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Screening of ionic liquids for acid gas removal

Giuseppe Andriania, Gianmaria Piob, Chiara Vianelloa,c, Ernesto Salzanob, Paolo Mocellina,c,\*

aUniversità degli Studi di Padova, Dipartimento di Ingegneria Industriale, Via Marzolo 9, 35131 Padova, Italia.

bUniversità di Bologna, Dipartimento di Ingegneria Civile, Chimica, Ambientale e dei Materiali, Via Terracini 28, 40131 Bologna, Italia.

cUniversità degli Studi di Padova, Dipartimento di Ingegneria Civile, Edile e Ambientale, Via Marzolo 9, 35131 Padova, Italia.

paolo.mocellin@unipd.it

To mitigate the environmental impacts of human activities, it is essential to carefully treat vented gaseous streams to prevent the release of undesired or toxic compounds into the atmosphere. These harmful chemicals can react with atmospheric elements such as oxygen or moisture, forming acids that pose significant environmental risks. Such acids can cause degradation of soils, contamination of surface and groundwater bodies, and damage to human-made structures and facilities. Therefore, developing and deploying technologies designed to remove the precursors of these polluting substances is paramount. Although several alternatives currently exist for this purpose, they often suffer from limitations such as low reusability, limited affinity for specific molecules, and inadequate absorbent capacity for effectively capturing the full range of pollutants in gaseous streams.

In light of these challenges, ionic liquids (ILs) have emerged as promising candidates for the selective and quantitative capture of pollutants. ILs offer distinct advantages, including their ability to be tailored for specific applications through chemical structure modifications, enhancing their selectivity and absorption capacity. However, to advance the development of sustainable capture technologies, it is crucial to thoroughly evaluate the absorption performance and physicochemical properties of ILs. This evaluation is necessary to identify the most effective ILs for pollutant capture and ensure that the selected materials can be used sustainably over the long term. In addition to their absorption efficiency, the thermal stability of ILs must be rigorously assessed to guarantee their durability and reliability in industrial applications.

Given these considerations, the present study undertakes a critical screening of various ILs for acid gas capture to identify the most suitable candidates for further research and potential industrial implementation, with a specific focus on SO2 capture. This screening process involves a comprehensive analysis of ILs based on key parameters, including their acid gas absorption capacity, the experimental conditions under which they operate (such as temperature and partial pressure of the treated gases), and their viscosity and density. By categorizing ILs according to these criteria, the study aims to pinpoint the ideal candidates for subsequent, more detailed thermal stability assessments, having the goal of advancing the field of acid gas capture by identifying ILs that demonstrate superior absorption performance and exhibit the necessary thermal stability for sustainable and long-term use in industrial processes.

* 1. Introduction

Reducing acid gas (AG) emissions is critical for sustainable development in our increasingly industrialized society. AGs encompass a variety of harmful species, including sulfur oxides (SOx), hydrogen sulfide (H2S), carbon dioxide (CO2), and nitrogen oxides (NOx), which can form acidic solutions upon contact with water, leading to severe environmental and public health issues (Halliday & Hatton, 2021) (Antonioni et al., 2016). Reducing AG emissions is crucial for mitigating the acidification of ecosystems, preventing the eutrophication of aquatic environments, reducing smog formation, and combating the intensifying effects of global warming (Tzimas et al., 2007). Among the methods for capturing AGs, chemical absorption is widely recognized and employed, with amine-based absorption (ABA) as the most prevalent approach (Mokhatab et al., 2019). Despite its widespread use, ABA suffers from several significant limitations, including high thermal energy demands for solvent regeneration, degradation and volatilization of amines, the potential for corrosion, and the formation of covalent bonds between certain AGs and amine molecules. These drawbacks underscore the need to develop alternative separation technologies that offer improved efficiency and selectivity for specific AGs.

