Assessment of the sustainability of intensified CO₂ capture schemes

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ARTICLE INFO

Keywords:

CO₂ capture
Monoethanolamine
Deep eutectic solvent
2030 agenda

ABSTRACT

The SDGs do address climate-related goals that are interconnected with the need to reduce greenhouse gas emissions. CO₂ capture involves the use of solvents such as Monoethanolamine (MEA), whose use, advantages, and disadvantages are well reported. Currently, there are alternative solvents that are theoretically more sustainable such as deep eutectic solvents (DES), however, a direct comparative with sustainable indicators is not always available. In this work, two schemes for the CO₂ capture process are evaluated and compared in a sustainable framework. Both schemes capture CO₂ from a combustion process to generate electricity. The first scheme considers Monoethanolamine (MEA) and the second scheme considers a DES (ChCl/ urea (1:2), considering in both schemes the use of natural gas, biogas, and coal as fuels that originate the CO₂ flux. The evaluation of both alternatives must be approached in a weighted manner and within a framework of sustainability. The results indicate that there is no single solution as the optimal solvent for CO₂ capture. It was observed that the choice of solvent is predominantly influenced by the type of fuel used in the combustion zone for electricity generation.

1. Introduction

Climate change is increasingly recognized as one of the most critical and complex challenges facing our planet today. The phenomenon of global warming, driven primarily by human-induced greenhouse gas (GHG) emissions, poses severe risks to ecosystems, weather patterns, and human societies worldwide. Greenhouse gases, including carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O), and water vapor, are pivotal in trapping heat within the Earth's atmosphere, thereby enhancing the greenhouse effect and leading to an overall increase in global temperatures [1]. According to recent estimates, the total annual GHG emissions amount to approximately 50 billion tons, measured in carbon dioxide equivalents (CO_{2eq}) [2]. This vast quantity of emissions contributes significantly to the rise in Earth's average surface temperature and underscores the urgent need for effective mitigation strategies.

Among the various greenhouse gases, CO₂ is the most significant due to its substantial impact on global warming. As depicted in Fig. 1, CO₂ emissions consistently outpace those of other greenhouse gases, highlighting CO₂'s central role in driving climate change. Research conducted by Al-Ghussain reveals that alterations in atmospheric CO₂ concentrations can have profound effects on global temperatures. For

instance, doubling or halving CO₂ concentrations in the atmosphere results in changes to the average land surface temperature of +3.8 °C or -3.6 °C, respectively [3]. This significant temperature sensitivity emphasizes the critical need to address CO₂ emissions as a central component of climate change mitigation efforts.

The primary source of CO₂ emissions is the energy sector, which encompasses a range of activities and processes that contribute to greenhouse gas emissions. Specifically, electricity generation is the largest contributor to CO₂ emissions, accounting for approximately 15,110.75 million metric tons of CO₂ annually, which represents about 47 % of total global emissions in 2020 [2]. This heavy reliance on fossil fuels for electricity generation exacerbates the problem, making it imperative to seek transformative changes in the energy sector's production structure. Given the substantial impact of electricity generation on CO₂ emissions, investigating and advancing CO₂ capture technologies in power plants is of paramount importance.

To address climate change effectively, it is essential to align strategies with the Sustainable Development Goals (SDGs) established by the 2030 Agenda. These goals provide a comprehensive framework for reorienting economic, political, and social activities towards environmental sustainability. Given the pressing need to mitigate climate

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<https://doi.org/10.1016/j.cep.2025.110222>

Received 10 October 2024; Received in revised form 19 January 2025; Accepted 11 February 2025

Available online 12 February 2025

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change, various strategies have been proposed to reduce CO₂ emissions, with carbon capture emerging as a critical solution for the industrial sector. The SDGs relevant to CO₂ capture include Goal 7, which aims to ensure access to affordable, reliable, sustainable, and modern energy; Goal 9, which focuses on promoting sustainable industrialization and fostering innovation; and Goal 12, which seeks to promote sustainable consumption and production patterns [4]. These goals underscore the necessity of integrating sustainability principles into CO₂ capture technologies to ensure that they are not only effective but also environmentally responsible.

Carbon capture and storage (CCS) technology is designed to separate CO₂ from emission sources, transport it to a storage site, and isolate it from the atmosphere over long periods [5]. This technology offers a promising approach to significantly reduce CO₂ emissions from industrial processes and address existing atmospheric CO₂. CCS technologies are generally categorized into three main types: post-combustion capture, pre-combustion capture, and oxy-combustion [6]. Among these, post-combustion capture (PCC) technology is the most developed and widely implemented, demonstrating high effectiveness in CO₂ capture processes. PCC techniques include chemical absorption, adsorption, membrane separation, cryogenics, hydrate formation, and microbial processes, each with distinct advantages and limitations.

Chemical absorption is one of the most established methods for CO₂ capture, particularly using alkanol amines. This method involves the use of solvents that react with CO₂ to form intermediate compounds, which are then desorbed through changes in pressure and temperature [7]. Alkanol amines, including primary, secondary, and tertiary amines, are commonly used as sorbents. Monoethanolamine (MEA) is the traditional solvent of choice due to its high capacity, cost-effectiveness, and rapid absorption rate. However, MEA presents several environmental concerns, including high toxicity and substantial energy consumption, highlighting the need for exploring alternative solvents [8,9].

The post-combustion capture based on chemical absorption has proven to be the most developed, studied, and competitive strategy since it has a fast absorption rate and high CO₂ capture efficiency [1,2]. The chemical absorption method is based on the use of chemical solvents that react with CO₂, generate intermediary compounds, and then perform CO₂ desorption by changing pressure and temperature parameters to recover the sorbent [3]. The main chemical sorbents are based on alkanol amines, which include primary, secondary, and tertiary amines [10]. The study of the different solvents has allowed the identification of their advantages and disadvantages as listed in Table 1.

The classic chemical solvent for CO₂ separation applications is aqueous monoethanolamine (MEA) due to its high CO₂ capture capacity, commercial availability, relatively low cost, fast absorption rate, and extensive research in industrial applications. While research has shown that process modifications could reduce energy consumption, it is not the only characteristic to evaluate when talking about process quality.

Table 1

Advantages and disadvantages of different chemical absorbents [11].

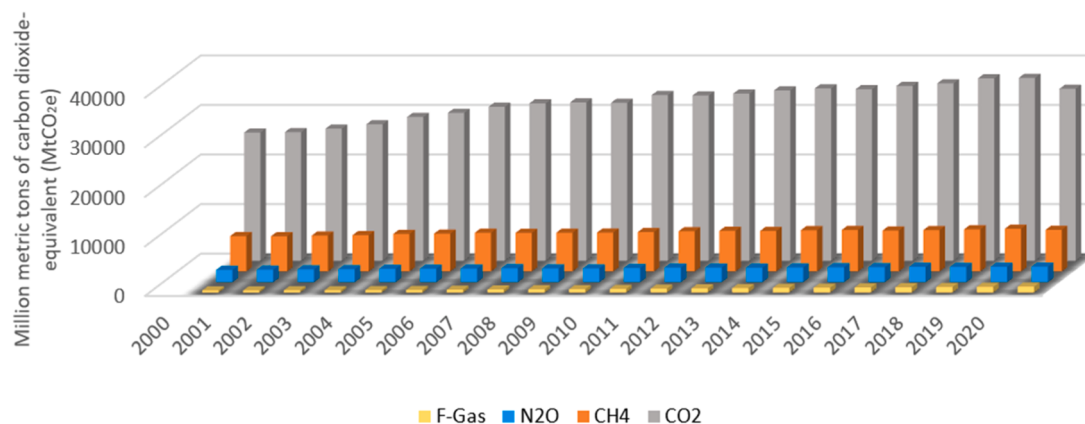
Absorbent	Advantages	Disadvantages
Organic amine solution	Excellent capture capacity and separation efficiency, and it is a well-researched technology.	The desorption process requires a significant amount of energy and is corrosive.
Ammonia	Low energy consumption, low corrosivity, and low cost.	Low average efficiency of aqueous ammonia in absorbing CO ₂ and high ammonia escape rates.
Sodium hydroxide solution	Strong CO ₂ absorption capacity and low cost.	Corrosive and high energy consumption for absorbent regeneration.

Despite its high efficiency, MEA is considered highly toxic, so its implementation entails a high environmental impact. Given these drawbacks, there is an opportunity to study new solvents that may be able to replace MEA in the CO₂ capture process.

