



Implementation of the Deep Eutectic Solvent, Choline Urea Chloride (1:2), to Evaluate the Sustainability of its Application During CO₂ Capture

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Abstract

Green chemistry aims to create chemical products and processes that decrease or eradicate the need for harmful substances. By using green solvents like Deep Eutectic Solvents (DES) it is feasible to devise new processes or modify current that adhere to the principles of circular economy and green chemistry. In this work is presented the potential use of choline urea chloride (1:2) as a solvent; taking the post-combustion capture of carbon dioxide (CO₂) as a case study. The results demonstrated that the methodology proposed allows the thermodynamic modeling of DES, obtaining a prediction of data very similar to the experimental trends previously reported. In addition, the use of DES in CO₂ capture allowed a 13.97% reduction in environmental impact and required 25.38% less energy than the traditional process based on amine absorption. This led to a reduction in global cost by 32.11% and 19.64% for equipment and services, respectively, and a 21.13% lower cost of operation.

Keywords CO₂ capture · Deep eutectic solvent · Process simulation · Sustainability

Highlights

- It was proved a methodology to implement DES thermophysical properties into the Aspen Plus process simulator.
- The simulation of the thermodynamic behavior followed the trend reported for experimental data.
- Through computer simulation, the use of DES was proved in an industrial application such as CO₂ capture.
- The sustainability of using DES as a solvent was compared against the conventional CO₂ capture process with amine-based solvents.

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Introduction

The carbon dioxide (CO₂) capture process has become an imperative need in our fight against climate change. However, it is not enough to simply capture this greenhouse gas; we must also ensure that the process is designed sustainably. Sustainability becomes a fundamental pillar in this context since it addresses a series of essential aspects for our planet and its future. First, CO₂ capture as part of a sustainable approach is a powerful climate change mitigation measure. Since carbon dioxide is one of the main contributors to global warming, effectively capturing and storing it will help reduce emissions and ultimately limit the rise in global temperatures. In addition, a sustainable perspective ensures that harvest methods are developed with respect for the environment. Some techniques may require the use of chemicals or technologies that, if not properly managed, could have a negative impact on biodiversity and ecosystems. Therefore, it is crucial to ensure that these processes are respectful of nature and minimize any environmental damage. Sustainability also seeks a responsible use of resources. Since CO₂ capture processes can require significant amounts of energy, materials, and water, it is essential to use these resources efficiently to minimize the carbon footprint and achieve greater process

efficiency. In this regard, long-term economic viability also plays a key role. A sustainable CO₂ capture process must be economically viable so that it can be implemented on a large scale and have a significant impact on reducing CO₂ emissions. Only then can we guarantee that these initiatives are sustainable and continue their contribution over time. On the other hand, the social aspect cannot remain in the background. A sustainable approach also considers social impacts, such as the health and well-being of local communities and the workers involved in the CO₂ capture process. Respect for people and the guarantee of their well-being are essential pillars of building a sustainable future. Ultimately, sustainability promotes innovation and the development of cleaner and more efficient technologies¹. The focus on sustainability drives the search for increasingly advanced and environmentally friendly solutions, which favors the transition towards a lower carbon economy and a more conscious and responsible society. To move towards environmentally friendly solutions in carbon capture processes, one of the primary difficulties is finding solvents that meet specific standards including biodegradability, recyclability, affordability, accessibility, and non-toxicity, in order to be considered a viable green option (Chandran et al. 2021). However, there are currently only a few solvents that meet these criteria and are classified as environmentally friendly. As a result, a new kind of green solvents, known as Deep Eutectic Solvents (DESs), is gaining traction in different scientific fields and being extensively researched for their sustainability.

DESs are created by blending two or more compounds that have a melting point lower than either of the individual components. DESs are usually formed by combining a quaternary ammonium halide salt, which acts as a hydrogen bond acceptor, with a hydrogen-donor molecule that interacts with it through hydrogen bonding to create a eutectic mixture (García et al. 2015). As green solvents, DESs are a viable alternative even to replace Ionic Liquids, since they have superior biodegradability, are non-toxic, have a lower cost, and maintain their thermodynamic advantages (Krishnan et al. 2020; Marchel et al. 2022). The DESs have a wide variety of applications such as solvents for the extraction and separation of various substances. In biodiesel and biomass processes, DESs have been applied to separate glycerol from biodiesel, and as cellulose pretreatment agents (Marcus 2019). Likewise, some DESs have high selectivity and efficiency to absorb CO₂ (K. Zhang et al. 2018).

An example that shows the potential use of a DES as green solvent is its application as a solvent in CO₂ capture plants. There are different DESs such as choline chloride/ethanolamine, choline chloride/L-arginine/glycerol, and choline chloride/1,2-propanediol (Y. Liu et al. 2021). However, it has been found that the combination of choline chloride with urea forms a liquid solvent called choline chloride:urea

(ChCl:U), also known as reline, which has a high capacity to selectively absorb CO₂ from gas mixtures (F. Liu et al. 2019).

Because DESs are relatively new solvents, their properties and potential uses have only been tested experimentally. In order to better explore these types of novel solvents in a rigorous but controlled environment, commercial simulators are useful. This work aims to present a strategy to implement the thermophysical properties of ChCl:U in the Aspen Plus process simulator, to guarantee the reliability of the calculation of the physicochemical properties, and to be able to present the potential use of choline urea chloride (1:2) as a solvent into a sustainable post-combustion capture of carbon dioxide. The objective is to verify that the use of eutectic liquids is more profitable, friendly to the environment, and safer than the conventional carbon capture process using amines as solvent.

Problem Statement

The urgent need to address global environmental damage and mitigate the effects of global warming and climate change is emphasized. The excessive emissions of greenhouse gases, particularly CO₂, have led to increased concentrations since the Industrial Revolution (Chen 2021). The burning of fossil fuels for energy has contributed significantly to anthropogenic CO₂ emissions, resulting in global warming. The current rate of global warming is alarming, with projections from the Intergovernmental Panel on Climate Change (IPCC) suggesting a likely increase of 274.5 K between 2030 and 2052 if the current trend persists (Djalante 2019).

Different technologies have been developed to address CO₂ emissions. These technologies can be classified as post-combustion CO₂ capture (CO₂ is captured from released gases after fuel combustion), pre-combustion CO₂ capture (converts fossil fuels into gas, collecting CO₂ while utilizing H₂ for energy production), and oxy-combustion (employs pure O₂ during combustion, resulting in the production of CO₂ and H₂O) (Dutcher et al. 2015). Post-combustion CO₂ capture is extensively researched due to its adaptability to existing CO₂ sources.

Several articles in the field of CO₂ capture focus on post-combustion techniques, including adsorption with chemical or physical solvents, separation using membranes, and adsorption with solid sorbents (Bhattacharyya and Miller 2017; Ochedi et al. 2021; Raganati et al. 2021; M. Wang et al. 2015a, b). It is noteworthy that traditional organic amine solvents are commonly used for CO₂ capture due to their reversible reaction with CO₂ (D'Alessandro et al. 2010; Shannon and Bara 2012). Amine-based technologies are currently considered the most advanced, cost-effective, and

efficient methods for industrial-scale CO₂ capture (Kenarsari et al. 2013) and are thus being further developed and implemented. However, traditional organic amine solvents used for CO₂ capture have drawbacks that conflict with sustainability principles. These solvents require high energy for regeneration, result in significant solvent loss, and have a negative environmental impact due to secondary pollution (Olajire 2010). The main waste product from post-combustion carbon capture is ammonia, produced through the oxidative degradation of amines. However, other volatile organic compounds and amines may also be released, including potentially carcinogenic substances like nitrosamines and nitramines, raising environmental concerns (Spietz et al. 2020).

In recent decades, alternative solvents like ionic liquids have been investigated for CO₂ capture (X. P. Zhang et al. 2012a, b). Early studies highlighted their advantageous properties such as high thermal stability, strong solubility, low vapor pressure, and designable structure (Mesbah et al. 2019). However, issues such as poor biodegradability, high toxicity, expensive precursors, and complex synthesis routes have limited their industrial applicability (Luo et al. 2021). In contrast, DESs, which share similarities with ionic liquids, have emerged as potential substitutes. DESs offer advantages like low cost, biodegradability, and ease of preparation (Florindo et al. 2019). Their sustainable properties make them promising candidates for future CO₂ capture applications, potentially replacing traditional organic amine solvents.

