CO$_2$ absorption using LIs functionalized with amino acids

Absorción de CO$_2$ utilizando LIs funcionalizados con aminoácidos

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Abstract

A comparative study was conducted on the absorption of carbon dioxide (CO$_2$) using ionic liquids (ILs) based on imidazole derivatives as cations and amino acids as anions ([BMI][AA] and [OMI][AA]). The synthesized ILs were characterized using Fourier Transform Infrared Spectroscopy, and absorption studies were conducted using a pressurized batch system under stable pressure and temperature conditions. Amino acids demonstrated an enhancement in the absorption of this acidic gas when compared to monoethanolamine, which is the most commonly used commercial absorbent compound.

Absorption, Ionic Liquids, CO$_2$

Resumen

Se realizó un estudio comparativo de la absorción de dióxido de carbono (CO$_2$) utilizando líquidos iónicos (LIs) basados en derivados de imidazol como cationes y aminoácidos como aniones ([BMI][AA] y [OMI][AA]). Los LIs sintetizados fueron caracterizados por espectroscopia infrarroja por Transformada de Fourier y los estudios de absorción se realizaron mediante un sistema de lotes presurizado a condiciones de presión y temperatura estables. Los aminoácidos mostraron una mejora en la absorción de este gas acido comparados con la monoetanolamina que es el compuesto absorbente comercial mas utilizado.

Absorción, Líquidos iónicos, CO$_2$

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Introduction

The main source of energy worldwide has been from non-renewable compounds which, when used, generate some acid gases, CO_2 being the one with the highest concentration and contributing to a large extent to the greenhouse effect. The production of this gas is not only increased by the above but also by other factors such as the burning of coal in power plants or those plants based on diesel, which are very frequent and harmful emission sources [1,2]. The economic development of countries, especially those in the process of growth, is linked to an increase in the demand for energy; therefore, projections indicate that the need for fossil fuels will continue to increase given the trend of a growing world population that demands more and more energy per inhabitant, making it increasingly necessary to develop new efficient and low-cost environmental technologies [3].

Among the main options for reducing CO_2 emissions are improving the efficiency of conventional power plants, having greater control over energy consumption, using a higher proportion of renewable resources, developing nuclear energy sources, and capturing and storing CO_2, the latter being the most feasible and possible option to develop at present, which is why it has assumed an important role in the last century [4-6].

Nature has its own method for capturing CO_2 using trees, resulting in its biological fixation; however, experts are more curious about developing new non-biological processes for CO_2 capture in large point sources, so several viable options for achieving this involving different processes are presented [7] Post-combustion, oxy-combustion and pre-combustion are the three main technological routes that are currently considered in CO_2 capture [8,9]. One of the advantages of post-combustion capture is that because capture occurs after combustion and before release to the atmosphere, it can be implemented in facilities that are still in operation [10].

Absorption is considered within this capture stage, in this process chemical solvents implement an acid-base chemical reaction and among the best known are alkane amines (generally called amines). Primary amines are more reactive with CO_2 but are also more corrosive and sensitive to degradation, so secondary and tertiary amines are used to reduce these characteristics [11-13].

LIs commonly known as "green solvents" have been noted as promising candidates for CO_2 adsorption removal due to their high thermal stability, negligible vapour pressure and adjustable physicochemical properties [14-20]. LIs exhibit a strong affinity to CO_2 that are derived by varying the cations or anions by the addition of functional groups, especially those containing amino groups [21-24]. The anion plays a key role in determining the solubility of CO_2 in LIs and by physically adsorbing CO_2, less energy is needed for the regeneration of LIs [25]. Therefore, it has been deduced that anions in conventional ILs have a greater impact on CO_2 solubility than cations [26-29]. It has been proven that when functionalised LIs are dissolved they can greatly reduce viscosity [30]. The adsorption capacity of most existing functionalised LIs solutions with a single functional group has been similarly reported for compounds with approximately 0.5 mol CO_2/mol LIs [31-33].

Therefore, in order to develop new materials that can function as CO_2 absorbers, LIs were functionalised by the addition of amino acid groups to take advantage of the advantages of LIs by improving the characteristics with respect to conventional amine absorbing composites.

Methodology to be developed

Synthesis of the functionalised LIs

Amino acid functionalised LIs [OMI][AA] and [BMI][AA] were synthesised in the laboratory by replacement and neutralisation reactions using Lysine [L], Arginine [A], Histidine [H] and Glutamine [G] as amino acids. The synthesis of the precursor LIs [R-MI][Br] starts with the alkylation of 1-methylimidazolium (0.1 mol) with the corresponding alkyl halide (0.11 mol) in an inert atmosphere for 48 hours with heating and magnetic stirring.

