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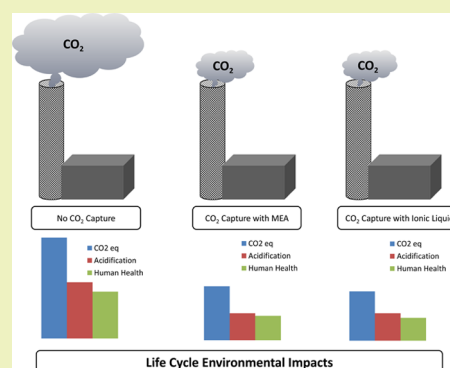
Life Cycle Environmental Implications of CO₂ Capture and Sequestration with Ionic Liquid 1-Butyl-3-methylimidazolium Acetate

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ABSTRACT: In this paper, for the first time, we present findings on the life cycle environmental implications of an ionic liquid (1-butyl-3-methylimidazolium acetate, [bmim]⁺[Ac][−])-based carbon capture and sequestration (CCS) process retrofitted to a coal-fired power plant. Ionic liquids are considered as promising alternative solvents to currently proven technology based on monoethanolamine (MEA). The results indicate that a CCS process with 90% CO₂ capture efficiency based on [bmim]⁺[Ac][−] reduces life cycle greenhouse gas (GHG) emissions by only 50%. This does not compare favorably with 75% life cycle GHG reductions offered by the MEA-based CCS process. In depth analysis reveals that despite lower total energy requirements for the ionic liquid process the increased need for electricity results in poor environmental performance. The insights gained from the results of the ionic liquid [bmim]⁺[Ac][−] CCS process is used to explore critical life cycle environmental performance parameters and trade-offs in the context of future design of functionalized ionic liquids for CO₂ capture. This work will aid energy system and policy analysts to understand the full implications of future ionic liquid-based CCS systems.

KEYWORDS: Ionic liquid, Carbon capture, Climate mitigation, Environmental evaluation, Process simulation



INTRODUCTION

In United States, electricity generation is the main source of CO₂ emissions with coal-fired and natural gas power plants contributing approximately 80% and 19%, respectively. Coal-fired power plant emissions comprise the largest stationary source of CO₂ emissions in the United States. In 2009, United States coal power plants generated 2.4 GtCO₂ out of the total United States CO₂ emissions of 6 GtCO₂ (about 40%).¹ In the foreseeable future, fossil fuel, in particular coal, will continue to dominate the power sector with CO₂ emissions projected to double by 2050 relative to 2007 emissions.² The correlation between increased CO₂ emissions and the greenhouse effect is widely accepted,³ and efforts aimed at CO₂ emission reductions are underway. Carbon capture and storage (CCS) is seen as a promising solution as it will allow the continual use of fossil resources while mitigating climate impacts.⁴ As current United States and global power production is dominated by pulverized coal power plants, the near term solution most likely to result in the largest CO₂ reductions would be retrofitting existing coal-fired power plants to capture and compress CO₂ for geologic storage.¹ CO₂ separation technologies currently available and widely used in industries are based on chemical reactions involving alkanolamine solvents such as monoethanolamine (MEA). Adoption of this absorption technology is currently hindered due to high operating costs as a result of high energy penalties for solvent regeneration¹ and high capital costs. CO₂ removal with an aqueous MEA solution involves chemisorption, with two amines reacting with CO₂ forming a carbamate salt.⁵ The molar uptake of CO₂ follows a stoichiometry of one

mole of CO₂ to two moles of amine. The enthalpy of reaction is estimated to be as high as −85 kJ/mol at 40 °C and the large heat of reaction results in large energy requirements to uncomplex the solvent in the regenerator.⁵ For a process involving 30 wt % MEA (aqueous) solution and 90% CO₂ removal, the energy requirement is estimated to be about 4 GJ/ton of CO₂ resulting in approximately 30% parasitic energy consumption and double the cost of electricity (COE).⁵ The U.S. Department of Energy (DOE) has set a goal of 90% CO₂ removal with less than 35% COE increase.⁶ The capture and regeneration steps of the MEA process are energy intensive, and this energy requirement has the largest influence on the cost driving researchers to focus on development of less energy intensive alternatives.⁷

Ionic liquid-based solvents are forerunners in current solvent research, development, and demonstration efforts.^{1,2} They are proposed as alternative solvents to the corrosive and degradation-sensitive amine solvents.^{8,9} Ionic liquids (ILs) are also attractive because they have negligible vapor pressure and, unlike volatile MEA, do not cause environmental concerns or raise costs associated with solvent replacement. ILs also have high decomposition temperatures, can operate over a wide liquid range, and can be chemically tuned.¹⁰ Some challenges associated with the use of IL for CO₂ capture include high viscosity, lower capacity due to high molecular weight, and cost.