Case Study Description

For this study, it was proposed a strategy for the implementation of a DES in the Aspen Plus process simulator. The considered case study was the design of CO₂ post-combustion capture proving that DES can be used as a sustainable alternative at the industrial level for CO₂ capture.

According to several authors, DESs, due to the nature of their components, are considered green solvents (Babaei and Haghtalab 2023; Hooshmand et al. 2023; Y. Liu et al. 2021). In addition, they have demonstrated high efficiency to capture CO₂, minimal environmental impact, limited tendency to evaporate, broad range of liquid states, compatibility with water, lack of flammability, absence of toxic qualities, compatibility with living organisms, capability to be broken down naturally, low vapor pressure, and good recyclability (Hooshmand et al. 2023; Wagle et al. 2016; S. Wang et al. 2015a, b; Q. Zhang et al. 2012a, b). There are different types of DESs reported in the literature that can be potentially used for CO₂ capture; nevertheless, due to its low cost, easy synthesis, and good biodegradation, the choline chloride:urea (ChCl:U) mixture in a 1:2 molar ratio has been

highlighted as an excellent solvent for CO₂ capture (Li et al. 2008; Xie et al. 2016; Y. Zhang et al. 2014). Moreover, the ChCl:U is has been extensively investigated. The ChCl:U eutectic mixture has exhibited excellent performance in very different areas such as catalysis, organic synthesis, electrochemistry, nanoparticle preparation, and carbon material production (S. Wang et al. 2015a, b). Due to the previously stated benefits and the existence of relevant experimental data for simulating DES in a chemical process, ChCl:U (1:2) has been selected as the primary DES for this work.

The proposed model presents a new design of CO₂ post-combustion capture using the physical absorption with ChCl:U (1:2) as an alternative green solvent. The proposed process consists of an absorption column (AC), two isobaric flash tanks (FT-1 and FT-2, respectively), and a regenerator column (DC), as shown in Fig. 1. The simulation of the process was carried out in the Aspen Plus process simulator.

For the simulation, it was considered 20,000 kmol/h as a feed flow of the flue gas when natural gas fuel is burned to produce energy. According to Luyben (2013), this flow represents the amount of combustion gases coming from a power plant. It was considered a constant feed flow of natural flue gas of 20,000 kmol/h with a composition of 0.03% mol of CO₂, 0.0589% mol of H₂O, 0.7665% mol of N₂, and 0.1446% mol of O₂ (Romero-García et al. 2022a, b; Songolzadeh et al. 2014). The first step of the process begins when the flue gas (F_1) enters into the AC, where CO₂ is separated by physical absorption using an aqueous solution of ChCl:U (1:2) as solvent (F_2). After the absorption, the clean gases such as nitrogen (N₂) and oxygen (O₂) are released into the atmosphere (F_3) and the CO₂ is concentrated in the bottom flow (F_4), which subsequently enters the second stage of the process.

The second and third stages of this intricate process closely adhere to a consistent methodology, employing an isobaric flash tank to facilitate the separation of residual nitrogen (N₂) and oxygen (O₂) within the F_4 feedstock. In the second stage, FT-1 is strategically employed, whereas the third stage implements FT-2. In these pivotal stages, clean gases emerge through F_6 and F_8 , respectively, while streams F_5 and F_7 are exclusively reserved for the efficient conveyance of carbon dioxide (CO₂) and the accompanying solvent. Notably, existing scientific literature hints at the theoretical feasibility of achieving CO₂ separation sans the requirement for flash tanks; however, this demands the execution of the separation process under the demanding constraints of cryogenic conditions (Fu et al. 2014; Yousef et al. 2018). While operating within such cryogenic environments undoubtedly delivers superior results in terms of achieving high CO₂ purity and remarkable recovery rates, it is imperative to recognize that the associated operational costs surge substantially. To navigate this financial challenge, our proposal advocates for an approach that positions us slightly below

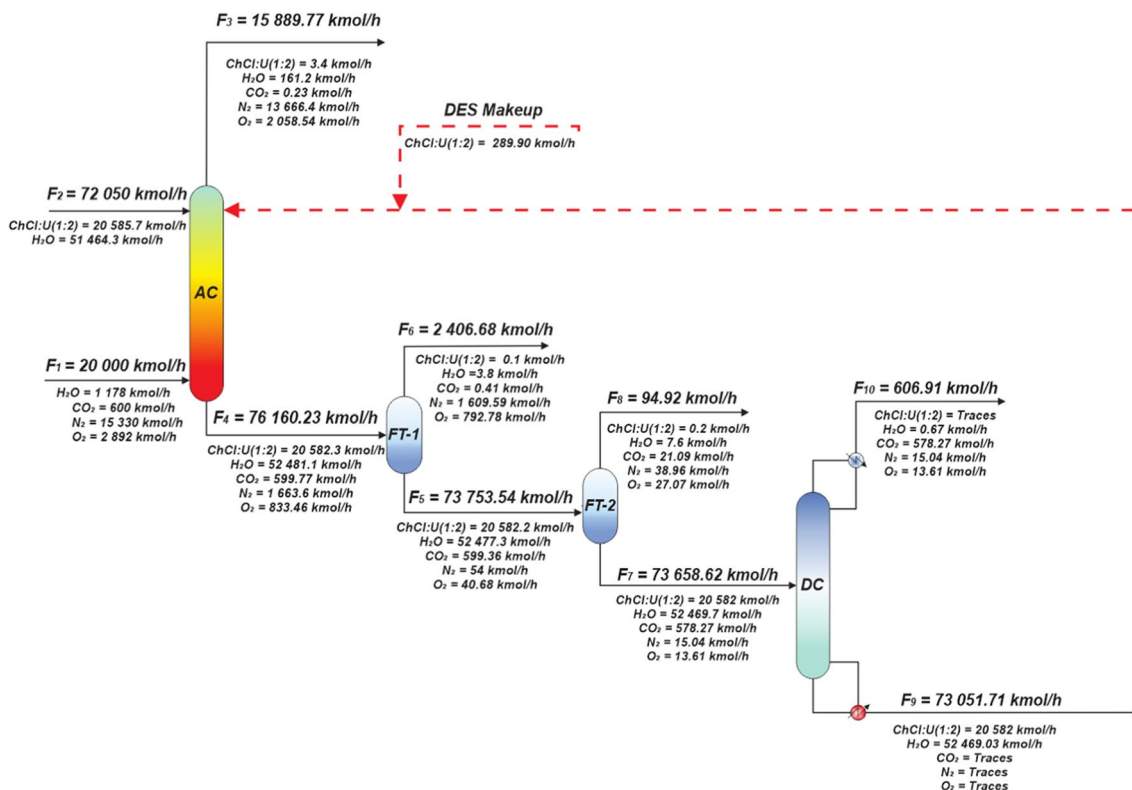


Fig. 1 Proposed scheme of post-combustion CO₂ capture process using ChCl:U (1:2) as solvent

the cryogenic threshold, ensuring a substantial CO₂ yield while sidestepping the rigors of full cryogenic conditions. A comprehensive sensitivity analysis, a cornerstone of our research endeavor, has yielded crucial insights into the most effective means of eliminating nitrogen (N₂) while maximizing CO₂ recovery. This analysis underscored the undeniable efficiency of employing a flash tank for the purpose. However, an important caveat arises: while it is theoretically plausible to carry out the separation within a single, albeit substantial, piece of equipment, such a colossal tank undeniably has consequences for equipment volume, subsequently leading to escalated operational and manufacturing expenditures. In light of this economic consideration, the most judicious and pragmatic strategy emerges as the deployment of two distinct units, each meticulously configured with unique operating conditions. This method ensures the simultaneous removal of both nitrogen (N₂) and oxygen (O₂) while concomitantly curtailing operational costs and diminishing energy consumption. In summation, this multifaceted process, spanning multiple stages, not only exemplifies an enhanced approach to N₂ and O₂ separation but also endeavors to alleviate the financial implications of cryogenic conditions. By tactically operating just below the cryogenic threshold, we strike a harmonious balance that allows for the generation of a substantial CO₂ yield without necessitating a plunge into the fiscal complexities associated with full

cryogenic conditions. Our sensitivity analysis, a key pillar of our investigation, has unequivocally pointed towards the preeminence of utilizing a flash tank for N₂ elimination. While the prospect of consolidating equipment into a single unit may appear alluring, the stark economic realities of such a choice render it less tenable. Embracing the deployment of two separate units, each tailored to specific operating conditions, emerges as the unequivocally optimal approach, skillfully harmonizing efficacy with cost-effectiveness in this pivotal separation process.