Although considered as a reference solvent, MEA is still hampered by its high capital cost and high energy consumption, some research has been carried out to reduce these limitations such as the one performed by Yin et al. [4]. Even though the use of MEA is relatively mature in its implementation, several studies have observed several disadvantages of its use. For example, Patricia Luis [5] mentions several consequences oriented to energy requirements during the MEA production process. The study by Zhang et al. [6] mentions that the use of MEA as a solvent can produce higher energy consumption, and higher environmental impact caused by solvent degradation. Cuellar-Franca and Azapagic [8] mention relevant consequences such as acidification and human toxicity. With these considerations, several proposals have been generated. For example, Alzahrani et al. [12] proposed the use of Solar Energy-Assisted Flue Gas Amine for CO₂ capture. On the other hand, Dong et al. also presented a coal-fired solar-assisted carbon capture power generation system integrated with an organic Rankine cycle for the same purpose. However, some disadvantages for the use of MEA persist.

Recent research has introduced deep eutectic solvents (DES) as a potentially greener alternative to traditional solvents like MEA. DESs are characterized by their selective CO₂ absorption capabilities and potentially lower environmental impact. Studies have demonstrated that DESs can achieve high CO₂ recovery rates, with some optimizing power generation plants to recover up to 95 % of CO₂ [13]. However, a comprehensive comparative analysis of DES and MEA, incorporating a broader range of sustainability indicators, is currently lacking.

The relevance of integrating sustainability into CO₂ capture processes cannot be overstated. As the world moves towards more stringent climate goals and environmental regulations, the adoption of CO₂

**Fig. 1.** Historical GHG emissions in the World, in million metric tons of carbon dioxide-equivalent (MtCO₂e) per year.

capture technologies must align with sustainability principles to ensure long-term effectiveness and minimal environmental impact. A sustainable CO₂ capture process not only addresses the immediate challenge of reducing CO₂ emissions but also contributes to broader environmental objectives, such as minimizing resource consumption, reducing waste, and limiting energy use.

A sustainable CO₂ capture process should consider several key factors:

Environmental Impact: The capture process itself should minimize negative environmental effects, including toxicity, resource depletion, and ecological disruption. Sustainable capture technologies should aim to reduce or eliminate harmful by-products and ensure that the overall environmental footprint is minimized.

Economic Viability: The economic aspects of CO₂ capture technologies are critical for their widespread adoption. Sustainable technologies should be cost-effective and economically feasible, considering both capital and operational costs. They should also provide a favorable return on investment and contribute to economic growth.

Energy Efficiency: Energy consumption is a significant factor in the overall sustainability of CO₂ capture processes. Sustainable technologies should be energy-efficient, reducing the amount of energy required for capture, separation, and transportation. This consideration is crucial for minimizing the overall carbon footprint of the capture process.

Scalability and Flexibility: Sustainable CO₂ capture technologies should be scalable and adaptable to different industrial applications. They should be capable of handling varying concentrations of CO₂ and be compatible with existing infrastructure (Fig. 2).

By evaluating these factors, this paper aims to provide a comprehensive analysis of the environmental, efficiency, economic, and energy parameters associated with CO₂ capture technologies using deep eutectic solvents (DES) and monoethanolamine (MEA). The study will involve a detailed comparative assessment of several indicators, including condition number (CN), individual risk (IR), total annual cost (TAC), eco-indicator 99 (EI99), mass loss index (MLI), and specific energy intensity (RSEI). These indicators will be analyzed on a per-unit basis, such as per kilogram of product, to standardize performance assessments across various scales and facilitate a more accurate comparison of each technology's feasibility and effectiveness.

In earlier studies, Romero-García [14] presented a comprehensive techno-economic analysis of CO₂ capture alternatives using

Monoethanolamine (MEA) as a solvent, underscoring its potential across a wide range of economic scenarios. Similarly, Martínez-Lomovskoi [15] investigated CO₂ capture processes with deep eutectic solvents, primarily assessing their economic and environmental performance. While these studies provide valuable insights into specific aspects of CO₂ capture technologies, there remains a critical need to evaluate these processes within a broader sustainability framework.

To truly advance the field, it is necessary to move beyond isolated economic and environmental analyses and adopt a more integrated approach. A holistic assessment should encompass not only the economic feasibility but also include metrics such as energy efficiency, process safety, and operational flexibility. These additional considerations are crucial to ensure that CO₂ capture technologies are not only cost-effective but also energy-efficient, inherently safer, and adaptable to varying operational conditions. A comprehensive sustainability evaluation will provide a more robust understanding of these technologies, supporting their advancement toward meeting global climate and sustainability goals.

The contribution of this paper lies in addressing the critical gap in existing research by providing a detailed and comprehensive comparative analysis of traditional and novel CO₂ capture technologies within a sustainability framework. By integrating a broad range of performance and sustainability indicators, this study aims to offer valuable insights into the development of CO₂ capture technologies that align with the principles of sustainability and the goals of the 2030 Agenda. The findings of this paper will contribute to advancing the field of CO₂ capture by highlighting the potential benefits and limitations of different technologies and providing guidance for future research and implementation.

2. Performance indexes and methodology

Creating or modifying processes towards sustainability necessitates a comprehensive evaluation of the proposed innovations to ensure that they align with the principles of sustainable manufacturing. This assessment must consider multiple dimensions, including economic viability, energy efficiency, environmental impact, and process safety and control. These dimensions are critical as they directly influence the sustainability and material efficiency of the manufacturing process. According to González and Constable [16] a systematic approach to sustainability must incorporate a balanced consideration of economic, environmental, and engineering factors. They emphasize that sustainability is not only about reducing environmental footprints but also about ensuring that processes are economically viable and operationally stable over time.

In this article, a set of carefully selected metrics was used to evaluate the sustainability of a process, encompassing various essential indicators. The Eco-indicator 99 (EI99) was chosen for assessing the environmental impact, as it provides a comprehensive analysis of the potential harm caused by a process to human health, ecosystem quality, and resource depletion. This aligns with the principles outlined by González and Lund [9] in their work on green metrics, where the importance of quantifying environmental impact using robust and well-established methodologies is underscored.

Economic feasibility was assessed through the Total Annual Cost (TAC), a metric that provides insight into the overall financial sustainability of the process, which is essential for ensuring that the innovations can be maintained and scaled in real-world applications. The dynamic behavior of the process was evaluated using the Condition Number, a critical indicator of the controllability and stability of the process, which are paramount for long-term operational success. Safety considerations, integral to sustainable process design, were measured using the Risk Index (IR), ensuring that the process not only performs well but also minimizes potential hazards to human health and the environment.

Energy efficiency, another cornerstone of sustainable design, was evaluated using the Specific Energy Intensity (RSEI), which reflects the

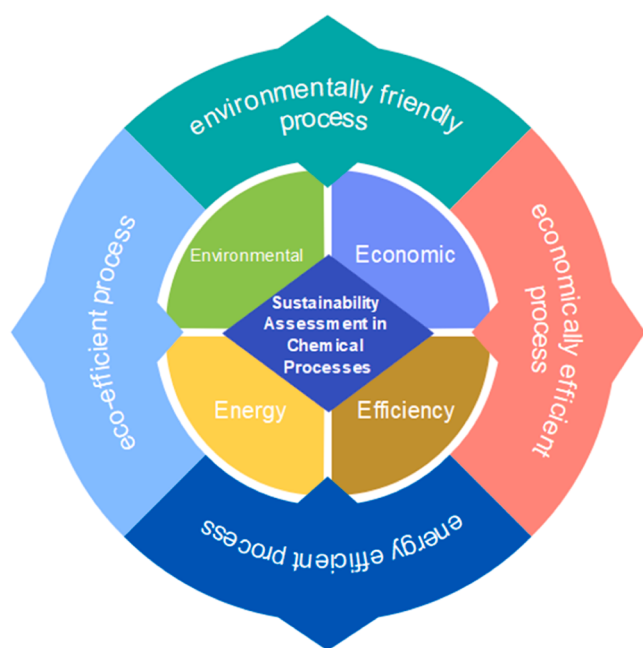


Fig. 2. Synergy in the analysis of sustainability indicators.

energy consumption relative to the production output. This metric is particularly relevant in light of global energy challenges and the need to minimize energy use while maintaining productivity. Lastly, the Mass Loss Index (MLI) was employed to measure process efficiency, ensuring that material wastage is minimized, thereby enhancing resource efficiency—a key aspect of green chemistry principles as discussed by González and Constable [16].