In response to these challenges, ionic liquids (ILs) have emerged as a promising class of materials for AG absorption, attracting considerable attention from researchers and industry alike. ILs are characterized by their low vapor pressure, highly tunable chemical structure, and exceptional thermal and chemical stability, along with their superior solvent properties (Zhang et al., 2013). These materials are salts with low melting points, sometimes even lower than room temperature, which enhances their practicality in various applications. ILs have demonstrated remarkable capabilities in dissolving a broad spectrum of AGs, including CO2 (Geng et al., 2021), H2S (Tian et al., 2014), NOx (Duan et al., 2011), and SO2 (Huang et al., 2006), with particularly high selectivity for SO2. The notable solubility of SO2 in ILs is attributed to the favorable physical and chemical interactions between the ionic components of the ILs and the gas molecules (Jiang et al., 2018). These interactions can be finely tuned by altering the chemical structure of the ILs through modifications to their anionic and cationic components, allowing for the optimization of absorption performance for specific AGs.

Moreover, despite the chemical nature of the interactions involved in AG absorption, ILs offer the advantage of easy regeneration without the need for phase changes, making them both efficient and cost-effective for repeated use. Given the wide variety of ILs that have been investigated for SO2 absorption, this study aims to provide a comprehensive review of the available ILs, evaluating their effectiveness as reactive solvents. The goal is to guide future research efforts toward identifying the most promising ILs for AG capture, with a particular focus on enhancing the efficiency, selectivity, and sustainability of these materials for industrial-scale applications.

* 1. Literature screening

The ionic liquids (ILs) analyzed for SO2 capture will be listed and compared to identify those that offer the best balance between absorption performance, operational constraints, and physicochemical and fluid dynamic properties. The absorption performance is represented by the SO2 adsorption capacity per gram of IL (gSO2/gIL). The operational constraints are defined by the temperature at which the absorption experiments were conducted (Tabs). Regarding the physicochemical properties of the fluid, the density of the IL (ρIL) has been considered representative. Eventually, the fluid dynamic property of the fluid has been expressed in terms of the liquid viscosity (μIL). For consistency, experiments with a partial pressure of SO2 of approximately 0.101 MPa were considered in this analysis.

The ILs will be ranked based on different properties because the understanding of gSO2/gIL alone is insufficient for determining the suitability of a material for full-scale applications. Experimental temperature and pressure provide insights into operational boundary conditions since both the mentioned variables can dramatically affect the absorption performances. Conversely, viscosity and density reflect the fluid’s resistance to flow. Thus, they are crucial aspects to consider for an effective further industrial implementation of the analyzed material. Table 1 summarizes the collected data and highlights the ILs with gSO2/gIL > 1. The meaning of the abbreviations used for the chemical compounds forming the different IL are only reported in the Nomenclature section.

Looking at the information reported in Table 1, the first evidence is the dependence of the absorption capacity with respect to the experimental temperature. Increasing Tabs, the value of gSO2/gIL tends to decrease since higher temperatures promote the stripping of the absorbed gas with respect to its absorption. However, different ILs tend to exhibit different dependences of gSO2/gIL as a function of Tabs. [TMG][Im], for instance, tends to have absorption performances that decrease less, increasing the absorption temperature if compared to [TMG][3-PyO]. This evidence encourages the applicability of [TMG][Im] on a full scale because the thermal effects related to absorption tend to increase the temperature of the absorbing fluid. Thus, using an IL less sensitive to temperature variation enhances the reliability of the involved material and operation. For the sake of clarity, only the most promising compounds with gSO2/gIL>1 have been reported in Table 1. Another interesting notice is related to the viscosity of the reported ILs. Substantially, all the μIL reported are above the reference value of water, but some have a viscosity value that makes their applicability practical impossible. More specifically, the higher the value of μIL, the higher the energy requirement associated with fluid flow. Thus, ILs like [TMG][3-PyO], [E3Py]Cl or [E2Py]Cl, which have μIL hundreds of times higher than the water value, will be practically unusable, even if they exhibit high absorption performance. Eventually, regarding the ρIL values, all of them are very close to the water reference value, so the density value will not affect dramatically the performance of the analyzed ILs. Water has been considered as a reference fluid since it is the most adopted and well-studied fluid for industrial applications.