To prove that the proposed model is more cost-effective, environmentally friendly, and safer than the traditional organic amine solvent capture, a comparison between both processes was carried out. Therefore, it is necessary to make a comparative analysis of the proposed process with ChCl:U (1:2) against the conventional capture process with monoethanolamide (MEA), from a sustainable point of view. Chemical absorption using MEA as solvent is used as a conventional solvent-based on amines for CO₂ capture. Moreover, the process modeling is made easier due to the well-known thermodynamic characteristics of the MEA. Hence, employing MEA for CO₂ capture serves as an ideal benchmark for the in-depth exploration of the process variables that have a direct influence on energy consumption, degradation, and environmental impact.

The conventional process used in this study for the comparative analysis is the one reported by Romero-García et al. (Romero-García et al. 2022a, b). For this particular case study, it was considered exclusively on the capture system, utilizing solely the capture system's topology as the basis for our simulation data. To maintain consistency of the comparison, it was considered that the flow rates were the same in both cases, necessitating adjustments to the operating conditions in order to attain purities and recoveries exceeding 95%. Consequently, the MEA model, following these necessary operating condition modifications, cannot be considered an optimized model. However, following a sensitivity analysis, MEA model does manage to fulfill the criteria of achieving purities and CO₂ recoveries greater than 95%. This approach ensures both systems under identical conditions for comparison.

To yield a sustainable process to be sustainable, it is crucial to assess various aspects including economic, environmental, and safety issues. Literature has demonstrated that combining different metrics can enhance the sustainability of the process by recognizing the interconnectedness between aspects like environmental impact, safety, profitability, and energy efficiency (Rafiei and Ricardez-Sandoval 2020). Jiménez-González and Constable (2011) suggested that these areas are necessary for assessing the green chemistry of a process.

Methodology

Database Implementation

The Aspen Plus process simulator was used to simulate the processes involved in CO₂ capture, which included CO₂, H₂O, N₂, O₂, and ChCl:U components. The thermophysical properties of CO₂, H₂O, N₂, and O₂ were calculated using

the property database of Aspen Plus. However, for ChCl:U, the thermophysical properties could not be obtained directly from the property database, so the database of ChCl:U was implemented into Aspen Plus. Experimental values and semi-empirical equations based on experimental data were loaded into the simulator to predict the thermophysical properties of ChCl:U. Parameter tuning was performed within Aspen Plus to obtain accurate predictions. The implementation of the ChCl:U database was carried out using the methodology proposed in Fig. 2. For more detailed information on loading DES properties into the Aspen Plus simulator, see the supplementary material.

According to Fig. 2, the first step is to define a new component by the user as ChCl:U. Once the component was created, it is necessary to define new scalar parameters for the component. The scalar parameters used for the component ChCl:U are shown in Table 1.

As well, with the critical pressure, critical temperature, and critical volume, it is possible to calculate the critical compressibility factor (Z_c) using the equation reported by Esmailzadeh et al. (2020), represented as shown in Eq. (1).

$$Z_c = \frac{P_c V_c}{RT_c} \quad (1)$$

Once the scalar properties are input, it is necessary to define some correlations dependent on temperature as well as to introduce the constants corresponding to the polynomial that can predict the desired property for the ChCl:U component. For this simulation, the Wagner equation was used to define vapor pressure, the heat capacity was defined by the ideal gas equation, the liquid surface tension was defined by the DIPPR equation, and the liquid viscosity was defined by the Andrade equation. The values used are shown in Table 2. Subsequently, the default methods in the Aspen Plus property system for pseudo-components were

Fig. 2 Steps for the implementation of the database of ChCl:U into Aspen Plus simulator

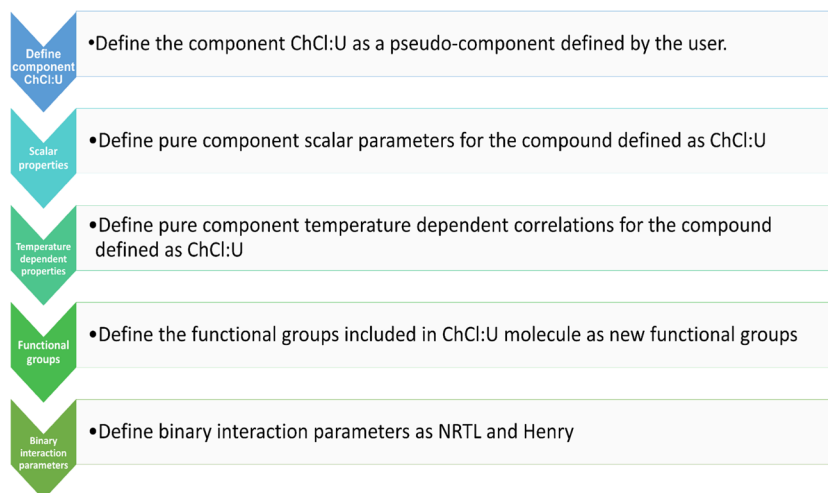


Table 1 Scalar parameters for the component ChCl:U

Property	Symbol	Value	Unit	Reference
Boiling temperature	T_B	445.6	K	Mirza et al. (2015)
Molecular weight	MW	86.58	g/mol	
Acentric factor	Ω	0.661	—	
Critical temperature	T_c	644.4	K	
Critical pressure	P_c	49.35	bar	
Critical volume	V_c	254.37	cm ³ /mol	
Density	P	1.076	g/cm ³	
Critical Compressibility factor	Z_c	0.2343	—	Calculated with Eq. (1)

Table 2 Correlations dependent on temperature for the ChCl:U component

Property	Wagner equation	Temperature unit	Property unit	Reference
Vapor pressure	$\ln p_r^{*l} = \frac{A_1(1-T_r)+A_2(1-T_r)^{1.5}+A_3(1-T_r)^3+A_4(1-T_r)^6}{T_r}$	K	—	Esmailzadeh et al. (2020)
A_1	A_2	A_3	A_4	
−6.133	−3.868	0.673	3.977	
Property	Heat capacity of the ideal gas equation			
Molar heat capacity	$C_p = C_1 + C_2T + C_3T^2$	K	J/molK	Leron and Li (2012)
	C_1	C_2	C_3	
	247.4	−0.5633	1.141×10^{-3}	
Property	DIPPR equation			
Liquid surface tension	$\gamma = C_1 + C_2T + C_3T^2$	K	N/m	Ma et al. (2018)
	C_1	C_2	C_3	
	0.09244	0.6043	0	
Property	Andrade equation			
Liquid viscosity	$\ln \eta_i^{*l} = C_1 + \frac{C_2}{T} + C_3 \ln T$	K	mPa/s	Ji et al. (2013)
	C_1	C_2	C_3	
	−443.7	26,670	62.14	

employed to calculate the viscosities, heat capacities, and surface tension of ChCl:U.

As the next step, it is necessary to define functional groups of choline chloride and urea as new functional groups. Moreover, it is necessary to input some binary interaction parameters. The Nonrandom Two-Liquid (NRTL) binary interaction parameters used for the simulation are shown in Table 3. As reported by Y. Liu et al. (2021) another important property to take into consideration during CO₂ capture with ChCl:U is the Henry's constant for physical-based DESs, which also were taken from Ma et al. (2018) and is shown in Table 4.

Before continuing with the simulation of the process, it is important to check that the simulator makes a correct prediction of the thermophysical properties of the ChCl:U

solvent. For this, a comparative analysis of the properties predicted by the simulator vs experimental data reported in the literature is carried out. The results of this validation are found in the “Validation of the Thermophysical Properties of ChCl:U” section.

Configuration of the CO₂ Capture System

For the CO₂ capture process, to model the thermodynamic properties of the DES system, it was considered the NRTL thermodynamic model. The choice of NRTL over other activity coefficients is because the NRTL model presents superiority in the simulation of liquid–liquid equilibria (Usman et al. 2021).

