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# Bio-based ionic liquids and poly(ionic liquid)s for CO<sub>2</sub> capture.

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In the century of the battle against the climate crisis, many different strategies have been considered to tackle the excess of greenhouse gases. Meanwhile long-term strategies – e.g. switch to carbon neutral energy sources and industrial process – will have larger impact, short-term can be readily applied to existing pollution sources. In particular, the carbon dioxide capture from flue gases attracted researchers' attentions in the last decades and many different separation processes have been developed. Among them, the so-called amine scrubbing is the most mature and marketable process. This capture process is well-known and have been applied for gas sweetening. Despite being very effective, it is not efficient from the energetic and environmental points of view, since amine solutions are toxic corrosive and the energetic toll for CO<sub>2</sub> release is too high.

The aim of this PhD project is to develop innovative ionic liquids (ILs) and polymerized ionic liquids (PILs) for carbon capture utilizing bio-molecule – choline and amino acids – in order to produce materials and CO<sub>2</sub> capture processes that better align to the principles of Green Chemistry. Indeed, (i) both choline and AAs are widely available from renewable sources at low cost and being biomolecules should result in highly biocompatible materials; (ii) the synthetic procedures reduced the use of hazardous and toxic reagent as much as possible; (iii) the capture processes demonstrated to set more favorable condition for CO<sub>2</sub> release respect amine scrubbing process.

The synthesis of the choline-based ILs have been conducted using an innovative method, which employ [Cho][Cl] as starting reagent and avoid [Cho][OH] through the whole process. Choline chloride indeed is a cheaper and less hazardous starting reagent. The ionic metathesis procedure employed, despite producing ILs with possible higher content of impurities, is suitable for scale up and the final products exhibited no issue due to the lower purity. Tested AAs focused on small group with similar structure, in order to assess the effect of different anion functionalization in the CO<sub>2</sub> capture process. The [Cho][AA] ILs were tested for CO<sub>2</sub> capture and release. In order to overcome the problem related to the high viscosity of ILs, dimethylsulfoxide (DMSO) was chosen as solvent because of its high boiling point (189°C), polarity and aprotic behavior. To characterize the CO<sub>2</sub> capture and release ability of this ILs, different experimental setups and techniques were developed – gravimetric measurements, batch reactor simulations, nuclear magnetic resonance (NMR) characterization and *operando* infrared spectroscopy (IR) experiments. To complete the whole characterization of the ILs, thermal degradation process was investigated and biocompatibility test on zebra fish – *Danio Rerio* – embryos were conducted.

Polymeric version of the [Cho][AA] ILs were also developed. Starting from [2-(Methacryloyloxy)ethyl] trimethylammonium chloride – [MACHo][Cl] for short – different PILs with AA were developed and tested for CO<sub>2</sub> absorption in solid phase. [MACHo][Cl] monomer merges choline and methacrylate structures and is claimed to be biodegradable and scarcely toxic.<sup>140</sup> The research on [MACHo]-based PILs aimed to explored different AA anions as well as different polymeric structures. In addition to neat [MACHo][Cl] polymer, the copolymerization with styrene and crosslinking with N,N'-methylene bis(acrylamide) were prepared. Further ionic exchange procedures were tuned depending on the solubility property of the starting PIL. All synthetic procedures aimed to avoid toxic and hazardous chemicals. Obtained PILs were identified and were tested for CO<sub>2</sub> and water absorption and desorption at 30°C by means of microbalance. In addition, PILs thermal properties were evaluated using differential scanning calorimetry (DSC) and thermos gravimetric analysis (TGA) coupled with evolved gas analysis (EGA) to assess main thermal degradation products.