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Cesari Laetitia, Christophe Castel, Eric Favre

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Membrane contactors for intensified physical solvent absorption biogas purification processes: a parametric study.

Introduction

Context :

Membrane contactors are one of the most promising technology for the intensification of gas-liquid absorption processes with the following advantages [1,2] :

- Large specific area of membrane module
- High expected intensification factors
- Independent choice of gas and liquid flowrates

Adressed questions :

- What is the maximal intensification factor of membrane contactors for physical absorption processes compared to packed columns ?
- What are the best flow configurations (i.e. liquid flowing inside or outside the fibers), module design (packing factor), membrane performances (mass transfer coefficient) ?
- What is the interplay between intensification and specific energy requirement ?
- How do solvent viscosity and gas solubility impact the intensification performances ?

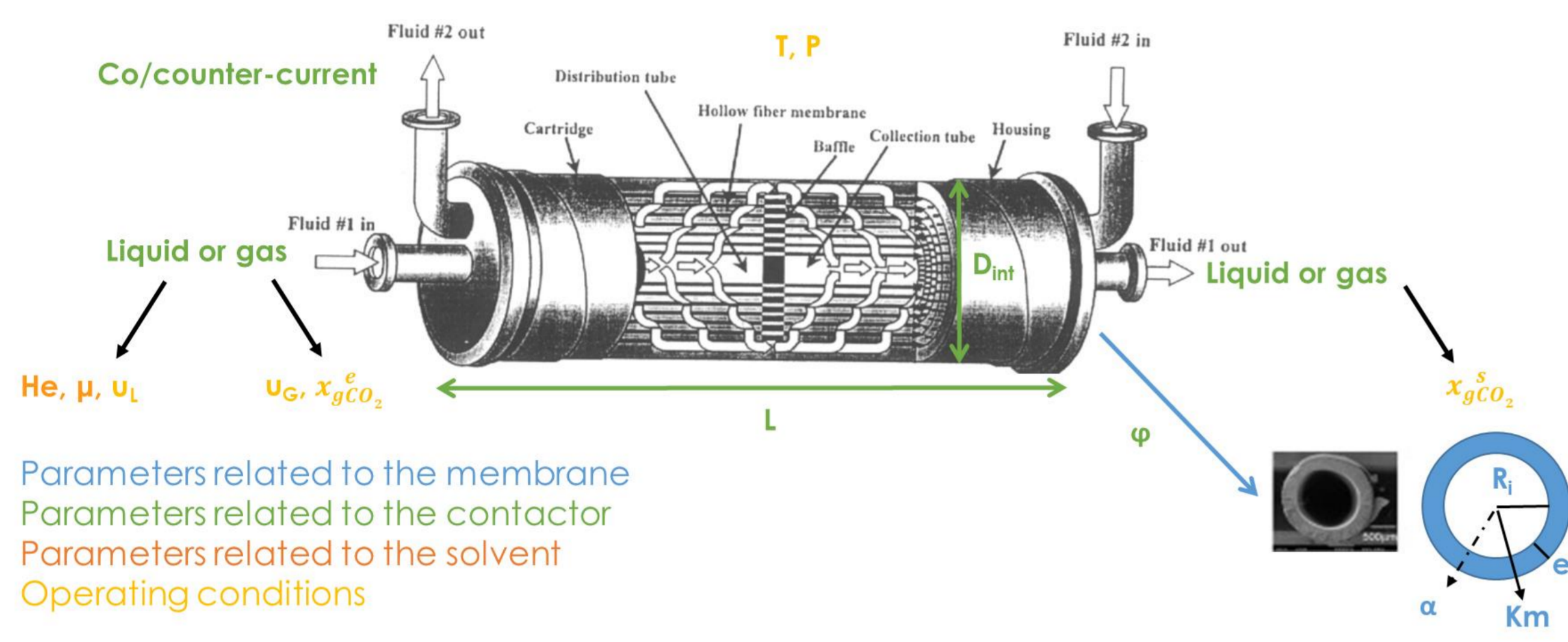


Figure 1 : membrane contactor and its parameters

Materials and Method

Equations :