Table 3 NRTL binary interaction parameters for the ChCl:U component

<i>I</i>	ChCl:U		ChCl:U		CO ₂		CO ₂		N ₂		CO ₂		H ₂ O		CO ₂		N ₂		O ₂	
<i>J</i>	N ₂	O ₂	O ₂	C	N ₂	O ₂	O ₂	C	O ₂	C	O ₂	O ₂	C	ChCl:U	H ₂ O	H ₂ O	H ₂ O	H ₂ O	K	K
Reference	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Aspen Plus	Ma et al. (2018)	Ma et al. (2018)	Ma et al. (2018)	Ma et al. (2018)	Ma et al. (2018)	Ma et al. (2018)	Ma et al. (2018)
<i>A_{II}</i>	0	0	0	0	0	0	0	0	0	0	0	0	0	1.61	10.064	10.064	0	0	0	0
<i>A_{JI}</i>	0	0	0	0	0	0	0	0	0	0	0	0	0	-0.48	10.064	10.064	0	0	0	0
<i>B_{II}</i>	0	0	0	0	0	0	0	0	-131.97785	-5.1212663	-1236.14	-538.95	-538.95	-538.95	-3268.14	-3268.14	143.351	182.272	182.272	182.272
<i>B_{JI}</i>	0	0	0	0	0	0	0	0	245.817302	4.26834851	3032.59	134.9	134.9	134.9	-3268.14	-3268.14	-36.5619	-107.244	-107.244	-107.244
<i>C_{II}</i>	0	0	0	0	0	0	0	0	0.3	0.3	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.3
<i>D_{II}</i>	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>E_{II}</i>	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>E_{JI}</i>	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>F_{II}</i>	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>F_{JI}</i>	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
<i>T_{lower}</i>	0	0	0	0	0	0	0	0	25	25	25	25	25	0	0	0	0	0	0	0
<i>T_{upper}</i>	1000	1000	1000	1000	25	25	25	25	25	25	1000	1000	1000	1000	1000	1000	1000	1000	1000	1000

Table 4 Henry interaction parameters for ChCl:U (Ma et al. 2018)

Component <i>I</i>	CO ₂	CO ₂	CH ₄
Component <i>J</i>	ChCl:U	Agua	ChCl:U
Temperature unit	K	K	K
Property unit	bar	bar	bar
<i>A_{II}</i>	242.95	159.87	16.16
<i>B_{II}</i>	-3055.05	-8741.55	1438.44
<i>C_{II}</i>	-47	-21.69	-4.42
<i>D_{II}</i>	0.13	0.0011	0.03
<i>T_{min}</i>	0	0	0
<i>T_{max}</i>	2000	2000	2000
<i>E_{II}</i>	0	0	0

For both capture processes, it is important to consider as a restriction that it must be recovered at least 95% of the CO₂ from the flue gas feed, with a purity above 95%. Under these conditions of purity, CO₂ can be marketed and reused in different applications at the industrial level. Of course, the industrial area of selling will depend on the purity achieved during the capture.

A base model was used for simulating the conventional process using MEA based on the simulation conditions reported by Romero-García (Romero-García et al. 2022a, b). During the absorption and desorption of CO₂, due to the reactions involved, several ionic species are formed as a result of the use of MEA as a solvent. Using the electrolyte Nonrandom Two-Liquid (e-NRTL) thermodynamic model, Nakagaki et al. (Nagy and Mizsey 2015) proposed that post-combustion capture can be effectively accomplished by properly modeling the thermodynamic parameters involved.

Moreover, for the proposed model with ChCl:U, to achieve the established recoveries and purities, it is necessary to identify which variables have a significant influence to reach the expected objective targets. As a first instance, it is important to point out the importance of water in the process. Different studies indicate the influence that water has on the behavior of the solvent ChCl:U (Di Pietro et al. 2021; Gageiro Machado et al. 2022; Hsu et al. 2014; Ma et al. 2018). Generally, due to the hygroscopic feature of DES, traces of water are unavoidable, which significantly affects the physicochemical properties of DES components. According to Di Pietro et al. (2021) data indicate that ChCl:U seems to maintain its structure as small interplays gradually occur between urea in the DES and the surrounding water. Moreover, viscosity and therefore CO₂ solubility can change because of water concentration. That is why the optimum range of ChCl:U ratio with water must be found so that the thermodynamics of the system with CO₂ are respected.

Therefore, as a preliminary study, a sensitivity analysis was performed on each process unit one by one. Table 5

shows the design and operation variables, as well as the ranges established in each unit of the process.

The strategy used corresponds to an iterative method, which consists of iterating “ n ” operation or design variables. While the rest of the variables that conform the system remain constant, to maximize CO₂ recovery and purity. This process was carried out equipment by equipment. For example, for the same condition of DES molar ratio, water molar ratio, temperature, and pressure the number of stages in the absorber in the reactor was varied so that it affects the CO₂ recovery and purity. Therefore, different number of

stage variations were made between the limits established in Table 5. Likewise, for the same condition of number of stages, DES molar ratio, water molar ratio, temperature in the absorber, and pressure were varied between the established limits reported in Table 5. The same process was applied to the other variables in the absorber until finding the combination of variables that allow reaching the recovery and purity of CO₂ that adjusted to the established restrictions. Figure 3 shows an illustrative diagram of the variable iteration process used as a strategy for the sensibility analysis, where the green boxes represent the variable that

Table 5 Design variables for the sensibility analysis

Process	Variables	Type of variable	Range	Units
Absorber	DES molar ratio	Continue	0.5–3	—
	Water molar ratio	Continue	0.5–3	—
	Number of stages	Discrete	20–100	—
	Temperature	Continue	315.15–343.15	K
	Pressure	Continue	1–14	bar
Isobaric flash tank	Pressure	Continue	1.1	bar
	Vapor fraction	Continue	0–1	—
Isobaric flash tank	Pressure	Continue	1.1	bar
	Vapor fraction	Continue	0–1	—
Desorber	Reflux ratio	Continue	0.5–2.5	—
	Number of stages	Discrete	10–30	—
	Feed stage	Discrete	2–29	—
	Pressure	Continue	1–14	bar

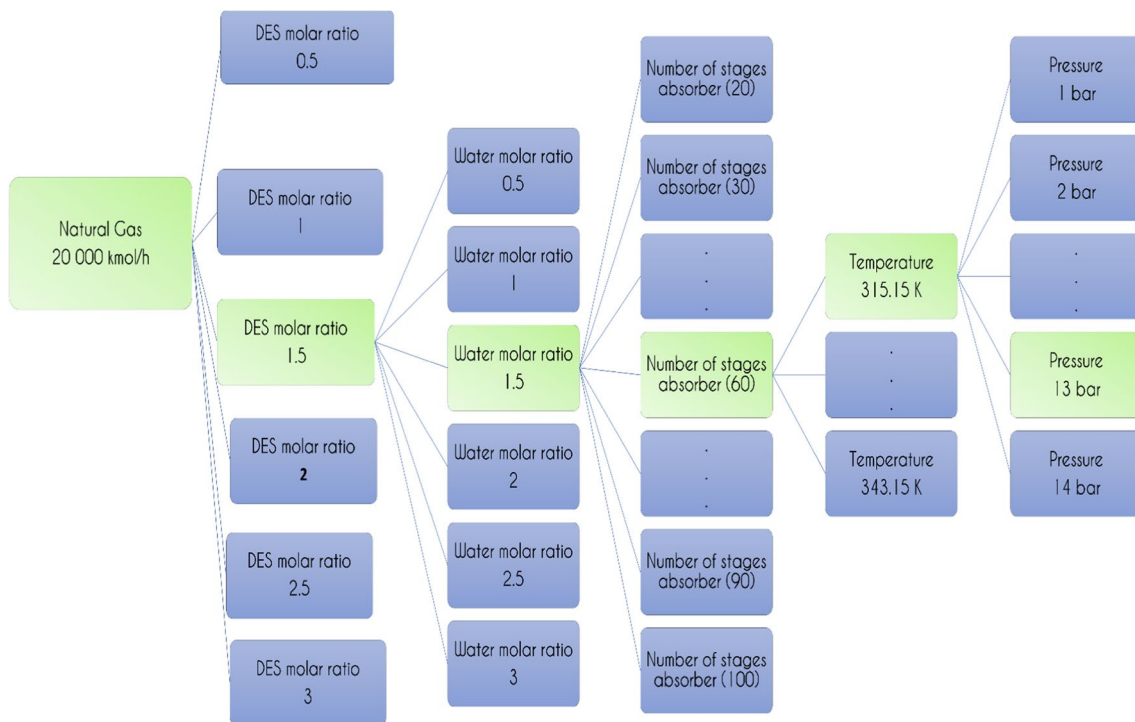


Fig. 3 Diagram of the variable iteration process

remains constant, while the blue ones represent the variable condition.

