineral resource scarcity and fossil fuel scarcity the impact score of MEA integration are more than 10 % higher than SPEI integration.

These results are in agreement with the results reported by An et al. (2019) who studied the influence of the integration of CCS technology into a cement plant. They also reported that, in the midpoint impact assessment, the integration of MEA-based CCS technology can decrease the global warming potential impact. However, the increase in the score of other environmental impacts was observed with the integration of CCS technologies. Similarly, García-Gusano et al. (2015) also reported a better performance score in terms of global warming impact when MEA-based CCS was integrated with the Spanish cement plant. They also reported that the integration of CCS technology can increase the impact potentials of photochemical eutrophication, ozone formation, acidification, ionising radiation, human toxicities, ecotoxicity, particulate matter, and land use by several times. Furthermore,

Galusnyak et al. (2022) also investigated the influence of the integration of calcium carbonate looping-based CCS unit into cement plant. They also reported that at a capture rate of 90 %, the global warming impact is around 69 % lower compared to the reference cement plant without CCS integration.

As mentioned in the Goal and Scope section, the environmental assessment study aimed to investigate whether integrating a CCS system could potentially reduce the climate change impact (i.e., global warming) without negative environmental impacts. The breakdown of the global warming impact is shown in Fig. 12. The results indicate that the main contributor to the global warming impact is the clinker production process in all three scenarios. However, the global warming impact score for the reference cement plant is almost 83 % higher than that of both the MEA-based and SPEI-based CCS system integrations. These results were expected as the capture rate of the CCS technologies is fixed at 90 %. Rolfe et al. (2018) also studied the influence of the integration of CCS technologies into a cement plant. They also reported that at a capture rate of 94 %, the global warming impact score

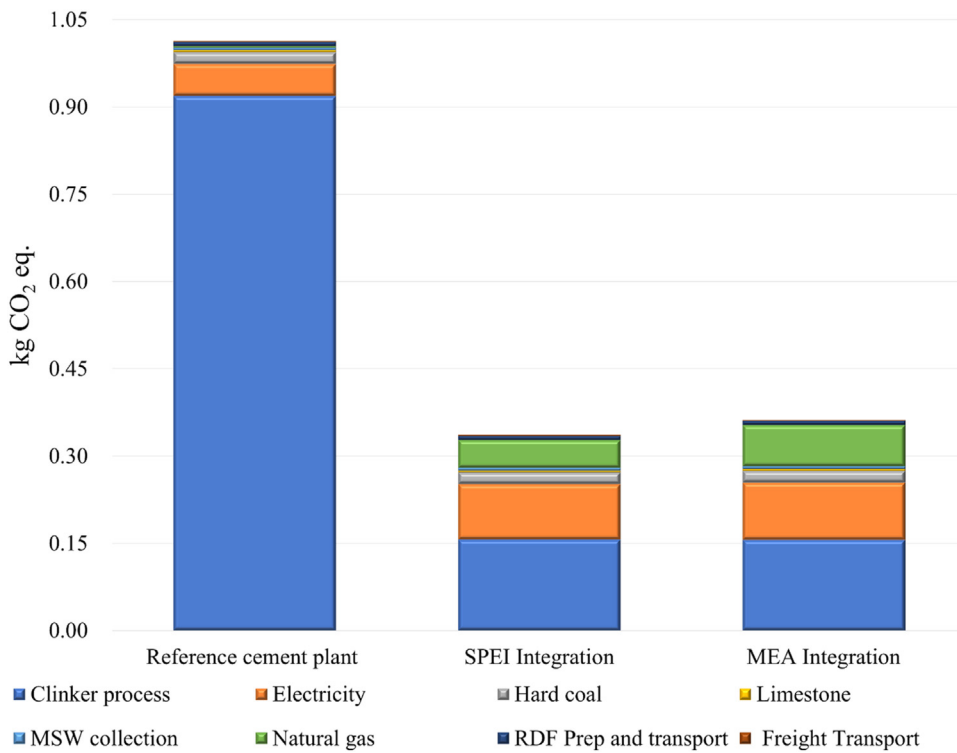


Fig. 12. Break down of global warming indicator, unit kg CO₂ eq. Method: ReCiPe 2016 Midpoint (H) V1.07 / World (2010) H.

is 90 % lower for the CCS integration compared to the reference cement plant.

In the case of CCS integration with the cement plant, after the clinker production process, the electricity and natural gas inputs have a higher contribution towards the global warming indicator compared to the reference cement plant. In the case of MEA integration, the contribution of natural gas and electricity was nearly 44 % and 97 % higher than that

of the reference cement plant, respectively. In the case of SPEI integration, the electricity and natural gas contribution to the global warming indicator was 3 % and 32 % lower in comparison to MEA integration.