- Henry coefficient : $He_i = \frac{C_i^{gas}}{C_i^{liquid}}$
- Absorbed CO₂ molar flow : $N_{CO_2} = Q_{G,0} \cdot y_{CO_2,0} - Q_{GL} \cdot y_{CO_2,L}$
- Transfer coefficient : Graetz equations
Resistance in series approach
- Differential equations : $\frac{d(Q_G \cdot y_i)}{dz} = K_G (C_i^{gas} - He_i \cdot C_i^{liquid}) a_m \Omega$
 $Q_L \frac{dC_i^l}{dz} = K_G (C_i^{gas} - He_i \cdot C_i^{liquid}) a_m \Omega$

Parameters :

	Symbol	Parameter	Value	Unit
Fixed parameters	$Q_{gas,0}$	Volume flow of gas at the entrance	0.0139	Nm ³ .s ⁻¹
	T	Temperature	298	K
	α	Selectivity	1	/
	D_{int}	Internal diameter of the module	0,17	m
	r_{int}	Hollow fiber internal radius	450	μ m
	e	Membrane thickness	50	μ m
	μ_g	Gas viscosity	10 ⁻⁵	Pa.s ⁻¹
	$D_{CO_2}^g$	Diffusion coefficient in gas phase	1.78 · 10 ⁻⁵	m ² .s ⁻¹
	$x_g^{CO_2} / x_g^{CH_4}$	Inlet CO ₂ /CH ₄ molar ratio	0,4 / 0,6	/
	$x_g^{CO_2}$	Outlet CO ₂ molar ratio	< 0,02	/
Variable parameters	ϕ	Module packing fraction	0.1 – 0.9	/
	u_i/u_g	Interstitial velocity ratio	0.1 – 20	/
	k_m	Membrane effective mass transfer coefficient	10 ⁻³ – 10 ⁻⁵	m.s ⁻¹
	P	Pressure	1 – 10	Bar
	/	Solvents	/	/
	/	Configurations	/	/
	/			