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Conventional ionic liquids absorb CO_2 based on a physical mechanism (no chemical reaction). Physical solubility and selectivity of CO_2 for different ionic liquids have been reported.^{11–14} Use of physical ionic liquids is mainly hindered by the low absorption capacity at the small partial pressures of CO_2 in post-combustion flue gas,² for example, 33 mols of IL $[\text{HMIM}]^+[\text{NTF}]^-$ will be required for every mole of CO_2 , and considering the high molecular weight of ILs, this would require a large volume.⁵ This resulted in researchers focusing on functionalized ionic liquids that allow chemical absorption. Examples include cation-functionalized ionic liquids,^{15,16} anion-functionalized ionic liquids,¹⁷ and carboxylate functionalized anions such as acetate.¹⁸

Energy requirements for CCS include the heat needed to regenerate the solvent and remove CO_2 , pumping requirements, electricity necessary for cooling the solvent to process temperature, and the energy needed to compress the separated CO_2 . It has been recognized that alternatives to aqueous amines for CO_2 capture must have a lower energy penalty and be less expensive.⁵ Toward this end, clear performance targets have been set by the DOE.¹⁹ Shiflett et al.²⁰ reported a 16% reduction in total energy for the IL process compared to the MEA process. Further, 11% reduction in total investment cost and 12% reduction in equipment footprint for the IL process were reported, making the ionic liquid process very attractive. However, it is important that alternatives be compared with respect to their environmental performance as the original motivation to develop CCS technology is GHG mitigation. To be an effective means of addressing climate change, it is essential that this environmental evaluation considers system-wide life cycle impacts rather than just reductions in energy penalty. It is possible that alternative CCS technologies with 90% CO_2 capture and similar parasitic energy requirements have widely differing life cycle climate and environmental footprints due to inherent differences in their processes and differences in the type of energy consumed (e.g., heat vs electricity). Such crucial information can only be derived from a holistic assessment of the energy and the environmental footprint using a life cycle assessment (LCA) framework.

In this work, for the first time, we report life cycle impacts of CCS using ionic liquids. We compare the environmental footprint of IL-based CCS technology with the MEA-based process and the reference scenario of no CO_2 capture. This work will aid energy system and policy analysts understand the full implications of future IL-based CCS technologies and effectively compare CCS with other mitigation strategies. Most importantly, this study will provide researchers working on development of task-specific ionic liquids for CO_2 capture with critical environmental performance parameters and trade-offs. This will enable them to tune new ionic liquid structures not only from a stand point of reducing energy penalty and cost but also for improving life cycle environmental and climate performance.

METHODS

CO_2 Capture System: Ionic liquid BMIM Acetate Process.

There are no commercial-scale CO_2 capture units based on ionic liquids as this technology is currently at the preliminary stages of development. This presents an enormous challenge in terms of accurate environmental performance estimation at an industrial scale. In this paper, we rely on process simulation to provide mass and energy balances for an industrial-scale CCS unit-based on IL. To our knowledge, the only process simulation for post-combustion CO_2 capture based on ionic liquids was published by Shiflett et al.²⁰ The IL

process was modeled using 1-butyl-3-methylimidazolium acetate ($[\text{bmim}]^+[\text{Ac}]^-$), which is believed to follow chemical absorption behavior.²¹ In this work, we utilize detailed mass and energy balance from Shiflett et al.²⁰ who performed the simulation for the IL CO_2 capture unit with Aspen Plus using experimental VLE data of CO_2 + $[\text{bmim}]^+[\text{Ac}]^-$.²¹ The composition of flue gas that enters the carbon capture unit is assumed to be 78% N_2 , 13% CO_2 , and 9% H_2O , by volume.

