MUTUAL INTERACTION OF COMPONENTS DURING MEMBRANE SEPARATION OF GASES

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INTRODUCTION

Membrane operations offer a viable solution for gas improving and separation. Specifically, they offer an effective solution for downstream syngas treatment after biomass gasification before transforming it into valuable compounds. As gasification is one of the perspective solutions for waste-to-fuels and waste-to-chemicals, it is very imporntat to describe the membrane separation process so it can be involved in the technologies.

The product of biomass gasifications consits of four main components – H_2, CO_2, CO, CH_4, along with minors (H_2S, COS, halogens) and possibly N_2 (when using air as gasification agent). A typical composition of woody-biomass air gasification is approx. 5 % H_2, 15 % CO, 17 % CO_2, 3 % CH_4 and 60 % N_2.

This study presents initial experiments with CH_4-N_2-CO_2 mixture to inspect the interaction of the components during multicomponent gas membrane separation.

EXPERIMENTAL SET UP AND METHODS

GAS MIXTURES

For this study, three-component mixture was chosen for the experiments, consisting of methane, nitrogen and carbon dioxide. As CO_2 is the most permeable, its concentration was fixed. The other two components N_2 and CH_4 were chosen as they have similar characteristics when processed in pure form..

Table 1 – Model mixtures used for the study.

#	$c_F(CO_2)$ [%mol]	$c_F(CH_4)$ [%mol]	$c_F(N_2)$ [%mol]
1	40	30	30
2	40	0	60
3	40	60	0

EXPERIMENTAL SETUP

The experiments were performed on laboratory membrane unit Ralex GSU-LAB-200 with interchangable membrane modules. For this study, a hollow fibre polyimide modul of following parameters was used. The unit allows to measure concentrations in each flow, total mass flow in each branch (feed, permeate and retentate) and temperatures. The scheme of the measuring systém is shown in the following figure. In table 2, parameters of the used polyimide membrane module are shown.

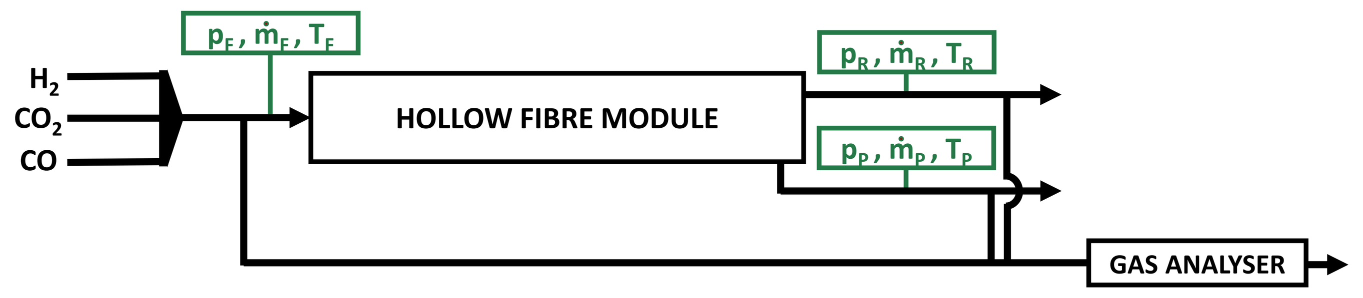

Figure 1 – Scheme of the measuring system. The measured values are mass flow (m_i), pressures (p_i) and temperatures (T_i) in each flow (F = feed, P = permeate, R = retentate). The gas analyser measures molar concentrations of each component (CO, CO_2 and H_2).

Table 2 – Parameters of the used polyimide hollow fibre module

Fibre length (mm)	Fibre diameter (mm)	Number of Fib. (-)	Total area (m²)
290	0,188	3000	0,820

USED EQUATIONS AND DEFINITIONS

Pressure difference Δp is defined as $\Delta p = p_R - p_P$ (bar)
where p_R and p_P are pressures in retentate (R) and permeate (P).