Together, these metrics provide a holistic evaluation framework that aligns with the multidimensional nature of sustainability. By incorporating these indicators, the analysis in this article is grounded in a robust theoretical foundation, ensuring that the proposed process innovations are not only theoretically sound but also practically viable, environmentally responsible, and economically feasible.

2.1. Eco-indicator 99 (EI99)

The Eco-indicator 99 (EI99) is associated with sustainability because it evaluates the environmental impact of a product or process across its entire life cycle, considering multiple environmental factors such as human health, ecosystem quality, and resource depletion. This comprehensive assessment helps identify and reduce significant environmental burdens, aligning with global sustainability goals like those in the UN's 2030 Agenda [17]. Eco-indicator 99 is one of the most widely used environmental impact estimation methods and consists of a quantitative analysis of the life cycle evaluated from beginning to end. The calculation is performed using Eq. (1). Where ω represents the damage weight factor (Pts/kg), C_i represents the impact value for each of the categories i , and α_j is the value of subcategory j (kg/year). This methodology accounts for the origin of raw material in processing and degradation. It is based on standard ecological indicators, which are numbers that express the total environmental burden of a product or a process. The higher the value of the indicator, the greater the environmental impact. Therefore, the best scenario is the lowest possible EI99.

$$EI99 = \sum_i \sum_j \omega \cdot C_i \cdot \alpha_j \quad (1)$$

2.2. Total annual cost (TAC)

The Total Annual Cost (TAC) is linked to sustainability because it ensures the long-term economic viability of a process by accounting for all costs, including capital, operating, and maintenance expenses. By focusing on minimizing TAC, companies are encouraged to optimize resource use, reduce waste, and improve energy efficiency, which aligns with sustainable practices. Additionally, TAC promotes a balance between short-term financial gains and long-term sustainability goals, ensuring that economic decisions support sustainable development [13]. The TAC assumes the annualization of the investment cost of the main process equipment over a 10-year amortization period. To calculate it, Eq. (2) is used. Where $C_{TM,i}$ is the capital cost of the equipment in dollars (\$), r represents the payback period in years, and $C_{ut,j}$ is the cost of cooling and heating services, in dollars per year (\$/year). The objective function evaluates the lowest annual cost of the process while taking into account the utilities used by the plant and various units. The best scenario is a total annual cost as low as possible to recover the investment.

$$TAC = \frac{\sum_{i=1}^n C_{TM,i}}{r} + \sum_{j=1}^n C_{ut,j} \quad (2)$$

2.3. Condition number (CN)

The Condition Number (CN) is a valuable metric for sustainability because it evaluates the controllability and stability of a process, which are essential for ensuring efficient, safe, and resilient operations [18]. A lower CN indicates greater process stability and easier control, leading

to more efficient operations with reduced resource waste, which is crucial for sustainable practices. Additionally, a low CN minimizes the risk of accidents or failures, aligning with sustainability by promoting safer processes that protect human health and the environment [19]. This metric also supports the 11th principle of Green Chemistry, which emphasizes real-time analysis and control to prevent pollution, further reinforcing its importance in sustainable process design and operation [20]. The Condition Number quantifies the sensitivity of the system to inaccuracies in process parameters and mode errors. Systems with small condition numbers present better control properties. Its calculation is performed as shown in Eq. (3), where (σ_*) is associated with the direction in which the system has more difficulty moving. On the other hand, the magnitude of (σ^*) indicates the easiest direction the system will move to.

$$\gamma^* = \frac{\sigma^*}{\sigma_*} = \frac{\text{maximum singular value}}{\text{minimum singular value}} \quad (3)$$

Although the Condition Number is reported numerically, its interpretation is qualitative. Its representation makes sense when compared with other designs. The design with the lowest value of condition number, compared to all comparative designs, is the one that presents the best control properties. The nitrogen (N_2) purity of the first column was monitored while the inlet flow of the flue gas to the absorber feed (first column) was perturbed. For the second perturbation, the CO_2 purity of the second column was monitored while perturbing by the reflux ratio. Finally, for the third perturbation, the solvent purity in the bottom flow from the second column was monitored while perturbing the reboiler duty in the same column.

2.4. Individual risk (IR)

The Individual Risk (IR) metric is crucial for sustainability because it evaluates the likelihood of accidents or harmful events affecting individuals, thus supporting the design of safer industrial processes. By quantifying potential risks to human health and safety, IR ensures that processes prioritize safety, reducing the chance of incidents that could harm people or damage the environment [21]. This focus on minimizing risks aligns with the broader goals of sustainability, including regulatory compliance and social responsibility. Moreover, IR supports Sustainable Development Goals (SDGs) such as Good Health and Well-being (SDG 3) and Decent Work and Economic Growth (SDG 8) by promoting safe working conditions and protecting human health, which are essential components of sustainable development [22]. The IR identifies the risk that a person faces based on his position, including the likelihood of an accident resulting in death or serious injury. The IR is defined as shown in Eq. (4). Where, f_i represents the recurrence that one accident will occur, and $P_{x,y}$ is the likelihood that the accident will occur in a particular location.

$$IR = \sum f_i P_{x,y} \quad (4)$$

In the methodology of this project, the instantaneous and continuous risk analysis was performed for each of the equipment involved in the CO_2 capture process that is in contact with the solvent of interest. Catastrophic events such as Boiling Liquid Expanding Vapor Explosion (BLEVE), Unconfined Vapor Cloud Explosion (UVCE), Jet Fire, Flash fire, and toxic explosion were evaluated using specific mathematical models for each case reported in the literature. The ideal scenario is the one with the lowest individual risk index (IR), as it would be a less risky process.

2.5. Mass loss index (MLI)

The Mass Loss Index (MLI) is associated with sustainability because it measures how effectively a process converts input materials into the desired product, highlighting resource efficiency and waste generation.

A lower MLI indicates that more of the input materials are being utilized productively, which reduces waste and enhances resource efficiency, aligning with sustainability goals. By identifying and reducing sources of waste, MLI supports waste reduction efforts, thereby minimizing environmental impact and lowering operational costs²¹. This metric also aligns with Sustainable Development Goals (SDGs) such as Responsible Consumption and Production (SDG 12), promoting efficient material use and contributing to global sustainability efforts²⁶. The mass loss index which is the amount of unwanted mass in the reactor outlet per product amount, includes direct emissions or resource use of the process under design [23].

$$MLI = \frac{\text{Total nonproduct mass out of process or process step}}{\text{mass of product}} \quad (5)$$

The ideal scenario would be a result of zero since product recovery would be greater than non-product recovery, that is, 100 % recovery. The worst case would be an MLI of 100 since we would be talking about having more non-products than the desired mass of products.

2.6. Specific energy intensity (R_{SEI})

The Specific Energy Intensity (SEI) is a key metric for sustainability as it measures the energy required to produce a unit of product, reflecting the efficiency of energy use in industrial processes. A lower SEI signifies better energy efficiency, which reduces overall energy demand and greenhouse gas emissions, aligning with sustainability goals to minimize environmental impact. Additionally, optimizing SEI helps lower energy costs, contributing to economic sustainability by achieving significant cost savings [13]. This metric supports Sustainable Development Goal 7 (Affordable and Clean Energy) by promoting more efficient energy consumption, thus advancing global efforts to improve energy efficiency and transition to cleaner energy sources [22]. The SEC is influenced by three main factors: the production process (including feedstock), the efficiency of the production process, and the type of products produced [24].

$$R_{SEI} = \frac{\text{Net energy used as primary fuel equivalent}}{\text{Mass of product}} \quad (6)$$

The ideal scenario would be a result of zero since the product recovery would be greater than the net energy used, but the lower the figure, the greater the product recovery and the lower the energy demand.

2.7. Material circularity index (MCI)

The Material Circularity Index (MCI) is a vital sustainability metric because it measures how effectively materials are reused and recycled within a system, reflecting the principles of a circular economy. By quantifying the proportion of material recovered and assessing how well products are designed for reuse, MCI promotes resource efficiency and waste reduction. Higher MCI values indicate better material recovery and recycling, which supports sustainability by minimizing new resource consumption and reducing environmental impact. This metric aligns with Sustainable Development Goal 12 (Responsible Consumption and Production) by encouraging efficient material use and recycling, thus contributing to more sustainable production practices and less environmental pollution. The Material Circularity Indicator of a product (MCIP) measures the extent to which the linear flow has been minimized and the restorative flow – maximized for the product component, and how long and how intensively a product is used compared to a similar industry-average product [25]. In this respect, the product is assigned a score between 0 and 1.

$$MCI = \frac{\text{Product mass recovered}}{\text{Total material consumption}} \quad (7)$$

Scale ranging from 0 is a linear process and 1 is a circular process.