The IL simulation was based on 90% CO_2 removal from flue gas, equal to 47000 t per year, with $[\text{bmim}]^+[\text{Ac}]^-$ as solvent. As the IL absorption process works under pressure, the flue gas is compressed to around 8 bar. The process as shown in Figure 1 consists of a packed

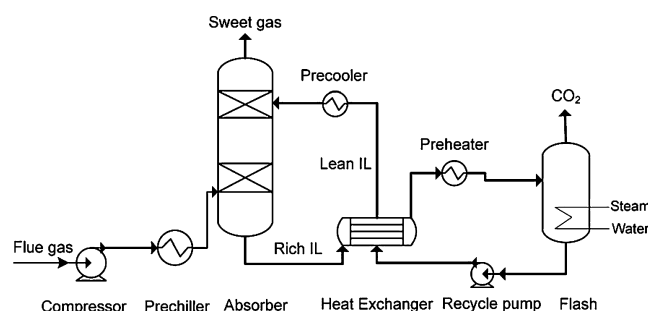


Figure 1. Typical process configuration of a carbon capture system.

absorption column for separating CO_2 from flue gas and a flash drum for solvent regeneration. The absorption column was modeled using 20 theoretical stages, and the stripping flash tank is a simple single-stage desorber. A heat exchanger was included between the two units to recover heat and reduce energy load. Heat energy (steam) is used in flash preheater and flash drum to release the chemically absorbed CO_2 from IL. Electricity is utilized in the absorber precooler that is a refrigeration machine to cool the solvent to 273 K. Other equipment using electricity include circulating pumps for ILs and a flue gas compressor. The authors optimized the operating conditions and flow rates to maximize CO_2 recovery and minimize energy consumption. A summary of the simulation results reproduced from Shiflett et al.²⁰ is presented in Table 5.

Life Cycle Inventory. A functional unit of 1 MWh electricity delivered to the grid was selected for this study. The system boundary for this study is shown in Figure 2. The baseline scenario was the electricity production from a 185 MW bituminous coal power plant without carbon capture and sequestration (CCS). The unit process inventory related to the reference case power plant and upstream emissions from coal mining and transportation were derived from the U.S. Life Cycle Inventory Database (USLCI).²² The energy requirements for the 30 mass percent MEA process were set to 4 $\text{GJ}_{\text{steam}}/\text{ton CO}_2$ using data from Koornneef et al.²³ On the basis of data provided by Koornneef et al.,²³ we assumed a loss of 2.34 kg MEA per ton of CO_2 captured, out of which 0.014 kg/ton CO_2 were direct emissions to the atmosphere. Because of MEA degradation by chemical reactions, formation of other pollutants like NH_3 , acetaldehyde, and acetone may occur. On the basis of estimates from Koornneef et al.,²³ we set the NH_3 formation amount to 0.21 kg/t CO_2 , and for acetaldehyde and acetone, we selected 0.008 and 0.001 kg/ton CO_2 , respectively, based on the analysis by Moser et al.²⁴ These emissions also go directly to the atmosphere from the top of the absorber. The total loss of MEA translated into a makeup solvent (MEA) requirement of 2.37 kg per MWh electricity. Emissions related to the production of the makeup MEA was derived using a combination of Ecoinvent²⁵ and USLCI²² data. We neglect the environmental impact of producing the initial charge of the MEA solution. It is well known that the CCS process with MEA scrubbing results in reduction of other pollutants such as SO_2 and NO_x .²⁶ Table 1 summarizes the assumed percentage reductions of other pollutants,²³ other emissions, and energy parameters for the MEA process.