As shown in Fig. 3, for each given condition a tree of possible combinations for the equipment is displayed. The same iteration process was carried out for the rest of the units of the process. In this way, more than 1000 preliminary designs are obtained; although they are not the optimal designs, they allow for illustrating the behavior of the variables that have greater weight to obtain the maximum CO₂ recovery and the highest purity. Therefore, it is important to carry out rigorous optimization under the simultaneous design-optimization strategy for the established objectives.

Sustainability Evaluation of the Process

Three different axes were taken into consideration for the comparative analysis: Total Annual Cost (*TAC*), which serves as a gauge of how economically efficient is the process. Individual Risk (*IR*) serves as a safety indicator by measuring the chance of a person will experience or be impacted by an accident on a regular basis. Eco-indicator 99 (*EI99*) is used to quantify the environmental impact resulting from the use of ChCl:U and MEA as solvents. These axes are required, according to Jiménez-González et al. (Jiménez-González and ConsTable 2011) to assess the sustainability of the process from a green chemistry perspective. Applying these indexes to distillation columns, Sánchez-Ramírez et al. (2019) and Contreras-Zarazúa et al. (2021) have demonstrated the sustainability of other processes.

Total Annual Cost

Guthrie's (1969) method, as modified by Ulrich (1984), is the foundation of the *TAC*. 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 correlations reported by Turton et al. (2008) are utilized to determine the cost of the units. Equation (2) displays the objective function that was employed.

$$TAC = \frac{\text{Capitalcost}}{\text{Paybacktime}} + \text{Operatingcost} \quad (2)$$

It is assumed that each process will operate for 8500 h per year during the plant's payback period of 5 years. The demand for very short payback times commonly applied to investments in energy saving ensures that the returns are relatively insensitive to assumptions about future energy costs (Pritchard 1982). Moreover, shorter paybacks mean more attractive investments, while longer payback periods are less desirable.

Eco-Indicator 99

An eco-indicator 99 (*EI99*) was employed to assess the effects on the environment. The life cycle assessment (LCA), on which the *EI99* methodology is based, provides the rationale for the hierarchical weighting in the relative evaluation of the damage.

A process, a product, or an activity can have its environmental load evaluated using the *EI99* by identifying and quantifying the material and energy used. Several authors have recently used this methodology to assess environmental impact (Errico et al. 2017; Gebreslassie et al. 2009).

The three main categories of impact considered by the *EI99* methodology are (1) resource depletion, (2) human health, and (3) ecosystem quality. The following elements are used to calculate *EI99*: steel is used to build equipment and ancillary equipment, while steam is used to generate heat and move to provide electricity. The associated data with these activities were taken from the standard databases (Goedkoop 1999).

Equation (3) defines the *EI99* as follows:

$$EI99 = \sum_b \sum_d \sum_{k \in K} \delta_d \omega_d \beta_b \alpha_{b,k} \quad (3)$$

where δ_d represents the normalization factor for damage of category d , ω_d represents the weighting factor for the damage of category d , β_b is the total amount of product b released per unit of reference flow due to direct emissions, and $\alpha_{b,k}$ is the damage in category k per unit of chemical product b released to the environment. An average European citizen's annual environmental loads are equivalent to one point on the *EI99* scale.

Individual Risk

The process safety quantification used the *IR* index. 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):

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

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. By using a Qualitative Risk Analysis (QRA), it is possible to determine the frequency and possibility of potential incidents and accidents, as well as any potential effects they may have. As a first step, the QRA methodology establishes the identification of the incident, which can be defined as any time material or energy is released during a process (Kumar 1996). Figure 4 shows the frequencies and potential accidents that might occur in

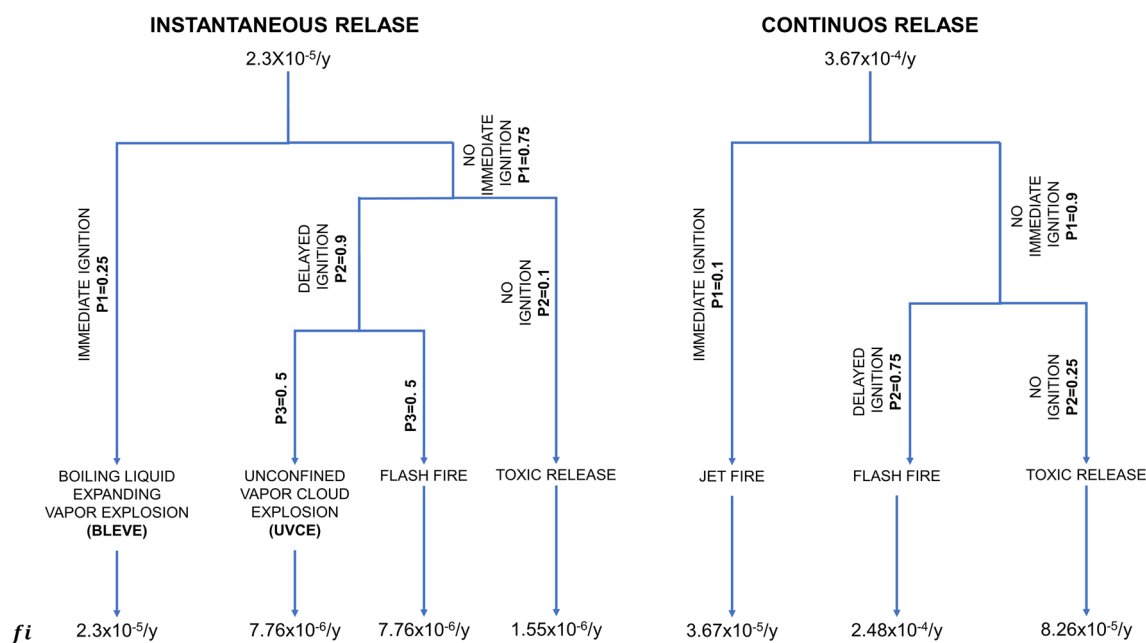


Fig. 4 Possible accidents and frequencies that can happen in a process

a process. Once the accident is identified, the second step of the QRA methodology is to identify the variable that causes the potential accident. According to Kumar (1996), thermal radiation (E_r) is a causative factor in the BLEVE, Jet Fire, and Flash Fire. The toxic release and the release concentration, generated by the overpressure (P_o), is the causative factor for the UVCE. The calculation for each accident's causative variables and likely accidents are displayed in the supplementary material section.

Results and Discussion

To make the results easier to read, the “Results and Discussion” section is divided into two sections. Firstly, the data from the validation of the thermophysical properties of the solvent are presented and then the data of the proposed case study are presented.

Validation of the Thermophysical Properties of ChCl:U

In order to proceed with the simulation, it is important to validate that the simulator is correctly performing the thermophysical properties of the ChCl:U pure solvent. To carry out the validation, different properties were analyzed at different temperatures. The properties analyzed were density (ρ), molar heat capacity (C_p), viscosity (η), and surface tension (γ). Each of the properties was compared by evaluating the percentage of error presented by the prediction made

by the Aspen Plus simulator, with respect to experimental data reported in the literature, and compared with the data obtained in ASPEN. The graphical comparison of the properties is presented in Figs. 5, 6, 7, 8, and 9.

Likewise, to support the obtained graphic results, the error percentage of each value obtained concerning the experimental data was calculated. Table 6 shows a summary of the comparison between the experimental data and the results predicted by the simulator for density, molar heat capacity, viscosity, surface tension, and weight percent of ChCl:U (1:2) at different temperatures. To see all the values obtained, consult the supplementary material section.