2.8. Global warming potential

Global Warming Potential (GWP) is a key sustainability metric as it measures the relative impact of various greenhouse gases on global warming compared to carbon dioxide (CO_2). By quantifying how much heat different gases can trap in the atmosphere over a specific period, usually 100 years, GWP provides a standardized method for assessing and comparing their climate change effects. This allows organizations to identify and prioritize the reduction of high-impact gases, supporting effective climate mitigation strategies. GWP is integral to Sustainable Development Goal 13 (Climate Action) as it aids in developing strategies to manage and reduce greenhouse gas emissions, contributing to global efforts to limit temperature rise and enhance climate resilience [26]. Global warming potential can be expressed as follows:

$$GWP = \frac{\text{Total mass of } \text{CO}_2 \text{ equivalents}}{\text{Mass of product}} \quad (8)$$

3. Case study

This case study presents the sustainability analysis by evaluating the different parameters in two scenarios of CO_2 capture in constant flow combustion. Two cases were studied: the use of Monoethanolamine (MEA) and the use of a deep eutectic solvent (DES) based on choline chloride (ChCl) and urea (a common fertilizer) in a 1:2 molar ratio. Monoethanolamine (MEA) has long been recognized as an effective and widely researched solvent for CO_2 capture, with proven efficiency in industrial applications due to its high capture capacity and fast absorption rate. Its extensive use in various CO_2 capture processes attests to its reliability and performance. However, MEA is not without its drawbacks; its desorption process is energy-intensive, and the solvent itself is known to have significant environmental impacts, including toxicity and degradation concerns, which contribute to a higher overall environmental footprint. On the other hand, deep eutectic solvents (DES) have emerged as a promising alternative, synthesized from environmentally benign chemicals such as choline chloride and urea. DES presents an attractive solution with its low toxicity, biodegradability, and minimal environmental impact. Furthermore, DES has shown potential in achieving high CO_2 capture efficiency, making it an appealing choice for sustainable CO_2 capture technologies. By combining environmental friendliness with promising operational performance, DES offers a green alternative to traditional solvents like MEA, addressing the growing demand for more sustainable CO_2 capture methods. For each case study the main fuels used in electricity production were considered, i.e. natural gas and coal, also biogas is considered as a green option biofuel, the flue gas compositions used for the different case studies are shown in Table 2. The choice of these fuels is due to the fact that they are the most widely used in the combustion zone in the geographical area referred to both case studies. To evaluate the different indicators within the sustainability framework, we used the three different scenarios for each combustion fuel for both MEA and DES. i.e. the most costly design, the least costly design, and the balanced scheme obtained from [14]. As mentioned in the introduction section, there are no works where a

Table 2
Fuel composition in mass and mole fraction [14].

		CH_4	C_2H_6	C_3H_8	i-C ₄ H ₁₀	N_2	CO_2
Natural Gas	Mass	0.96	0.018	0.004	0.001	0.008	0.009
	Mole	0.98	0.009	0.001	0.0004	0.004	0.003
Biogas	Mass	0.6	–	–	–	0.02	0.38
	Mole	0.8	–	–	–	0.015	0.185
Coal		C	H	O	N	S	
	Mass	0.782	0.052	0.136	0.013	0.017	
	Mole	0.51	0.41	0.066	0.007	0.004	

comparative evaluation of CO₂ capture schemes using DES or MEA as solvent was addressed.

2.1. Case study 1: monoethanolamine (MEA) as a solvent

According to the work presented by Romero García et al. [14]. The CO₂ capture plant used as solvent an aqueous solution of monoethanolamine (MEA) at 30 wt% and a continuous flow. The process consists of an absorption column and a desorption column at the end of the process (Fig. 3).

The results obtained by Romero-García et al. [14] were presented in Pareto fronts as shown in Fig. 4 which is an extract of their results. Their work highlights that the use of Biogas increases the economic impact but also this impact is diminished due to the high recovery of CO₂. Also, because of some operative variables, the system would not present good controllability. In the case of coal, there is a trend between EI99 and CN, when both are low the ROI is also low, so the process is not economically profitable when there is good controllability and good environmental impact.

2.2. Case study 2: use of a deep eutectic solvent (DES)

To implement environmentally responsible CO₂ solvents, the use of deep eutectic solvents (DEPs) for CO₂ capture was proposed by [15] claiming some advantages over amines. Particularly, the advantages are due to the non-toxic and non-corrosive nature of many of them and their high thermal and oxidative stability. [15], carried out the design and optimization of a carbon capture plant taking up the case study of [14] but now using novel green DES aqueous ChCl/ urea (1:2). The process is composed of an absorber and desorber as the traditional process but with the implementation of two flash tanks to treat the DES before desorption (Fig. 5).

The results obtained by Martinez-Lomovskoi et al. [15] are presented in pareto fronts as the extract shown in Fig. 6, where scenarios using fuels such as natural gas (NG), biogas (BG), coal (C), and associated gas (AG) were shown. The results of the design and simulation of CO₂ capture with the use of deep eutectic solvents, obtained reveal that the tendency towards the selection of the design with a lower eco indicator 99 (EI99) causes the total annual cost (TAC) to increase. The Coal process provides the lowest energy use per unit of CO₂ captured when compared to each of the gaseous fuels as the energy savings are reflected in the TAC value of coal which is 12.0 %, 27.3 %, and 32.4 % lower per ton of CO₂ captured, compared to GA, NG, and BG, respectively. In terms of environmental impact, the Coal process presents a 20.1 %, 15.7 %, and 30.2 % lower value per ton of CO₂ captured compared to GA, NG, and BG, respectively. The flash stages (Flash 1 and Flash 2) play a critical

role in preconcentrating the effluent from the initial absorption stage, as depicted in Fig. 5. These stages are designed to mitigate separation conditions that would otherwise result in increased operational costs, such as the need for vacuum operation or the use of low condenser temperatures.

In the process, the gaseous effluent enters the bottom of the absorption column in a countercurrent flow relative to the solvent. Following the absorption process, a stream rich in O₂ and N₂ exits the dome of the absorption column, while the deep eutectic solvent (DES), now enriched with CO₂ as well as a certain proportion of O₂ and N₂, exits from the column's bottom. To effectively remove a significant amount of O₂ and N₂ from the DES-rich stream, and to prevent the necessity of harsh separation conditions in the DES regeneration column, two flash separation units are incorporated into the system. In both flash units, O₂, N₂, and trace amounts of CO₂ are released in the 'upper' stream, thereby minimizing their impact on the overall CO₂ recovery. Ultimately, in the DES recovery column, CO₂ is expelled through the dome, and high-purity DES is recovered at the bottom to be reintroduced into the absorption column.

As mentioned so far, the suitability of using MEA and DES as solvents has been already explored. However, a comparative analysis has not been performed to select the best solvent for CO₂ capture or develop a wider picture of the correlation between solvent and fuel in the combustion stage. Therefore, taking this as a preamble, in this article an evaluation of the parameters of the three selected designs of each fuel is performed.

From the Pareto front, three designs were selected, the most expensive (high), the least expensive (low), and the intermediate data (central), flow data were retrieved for each design, depending on the fuel used (natural gas, coal, and biogas). All schemes were simulated in Aspen Plus. In the context of the Pareto front presented in Fig. 6, the figure evaluates the relationship between Total Annual Cost (TAC) and Environmental Impact (EI99). The observed trend indicates an inverse correlation, where an increase in TAC corresponds to a decrease in environmental impact, and vice versa. This trade-off can be attributed to two key factors. First, TAC is influenced by both capital and operational costs, while EI99 is highly sensitive to the amount of heat utilized in the process. This interplay results in the region where the highest TAC and lowest EI99 values are observed, which corresponds to process designs characterized by low energy consumption but compensating for this with larger equipment sizes. Specifically, these processes exhibit "low" energy consumption but require larger columns with greater diameters.