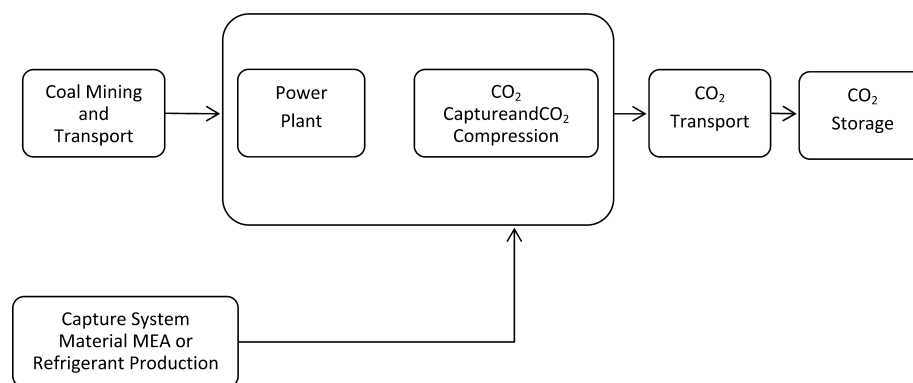


Figure 2. System boundary for CCS life cycle assessment.

Table 1. Performance Parameters of CCS–MEA Process

parameter	unit	value
CO ₂ removal	%	90
SO ₂ removal	%	90
NO _x removal	%	1.25
HCl removal	%	95
HF removal	%	90
PM removal	%	50
MEA emission	kg/t CO ₂	0.014
NH ₃ emission	kg/t CO ₂	0.21
heat requirement	GJ _{steam}	4
electricity requirement	kWh/t CO ₂	23.6
power equivalent factor	GJ _e /GJ _{steam}	0.20

As for the ionic liquid process, we assumed no degradation and loss with complete reuse of the ionic liquid without any makeup requirements. This is reasonable because ionic liquids are essentially nonvolatile and have high thermal stability. In addition, we assumed the same percentage reduction of other pollutants as in the case of MEA. Energy requirements for the IL capture process were derived from process simulation results of Shiflett et al.²⁰ As for cooling requirements (for IL), we selected industrial refrigeration using ammonia as the refrigerant. We assumed a specific charge of 5 kg per KW cooling load for ammonia based on data provided by Poggi et al.²⁷ We assumed a 7% refrigerant loss per KW electricity per year based on data from the IPCC report.²⁸ Because we assumed no vaporization of ionic liquid during the process and that the IL will be recycled and no makeup solvent is needed, we can neglect the environmental impacts of IL production phase.

After capture, the CO₂ flow needs to be first dehydrated and then compressed by multi-stage compressors to 11 MPa to facilitate transport via pipeline. Electricity requirement for running these four-stage compressors comes from the power generation process. For the injection of CO₂ into underground reservoirs, it needs to be compressed further to 15 MPa by two-stage compressors running with electricity from the grid. Energy requirements for the compressors were calculated using eq 1²⁹

$$W = \frac{ZRT_1}{M} \frac{N\gamma}{\gamma - 1} \left[\left(\frac{p_2}{p_1} \right)^{\gamma - 1/N\gamma} - 1 \right] \text{ and } E = \frac{W}{\eta_{is}\eta_m 3600} \quad (1)$$

where W is the specific work (kJ/kg CO₂); E is the specific electricity requirement (kWh/kg CO₂); Z is the compressibility factor (0.9942); R is the universal gas constant (8.3145 J/(mole K)); T_1 is the suction temperature (313.15 K for MEA process and 344 K for IL process); γ is the specific heat ratio (C_p/C_v) (1.293759); M is the molar mass (44.01 g/mol); p_1 is suction pressure (MPa) (transport = 0.101325, injection = 10.7); p_2 is the discharge pressure (MPa) (transport = 11, injection = 15); N is the number of compressor stages (transport = 4, injection = 2); η_{is} is the isentropic efficiency (80%); and η_m is the mechanical efficiency (99%).²³ The electricity needed for CO₂ compression was calculated as 111 and 119 kWh/t CO₂ for the MEA and IL process, respectively. As pointed out before, transportation of captured CO₂ is assumed to be carried out by a 50 km length pipeline. Fugitive emissions (leakage) of CO₂ during operation were estimated with the use of the methodology developed by the IPCC.²⁸ As the main goal of this study is to compare the CCS process with two solvents, we decided not to consider construction and dismantling phases for CCS plants.