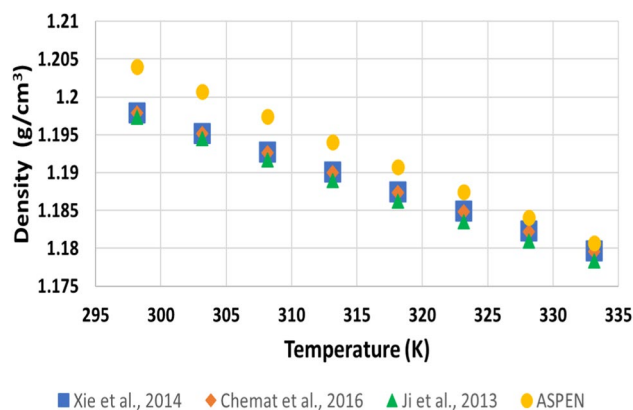


Fig. 5 Density from experimental data from the literature: Xie et al. (2014) (blue square), Chemat et al. (2016) (orange diamond), and Ji et al. (2013) (green triangle) against ASPEN prediction (yellow circle)

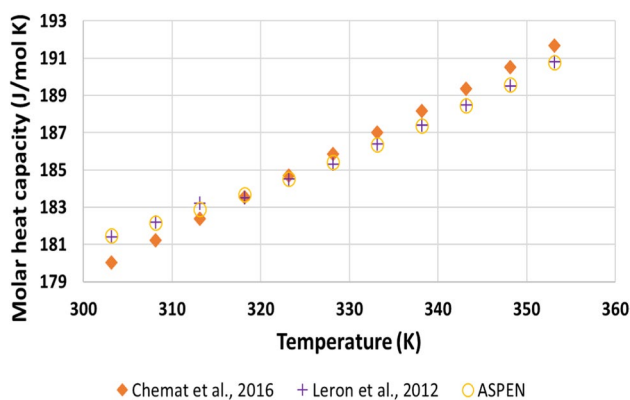


Fig. 6 Molar heat capacity from experimental data from the literature: Chemat et al. (2016) (orange diamond) and Leron and Li (2012) (cross) against ASPEN prediction (yellow circle)

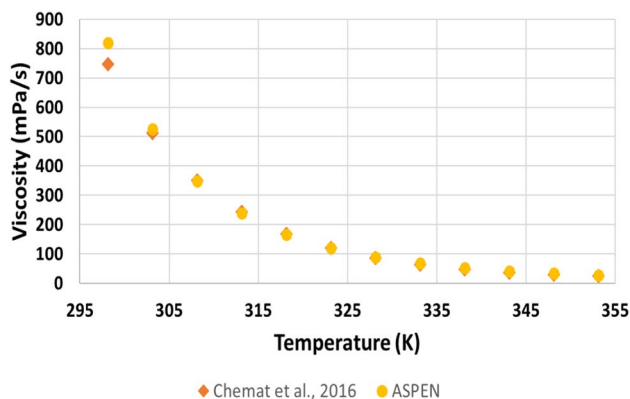


Fig. 7 Viscosity from experimental data from the literature: Chemat et al. (2016) (orange diamond) against ASPEN prediction (yellow circle)

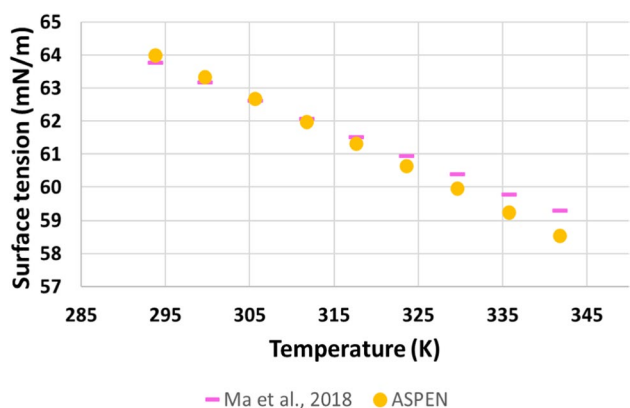


Fig. 8 Surface tension from experimental data from the literature: Ma et al. (2018) (minus sign) against ASPEN prediction (yellow circle)

In Table 6 only the two highest values (bold) and the two lowest values (italic) of the error percentage obtained

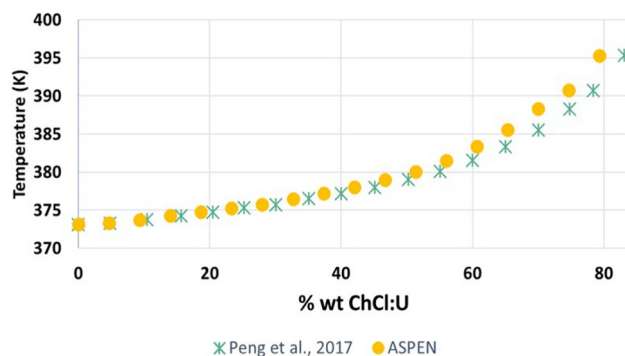


Fig. 9 Solvent weight percentage from experimental data from the literature: Peng et al. (2017) (green asterisk) against ASPEN prediction (yellow circle)

are presented for each data set analyzed. This is to show the lower and upper limits; for any other measurement analyzed the percentage of error is within these limits presented. Figure 5 shows that at low-temperature values (298 K) the predicted density values move away in the order of 0.005 units from the experimental density values. As the temperature increases, the predicted values are more similar to the experimental ones. Even though a graphic deviation is visually observed, the percentage of error between the predicted density data and the experimental density data is less than 0.6% for all sets of data analyzed. In Fig. 6, the values are graphically spliced, and the values predicted are very close to the experimental ones. However, at lower temperatures, a small deviation of the order of magnitude of 1 unit is visually perceived concerning the experimental data reported by Chemat et al. (2016). Despite this deviation, when evaluating the error percentage, it is found that when analyzing this data set, at low temperatures the highest error percentage is less than 1%. This trend changes around 320 K and 340 K, where the error rate is the lowest for this data set below 0.1%. When comparing the heat capacity prediction data with respect to the data reported by Leron et al. (Leron and Li 2012), it is visually observed in Fig. 6 that the prediction follows the same trend as the experimental values in all the temperature range. This is verified in Table 6 showing that the percentage of error obtained for this data set is less than 0.1% for all the heat capacity values evaluated.

In the case of viscosity, it is possible to see in Fig. 7 that the predicted data is consistent with the experimental data. When evaluating the percentage of error, it can be seen in Table 6 that for viscosity the highest percentage of error is around 10%. Despite being a relatively high value according to Brook and Arnold (2018) the maximum limit according to the design of experiments is 10%. Therefore, it can be said that the prediction of viscosity values at different temperatures is correct and are in agreement with the experimental values reported.

Table 6 Comparison between the experimental data and the results predicted by the simulator for density, molar heat capacity, viscosity, surface tension, and weight percent of ChCl:urea (1:2) at different temperatures (Chemat et al. 2016; Ji et al. 2013; Leron and Li 2012; Ma et al. 2018; Peng et al. 2017; Xie et al. 2014)

Simulation data		Experimental data						
Aspen Plus		Chemat et al. (2016)			Xie et al. (2014)		Ji et al. (2013, p. 201)	
Temperature (K)	ρ (g/cm ³)	ρ (g/cm ³)	% Error	ρ (g/cm ³)	% Error	ρ (g/cm ³)	% Error	
298.15	1.204	1.198	0.507	1.198	0.507	1.197	0.559	
303.15	1.201	1.195	0.459	1.195	0.459	1.194	0.520	
328.15	1.184	1.182	<i>0.160</i>	1.182	<i>0.151</i>	1.181	<i>0.266</i>	
333.15	1.181	1.180	<i>0.097</i>	1.180	<i>0.088</i>	1.178	<i>0.203</i>	
Aspen Plus		Chemat et al. (2016)			Leron and Li (2012)			
Temperature (K)	C_p (J/mol K)	C_p (J/mol K)	% Error	C_p (J/mol K)	% Error			
303.15	181.49	180.05	0.802	181.40	0.051			
308.15	182.16	181.21	0.005	182.20	0.020			
313.15	182.89	182.37	0.003	183.20	0.168			
318.15	183.68	183.54	<i>0.001</i>	183.50	0.097			
323.15	184.52	184.70	<i>0.001</i>	184.50	<i>0.011</i>			
338.15	187.39	188.18	0.004	187.40	<i>0.006</i>			
Aspen Plus		Chemat et al. (2016)						
Temperature (K)	η (mPa/s)	η (mPa/s)	% Error					
308.15	349.03	351.46	<i>0.690</i>					
323.15	120.45	119.81	<i>0.536</i>					
343.15	40.96	37.039	10.587					
348.15	32.96	29.79	10.642					
Aspen Plus		Ma et al. (2018)						
Temperature (K)	γ (mN/m)	γ (mN/m)	% Error					
305.65	62.67	62.63	<i>0.072</i>					
311.74	61.99	62.06	<i>0.110</i>					
335.76	59.25	59.77	0.876					
341.74	58.55	59.29	1.248					
Aspen Plus		Peng et al. (2017)						
Temperature (K)	% w ChCl:U	% w ChCl:U	% Error					
373.3	4.67	4.8	<i>2.778</i>					
375.73	28.00	30.1	<i>6.977</i>					
376.49	32.67	35.1	<i>6.933</i>					
374.26	14.00	15.6	10.256					
395.33	79.33	83.1	<i>4.533</i>					

*numbers in bold for the maximum error values, and in italics for the minimum error values

In the case of surface tension, it is possible to see in Fig. 8 that at low temperatures the predicted data are consistent with the experimental data. As the temperature increases, the predicted data present a slight deviation of the order of between 0.29 and 0.74 units of magnitude. However, when evaluating the percentage of error, it can be seen in Table 6 that for surface tension the highest percentage of error is around 1.2%. Thus, the prediction of surface tension values at different temperatures is correct and consistent with the experimental values.

