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# Graphene Oxide Doped Ionic Liquid Ultrathin Composite Membranes for Efficient $CO_2$ Capture

Madhavan Karunakaran*, Luis Francisco Villalobos, Mahendra Kumar, Rahul Shevate, Faheem Hassan Akhtar, and Klaus-Viktor Peinemann

Advanced Membranes and Porous Materials Center, 4700 King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia

*Corresponding Author (madhavan.karunakaran@kaust.edu.sa)

## S1. Membrane Characterizations

Fourier transform infrared (FT-IR) spectra of the GO and GO/ILs were recorded on a Nicolet iS10 spectrometer (Thermo Scientific Co.) in the frequency range of 4000 to 600 $cm^{-1}$ with a resolution of 4 $cm^{-1}$ at 16 scans. All scans were recorded at ambient temperature. Raman spectra were recorded on a Horiba LabRam Raman spectrometer equipped with a 50×Objective lens (Olympus, NA 0.75) at a wavelength of 532 nm. The pristine GO and GO complexed ionic liquid powders were directly spread over the glass substrate, and then the laser beam was focused on the powder surface to record Raman spectra. Atomic Force Microscopy (AFM) analysis was performed using Agilent microscope (Model 5400) in a tapping mode using a silicon nitride tip with a resonance frequency in the range from 76 to 263 kHz. The GO and GO/IL solutions were deposited on the silicon wafer and the thickness of single layer GO sheets were measured. In the case of membrane samples, small pieces of membranes were placed onto the glass plate and images were recorded direclty.

Scanning electron microscopy (SEM) images of the dense GO based membranes were recorded on a Field emission scanning electron microscope (FESEM, FEI Quanta 200 series). Imaging was carried out at an acelerating potential of 5 kV with a working distance of 10 mm. The dried membrane samples were mounted on the aluminum stubs with the help of aluminum tape and coated with gold before recording their SEM images. For cross-section morphology analysis, membranes were fractured in liquid nitrogen and their SEM images were recorded. Ultrathin layers of GO on the surface of the membranes were detected using TEM. Both the IL and IL/GO membranes were embedded in a low-viscosity epoxy resin (Agar R1165) and cured for 24 h at 60°C. About 100 nm ultrathin sections of membrane were prepared using an ultramicrotome (Leica EM UC6) and then placed on180-mesh copper grids. Images were obtained using a Titan FEI transmission electron microscope operating at 300 kV. Wide angle X-ray diffraction (WAXD) measurements were conducted on a Bruker D8 Advance X-ray diffractometer with Cu-K radiation ($\lambda$ = 1.54 Å) operated at 40 kV and 40 mA. The 2-theta angular region between $3^o$ and $50^o$ was explored at a scan rate of $5^o$/min. Nitrogen adsorption isotherms for GO and complexed GO-IL samples were recorded on a Micromeritics ASAP 2020 physisorption analyzer at 77 K. The specific surface area of the sample is calculated by Brunauer-Emmett-Teller (BET) method, using the $N_2$ adsorption data in a relative pressure ($P/P_o$) range of 0.05 – 0.22.

**Table S1:** Gas permeation of PAN, PAN/[EMIM][Ac] and PAN/[EMIM][$BF_4$].

| | $CO_2$ (GPU) | $N_2$ (GPU) | $CO_2/N_2$ |
|---|---|---|---|
| PAN | 26000 | 32000 | 0.8 |
| PAN/[EMIM][Ac] | 75 | 3.75 | 20 |
| PAN/[EMIM][$BF_4$] | 150 | 6.82 | 22 |