Impact Assessment. Impact assessment methods are used to translate the inventory data into environmental impacts through the use of characterization factors. These characterization factors represent relative impacts of different chemicals (as in the case of global warming potential) or are a function of fate, exposure, and effect of the chemicals (as in the case of human health and ecotoxicity). In this study, we adopted the impact assessment methodology based on Tools for the Reduction and Assessment of Chemical and other Environmental Impacts (TRACI) developed by the U.S. Environmental Protection Agency.³⁰ The following impact categories were considered: (1) global warming potential (GWP), (2) acidification (AP), (3) eutrophication (EP), (4) smog formation (smog), (5) human health criteria (HH criteria), (6) human health cancer (HH cancer), (7) human health noncancer (HH noncancer), and (8) ecotoxicity (EcoTox). We applied classification and characterization steps to relate emissions in the inventory to relevant impact categories and to identify characterization factors based on the media where the emissions occur.

Table 2. Environmental Impacts of Reference Case (w/o CCS), CCS–MEA, and CCS–IL (Functional Unit: 1 MWh Electricity Delivered to the Grid)

	GWP (kg CO ₂ equiv)	acidification (kg H+ moles equiv)	HH Criteria (kg PM10 equiv)	eutrophication (kg N equiv)	smog (kg O ₃ equiv)	ecotoxicity (CTU eco)	HH (CTU cancer)	HH (CTU noncancer)
w/o CCS	1071	481	1.39	0.133	74.7	1.30	250×10^{-7}	1.66×10^{-5}
CCS–MEA	281	265	2.12	0.245	117.3	2.76	4.71×10^{-7}	2.65×10^{-5}
CCS–IL	573	463	5.55	0.396	216.1	4.75	8.63×10^{-7}	5.29×10^{-5}

RESULTS AND DISCUSSION

This section summarizes the key findings related to the life cycle environmental impacts for the functional unit of 1 MWh electricity generated from a power plant using an IL ([bmim]⁺[Ac][−])-based CCS system compared to a power plant using a MEA-based CCS system and the reference case of no CO₂ capture. The total scores related to the eight impact categories are presented in Table 2. The impact profiles related to the three cases are shown in Figure 3, with the reference case

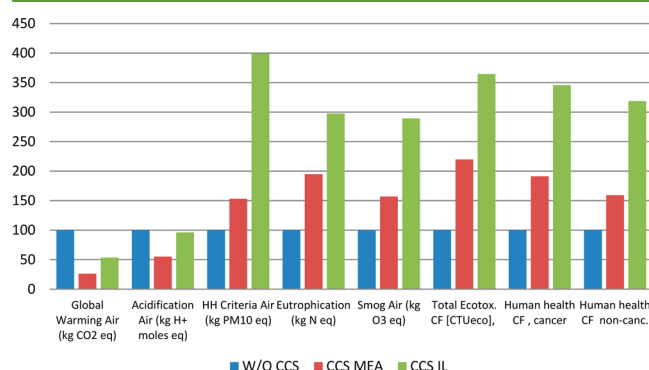


Figure 3. Comparison of scaled impacts of reference case (w/o CCS), CCS-MEA, and CCS-IL.

of no CO₂ capture set at 100% and the other two displayed at a level relative to the former. Comparison of the results shows that the GWP and AP decreased for the two cases with CCS in relation to the reference case, while all other impacts increased significantly. Compared to the reference case, CCS-MEA resulted in 75% reduction in GWP, while CCS-IL shows only about a 50% reduction. This is due to the fact that the CCS-IL process requires more coal equivalent energy than the CCS-MEA process. As for acidification potential, a 50% reduction is achieved through the MEA process, while a small reduction is achieved for the IL process. This reduction is due to the fact that SO₂ (a strong acid gas) concentration is significantly reduced by the modified FGD control required²⁶ for the CCS cases. The unit process breakdown of the global warming potential and acidification potential are presented in Figures 4 and 5, respectively.

Previous modeling and simulation studies²⁰ have shown that the CCS system based on IL [bmim]⁺[Ac][−] reduces parasitic energy consumption by 16% and investment by 11% in comparison to a CCS system based on the traditional solvent