The anion exchange reaction was performed based on the metathesis of halide salts with potassium hydroxide (KOH) in ethanol at low temperatures (below 10°C) in an equimolar fashion to form the precursor LI [R-MI][OH], which was subsequently filtered to remove excess salt in the sample and neutralised with the corresponding AA under constant stirring at room temperature. The ethanol was then distilled under vacuum to remove it from the [R-MI][AA] solutions before use. The proposed reaction mechanism can be seen in figure 1. The nomenclature used throughout this study is presented in table 1.
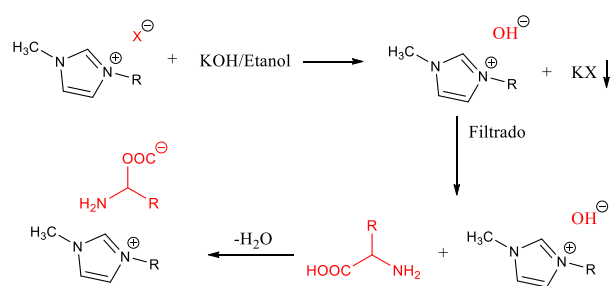
Figure 1 Reaction scheme for the synthesis of LIs functionalised with amino acids

No.	Cation	Anion	Identification
1	1- Butil, 3- Metilimidazol	Lysine	[BMI][L]
2		Arginine	[BMI][A]
3		Glutamine	[BMI][G]
4		Histidine	[BMI][H]
5	1-Metil, 3- Octilimidazol	Lysine	[OMI][L]
6		Arginine	[OMI][A]
7		Glutamine	[OMI][G]
8		Histidine	[OMI][H]

Table 1. Amino acid-functionalised ILs in this study

CO_2 absorption with LIs-Aas

The absorption tests were carried out at 303 K and a pressure of 3 atm. The apparatus consists of a stainless steel gas tank under constant stirring with a digital manometer connected, the diagram is shown in figure 1. The temperature is controlled by a thermostat in the heating bath ($\pm$ 0.1 K variation). The variation in gas pressure was approximately $\pm$0.001 bar over the pressure range used. Approximately 10 g of pure [R-MI][AA] or dissolved in water was measured into the reservoir and hermetically sealed. The first step consists of removing the air in the system by means of a vacuum pump to which it is connected for about 15 minutes. Subsequently the CO_2 is loaded into the tank and starts the absorption process with agitation. The system is considered as an equilibrium state once the pressure remains constant.

The concentration of absorbed CO_2 is determined using the ideal gas equation of state due to the pressure conditions handled. The variation in the absorption measurement results is approximately 0.02 mol CO_2/ mol LI.
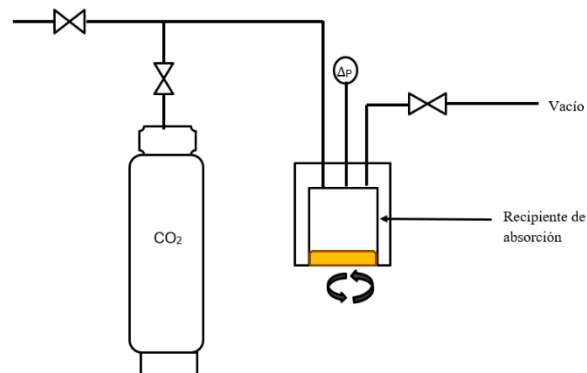
Figure 2 Schematic of the absorption equipment used

Figure 2 shows a very marked signal concerning the ionic liquid [BMIM+] centred at 3378 cm^{-1} which is attributed to the quaternary ammonium salt formed with the halogen (-N+R4X-), the formation of this band is due to the hydrogen bridge interaction formed between the acid hydrogen of the imidazole ring and the halogen ion, which weakens the C-H bond and decreases the location frequency resulting in an increase in signal intensity and a broadening of the band [34].

The next band is observed in all LIs functionalised with AAs, between 3500-3100 cm-1, this signal is attributed to the -NH groups and becomes broader because it overlaps with the -OH stretch in the amino acid structure [35]. The bands located at 3200-3000 cm^{-1} and 1166 cm-1 correspond to the vibrational modes of the imidazole ring, symmetric and asymmetric stretches of the C-C, N=C, C=C and N-C groups [36]. In LIs-AAs, at 1590 cm-1 a pronounced stretching is observed and at 1376 cm^{-1} a weaker one attributed to the C=O group of the ("CO_2") [37]. At 1660 cm^{-1} and between 1550-1300 cm-1 asymmetric bending bands of NH^{3+} are present [37] When comparing the spectrum of [BMIM+] with the [BMIM+][AA-] the differences are remarkable, which confirm the functionalisation of the precursor LI with the AAs, since the appearance of the characteristic band of the imidazole ring resonance near 3000 cm^{-1} is detected, which continues to be present even with the incorporation of these compounds.