Table 1: Summary of analysed ionic liquids and related characteristics.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Type of IL | gSO2/gIL | Tabs | μIL | ρIL | References |
| [-] | [K] | [mPa s] | [kg/m3] |
| [TMG][3-PyO] | 1.374 | 293 | 2428 | 1085 | (Cui et al., 2021) |
| 1.206 | 303 | 627 | 1076 |
| 1.069 | 313 | 214 | 1068 |
| 0.916 | 323 | 89 | 1059 |
| 0.748 | 333 | 43 | 1050 |
| [TMG][Im] | 1.318 | 293 | 8.9 | 997 | (Shang et al., 2011)(Ye et al., 2016)(Chen et al., 2015) (Lei et al., 2014) |
| 1.108 | 313 | 5.4 | 990 |
| [E2Py]Cl | 1.155 | 293 | 11800 | 1184 | (Wang et al., 2014) |
| [TMG][2-PyO] | 1.145 | 293 | 233 | 1085 | (Cui et al., 2021) |
| [Emim][SCN] | 1.130 | 293 | 28.27 | 1120 | (Yang et al., 2013)(Wang et al., 2013) |
| [C2Mim][SCN] | 1.130 | 293 | NA | NA | (Wang et al., 2014)(Wang et al., 2013) |
| [Et2NEMim][Tetz] | 1.102 | 293 | NA | NA | (Ren et al., 2018)(Yang et al., 2013) |
| [P4442][4-PyO] | 1.080 | 293 | 881 | 973 | (Cui et al., 2021) |
| [E3Mim]Cl | 1.057 | 293 | NA | NA | (Wang et al., 2014) |
| [Bmim][HB(Im)3] | 1.055 | 293 | 1568 | 1300 | (Ren et al., 2018)(Zhang et al., 2016) |
| [E3Py]Cl | 1.050 | 293 | 9600 | 1178 | (Wang et al., 2014) |
| [P4442][3-PyO] | 1.035 | 293 | 543 | 987 | (Cui et al., 2021) |

* 1. Data elaboration

Figure 1 has been reported to elaborate further on the analyzed data. Here, the x-axis reports 1/μIL as a self-standing indicator of operational expenditures (OPEX), whereas the y-axis reports gSO2/gIL as a capital expenditure (CAPEX) indicator. More specifically, 1/μIL and gSO2/gIL have been chosed as representative costs indexes. More specifically, the 1/μIL is inversely proportional to OPEX, since the higher the 1/μIL, the lower the OPEX. On the contrary, gSO2/gIL is inversely proportional to CAPEX, since the higher gSO2/gIL, the lower the CAPEX. As a matter of fact, the higher μIL, the higher the pressure drops related to fluid flow, and conversely, the higher the expenditure related to the energy that must be supplied to pumps. Regarding gSO2/gIL, the higher its value, the lower the amount of IL that must be involved to capture a fixed amount of SO2 from a process stream. Consequently, lower quantities of ILs mean smaller equipment to be involved in absorption operations. Since the CAPEX linked to purchasing industrial equipment is substantially related to the equipment volume, the link between CAPEX and gSO2/gIL becomes evident. Eventually, Figure 1 can be represented as a valuable tool to visualize the absorption performances of the analyzed materials.



Figure 1. Graphical summary of ionic liquids with gSO2/gIL > 1 in terms of CAPEX vs OPEX indexes.

