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### **ACCEPTED MANUSCRIPT**

## The performance of affordable and stable cellulose-based poly-ionic membranes in $CO_2/N_2$ and $CO_2/CH_4$ gas separation

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#### Abstract

The majority of commercial membrane units for large-scale natural gas sweetening are based on cellulose acetate (CA). However, the low selectivity and risk for and plasticisation affect adversely the performance of CA-based systems. Herein, we present a new class of CA-derived poly(ionic liquid) (PIL) as a thin film composite (TFC) membrane for CO<sub>2</sub> separations. CA is modified with pyrrolidinium cations through alkylation of butyl chloride, substituting the group in the polymer backbone, and further anion exchange to hvdroxvl bis(trifluoromethylsulfonyl)imide, P[CA][Tf<sub>2</sub>N]. The synthesised PIL material properties are extensively studied. The  $CO_2$  separation performances of the newly synthesised materials is evaluated by gravimetric gas sorption experiments, single gas time-lag experiments on thick membranes, and mixed-gas separation experiments on TFC membranes. The results are compared to the parent material (CA) as well as a reference PIL (poly(diallyldimethyl ammonium) bis(trifluoromethylsulfonyl)imide (P[DADMA][Tf<sub>2</sub>N])). The ideal CO<sub>2</sub>/N<sub>2</sub> sorption selectivity of P[CA][Tf<sub>2</sub>N] is constant up to 10 bar. The single gas transport measurements in P[CA][Tf<sub>2</sub>N] reveal improved ideal CO<sub>2</sub> selectivity for the CO<sub>2</sub>/N<sub>2</sub> gas pair and increased CO<sub>2</sub> permeability for the CO<sub>2</sub>/CH<sub>4</sub> gas pair compared to the reference PIL. Mixed-gas permeation tests demonstrated that P[CA][Tf<sub>2</sub>N]-based membranes with a 5 µm thick selective layer has a two-fold higher CO<sub>2</sub> flux compared to conventional CA. These results present CA modification into PILs as a successful approach promoting the higher permeate flows and improved process stability in a wide range of concentrations and pressures of CO<sub>2</sub>/N<sub>2</sub> and CO<sub>2</sub>/CH<sub>4</sub> gas mixtures.

#### Key words

Cellulose acetate, poly(ionic liquid), poly(diallyldimethyl ammonium) bis(trifluoromethylsulfonyl)imide (P[DADMA][Tf<sub>2</sub>N]), thin-film composite membrane, time-lag, ideal sorption selectivity, gas separation,  $CO_2$  capture, flue gas, biogas.

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