Conversely, the region where TAC is lower but EI99 increases is primarily associated with equipment designs that follow a different approach, contrasting with the previously explained design philosophy. In this part of the Pareto front, processes typically feature fewer equilibrium stages in their columns but require higher energy consumption. In the middle section of the Pareto front, both TAC and EI99 values are closer to the origin, suggesting that the most optimal solutions, in terms of both cost and environmental performance, are found in this area.

Although all the designs presented on the Pareto front meet the recovery and purity constraints, there may be variations in terms of TAC, EI99, and equipment design.

In summary, the evaluation of the different parameters involved the use of three different platforms (Fig. 7). For the condition number (CN) calculation, perturbations were performed on each of the designs from the Aspen Plus platform, and these results were evaluated in MATLAB. For the evaluation of the other parameters, the data collected from the different Aspen Plus designs were used to be used in the different methodologies for each calculation, programmed in Excel.

4. Results

This section details the results of the sustainability assessment of different CO₂ capture processes, alongside a critical discussion of the values obtained and their broader implications for the choice of solvent.

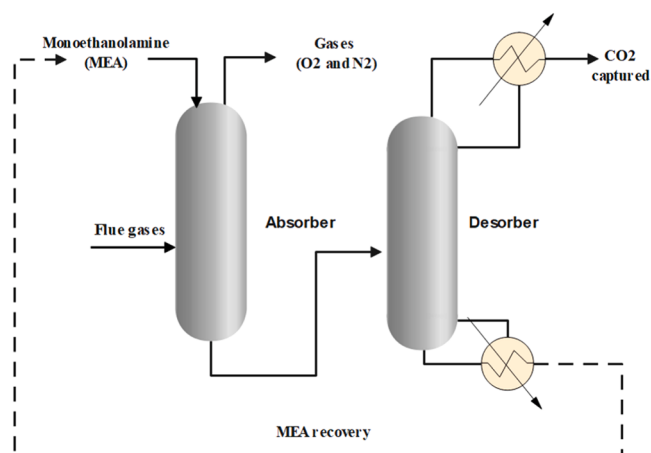


Fig. 3. The post-combustion capture process (PCC) using MEA.

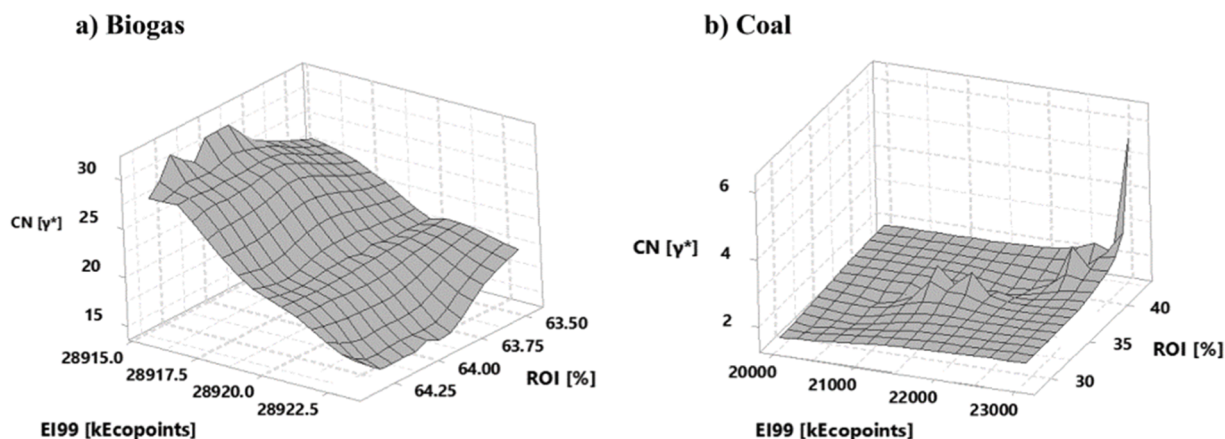


Fig. 4. Pareto fronts of MEA used as a solvent.

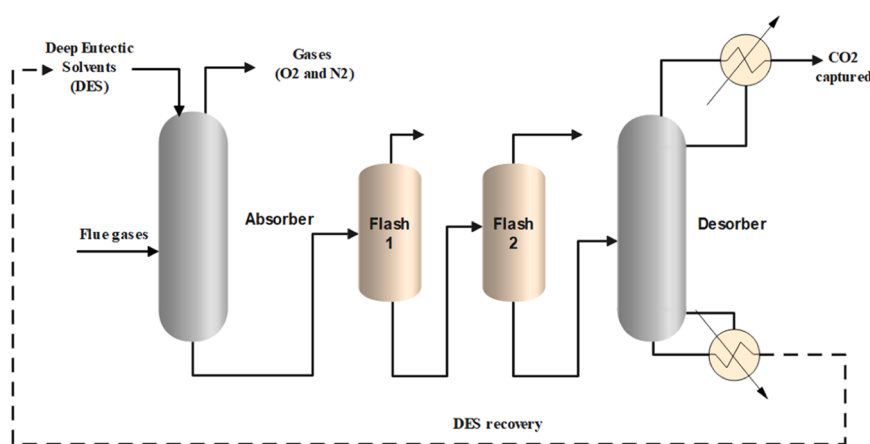


Fig. 5. The post-combustion capture process (PCC) using DES.

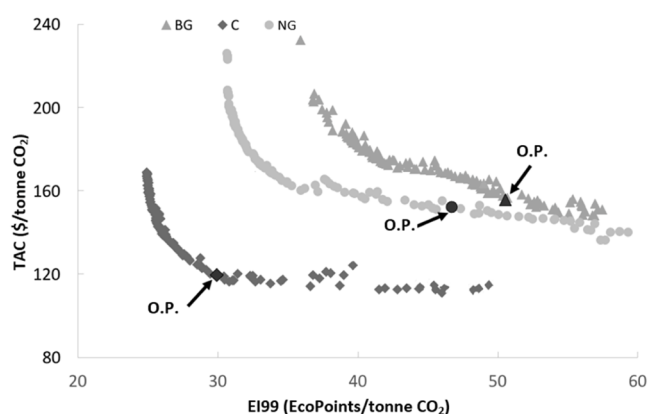


Fig. 6. Pareto fronts of DES used as a solvent.

The first case study evaluates natural gas as the combustion fuel, with the results displayed in Fig. 8.

In terms of process controllability, measured by the condition number (CN), designs using Monoethanolamine (MEA) as a solvent consistently exhibited the smallest CN values across all schemes. This suggests that MEA-based systems offer greater operational stability, allowing for more precise control under varying conditions. From an industrial perspective, this feature is highly valuable as it reduces the likelihood of operational inefficiencies or disruptions, which can result in additional costs or safety concerns. The observation that the lowest CN values

coincided with the lowest total annual cost (TAC) highlights a key relationship: better-controlled processes are often more cost-effective due to their ability to minimize deviations, reduce energy consumption, and require fewer corrective measures.

When examining the total annual cost (TAC) per kilogram of CO₂ captured, MEA-based processes demonstrated a significant cost advantage over those using Deep Eutectic Solvents (DES). The TAC for MEA schemes was approximately 43.4 % lower than that for DES-based schemes. This is a substantial difference, underlining the economic efficiency of MEA in large-scale applications. The Eco-indicator 99 (EI99), which assesses the overall environmental impact, followed a similar trend, with MEA outperforming DES by approximately 39.7 %. The primary factor contributing to these differences is the reboiler heat duty, a critical component of the energy demand in CO₂ capture processes. MEA-based schemes required a reboiler heat duty of 5536.6 kJ/kg CO₂ captured, significantly lower than the 8735.9 kJ/kg CO₂ required by DES-based schemes. The lower energy demand not only reduces operating costs but also translates into a smaller environmental footprint, as less energy is consumed, resulting in lower greenhouse gas emissions and reduced environmental degradation.

