Natural gas sweetening using high pressure resistant CO2 selective PVAm/PVA membranes

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The most widely used technology for natural gas sweetening nowadays is amine absorption; a technology using large columns for absorption and desorption, and with a fairly high capital costs and energy consumption. An alternative to this could be membrane technology for the same application where no chemicals or solvents are needed, and with modular solutions which will have a much smaller footprint. There are already membrane processes installed for this separation many places in the world, but the main problems that limit the use of membranes are their relatively poor performance at high pressures. Today commercial membranes for this separation are in general conventional polymeric membranes based on solution-diffusion mechanism, which exhibits relatively low selectivity and flux (e.g. CO_2/CH_4 selectivity of only about 12-15 with CO_2 permeance up to 0.20 $m^3(STP)/m^2 \cdot h \cdot bar$). The required separation performance that could make membrane competitive to amine adsorption process will need a CO_2/CH_4 selectivity of ~40 with CO_2 permeance of at least 0.3 $m^3(STP)/m^2 \cdot h \cdot bar$ according to Baker [1]. In the past 10 years, our group has developed and patented a series of facilitated transport membranes with amino groups as fixed-site-carrier (FSC) to enhance CO_2 transport, including a polyvinylamine/polyvinylalcohol (PVAm/PVA) blend membrane. In this membrane, the reversible reactions of CO_2 with amino carriers in PVAm facilitate the CO_2 transport, resulting in both high CO_2 permeance and CO_2/CH_4 selectivity, while the entanglement of PVAm with a mechanically robust polymer PVA enhances polymeric network. Currently a selectivity of CO_2/CH_4 up to 50 and CO_2 permeance up to 0.45 $m^3(STP)/m^2.h.bar$ have been documented.

The current work focuses on the upscaling of the membrane for pilot scale permeation test and the optimization of the PVAm/PVA blend FSC membrane to improve its high pressure resistance for high pressure separations. In this study commercially available PVAm with high molecular weight (e.g, PVAm MW 340,000) was used in the PVAm/PVA blend membrane. The mechanical properties and separation performances of the PVAm/PVA blend membranes are being tested. Membranes with upscaled size (300×300mm) were prepared. CNTs were introduced to the blend membrane to further reinforce the FSC membrane. The functions of the

CNTs in this membrane include the reinforcement of the membrane mechanical properties and the improvement of the membrane swelling capacity (and hence CO_2 separation efficiency) at high pressures due to the nano spacer effect of the CNTs. PVAm/PVA blend membranes with optimized selective layer thicknesses at pressures 5bar, 20bar, 40bar, 60bar and 80bar were investigated in a pilot scale high pressure permeation rig. The pilot scale module and operation parameters were optimized, including feed gas flowrate (20-200 ml/min) and sweep gas flowrate (0-20ml/min), feed pressure (2.0-90bar), sweep gas pressure (1.0-7.0bar), relative humidity (30-100%), separation temperature (RT-60^{o}C).

References

[1] R.W. Baker, Future Directions of Membrane Gas Separation Technology, *Ind. Eng. Chem. Res.*, **41** (2002), p. 1393-1411.