Based on the data presented in Table 1, [TMG][Im] emerges as a promising candidate for industrial-scale application for selective and efficient SO2 capture, as confirmed by Figure 1. Although [TMG][Im] ranks second in terms of gSO2/gIL, surpassed only by [TMG][3-PyO], it offers significantly lower viscosity - 300 times less - while maintaining nearly the same density as [TMG][3-PyO]. This indicates that despite capturing 0.056 gSO2/gIL less at 298 K, [TMG][Im] is much easier to handle. Furthermore, [TMG][Im] demonstrates a more stable absorption performance with respect to Tabs compared to [TMG][3-PyO], indicating a more reliable capability for SO2 capture as the absorption temperature increases.

When comparing the absorption performance of [TMG][Im] with conventional amines, it is important to note that the absorption capacity of amines varies depending on the specific type selected. Generally, amine-based technologies can capture up to 1 gSO₂/gAMINE under the most favorable conditions (Kohl & Nielsen, 1997). However, unlike ionic liquid technologies, amines are typically used in aqueous solutions with a maximum concentration of 30% w/w (Laribi et al., 2018). This reduces the absorption capacity to approximately 0.3 gSO₂/gSOLUTION. Additionally, the absorption performance of amines decreases over time due to degradation, leading to reduced efficiency with prolonged use.

An intriguing feature of the analyzed ILs is their exceptionally high selectivity for SO₂ sorption in the presence of CO₂ (Geng et al., 2021) (Huang, et al., 2014). The selectivity can reach values approaching unity, indicating that ILs technology can be effectively integrated with existing acid gas removal solutions susceptible to SO₂ poisoning. For example, the potassium carbonate system developed by the Giammarco-Vetrocoke process (Ullmann, 2011) can be deactivated in the presence of SO₂ due to competitive chemical reactions (Wappel et al., 2011). By pre-treating a gas mixture with ILs to selectively remove SO₂, the subsequent potassium carbonate unit can operate without the risk of poisoning, thereby ensuring reliable CO₂ absorption in the second stage. Conversely, a 30% w/w amine solution has a viscosity of approximately 2.6 mPa·s at 298 K (Hsu & Li, 1997). This means that while the amine solution captures only 23% of the SO₂ absorbed by [TMG][Im], its viscosity is 30% that of [TMG][Im]. Despite this, the higher stability of ILs compared to amines suggests that over long-term use, the advantages of their superior absorption capacity and thermal stability will outweigh the disadvantages associated with higher costs due to fluid flow resistance (Dutcher et al., 2015). In addition, the adoption of multi-stage treatment systems for the removal of acid gases, will be needed in many situations to obtain the required removal efficiency (Dal Pozzo et al., 2020). Ultimately, SO2 is captured by ILs through reversible chemical bonds (Shang et al., 2011), enabling a sustainable and non-invasive regeneration of the solvent.

* 1. Current limitations

An in-depth examination of the data in Table 1 reveals certain limitations in the current scientific literature regarding ionic liquids (ILs). Notably, for several ILs, critical information about viscosity (μIL) and density (ρIL) remains incomplete across the compounds analyzed. This gap underscores the need for more comprehensive and systematic investigations into the physicochemical properties of these materials. The lack of available data becomes particularly evident when considering essential thermal properties such as heat capacity (Cp) and thermal conductivity (kT), which are crucial for accurate modelling and simulation of the thermal behavior of materials. The absence of this data constrains the potential for further developments and detailed analyses, limiting the reliability of predictions and optimizations for practical applications.

Moreover, significant gaps in knowledge also emerge when considering the thermal stability of ILs. To date, the decomposition behavior of these substances has not been thoroughly explored. Specifically, standard values typically obtained through calorimetric analysis—such as onset temperature (Tonset) and pressure (Ponset), peak temperature (Tmax) and pressure (Pmax), decomposition activation energy (Ea), pre-exponential factor (kk∞), and reaction enthalpy (ΔHr)—are still largely unavailable. The lack of this fundamental data hampers the ability to fully assess the safety and efficiency of ILs in various applications.

