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The study of the selectivity of methane over carbon dioxide and inert gases using composite inorganic membranes

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OUTLINE

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GREENHOUSE GASES

- Gaseous hydrocarbons that are prevalent under increased pressure include solution gases in oil reservoirs.
- These gases are usually conserved but in some cases they are flared (Fig. 1).
- The main gas in Natural gas is methane.
- Flaring brings about the emissions of carbon dioxide which is one of the main gases responsible for global warming.

- Greenhouse gases that include carbon dioxide, methane and other non-methane hydrocarbons are responsible for global warming.
- The distribution of emissions worldwide (Fig. 2) shows USA to be the main contributor.

Fig. 1: Natural gas flaring

Fig. 2: Cumulative CO2 emissions, 1751 – 2012 (courtesy Hansen, J., et al. (2013))
Different sectors that contribute to the emission of greenhouse gases in 2010 are presented in Fig. 3.

The highest contributors are industry, Agriculture, Forestry and Other Land Use (AFOLU), electricity and heat production.

There are regulations in place in many countries for the emissions of these gases into the atmosphere.

Focussing on natural gas as it is an important fuel gas that can be used for power generation as well as a raw material in petrochemical industries.

There are various measure in place for the treatment of natural gas and its transportation to pipelines as well storage facilities.

Membrane Technology has only about 5% of the market for processing natural gas in the United States.

This percentage is expected to rise as better carbon dioxide selective membranes are developed.
The choice of membrane material is dependent on the application of the membrane. The basic support used in this work is alumina support that is macro-porous with a large pore size of 6000 nm. It has an intermediary layer consisting of titanium oxide. The outer layer can be modified with various components such as metals, silica, zeolites or it can be mixed matrix and contain a mixture of polymer incorporated in the zeolitic pores.
There are different procedures that can be used for the modification of a membrane support. The dip-coating process (Fig. 6) was used in this work and it involves the preparation of a nano-particulate suspension and immersing the membrane into the suspension for a period of time.

Chemical vapour deposition (Fig. 7) is used for high quality modification of membranes. The nano-particle thin film is deposited as a vapour on the membrane support. This is a future methodology to be used in my research work.
GAS SEPARATIONS

Gas flow through a membrane can be a pressure driven process. The feed gases are fed into the membrane, the gases that don’t go through the membrane are called the retentate and the gases that go through the membrane are referred to as the permeate (Fig. 8).

The flux (J) of a gas is the amount passing per unit time and per unit of membrane surface area. Flux has an impact on the economic feasibility on the use of membrane technology since high flux for a low membrane surface area reduces the capital costs of operation.

Selectivity is the partitioning of a component into permeate and feed. This gives an idea of how a particular component moves through the membrane in relation to another component. Selectivity strongly determines the technical feasibility of a membrane separation process.
The type of membrane material, pore size and nature of the components to be separated determines the type of transport mechanism through the membrane. A membrane can separate components using a combination of flow mechanisms.

- Poiseuille’s flow separate gases based on their viscosities.
- Knudsen flow is dependent to the molecular mass of the gas components.
- Surface flow involves the adsorption of a component on the surface of the membrane.
- Different gases have different condensation pressures, hence capillary condensation uses this difference to separate gases.
- Various gases have different kinetic diameters hence molecular sieving is possible when the pore size of the membrane support is in the nano scale.

Fig. 9: Mechanism of gas transport through a membrane
AIM AND OBJECTIVES

- Prepare and characterise a silica and zeolite membrane on an alumina support
- Determine the flux of methane and CO$_2$, through the prepared membranes
- Determine the separation factors of the gases through the membrane in relation to methane
EXPERIMENTAL RIG

- Zeolite membrane was fabricated using the dip-coating method (Fig. 10)
- Characterisation was done by carrying out nitrogen adsorption-desorption measurements using the Quantachrome gas analyser (Fig. 12)
- Permeation tests were carried out using a membrane reactor (Fig. 11).
The molar flux of both gases increased linearly with increase in pressure. This is in agreement with literature. At pressures above $6 \times 10^5$ Pa the slope remained constant and hence this could indicate the contribution due to viscous flow (previous work) being prevalent for the silica membrane at a higher pressure. The molar fluxes are linear functions of the pressure drop across the zeolite membrane.
RESULTS AND DISCUSSION

- CH$_4$/CO$_2$ selectivity decreases with increasing operating temperature in the silica membrane.
- CH$_4$/CO$_2$ obtained from the zeolite membrane is much higher than that of the silica membrane. The selectivity also increases with increase in temperature.
- At a higher temperature of about 500K the selectivity of the membrane to CH$_4$ over CO$_2$ falls to its room temperature value in the case of zeolite membrane.

Fig. 15: Effect of temperature on CH$_4$/CO$_2$ selectivity of silica membrane

Fig. 16: Effect of temperature on CH$_4$/CO$_2$ selectivity of zeolite membrane
The hysteresis isotherms in Fig. 17 implies that both membranes are mesoporous and could undergo capillary condensation during hysteresis.

Table 1 shows the desorption summary of both membrane membrane.

- Table 1: Desorption summary of (a) silica membrane and (b) zeolite membrane

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CONCLUSION

The results obtained have shown that it is possible to use a mesoporous membrane to selectively remove carbon dioxide from methane to produce pipeline quality natural gas. The zeolite membrane has given better separation factor than the silica membrane.

FUTURE WORK

There is a need for further study of the transport mechanism of methane through the zeolite membrane since this is essential for the separation of other hydrocarbons that could be present as impurities.