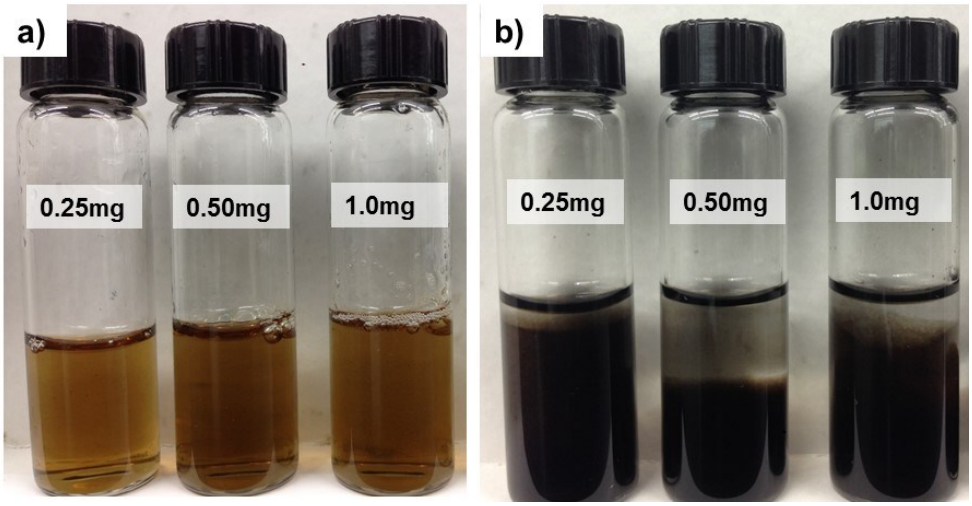


**Figure S1.** GO dispersed in 20-wt% [EMIM][Ac] in DI water a) 10 min, b) after 24 h.

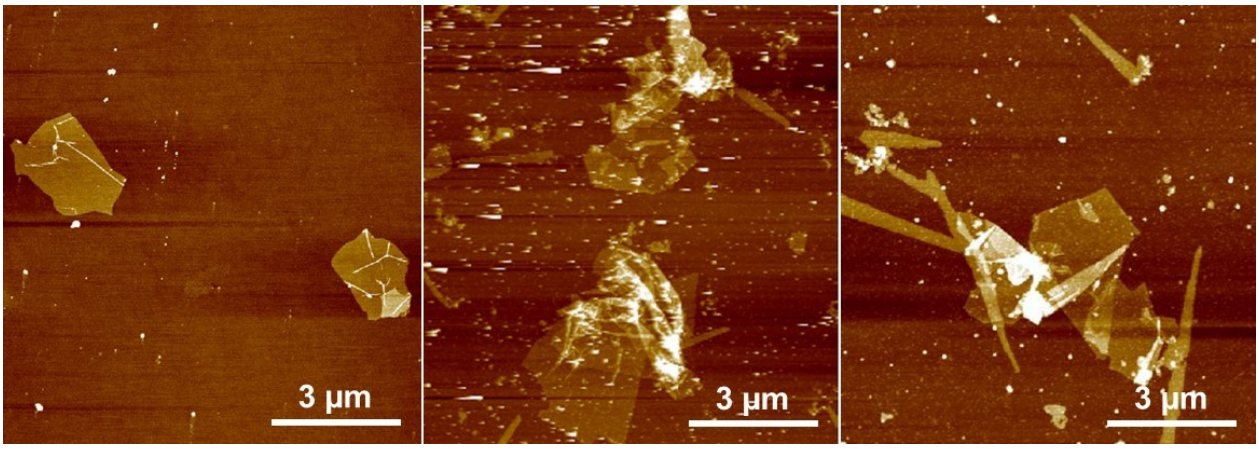


**Figure S2.** AFM images of : a) GO, b) [EMIM][Ac]/GO and c) [EMIM][$BF_4$]/GO.