Additional sustainability indicators, such as the mass loss index (MLI) and specific energy intensity (RSEI), also favored MEA. The MLI, which measures the efficiency of material usage, showed that MEA schemes had an average value of 15.1 %, compared to 47.9 % for DES schemes. This stark contrast indicates that MEA-based processes are more efficient in converting input materials into the desired product, with less waste generated. The RSEI, which quantifies the energy required per unit of CO₂ captured, further demonstrated the superiority

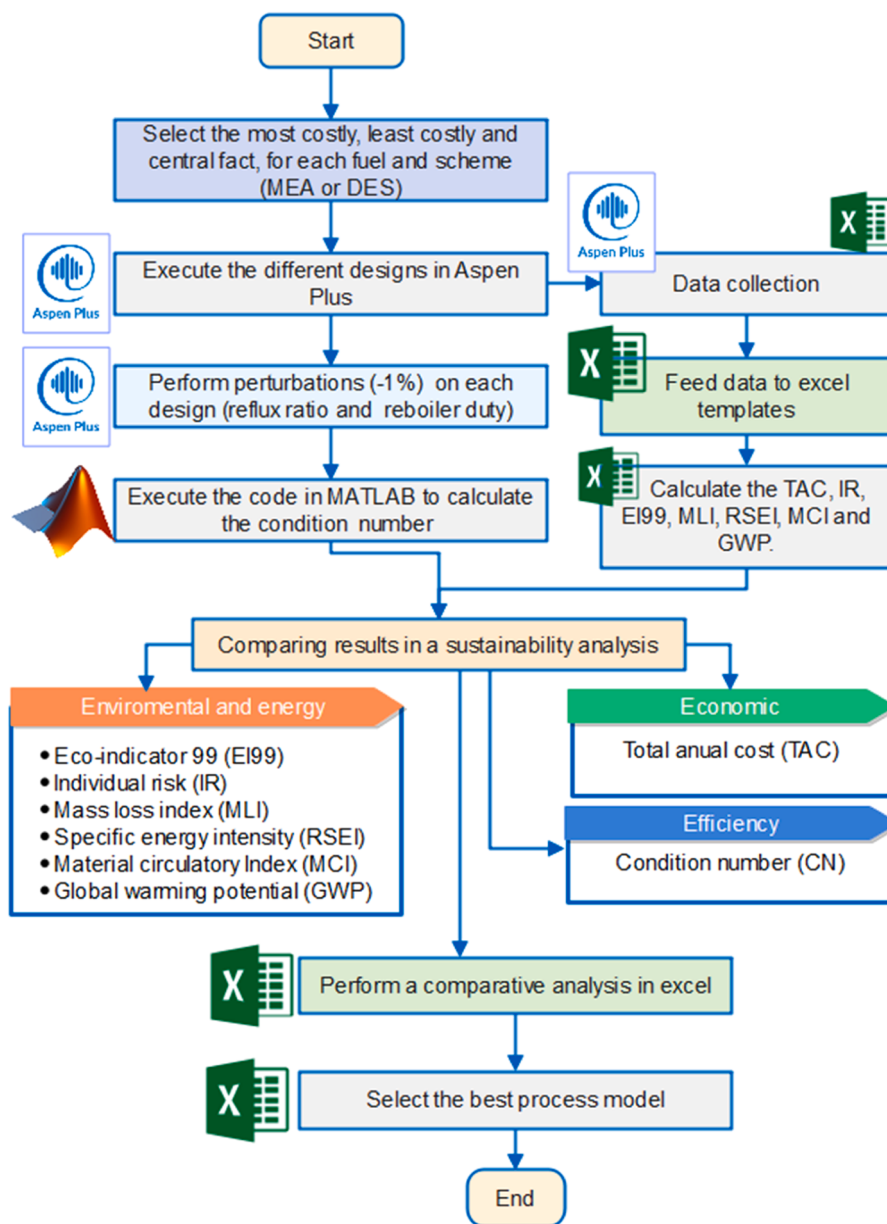


Fig. 7. Methodology for the evaluation of the different parameters.

of MEA, with an average value of 0.084 kg/kg CO₂ captured compared to 0.212 kg/kg CO₂ for DES. These results underscore MEA's strength in material and energy efficiency, both of which are crucial for minimizing resource consumption and lowering operational costs in large-scale implementations.

A detailed comparison of the best designs from the natural gas combustion schemes reveals a clear advantage for MEA in terms of cost efficiency, environmental impact, and energy consumption. While DES-based schemes were initially expected to offer better controllability due to the inherent green chemistry properties of the solvent, the results suggest otherwise. MEA not only provided better overall performance but also outperformed DES in the specific area of process controllability, which is crucial for ensuring the stability and safety of large-scale industrial operations. The superior performance of MEA is largely attributed to its lower reboiler heat duty, which reduces both operational energy requirements and associated costs. Although DES holds potential as a sustainable alternative due to its environmentally friendly composition, it appears that further optimization is necessary for it to become a competitive option in industrial CO₂ capture applications.

In contrast, the results for coal-fired electricity generation, shown in Fig. 9, present a more balanced comparison between MEA and DES. Once again, MEA-based designs exhibited the smallest condition numbers, indicating better control properties. This is particularly important for coal-based processes, where operational stability is often more challenging due to the nature of the fuel and the associated emissions. The larger column diameters observed in MEA schemes likely contribute to this improved controllability, as they provide greater resilience to process disturbances. This increased resilience allows MEA schemes to maintain operational stability even under varying conditions, further enhancing their suitability for large-scale industrial applications.

The lowest TAC per kilogram of CO₂ was found at the center point of the Pareto front for MEA-based schemes, with reductions of 3.3 % and 74.2 % compared to the high and low point designs, respectively. The DES-based schemes, while effective, showed a smaller TAC reduction of 9.1 %, indicating that MEA remains the more cost-efficient option in this context. The EI99 for MEA-based schemes was also approximately 9.1 % lower than for DES-based schemes, a difference that can be traced back

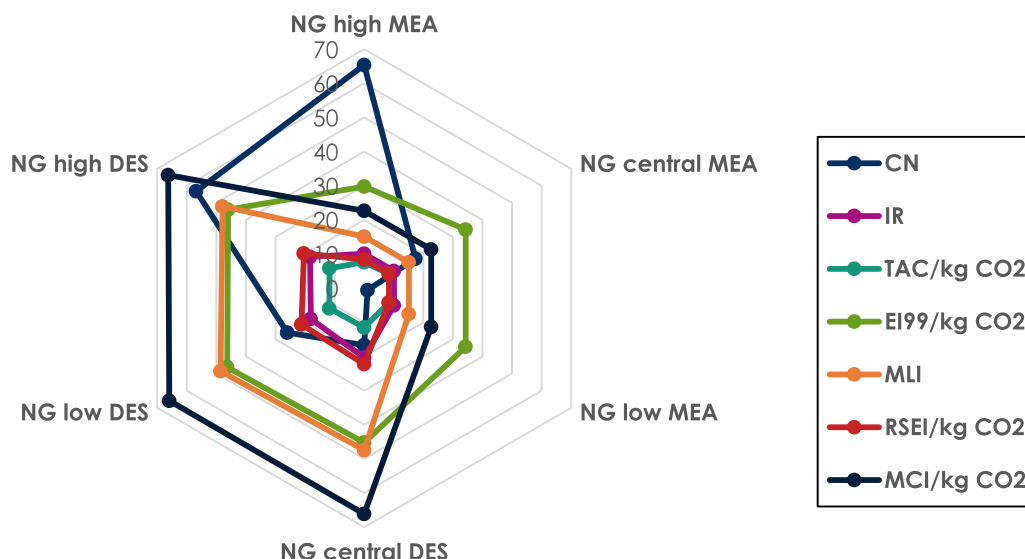


Fig. 8. Radial graphs for CO₂ capture with MEA and DES as solvents, using natural gas (NG) as fuel. Evaluating the indicators of condition number (CN), individual risk (IR), total annual cost (TAC), eco-indicator 99 (EI99), mass loss index (MLI), and specific energy intensity (RSEI).