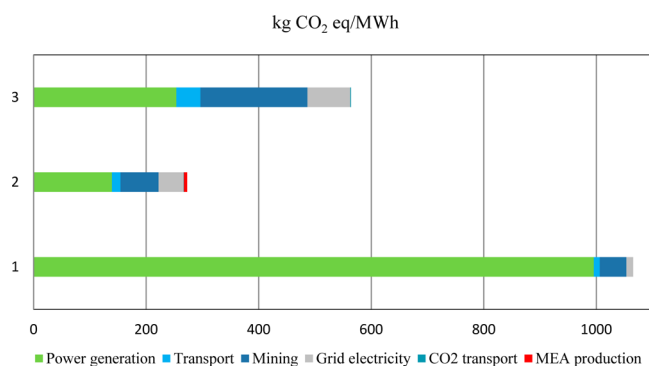


Figure 4. Unit process breakdown of global warming potential (GWP). 1- W/O CSS; 2- CCS MEA; 3- CCS IL

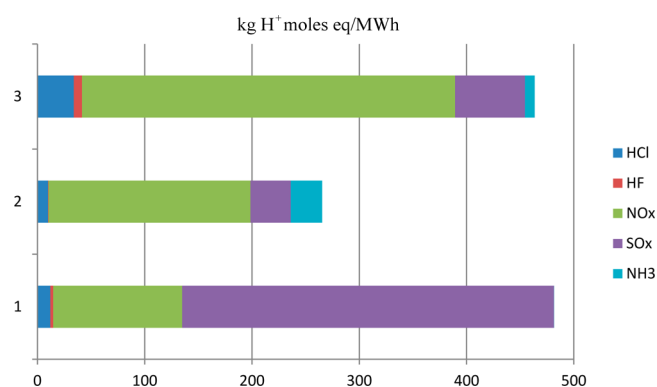


Figure 5. Unit process breakdown of acidification potential (AP). 1- W/O CSS; 2- CCS MEA; 3- CCS IL.

monoethanolamine. The net energy requirements for the two CCS processes for a capacity of 47,000 t reported by Shiflett et al.²⁰ are shown in Table 3. As shown in the table and as

Table 3. Total Energy Balance of CCS-MEA and CCS-IL

	MEA	IL
heat (kW)	11,627	7145
electricity (kW)	13	2645
total	11,640	9790

concluded by Shiflett et al.,²⁰ there is indeed a significant reduction (16%) in total energy required for the IL process with a similar capacity to capture the same amount (90%) of CO₂ as that of the MEA process. This reduction combined with the lower investment costs makes the IL process very attractive in relation to the MEA process from a technical and economic viewpoint.

However, the findings from this study show that life cycle greenhouse gas emissions from a power plant with 90% post-combustion CO₂ capture using ionic liquid [bmim]⁺[Ac][−] is lowered by only 50% in relation to no CO₂ capture. In comparison, 90% capture based on the traditional solvent, 30% aqueous monoethanolamine, reduces greenhouse gas emissions by 75%, indicating that the MEA process offers significantly higher climate mitigation than IL process. This contradiction is essentially due to the difference in the forms of energy required for the two CCS systems. As shown in Table 3, even though the overall energy balance is more favorable for the IL process, the electricity needed for this process (2645 kW) is much higher than the MEA process (13 kW). The power equivalent factor for electricity production in thermal power plants is around 0.20 GJ_{electricity}/GJ_{steam}.²³ This factor captures the electrical equivalent of heat associated with the loss of steam turbine output compared to the heat of steam implying that the energy penalty associated with the electricity use is higher than the use of steam. This, from an LCA perspective, translates into an increase in the net amount of coal needed to produce an energy equivalent of electricity compared to an energy equivalent of steam. The overall coal requirements taking into account the energy penalties for the two CCS processes are shown in Table 4.

It is clear from the above analysis that the increased electricity demand for the IL process results in higher life cycle GWP in comparison to the MEA process. In the case of the MEA process, the majority of the energy requirement is for heating the stripper reboiler to regenerate the solvent, while

Table 4. Coal Consumption for Different Scenarios

	kg coal/MWh net electricity
w/o CCS	442
CCS–MEA	631
CCS–IL	1777

some electricity (13 kW) is required to run the absorption and stripping pumps.²⁰ For the ionic liquid process, steam is required for the flash tank and flash preheater (for regeneration), while electricity (2645 kW) is required to run the flue gas compressor, refrigeration machine, and IL recycle pump.²⁰ More in depth treatment reveals that the electricity consumption in the CCS–IL case is primarily for refrigeration purposes related to cooling the IL to the required low process temperature and for compressing the flue gas to the required high operating pressure. Table 5 summarizes optimized