The integration of both CCS technologies has a higher impact score on fossil resource scarcity, as shown in Fig. 13. The main contributors to the higher impact in the case of CCS integration were electricity and natural gas input. The electricity and natural gas consumption was high-

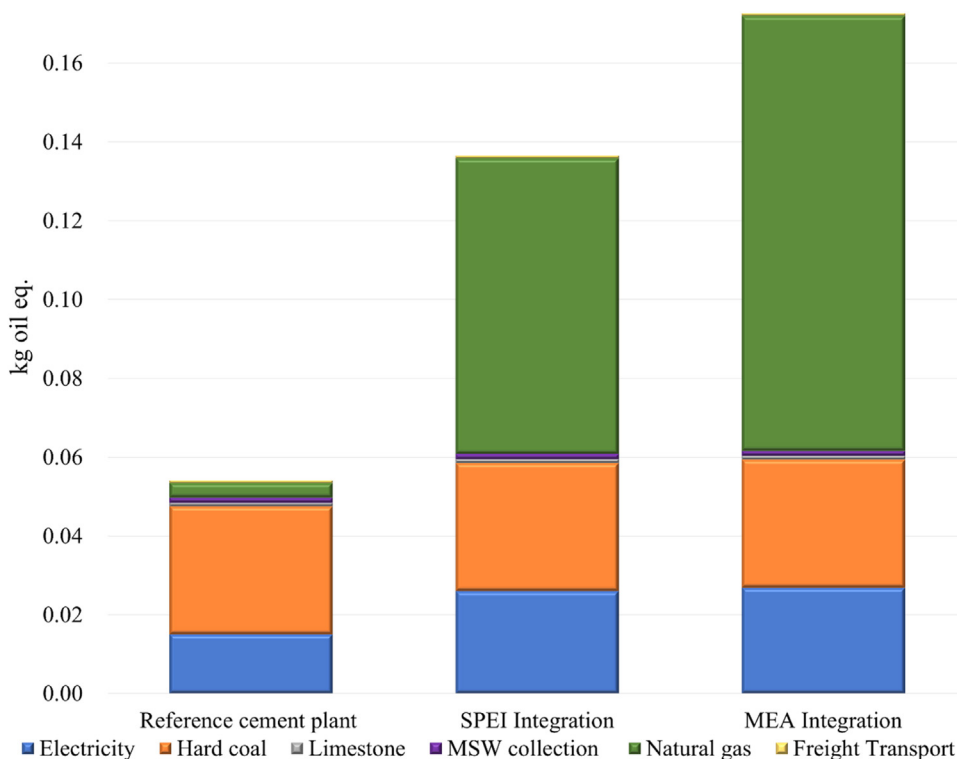


Fig. 13. Break down of fossil resource scarcity indicator, unit kg oil eq. Method: ReCiPe 2016 Midpoint (H) V1.07 / World (2010) H.