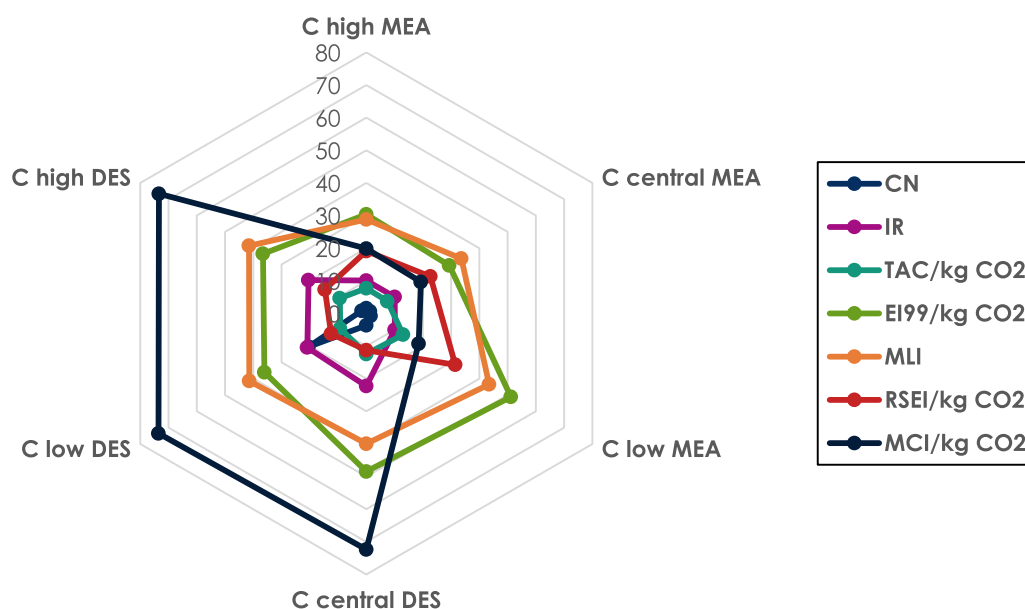


Fig. 9. Radial graphs for CO₂ capture with MEA and DES as solvents, using coal (C) as fuel. Evaluating the indicators of condition number (CN), individual risk (IR), total annual cost (TAC), eco-indicator 99 (EI99), mass loss index (MLI), and specific energy intensity (RSEI).

to the energy-intensive nature of DES processes. The average reboiler heat duty for DES was 7520.8 kJ/kg CO₂ captured, compared to 6751.4 kJ/kg CO₂ for MEA processes. Although this difference may appear moderate, it has significant implications for both cost and environmental impact, as higher energy consumption directly increases both the TAC and EI99.

Interestingly, when evaluating specific energy intensity (RSEI), DES exhibited better performance than MEA. This indicates that while DES processes may require higher energy input overall, they are more efficient in utilizing that energy for the specific task of CO₂ capture. This suggests a potential niche for DES in applications where energy efficiency is the primary concern, though it must be weighed against the higher costs and environmental impacts associated with its use. This trade-off highlights the importance of considering the full spectrum of sustainability metrics when selecting a solvent for CO₂ capture.

For biogas-based CO₂ capture (Fig. 10), DES outperformed MEA in

several key areas. DES-based schemes exhibited TAC reductions of approximately 60.7 % compared to MEA schemes, along with a 53 % reduction in EI99. These substantial improvements in DES's performance can be attributed to the lower reboiler heat duty in DES schemes (4×10^8 kJ/h), compared to 4.7×10^8 kJ/h in MEA schemes. This lower energy requirement leads to reduced operating costs and a smaller environmental impact, making DES a more attractive option for biogas-based applications. However, despite DES's cost and environmental advantages, MEA-based schemes generated fewer by-products and demonstrated a lower MLI, indicating greater efficiency in material usage.

While DES offers a clear advantage in terms of cost and environmental impact in biogas-based processes, MEA remains more efficient when considering energy investment for CO₂ capture. This highlights a key trade-off between the two solvents: DES excels in reducing overall economic and environmental burdens, while MEA provides better

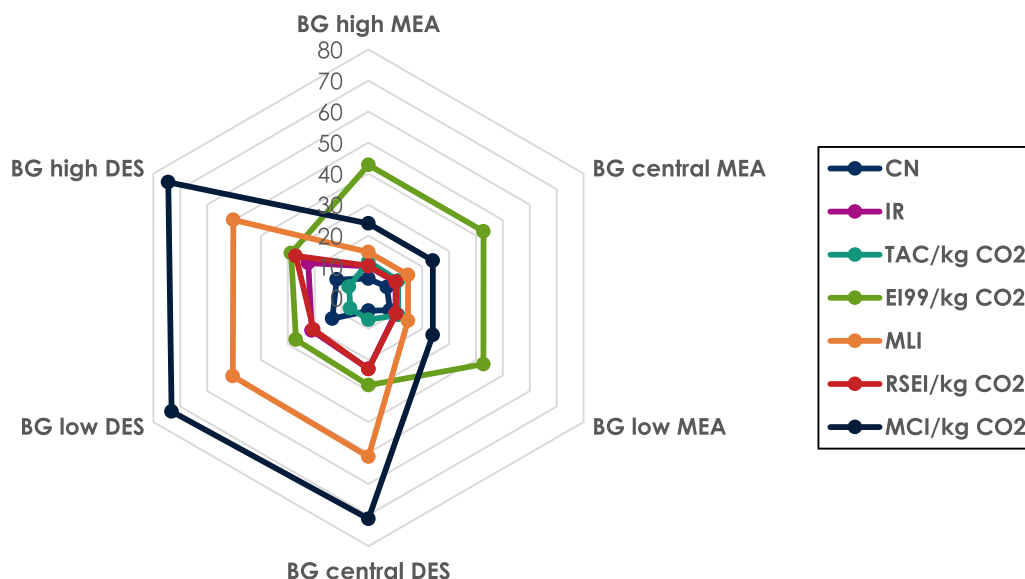


Fig. 10. Radial graphs for CO₂ capture with MEA and DES as solvents, using biogas (BG) as fuel. Evaluating the indicators of condition number (CN), individual risk (IR), total annual cost (TAC), eco-indicator 99 (EI99), mass loss index (MLI), and specific energy intensity (RSEI).

control over energy and material flows, resulting in higher overall process efficiency.

To further refine the understanding of these trends, a comparative analysis was conducted by selecting the best designs for each solvent and fuel combination. In general, DES emerged as the more sustainable option across most indicators, particularly in biogas-based processes. However, MEA consistently demonstrated superior energy use and process integration, which is critical for large-scale operations that demand high levels of stability and control. This suggests that while DES may be more suitable for niche applications where environmental sustainability is the primary concern, MEA remains the better option for industrial-scale CO₂ capture due to its balanced performance across economic, environmental, and operational metrics.

The circulatory material index (MCI), which measures the degree to which materials are reused within the process, also provided important insights. For DES-based schemes, the MCI ranged from 0.65 for natural gas processes to 0.74 for biogas processes, indicating a greater degree of material recirculation and sustainability. In contrast, MEA-based schemes exhibited MCI values between 0.18 and 0.23, reflecting a more linear process with lower material recovery. These results suggest that DES, despite its higher energy requirements, may offer better long-term sustainability in processes where material efficiency and waste reduction are prioritized.

Based on these findings, it is evident that there is no one-size-fits-all solution for CO₂ capture. MEA is the better option for natural gas and coal-fired electricity generation, offering lower costs and environmental impacts. DES, on the other hand, excels in biogas-based processes, delivering significant reductions in both TAC and EI99. This underscores the importance of tailoring solvent selection to the specific fuel used in the process, ensuring that trade-offs between cost, energy efficiency, and environmental sustainability are appropriately balanced.

One important consideration is the inherent safety of DES-based processes. As shown in Figs. 8–10, DES schemes consistently exhibited higher process risks compared to MEA. This is largely due to the additional equipment required for DES-based processes, such as the flash tanks, which introduce greater complexity and increase the potential for operational failures. Although DES solvents are less corrosive and more environmentally friendly, their use in large-scale industrial processes must be carefully managed to mitigate these risks.

Table 3 presents the calculated data for the Global Warming Potential (GWP) associated with the use of DES and MEA. These data provide clear insights into the long-term climate impact of each solvent option. Specifically, the use of MEA results in an increased GWP when natural gas (NG) and biomass gasification (BG) are utilized as fuels. In contrast, the application of DES as a solvent lead to a reduction in GWP when coal is used as the fuel.

Based on the sustainability indicator classification proposed by Ruiz-Mercado [27], the indicators assessed in this study can be categorized into six key dimensions: environmental impact, efficiency, energy consumption, economic feasibility, safety, and controllability. Upon evaluating all the indicators, it becomes possible to identify the most feasible solvent considering these criteria, as well as to assess the influence of the fuel type used. Specifically, the optimal solvent for CO₂ capture is determined by the combined analysis of all evaluated indices, along with the nature of the fuel utilized (See Fig. 11). While these findings focus on comparing the use of ChCl:U as an eutectic solvent with MEA as a conventional solvent, they provide valuable insights into the broader considerations involved in selecting an appropriate solvent for the CO₂ capture process.

An interesting point to consider is the role that the CO₂/C composition plays in each fuel. In a comparative manner, it is possible to assess the amount of carbon (C) supplied to the combustion chamber for each

Table 3

Global warming potential index (GWP) in CO₂ capture process with MEA and DES as solvents, using natural gas (NG), coal (C) and biogas (BG) as fuel in 100-year time horizon.