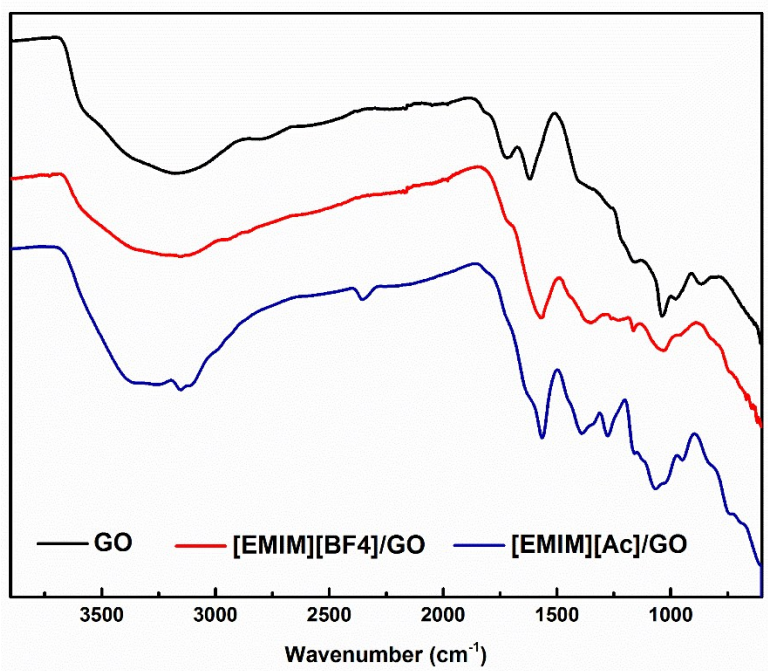


**Figure S3.** FTIR spectra of: a) GO, b) [EMIM][Ac]/GO and c) [EMIM][$BF_4$]/GO.

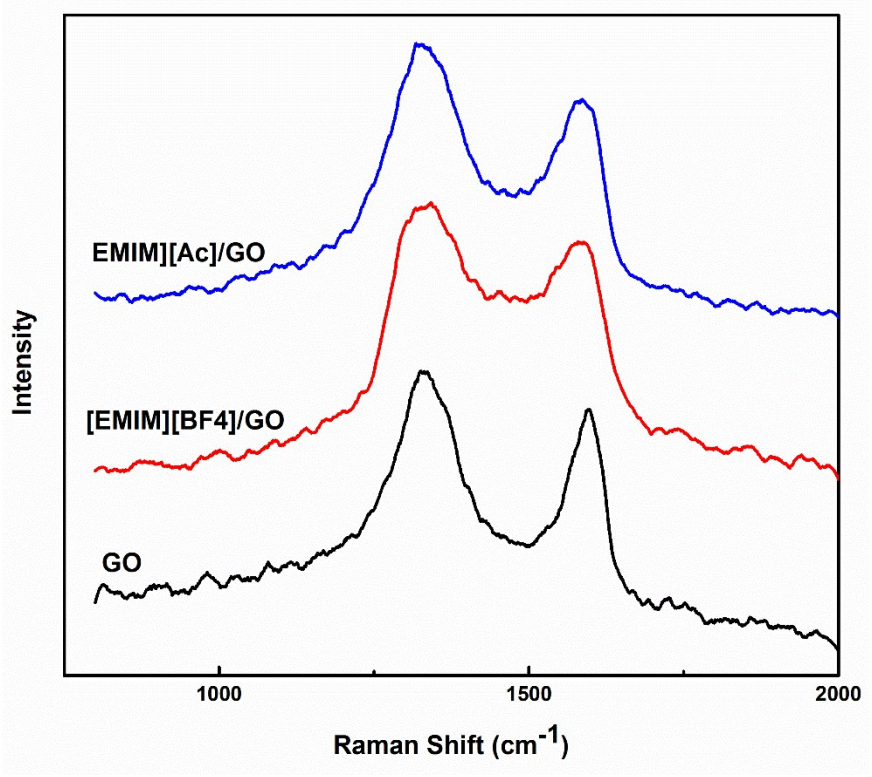


**Figure S4.