It is also possible to observe that the compound with a linear anion and with a large amount of NH_2 groups such as lysine, has a good absorbance due to the fact that these groups are less attracted to other electronegative elements, which allows a greater dispersion of the anions and a better interaction between them. [38].

Results

Characterisation

The FTIR spectra of the synthesised ionic liquids are described below:
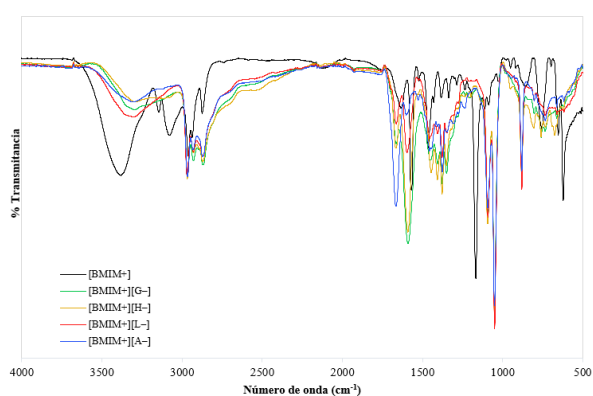
Figure 3 FTIR spectrum of the [BMIM+] [AA-] family of LIs

Figure 3 again shows a signal for the [OMIM+] LI centred at 3398 cm-1 that is attributed to the quaternary ammonium salt formed with the halogen (-N+R4X-) [35]. The bands observed between 3500-3100 cm^{-1} are signals attributed to the -NH groups with the -OH stretch in the AA structure of LIs functionalised with the same [35]. The bands located at 3200-3000 cm^{-1} and 1166 cm^{-1} correspond to the vibrational modes of the imidazole ring, symmetric and asymmetric stretching of the C-C, N=C, C=C and N-C groups [36]. On the other hand, in LIs-AAs, at 1570 cm-1 a pronounced stretching is observed and at 1350 cm-1 a weaker one, attributed to the C=O group of the CO_2. [37]. At 1674 cm-1 and between 1550-1300 cm-1 asymmetric NH^{3+} bending bands are present [37] Comparing the spectrum of [OMIM+] with the [OMIM+][AA-] again confirms the functionalisation of the precursor LI with each AA, as the appearance of the characteristic band of the imidazole ring resonance near 3000 cm^{-1} is detected in all structures.

It is possible to observe a shift and amplitude in the signals near 3200 cm^{-1}, which is attributed to the increase of the alkyl chain present in the cation, which presents a higher number of C-H bonds and therefore increases the absorbance [38].
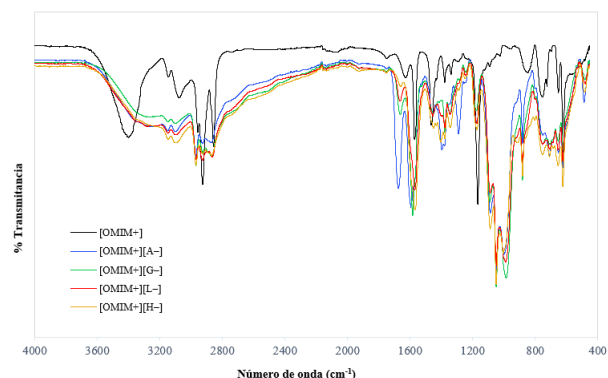
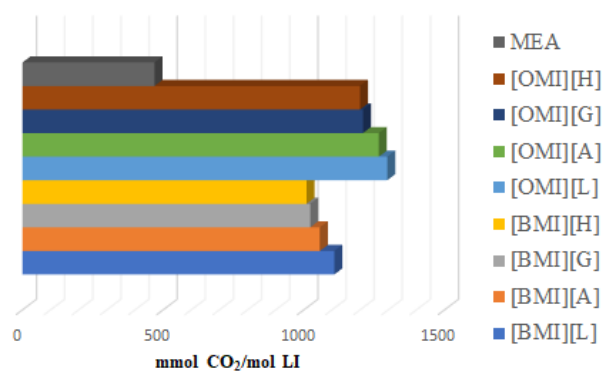**Figure 4** FTIR spectrum of the [OMIM+][AA-] family of LIs

CO_2 adsorption with ionic liquids

Solubility tests were performed on pure and dissolved LIs in water, in all cases, the absorption capacity of the LIs was higher in aqueous solution due to the fact that their viscosity is reduced when diluted. At the end of the absorption tests carried out with the LI-water solutions, especially those of [BMI][AA], a gas release was observed, which is attributed to the fact that not only chemisorption but also physisorption takes place during the absorption process. It should be noted that the solubility of the materials is more promising than with pure MEA. The absorption results can be seen in detail in table 2.

| No.	Identification	(mmol CO_2/ mol LI)	
		LI puro	LI in aqueous solution (35%)
1	[BMI][L]	821.47	1109.38
2	[BMI][A]	788.58	1057.80
3	[BMI][G]	761.94	1023.85
4	[BMI][H]	748.32	1010.70
5	[OMI][L]	719.25	1298.41
6	[OMI][A]	708.47	1268.43
7	[OMI][G]	902.62	1212.20
8	[OMI][H]	891.55	1200.45
9	MEA	-----	470.54

Table 2 Absorption results of the amino acid functionalised LIs

Figure 4 presents a comparison between the adsorption capacity of MEA and functionalised LIs-AA. It can be clearly seen how CO_2 solubility is favoured with these compounds, especially with lysine anions.