	MEA			DES		
	Natural Gas	Coal	Biogas	Natural Gas	Coal	Biogas
High point	65.83788	33.77784	67.5683	42.29183	57.6167	42.80369
Central point	65.90676	29.19377	67.56825	42.26607	57.58439	42.89697
Low point	65.90676	22.77071	67.56838	42.29113	57.59642	42.88741

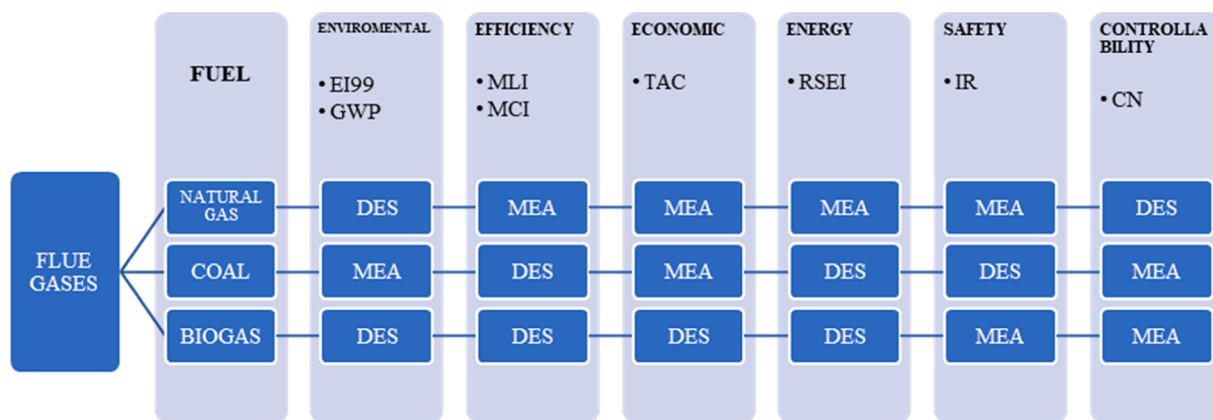


Fig. 11. Solvent recommendation for use in the CCP process based on the evaluation of sustainability indicators.

fuel (see Table 4). From this, it is apparent that the fuel with the lowest effective carbon content (coal) demonstrated better controllability in the evaluated designs. Additionally, a lower impact was observed in the indicators categorized as environmental, though there was a tendency for higher values in those grouped under energy usage. On the other hand, as the net carbon input increases, both the Total Annual Cost (TAC) and the environmental impact also increase, while process controllability decreases. Overall, it could be assumed that capture processes working with a lower effective carbon content in the capture process perform better compared to those fed with a higher amount of effective carbon.

5. Conclusions

This study evaluated the performance of two solvents, monoethanolamine (MEA) and a deep eutectic solvent (DES), for CO₂ capture from combustion processes using natural gas, coal, and biogas as fuels. The results highlight the potential for these solvents, while also pointing to areas for further exploration to enhance sustainability and efficiency in CO₂ capture technologies.

Our analysis revealed that MEA offers significant advantages in economic performance and environmental impact when natural gas is used, with a 43.4 % lower total annual cost (TAC) and a 39.7 % reduction in the Eco-indicator 99 (EI99) compared to DES. MEA also demonstrated a 15.1 % lower Material Loss Index (MLI) and superior controllability, as indicated by a lower condition number and reduced Relative Sustainability Energy Intensity (RSEI). For coal-fired processes, MEA showed similar trends, with a 9.1 % reduction in TAC and a 9 % decrease in EI99, and an MLI 13.7 % lower than that of DES. Conversely,

DES showed its strengths in biogas-based CO₂ capture processes, outperforming MEA with a 60.7 % lower TAC and a 53 % reduction in EI99. DES also exhibited lower condition numbers, indicating improved controllability under these conditions. The MLI and RSEI values for DES were similar to those of MEA, suggesting comparable energy requirements.

Despite some challenges, both MEA and DES have proven to be effective solvents for CO₂ capture, with their performance varying depending on the fuel used. MEA, while competitive in many key metrics, faces the significant challenge of CO₂ emissions during its production, which could diminish its environmental advantages. In contrast, DES—thanks to its renewable and biodegradable nature, as well as its favorable performance indicators—represents a promising alternative for sustainable CO₂ capture processes.

As the field of CO₂ capture continues to evolve, there are several promising avenues for future research. One of the key areas for further exploration is the development of next-generation solvents that can combine the strengths of both MEA and DES while mitigating their individual weaknesses. For MEA, future research may focus on reducing its energy consumption, minimizing solvent degradation, and finding ways to lower its environmental footprint during production. For DES, optimization could focus on enhancing its performance in applications beyond biogas, such as in natural gas and coal-fired processes, by reducing energy demands and improving scalability for industrial use. Another exciting area for future breakthroughs lies in the integration of advanced solvent technologies with renewable energy systems. Coupling CO₂ capture with renewable energy sources, such as solar or wind power, has the potential to dramatically reduce the overall carbon footprint of these processes. Additionally, further exploration of process intensification techniques, such as the use of modular reactors or novel heat integration methods, could result in more energy-efficient and cost-effective CO₂ capture systems.

Finally, as the ultimate goal of CO₂ capture technologies is to achieve a negative carbon footprint, further research should focus on understanding the full lifecycle impacts of both MEA and DES, including their production, use, and end-of-life scenarios. Future work will need to explore circular economy principles, such as solvent recycling, to ensure that the environmental benefits of CO₂ capture processes are maximized.

Thus, this study highlights the comparative performance of MEA and DES for CO₂ capture from natural gas, coal, and biogas combustion. While MEA generally provides better economic and environmental outcomes for natural gas and coal applications, DES proves to be more effective for biogas, offering significant reductions in both total annual cost and environmental impact.

However, the future of CO₂ capture lies in advancing the performance of solvents like DES and MEA while developing new technologies that further optimize energy efficiency, reduce environmental impacts, and improve scalability. The next wave of research and technological

Table 4

Relationship of the composition of the fuel used and the effective mass of carbon in the effluent.

Feed (1000 Kmol/h)	Composition (mol)		Effective carbon mass (kg/h)	Total carbon mass (kg/h)
Natural gas	CH ₄	0.98	11,760	12,067.2
	C ₂ H ₆	0.009	216	
	C ₃ H ₈	0.001	36	
	i-C ₄ H ₁₀	0.0004	19.2	
	N ₂	0.004	–	
	CO ₂	0.003	36	
Biogas	CH ₄	0.8	9600	11,820
	N ₂	0.015	–	
	CO ₂	0.185	2220	
Coal	C	0.51	6120	6120
	H	0.41	–	
	O	0.066	–	
	N	0.007	–	
	S	0.004	–	

breakthroughs will likely focus on novel solvents, hybrid systems, renewable energy integration, and process intensification—bringing us closer to achieving a truly sustainable and carbon-negative future.

The choice between MEA and DES should not only be based on their current performance in CO₂ capture but also consider their future potential. DES, with its renewable, biodegradable materials and lower associated CO₂ emissions, aligns more closely with the goal of achieving a negative carbon footprint. Meanwhile, future innovations in MEA-based technologies could maintain its competitive edge. Continued research and development will be key to optimizing these solvent systems and exploring new technologies that advance sustainability in CO₂ capture processes.

Declaration of generative AI and AI-assisted technologies in the writing process

During the preparation of this work the author(s) used chatGPT in order to correct style and grammar. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

CRediT authorship contribution statement

Melanie Coronel-Muñoz: Writing – original draft, Validation, Software, Methodology. **Ana Gabriela Romero-García:** Writing – original draft, Supervision, Conceptualization. **Brenda Huerta-Rosas:** Writing – original draft, Supervision, Software, Conceptualization. **Eduardo Sánchez-Ramírez:** Writing – original draft, Validation, Supervision, Methodology, Conceptualization. **Juan José Quiroz-Ramírez:** Writing – original draft, Supervision, Software, Methodology, Conceptualization. **Juan Gabriel Segovia-Hernández:** Writing – original draft, Supervision, Conceptualization.

Declaration of competing interest

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

Acknowledgements

The authors acknowledge the financial support provided by Universidad de Guanajuato and CONAHCYT.

Data availability

Data will be made available on request.

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