Moreover, Fig. 9 shows an analysis of the composition of the solvent (weight percent) at different temperatures. In this case, it is observed that graphically the data prediction presents the same trend as the experimental data. In this case, the biggest error percentage for this data set is 10%. Emphasizing that according to Brook and Arnold, the maximum error limit allowed for the design of experiments is 10% (Brook and Arnold 2018). Therefore, at high concentrations of ChCl:U, it presents the condition of being in the maximum allowed limit of error. However, for the CO₂ capture process, it is needed a concentration of 30% by weight of

the solvent. So, in these conditions of concentration of the solvent, the behavior of the solvent is closer to the established in the experimental data. Therefore, according to the results obtained in the validation, the simulator is capable of correctly predicting the thermophysical properties of the solvent ChCl:U.

CO₂ Capture System Analysis

This section presents the results obtained from the case study of the post-combustion CO₂ capture system using ChCl:U as solvent. Firstly, from the sensibility analysis for the CO₂ capture system using ChCl:U as the solvent, it was possible to achieve a recovery of 96.38% of the CO₂ contained in the flue gas with a purity of 95.28%. The conditions that allow us to obtain the aforementioned recovery and purity are shown in Table 7.

Certainly, let us further elaborate on the nuances of the study and the reasoning behind the chosen conditions:

In the realm of process optimization and comparative studies, it is essential to recognize that achieving an “ideal” model is often a complex and multi-faceted endeavor. While we did not explicitly engage in a formal optimization process within this study, it is crucial to emphasize that our operating conditions were not arbitrarily selected. Rather, they

were meticulously crafted through a comprehensive sensitivity analysis, which served as the bedrock of our decision-making process. This analysis was instrumental in ensuring that the specified purity and recovery thresholds for CO₂ were met without the necessity of venturing into the intricate realm of cryogenic conditions.

Moreover, the decision to uphold comparability with the reference MEA system, as detailed in the study by Romero-García, Ramírez-Corona, et al. in 2022, was paramount for establishing a rigorous basis for our comparative analysis. Their reference system incorporates an optimized model of the MEA capture system, intricately integrated with a power plant. In contrast, our study centered exclusively on the capture system, with our simulation data based solely on the topological attributes of this system.

To ensure a fair and meaningful comparison, it was imperative to maintain consistent flow rates between both cases. This alignment, however, necessitated a recalibration of the operating conditions to guarantee the attainment of purity and recovery rates exceeding 95%. It is vital to stress that these adjustments were undertaken not as a means to optimize our model but rather to create an equitable basis for comparison. Hence, it is essential to clarify within the manuscript, particularly in the “Case Study Description” section, that the MEA model, subsequent to these essential operating

Table 7 Values of the variables for the sensibility analysis of the process using ChCl:U in comparison with the conventional process

Equipment	Variables	Unit	ChCl:U model Value	MEA model Value
Absorber	Number of stages	—	30	29
	Flue gas feed stage	—	30	29
	Solvent feed stage	—	1	1
	Temperature	K	333	313
	Pressure	bar	13	1
	Flue gas molar feed flow	kmol/h	20,000	20,000
	CO ₂ in flue gases	kmol/h	600	600
	Solvent feed flow	kmol/h	72,050	44,038
	DES percent	%w	0.3	—
	Water percent	%w	0.7	—
	DES:water ratio	—	1:2.5	—
Isobaric flash tank	Pressure	bar	1.09	—
	Vapor fraction	—	0.0316	—
Isobaric flash tank	Pressure	bar	1	—
	Vapor fraction	—	0.001208	—
Desorber	Number of stages	—	16	24
	Feed stage	—	6	3
	Reflux ratio	—	1.64	2.33
	Pressure	bar	1	1
	Thermal duty	GJ/h	271.4	363.74
	CO ₂ captured	kmol/h	578.27	594
	CO ₂ recovery	%	96.38%	99%
CO ₂ purity	%	95.28%	97.6%	

condition adaptations, should not be classified as an optimized model per se. Nevertheless, through a meticulous sensitivity analysis, our MEA model indeed demonstrates its capability to meet the stipulated criteria of achieving purity levels and CO₂ recoveries exceeding 95%. This methodical approach ensures that both systems operate under identical conditions, thereby fortifying the robustness and integrity of our comparative analysis.

From the results, it is important to highlight that during the absorption the variables that have a significant impact on the process performance are the pressure, temperature, and DES:water ratio. In regards to temperature, ChCl:U presents a good CO₂ absorption as long as it is operated within the established temperature ranges between 313.15 and 343.15 K. Regarding pressure, it has a significant impact on the operation. For example, if the process is settled to a constant pressure and then the temperature is varying from 313.15 to 343.15 K, CO₂ recoveries will not change significantly. Otherwise, when the temperature is settled constant and pressure is varied, CO₂ recovery will have a significant change. The ChCl:U and water ratio is another variable to take into consideration because it strongly affects CO₂ solubility. This can be explained due to the high viscosity of ChCl:U. Based on certain articles, adding water can greatly reduce ChCl:U's viscosity (Xie et al. 2014, p. 2), which in turn reduces the solubility of CO₂. Due to the decreased friction losses, the low viscosity of aqueous ChCl:U will improve CO₂ mass transfer and lower pumping expenses. However, the reduced CO₂ solubility will demand more solvent and a large absorption tower, which will degrade the process' overall performance (Ma et al. 2018).

Drawing upon the extensive sensitivity analysis conducted within this research, our findings elucidate that achieving a CO₂ recovery rate surpassing 95% with a purity level exceeding 95% while simultaneously effecting the separation of CO₂ from residual traces of N₂ and O₂ necessitates a substantial perturbation in the system's pressure dynamics, particularly when employing flash tanks. While it is theoretically conceivable to execute CO₂ separation using relatively simpler distillation columns (Fu et al. 2014; Yousef et al. 2018), thereby obviating the necessity for flash tanks, our investigation concurs with prior research in asserting that the attainment of a molar purity of CO₂ exceeding 95% inexorably compels the operation of the entire process under the formidable constraints of cryogenic conditions. This crucial observation underscores a conundrum with far-reaching implications, transcending mere scientific curiosity.

The decision to embrace cryogenic conditions, yielding commendable purity and recovery rates for CO₂, reverberates across several dimensions, each laden with its own set of considerations. From an economic standpoint, the costs associated with cryogenic operations are substantial and impact not only capital expenditure but also ongoing

operational expenses. Moreover, the imposition of cryogenic conditions introduces inherent safety risks and necessitates rigorous safety measures, further escalating the operational burden. Additionally, the environmental footprint of cryogenic processes, marked by energy-intensive refrigeration requirements, must be scrutinized given the contemporary emphasis on sustainability.

Hence, in light of these multifaceted challenges, the model proposed in this research advocates for a prudent alternative: the strategic incorporation of two flash tanks within the process architecture. These flash tanks, thoughtfully configured and positioned, play a pivotal role in inducing a significant pressure drop, thereby facilitating the effective separation of CO₂ while artfully circumventing the need for cryogenic conditions. This decision to navigate just below the cryogenic threshold ensures the preservation of economic feasibility, safety, and environmental responsibility while delivering CO₂ recovery and purity levels that rival those achieved through the rigors of cryogenic distillation.

In summation, this study underscores the pivotal role of pressure dynamics in CO₂ separation and highlights the multifaceted implications of operating under cryogenic conditions. By advocating for a well-calibrated implementation of flash tanks, our proposed model offers a balanced approach that not only rivals the performance of cryogenic distillation but also navigates the complex terrain of economic viability, safety, and environmental responsibility in the pursuit of high-purity CO₂ recovery.

Finally, for the desorption column, it is important to highlight that it is developed in a conventional distillation to recover the solvent as well as to obtain pure CO₂. In this step, the solvent amount has a direct influence on the reboiler duty, as the higher the solvent amount the reflux ratio will increase and therefore also the energy requirements for the capture. As preliminary conclusions, from the sensibility analysis, it was possible to see the considerable impact that every variable has over the complete process. Although the sensitivity analysis allows finding combinations of the variables in which the expected recovery and purity are achieved, it is necessary to submit the process to some optimization method that allows finding an optimal design.

