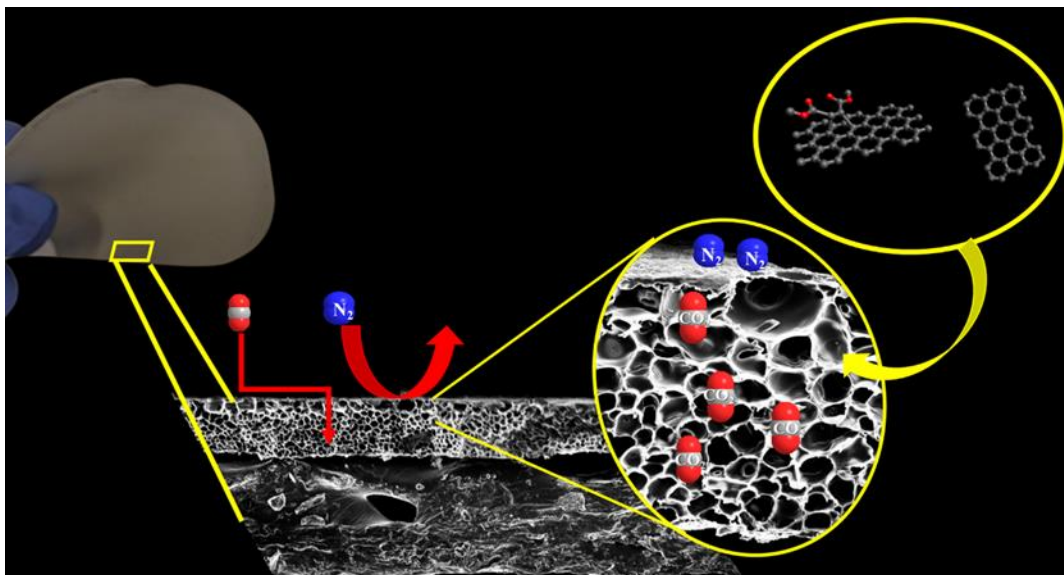


Thesis summary



The relentless progress of human society, coupled with globalization, escalating emissions, and the apathy of political and economic forces, has brought our world to the brink of catastrophe. As signs of climate change become increasingly evident, institutions are finally recognizing the urgency and pushing for solutions before our familiar world undergoes irreversible transformation. In light of this, my thesis project is dedicated to exploring innovative materials for efficient separation of CO₂ from other pollutants, aligning with the overarching concept of a Circular Economy.

To achieve this objective, I conducted an in-depth study on Mixed Matrix Membranes (MMMs), focusing on the functionalization of graphene-based fillers. This involved both adapting existing methods from the literature and developing novel functionalization approaches. Over the course of my research, I successfully developed two new inorganic fillers: one containing Diethyl maleate (GEM) and the other containing acetyl dicarboxylate (M). These fillers exhibited remarkable chemical and physical properties, such as ethanol dispersibility in case of GEM, this nanocomposite can be dispersed in low-boiling polar solvents such as ethanol at a concentration of 1.10 mg/ml. The dispersion exhibits excellent stability, as confirmed by zeta potential studies, which revealed a relatively stable dispersion with a mean zeta potential ranging from 30 mV to 28 mV after one year. When a radical initiator is utilized, the nanocomposite exhibits enhanced stability in ethanol, reaching a mean zeta potential of 40 mV and maintaining it over an extended period.

The second graphene-based filler involves a novel Diels-Alder cycloaddition reaction occurring on the surface of graphene. The obtained data confirmed an increase in defects on the carbonaceous material's surface through Raman spectroscopy. X-ray diffraction analysis revealed

a change in the crystalline structure before and after the reaction. This reaction can be completed in a short time using microwave irradiation. Remarkably, this material exhibits interesting physical characteristics, including fluorescence and conductivity, thereby opening doors to new applications in the field of energy.

Simultaneously, I investigated the optimal concentration of exfoliated graphene in glassy polymer, specifically PSU, and an ionic liquid (BMIM Succ.) for enhanced CO₂ separation.

Analytical instrument data indicates that the presence of 1% to 2% graphene significantly improves membrane permeability, with an increase of 184% and 207% compared to pure PSU membranes for 1% graphene and 2% EG, respectively, at a CO₂ pressure of 0.4 bar (40% relative pressure).

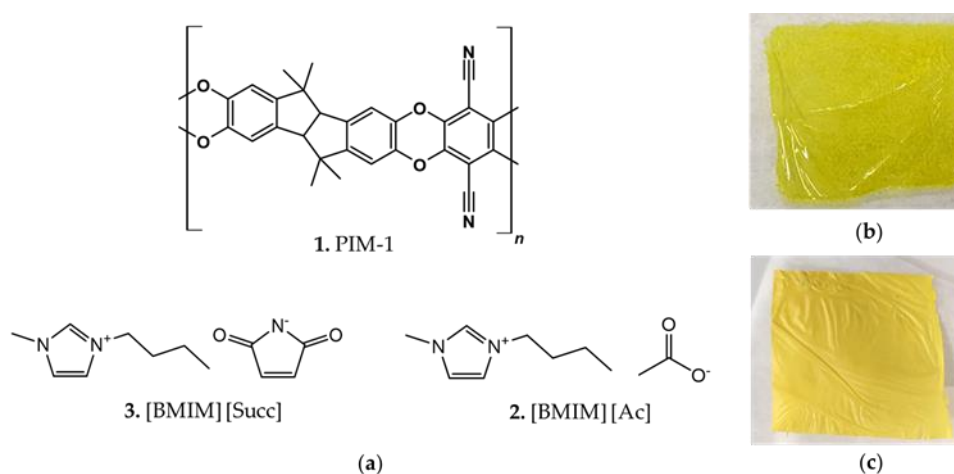


Figure 1 PIM- Membranes

The presence of the ionic liquid BMIM Succimidate decreases the permeability of graphene-containing membranes. Additionally, I explored the application of the highly innovative polymer PIM-1, which demonstrated exceptional CO₂ permeability. In the case of PIM-1-based membranes, the high solubility of CO₂ in the polymer posed challenges in conducting realistic comparisons using the available analytical tools. Incorporating graphene functionalized with TCNE significantly reduces the solubility of CO₂, enabling evaluation and comparison of mixed matrix membranes prepared with PIM-1. A screening study was done with PIM-1 and BMIM Succ. and BMIM Ac. ionic liquids, we observed how BMIM Succ has a negative influence on CO₂ diffusion and solubility for PIM-1, and how this decreases when the concentration of the ionic liquid is very high. BMIM Ac on the contrary proved to be better as an ionic liquid, increasing the solubility to CO₂ for these membranes, in both cases the optimal concentration that best affects the solubility to the gas is 25%, above 25% a lowering of membrane performance is observed.

Furthermore, I delved into the structural aspects of the membranes themselves, devising a multilayer configuration. This design featured a central body of PSU covered with either graphene

materials or PIM-1, thereby creating distinct pore types. These specialized pores facilitated the concentration of CO₂ on the membrane surface, enabling efficient separation, intriguing data was obtained. Membranes consisting of a PSU-based core body covered with a thin layer of either PIM-1 or PIM-1 mixed with GTCNE exhibit improved performance compared to PSU alone. The presence of the thin layer enhances CO₂ absorption when PIM-1 is present on the surface, while the inclusion of GTCNE in the PIM matrix facilitates preferential CO₂ diffusion toward the middle part of the PSU