Despite these limitations, [TMG][Im] currently stands out as a promising candidate for large-scale implementation when evaluated against CAPEX (capital expenditure) and OPEX (operational expenditure) criteria. However, to ensure its suitability for industrial use, further research must be conducted to characterize its physicochemical properties and decomposition behavior fully. Such investigations would enable more detailed and accurate analyses, including thermal stability assessment (Andriani et al., 2024a), which are critical for ensuring the safe and efficient use of [TMG][Im] in process equipment (Andriani et al., 2024b), its secure storage in industrial facilities, and its behavior during potential accidental shutdowns (Andriani et al., 2024c). Addressing these gaps in knowledge is essential to advancing the practical application of ILs in various industries (Andriani et al., 2024d), where reliable data on thermal properties and decomposition characteristics are crucial for safe and effective operation (Andriani et al., 2024e).

* 1. Conclusions

A comprehensive review of the existing literature on the application of ionic liquids (ILs) for the selective and efficient capture of sulphur dioxide (SO2) has been undertaken. This review highlights the growing recognition of ILs as a viable technological alternative to conventional acid gas capture materials. The advantages of ILs, including their high regeneration properties, low volatility, selective SO2 absorption, and customizable molecular structures, make them particularly attractive for this purpose. Among the various ILs examined, [TMG][Im] has emerged as the most promising candidate for further investigation. It demonstrates an optimal combination of absorption efficiency, appropriate viscosity, suitable density, and stable performance in capturing SO2 across various operational temperatures.

The identification of [TMG][Im] as the leading candidate is based on its superior performance CAPEX and OPEX metrics compared to other ILs, making it a compelling choice for future studies. However, to ensure that [TMG][Im] meets the stringent requirements for industrial-scale application, it is crucial to conduct additional research. This should include a detailed thermal stability assessment to evaluate its robustness under different operating conditions. Such studies are essential to confirm the reliability and safety of [TMG][Im] in large-scale separation processes and storage systems. Understanding the thermal behavior of [TMG][Im] is particularly important, as it directly impacts its performance during extended operational periods and under varying temperature regimes.

In summary, while [TMG][Im] shows great potential for SO2 capture, further experimental validation is necessary. Investigating its thermal stability and decomposition characteristics will ensure that [TMG][Im] can be effectively and safely implemented in industrial applications. These additional studies will contribute to a deeper understanding of the material's capabilities and limitations, ultimately guiding its development and optimization for practical use in SO2 separation technologies and secure storage solutions. Further efforts will focus on a detailed experimental thermal analysis to assess the stability of the most promising compounds.

Nomenclature

TMG – 1,1,3,3-tetramethylguanidinium

ABA – amine-based absorption

AG – acid gas

Cp – heat capacity

kT – thermal conductivity

Im – imidazole anion

Tonset – onset temperature

Ponset – onset pressure

Tmax – peak temperature

Pmax – peak pressure

Ea – activation energy

kk∞ – pre-exponential factor

ΔHr – reaction enthalpy

2-PyO – 2 pyridinolate anion

3-PyO – 3 pyridinolate anion

4-PyO – 4 pyridinolate anion

E2Py – 2 ether-functionalized pyridinium

E3Py – 3 ether-functionalized pyridinium

Cl – cloride

Emim– 1-Ethyl-3-methyl-imidazolium

SCN – thiocyanate

C2mim – 1-ethyl-3-methylimidazolium

Tetz – tetrazolate

Et2NEmim – 1-(2-diethyl-aminoethyl)-3-methylimidazolium

P4442 – tri-n-butyl(ethyl)phosphonium

E3mim – 1-[2-[2-(2-methoxy)ethoxy]ethoxy]ethyl-3-methyl imidazolium

Bmim – 1-butyl-3-methylimidazolium

HB(im)3 – hydrotris(imiazolyl)borate

ILs – ionic liquids

gSO2/gIL – grams of SO2 adsorbed per gram of IL

Tabs – absorption temperature

ρIL – ionic liquid’s density

μIL – ionic liquid’s viscosity

CAPEX – capital expenditure

OPEX – operational expenditures

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