** Raman spectra of: a) GO, b) [EMIM][Ac]/GO and c) [EMIM][$BF_4$]/GO.

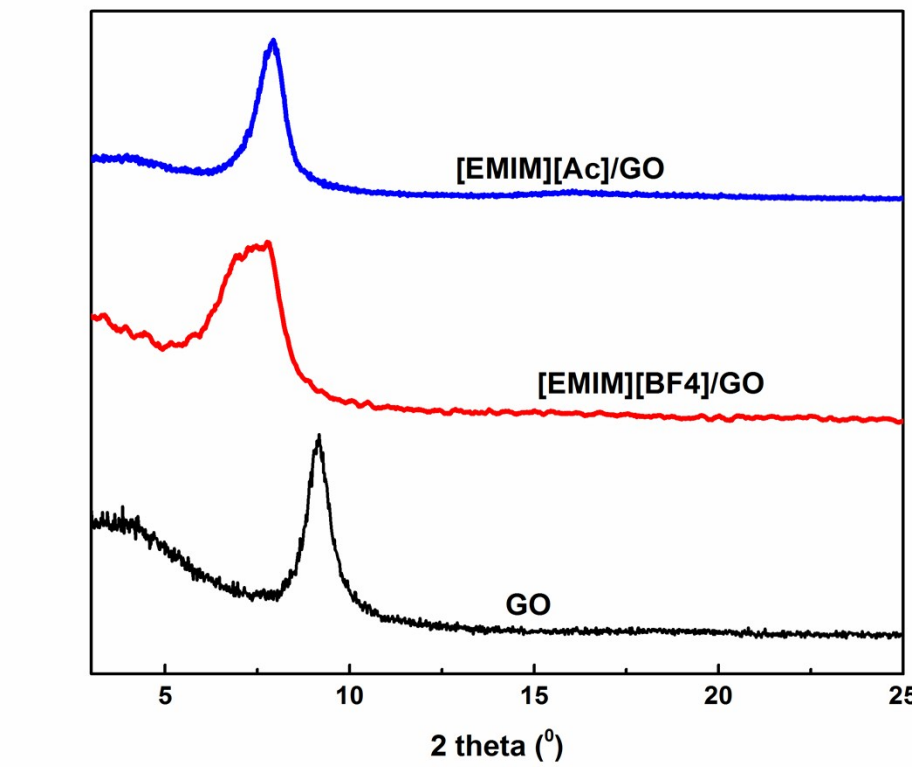


**Figure S5.** The WXRD patterns of: a) GO, b) [EMIM][Ac]/GO and c) [EMIM][$BF_4$]/GO.

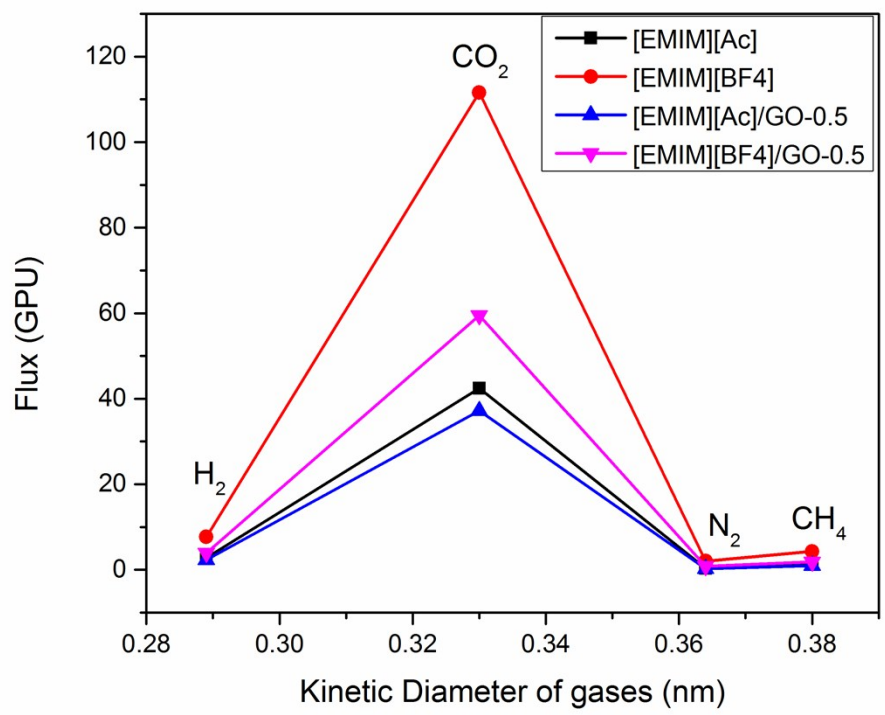