Permeability P_i is defined as $P_i = \frac{\dot{n}_{i,P} * L}{A * \Delta p}$ (Barrer)
where n_i is molar flows of component *i* in permeate, L is length of the module, A its area and Δp is press. diff.

Stagecut θ is defined as $\Theta = \frac{\dot{n}_{tot,P}}{\dot{n}_{tot,F}}$ (Barrer)
where $n_{tot,P}$ and $n_{tot,R}$ are total molar flows of all components in permeate and retentate flow respectively.

Component recovery R_i is defined as $R_i = \frac{\dot{n}_{i,P}}{\dot{n}_{i,P} + \dot{n}_{i,R}}$ (-)
where $n_{i,P}$ and $n_{i,R}$ are molar flows of component *i* in permeate and retentate flow.

Table 3 – Measured permeabilities of pure components for further evaluation

Component	Carbon dioxide CO_2	Methane CH_4	Carbon monoxide CO
Permeability (Barrer)	343 ± 11	13.4 ± 1.1	13.2 ± 1.0

Permeabilities were measured at different permeate pressures (1, 2 and 4 bar) and at different pressure differences (ranging 1-6 bar with 1 bar increments for N_2 and CH_4 and 0-1 bar with 0,1 increments for CO_2 As can be seen, nitrogen N_2 and methane CH_4 have very close permeabilities and thus offer interesting comparison when processed in mixtures.

RESULTS

CO_2 PERMEABILITY

If only permeabilities of components are taken into account, the CO_2 permeability in all three cases should be very close – 40 % of the mixture is always CO_2 with pure component permeability 343 Barrer and 60 % of the mixture is made up by components with pure components permeability approx. 13 Barrer. See following figures for indication of deviations.