Table 5. Summary of Operating Conditions

	MEA		IL	
	T (K)	P (kPa)	T (K)	P (kPa)
absorber				
flue gas inlet	312	112	312	791
vent gas outlet	313	101	280	618
absorbent inlet	308	136	273	791
absorbent outlet	328	112	296	618
stripper unit				
CO ₂ outlet	329	143	344	108
absorbent inlet	361	239	344	446
absorbent outlet	401	164	344	108

operating conditions obtained from Shiflett et al.²⁰ of all main streams for the two processes. We can see that the absorbent inlet temperature requirement for the IL process is 0 °C, which demands an industrial refrigeration unit needing a large amount of electricity and refrigerant. However, to meet the same capacity with the MEA solvent, the inlet temperature can be as high as 30 °C, which is assessable with air fan coolers and does not require refrigeration. Similarly, the IL process requires a high pressure of 8 bar, compared to the atmospheric pressure for MEA, needing significant amount of electricity for flue gas compression. It is worth noting that even though the difference between the recycle solvent temperature and solvent inlet temperature is higher for the MEA process (93 °C) in comparison to the IL process (71 °C) the fact that the inlet temperature for the IL process is 0 °C acts as a big disadvantage. It appears that a key parameter for future ionic liquid development is to attain a solvent inlet temperature of above 300 K and operate close to atmospheric pressure. There are indeed several promising candidates that show similar or higher CO₂ solubility than [bmim]⁺[Ac][−] even at temperatures above 300 K.

It is estimated that current solvent absorption processes for CO₂ separation and compression require 2.5–4.32 GJ/ton CO₂ for 90% removal,^{2,23} while the theoretical minimum work required is 0.44 GJ/ton, which indicates significant potential for process improvements through the development of new solvents. ILs due to their vast tunability opportunities⁵ and availability of unlimited structural combinations³¹ offer a good platform. Current research efforts in the development of task specific ionic liquids for CO₂ capture are directed at reducing regeneration energy (heat) needed through lowering reaction

enthalpies while maintaining high loading capacities.³² Thermodynamically, there exists a trade-off between higher CO₂ capacities at a particular temperature and lower regeneration energy. From a climate mitigation viewpoint, our analysis shows that in addition to keeping parasitic energy requirements low specific reductions in the use of higher energy forms (electricity) are also important. This means that future ionic liquids should have high loading capacities at moderate process temperatures (25–30 °C) (to eliminate refrigeration needs) and low operating pressures (1 atm) (to eliminate compression needs) and at the same time have low reaction enthalpies to reduce regeneration energy. Several promising functionalized ionic liquids that could potentially meet the above requirements are currently being researched.³² In addition to solvent design, process optimization, proper equipment design, and heat integration can result in significant improvements.

Finally, with respect to key metrics, the DOE requires 90% CO₂ removal from flue gas with the cost of electricity increase less than 35%. This just relates to the cost per ton of CO₂ removed. Usually, because the purpose of adding a capture unit is to reduce the CO₂ emissions per net kWh delivered, the cost of CO₂ avoidance (eq 2) is used as an indicator

$$\text{cost of CO}_2 \text{ avoided (\$/ton CO}_2\text{)} = \frac{(\$/\text{kWh})_{\text{capture}} - (\$/\text{kWh})_{\text{reference}}}{(\text{ton CO}_2/\text{kWh})_{\text{reference}} - (\text{ton CO}_2/\text{kWh})_{\text{capture}}} \quad (2)$$

However, the real purpose of CCS is climate mitigation, and hence, we propose that different CO₂ control technologies be compared using the economic indicator that captures the cost of life cycle GHG avoidance (eq 3)

$$\text{cost of GHG avoided (\$/ton CO}_{2\text{equiv}}\text{)} = \frac{(\$/\text{kWh})_{\text{capture}} - (\$/\text{kWh})_{\text{reference}}}{(\text{ton life cycle CO}_{2\text{equiv}}/\text{kWh})_{\text{reference}} - (\text{ton life cycle CO}_{2\text{equiv}}/\text{kWh})_{\text{capture}}} \quad (3)$$

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Notes

The authors declare no competing financial interest.

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