**Figure S6.** Gas permeability of IL/GO membranes as a function of kinetic diameters of gases.

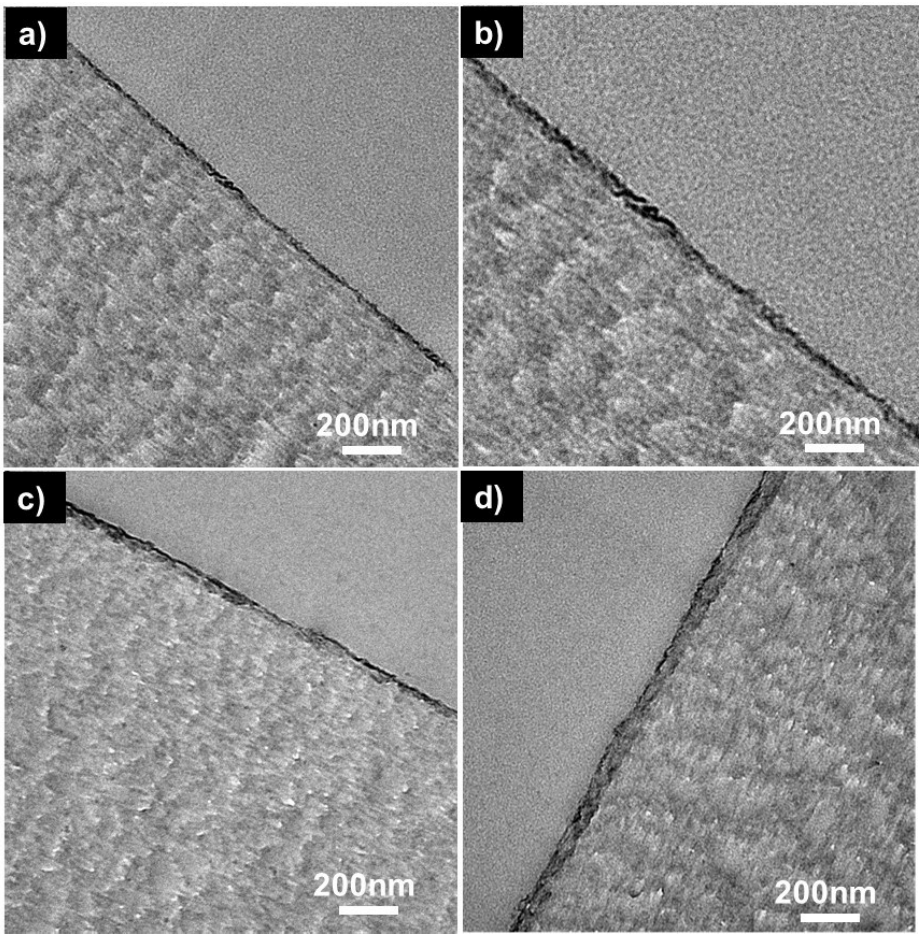


**Figure S7.** TEM images of: a) [EMIM][Ac]/GO-0.25; b) [EMIM][Ac]/GO-0.5, c) [EMIM][Ac]/GO-1 and d) [EMIM][Ac]/GO-2.