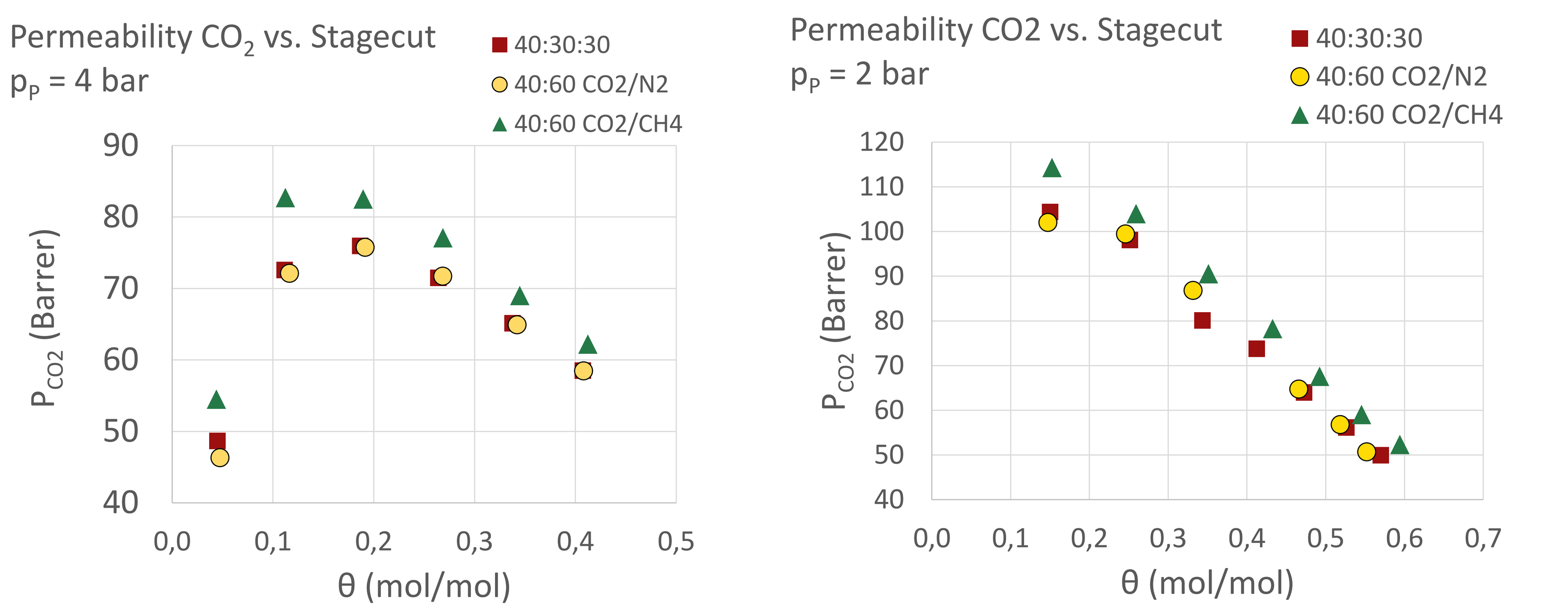

Figure 2 – Permeability of CO_2 in different mixtures on molar stagecut. As can be seen, the characteristics deviate for CO_2-CH_4 mixture.

CO_2 RECOVERY

Similarly as in previous case, the recovery of CO_2 differs for mixtures with N_2 and without N_2. The mixture of CO_2 and CH_4 shows deviation from the trend of mixtures with N_2. However, with increasing permeate pressure the differences decrease.

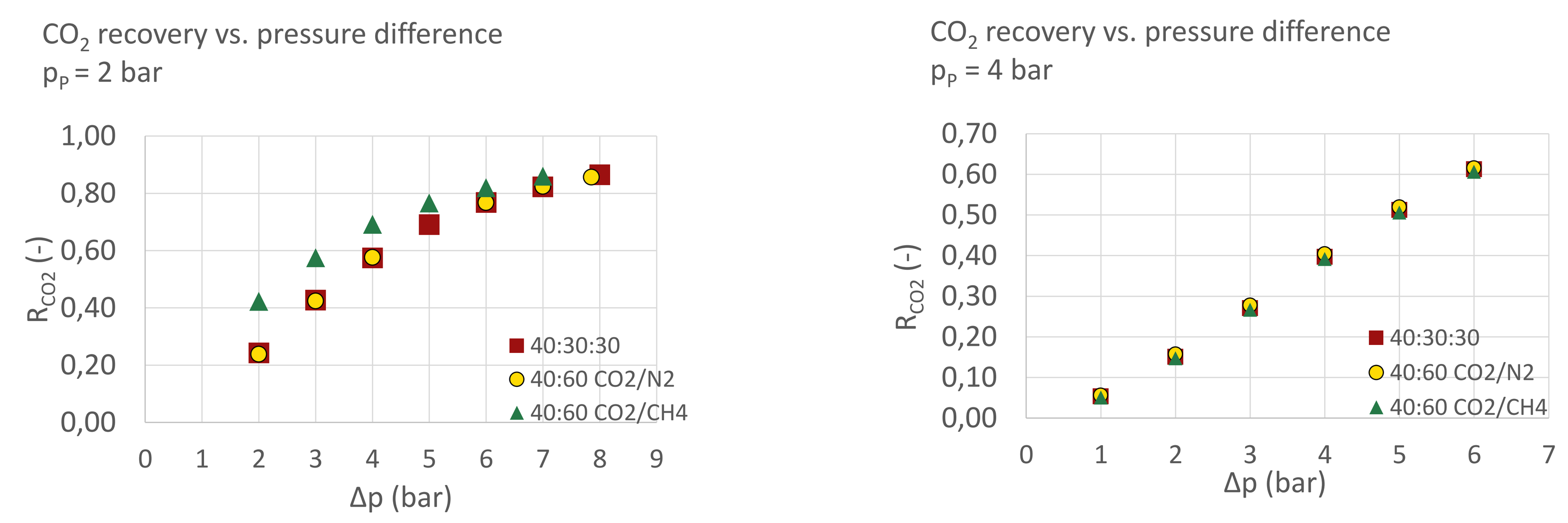

Figure 3 – CO_2 recovery for all three mixtures – differences lower for higher permeate pressure.