Table 1 : Fixed and variable parameters in this work

Main results

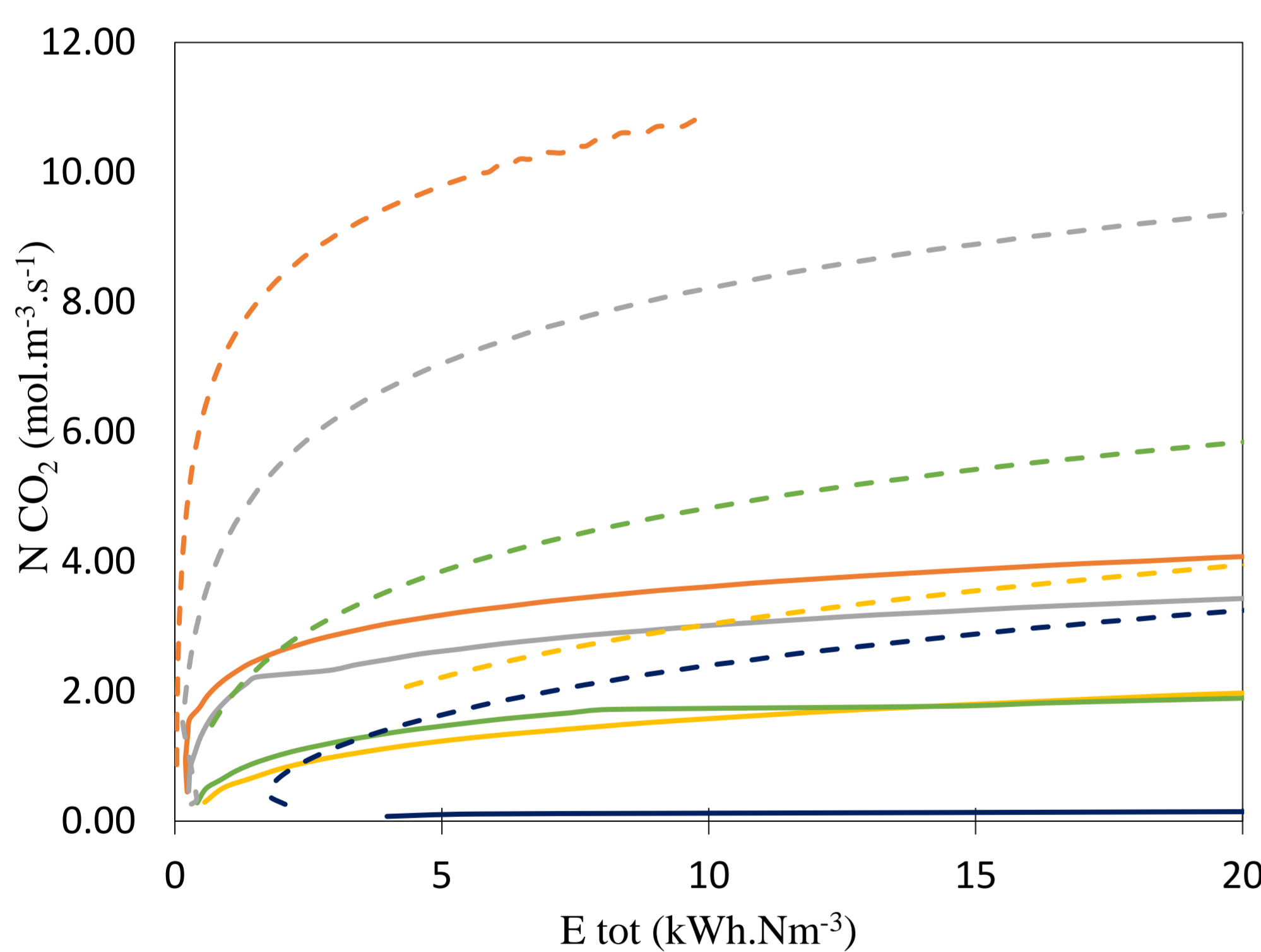


Figure 2: Maximum CO₂ flow for each solvent with k_m value fixed at 10⁻³ m.s⁻¹.

Blue: water; orange: methanol; grey: NMP; yellow: selexol; green : PC., Continuous lines: liquid in conf-iguration (compacity of 0.9); dotted lines: liquid out configuration (compacity of 0.5)

Influence of solvent properties

Performance of the solvents decreases as follows :
methanol > N-methyl-2-pyrrolidone (NMP) > Propylenecarbonate (PC) > selexol > water
Key factors : low CO₂ solubility in water, high viscosity of selexol.

Influence of membrane performances

Decrease of the k_m value decreases the transferred molar flow.
Methanol and NMP are the most efficient solvents and the most affected.
For water, limitation is made by the liquid side instead of the membrane

Influence of flow configuration (liquid in or liquid out)

Optimum of the compacity value in liquid-out configuration. Value is dependent of the solvent used.
Always higher value of compacity is better in liquid-in configuration.
Strong dependence of the correlation used

Comparison with packed columns: Intensification possibilities

High value of intensification factors
Intensification factor independent of the inlet pressure

Solvent	Pressure bar	Z m	N CO ₂ mol.m ⁻³ .s ⁻¹	Etot kWh.Nm ⁻³	IF -
Methanol	1	3.35	3.18	5.03	5.68
	5	0.67	15.9	6.69 10 ⁻²	5.76
	10	0.34	31.8	8.67 10 ⁻²	6.05
NMP	1	3.64	2.93	8.75	7.92
	5	0.73	14.7	8.6 10 ⁻²	8.40
	10	0.36	29.3	8.91 10 ⁻²	8.83
PC	1	5.72	1.86	18.3	7.44
	5	1.14	9.31	0.142	7.82
	10	0.57	18.6	9.61 10 ⁻²	8.23
Selexol	1	5.47	1.95	19.1	13.93
	5	1.09	9.75	0.211	15.00
	10	0.55	19.5	0.105	15.98
Water	1	44.3	0.24	89.9	2.00
	5	8.86	1.20	0.298	1.82
	10	4.43	2.40	0.118	1.78

Table 2 : Influence of the pressure in liquid-in configuration ($u_i/u_g = 1$, $\phi = 0.9$, $k_m = 10^{-3}$ m.s⁻¹).

Conclusion and perspectives

This study intended to explore the **possibilities of process intensification** thanks to membrane contactors for absorption processes based on physical solvents. A **systematic parametric study** has been performed for CO₂ absorption in five different solvents showing a broad range of solubility and viscosity.

The results obtained here show however that **solvent viscosity** strongly impacts the interest and feasibility of membrane contactors compared to packed columns. Because the **solubility / viscosity interplay** plays a key role in the maximal intensification performances, a parametric study on operating temperature would be interesting. It is known that a lower temperature generally increases solubility but also viscosity. The intensification / energy trade-off is thus likely to strongly depend on the solvent temperature. The influence of solvent volatility, which has not been taken into account in this study, should also be considered.

It is expected that this exploratory parametric study will stimulate further researches on intensified processes with physical solvents.

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