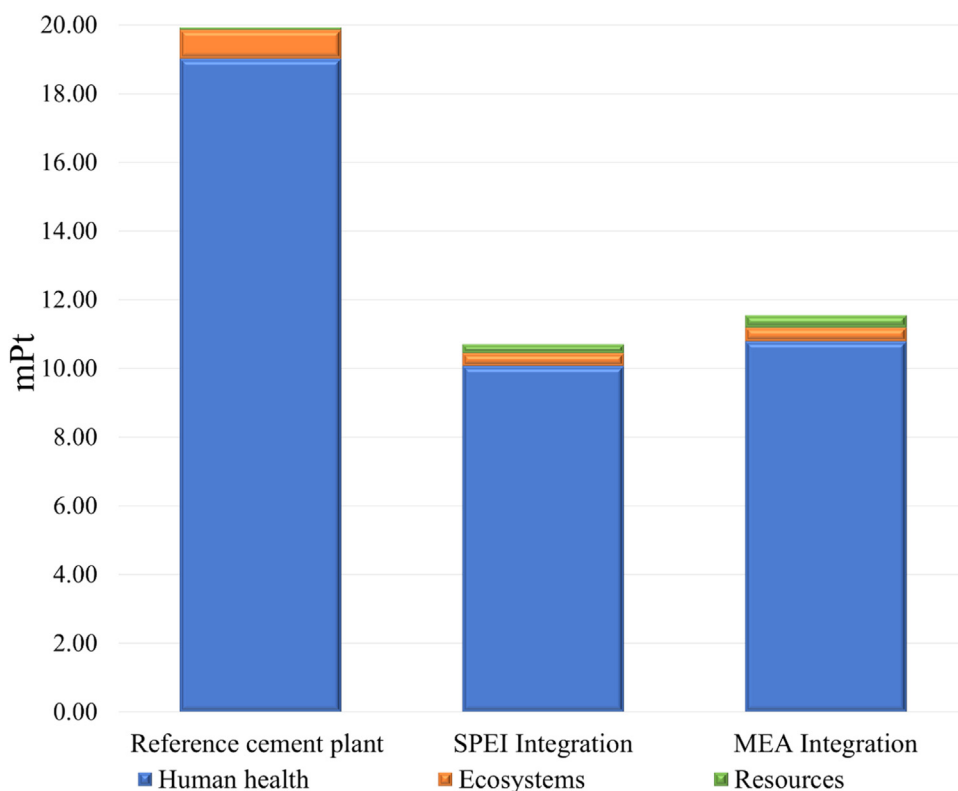


Fig. 14. Endpoint single score assessment. Method: ReCiPe 2016 Endpoint (H) V1.07 / World (2010) H/A.

est in the MEA integration. Therefore, when MEA integration was compared to the reference case cement plant, the electricity and natural gas input resulted in 44 % and 97 % higher scores, respectively. Meanwhile, the SPEI integration resulted in 3 % and 32 % lower scores of electricity and natural gas consumption compared to the MEA integration. An et al. (2019) reported that most of the environmental impacts in the case of CCS integration are linked to heat and power requirements, therefore, resulting in a higher impact score.

5.2.2. Endpoint indicator

The endpoint single score results are presented in Fig. 14. The integration of CCS technologies results in a lower impact on human health and the ecosystem. For human health and the ecosystem, the base case assembly exhibits values of 18.94 and 0.85 mPt, respectively. Meanwhile, the lowest values are exhibited by SPEI integration at 9.99 and 0.37 mPt, respectively, for the human health and ecosystem indicators. However, the values for the resource indicator are at 0.35, 0.26, and 0.06 mPt, respectively for MEA integration, SPEI integration, and the reference cement plant.

Similarly, damage assessment results show that in the case of MEA integration, the impact on human health and damage to the ecosystem is almost 44 % and 53 % lower, respectively, compared to the reference cement plant. However, due to the utilization of higher electricity and natural gas, the impact of fossil fuel depletion is higher in the case of CCS integration. In the case of the reference cement plant, the resource depletion impact is almost 82 % lower compared to MEA integration.

The SPEI integration has a lower endpoint impact compared to the MEA integration in all three categories, i.e., resources, ecosystem, and human health. In the case of SPEI integration, the impact on human health, ecosystem, and resources is almost 7 %, 9 %, and 26 % lower than MEA integration. This is due to the requirement for a lower amount of electricity and natural gas, and lower emissions. A similar trend of results has been presented by Singh et al. (2012). They studied the influence of the integration of CCS into various processes. They also reported that the integration of CCS technology can significantly reduce

the impact score of human health and ecosystem score thereby showing a positive impact. However, the impact score of the resource indicator was reduced for the resources because of the utilization of more energy to carry out the CO₂ capture operation.

6. Conclusion

This study aimed to evaluate the technical and environmental assessment of the integration of carbon capture and storage technologies into the cement plant. A comparison of conventional MEA-based carbon capture technology and SPEI-based carbon capture technology was performed. The main conclusions from this study are the following:

From the technical point of view, integration of both processes can eliminate the CO₂ from the flue gas to up to 90 %. The equivalent CO₂ emissions avoided were at 742.9 and 749.3 kg CO₂/tonne clinker for the MEA-based carbon capture process and SPEI-based carbon capture process, respectively. However, the requirement for electricity and natural gas increased for both carbon capture technologies. This was due to the additional energy required to operate CO₂ capture technologies. According to the results of SPECCA the SPEI-based carbon capture unit integrated with the cement plant showed better performance than MEA-based carbon capture process integration. The SPECCA of MEA integration was at 6.5 GJ/tonne CO₂ and for SPEI integration at 4.3 GJ/tonne CO₂. The life cycle impact assessment results also indicated that this was the key component that reduced the environmental impact of SPEI integration.

From an environmental assessment perspective, the SPEI-based carbon capture process integration showed better environmental performance compared to MEA-based integration in all impact categories. The MEA-based carbon capture process integration had a higher score in the global warming indicator (still lower than the base case) due to the higher energy requirement for sorbent regeneration, resulting in greater emissions for energy generation. Additionally, the MEA-based carbon capture process integration had a score almost 21 % higher for fossil re-

source scarcity due to the increased utilization of electricity and natural gas for capture process operation.

Also, it can be depicted from the single score endpoint results, overall, the integration of CCS technologies into cement plants have a positive environmental impact in terms of human health and ecosystems. The results show that greater benefits can be achieved by integrating the SPEI-based carbon capture process integration compared to MEA-based carbon capture process integration.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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