CO_2 FLOW THROUGH THE MEMBRANE

Another criteria is the amount of CO_2 permeating through the module per second. As can be seen in the following figures, the trend with changing permeate pressure is opposite for mixture with N_2 only. With increasin permeate pressure, the flow of CO_2 increases whereas in mixtures with CH_4 (binary and tertiary), the increasing pressure drop lowers the curves for CO_2 permeating flow. (Lines in figures are just to highlight the trend and have no physical meaning.)

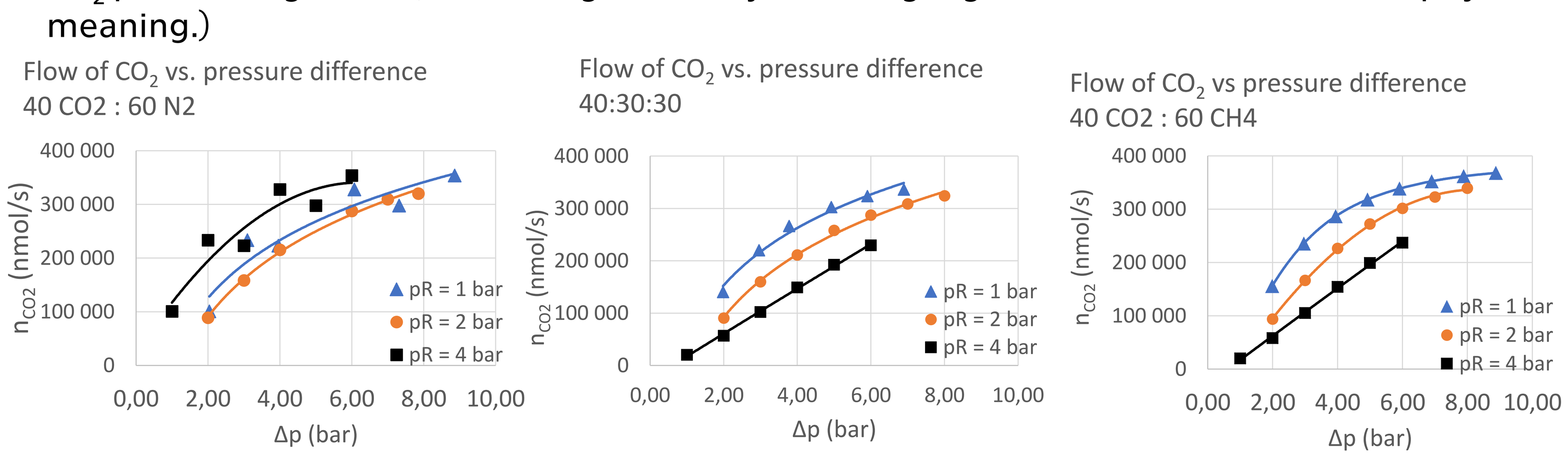

Figure 4 – Total CO_2 flow through the membrane in all three cases. For mixture with N_2 – oposing trend when increasing permeate pressure p_P.

CONCLUSIONS AND DISCUSSION

As seen in the figures above, the composition of permeate and the process of multicomponent gas separation using membrane modules is affected by the composition of the feed gas despite its components having similar permeabilities.

From several points of view (permeability and recovery of CO_2), the characteristics were similar for mixtures with all three components /40:30:30) and the mixture with nitrogen only /40:60 N_2). However, for total flow of the CO_2 through the membrane, an anomaly occured when increasing the permeate pressure and the mixture with N_2only showed opposite trend from the other two.

In conclusion:

- the components affect each other during membrane seprataion
- pressure conditions affect the level of interaction (different results for different p_P)
- more parameters than Permeability of pure components affect the process of separation

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