To evaluate the sustainability of the ChCl:U processes, it was evaluated the overall performance against the conventional CO₂ capture process with MEA. Both processes were evaluated with three different indexes: *TAC* to evaluate the economics of the processes, *EI99* to evaluate the environmental footprint, and *IR* to evaluate how safe are the processes. The results obtained from this analysis are shown in Table 8, while the trend of the results can be explained according to the values of the design and operation variables shown in Table 7.

To make a better analysis, the economic index was split into equipment cost, utility cost, solvent cost, and operation

Table 8 Economic, environmental, and safety comparison of the proposed model using ChCl:U against the conventional process

	MEA conventional process		ChCl:U proposed process	
Equipment cost (USD)	Absorption column	17,341.92	Absorption column	17,894.39
			Flash 1	4570.40
	Desorption column	139,378.03	Flash 2	2261.16
	Total	156,719.95	Desorption column	81,665.21
Utility costs (USD)	Refrigerant (R290)	21,625,265.16	Total	106,391.16
	Medium/high water vapor	68,342,501.8	Refrigerant (R1150)	4,167,750.75
	Total	89,967,766.96	Medium/high water vapor	68,128,341.534
Solvent cost (USD)	2,032,225.75		Total	72,296,092.28
Operation cost (USD)	91,833,480.61		89,629,595.00	
Energy (GJ/h)	363.74		72,422,748.43	
<i>EI99</i> (kEcopoints)	22,550.17		271.4	
<i>IR</i> (IR/kg total flow)	1.335E – 11		19,398.46	
			7.16E – 11	

cost. Analyzing the distribution of costs, it is possible to see that globally, the equipment cost for the conventional process is 32% more expensive than the proposed model. The proposed process would be expected to be more expensive because it contains two extra process units. However, in the case of the conventional process, the desorption column is bigger than the one from the proposed process. So, by being bigger in dimensions, the costs of the equipment increase. In this case, the cost of this equipment is big enough to exceed the cost of the two flash equipment and the desorption column corresponding to the proposed process.

Concerning the utility costs, it is possible to see that the conventional process is more expensive. This can be explained since to achieve the recovery obtained in the conventional process, it is necessary to increase the heating and cooling requirements. As well, this increment influences the energy requirements for the process which impacts directly the cost.

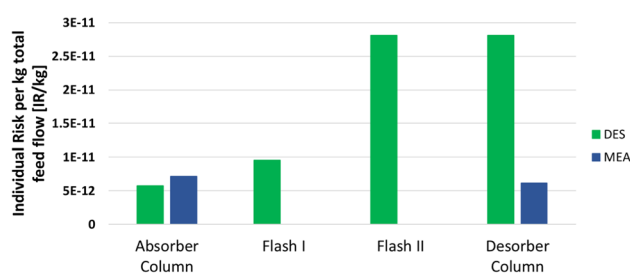
Regarding the solvent's cost, it is evident that producing ChCl:U is more expensive than the cost of MEA. Although ChCl:U components are common compounds, to reduce processing costs it is necessary to increase the demand for this solvent. Unlike MEA, which has several industrial applications, its demand is common, and therefore, there are standard costs for this solvent. Although in some sections the costs of the proposed process were lower, the cost of solvent is big enough to globally exceed the total cost of the conventional process.

About the environmental index, the proposed process has a lower environmental impact compared with the conventional process. This is due to different factors; one of them is the selection of the solvent in each process. For the conventional process, since MEA is highly toxic, there is an increase in health damage compared to the use of ChCl:U, which due to the nature of its components is not toxic. On

the other hand, having a high energy requirement in the conventional process, it will considerably affect the sum of the environmental impact. Moreover, even when the proposed model has more equipment units, the conventional process is dimensionally bigger, and therefore, it required more steel impacting directly on the environmental indicator.

The safety index was normalized to the individual risk per kilogram of the total flow that enters the process. This is because the *IR* is affected due to the amount of mass; in that way, to make a comparison with the result obtained, it is necessary to evaluate the index per kilogram. Figure 10 presents the individual risk index, sorted by process unit, giving an overview of how each unit affects the process safety index.

According to Fig. 10, it can be seen graphically that for the absorption column, the *IR* index presented by the process with DES is relatively lower than the process with MEA. This could be explained since in both cases, according to Table 7, there is no considerable difference in the size of both columns. It could be expected that the absorption column, in the case of the process with DES, as it has higher pressure and temperature conditions, would have a higher contribution to the risk index; however, these conditions repeat until the later stages of the process. In the case of flash tanks,

**Fig. 10** Comparison of the breakdown of the risk index by process unit for the DES process against the MEA process

according to Table 7, very large pressure drops are occurring so this condition directly affects the weighting of the safety index. As in the flash tanks, the desorption column intends to maintain a large pressure drop compared to the first stage of absorption. Although this column is smaller in dimensions than the desorption column of the process with MEA, the condition of global pressure drop in the process has a notable impact on the weighting of the risk index of the process.

Globally, the results show that there is more risk of an accident occurring in the proposed process. This can be explained due to the values of the operating variables as well because it is a process with more operation equipment. In addition to the analysis, the conventional process is operating with two columns, while the proposed process has 2 columns and 2 flash tanks. When a process contains more process units, it will be considered more dangerous. Moreover, the proposed model is operating at high pressure (13 bar) during the absorption and then it is necessary to have a big pressure drop (1 bar), while in comparison with the conventional process operating pressure stays constant at low pressure (1 bar) during all the capture process. The range of operational conditions affects the safety index.

Conclusion

This study explores the potential application of choline urea chloride (1:2) as a sustainable solvent in the context of post-combustion carbon dioxide (CO₂) capture. When comparing some properties of DES at different temperatures, the values predicted by the simulator manage to adjust very approximately to the values reported in the literature. Reporting an error percentage of less than 1% for most of the compared data, in the case of the solvent composition analysis, the maximum error obtained is 10%, so according to Brook and Arnold (2018), it is the maximum limit value allowed for the design of experiments.

From the comparison of the CO₂ capture with ChCl:U concerning the conventional capture process with MEA, it can be concluded that under the same flue gas flow conditions, the proposed model turns out to be a competitive option. From an economic point of view, the proposed model presents lower costs of equipment, operation, and utilities. It also requires less energy consumption compared to the traditional model with MEA. Regarding the costs of the solvent, ChCl:U turns out to be more expensive. So, to reduce production costs, it is necessary to increase the demand for this solvent at the industrial level. In the case study presented, the model using the ChCl:U eutectic mixture as solvent presents a good performance globally. For this case study, through a sensitivity analysis, the values of the variables that allowed obtaining a CO₂ recovery greater than 95% with a purity greater than 95% were found. Regarding

the environmental aspect, it is possible to obtain a process whose environmental impact is less than the traditional process with MEA. Thus, it is verified that the application of DES as green solvent effectively leads to the improvement of existing processes, reducing the environmental impact. Moreover, through the use of green solvents like DES, it is possible to obtain sustainable processes such as the case of CO₂ capture technologies that meet the objectives of the 2030 Agenda. For this reason, it would be interesting to study the industrial application of these solvents in other processes, different from CO₂ capture.

Nomenclature AC: Absorption column; Ω : Acentric factor; BLEVE: Boiling Liquid Expanding Vapor Explosion; T_B : Boiling temperature; CO₂: Carbon dioxide; ChCl:U: Choline chloride:urea; Z_c : Critical compressibility factor; P_c : Critical pressure; T_c : Critical temperature; V_c : Critical volume; DES: Deep eutectic solvent; P : Density; DC: Desorber column; *EI99*: Eco-indicator 99; FT-1: Flash tank number 1; FT-2: Flash tank number 2; R : Ideal gas constant; *IR*: Individual Risk; IPCC: Intergovernmental Panel on Climate Change; LCA: Life cycle assessment; C_p : Molar heat capacity; MW: Molecular weight; NRTL: Nonrandom Two-Liquid; QRA: Qualitative Risk Analysis; γ : Surface tension; TAC: Total Annual Cost; UVCE: Unconfined Vapor Cloud Explosion; η : Viscosity

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Data Availability The authors declare that the data supporting the findings of this study are available within the paper and its Supplementary Information files. Should any raw data files be needed in another format they are available from the corresponding author upon reasonable request. Source data are provided with this paper.

Declarations

Conflict of Interest The authors declare no competing interests.

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