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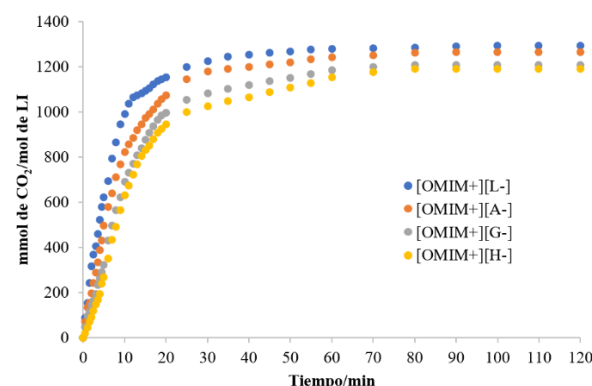
Figure 5 Absorption capacity of LIs-AA in 35% mass aqueous solution

The influence of the LIs cations on CO_2 solubility was studied with a similar anion between the butyl and octyl chains. Figure 5 shows that lysine greatly favours the absorption of CO_2, this is attributed to its solubility given its molecular structure which makes it favourable to the interaction with the gas.
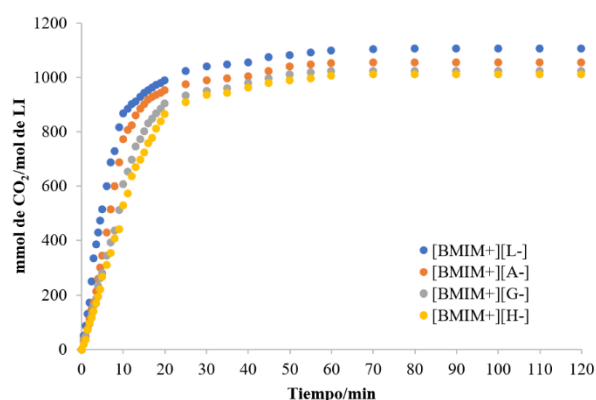

Figure 6 Variation in the absorption capacity of diluted [BMIM+][AA-]

Figure 6 shows that the absorption capacity is favoured in a similar way to that of lysine, despite the fact that the LIs with the longest alkyl chain are present in this one. A comparison between both figures shows that the absorption capacity is favoured in a similar way to the length of the chain, this can be attributed to the fact that the longer the alkyl chain, the greater the dispersion force of the cation, improving the interaction with CO_2 [39,40].

Figure 7 Variation of the absorption capacity of diluted [OMIM+][AA-]

The excellent properties of LIs and their ability to synthesise them highlight their importance in adsorption processes. In some cases it is possible to appreciate that the reaction kinetics of LIs-AA is higher compared to conventional amines. Figure 7 shows the comparative kinetics of lysine-functionalised ILs compared to MEA, and it is possible to appreciate that the reaction kinetics is faster with respect to these materials.
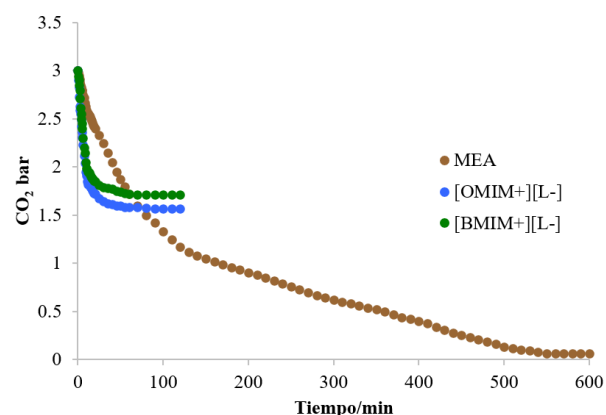
Figure 8 Absorption kinetics of lysine derivatives compared to MEA

Acknowledgement

Conclusions

The [OMI]-derived ILs obtained the most promising results compared to [BMI]-derived ILs, because the absorption is favoured with the length of the alkyl chain, obtaining the best results with the lysine amino acid since the amount of amino groups (especially primary) in the anions as well as the structural arrangement favours the absorption.

The solubility of CO_2 was favoured with dilution of these LIs-AA in water, which is mainly attributed to physisorption processes.

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