Membrane separations and energy efficiency

Christophe Castel, Eric Favre[*]

LRGP-CNRS Université de Lorraine, 1 rue Grandville, 54001 Nancy, France

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ABSTRACT

Membrane processes are usually considered to offer very promising performances in terms of energy efficiency for industrial separations. This statement particularly holds for homogeneous gas and liquid separations which are traditionally performed thanks to unit operations based on a phase change (distillation, evaporation, condensation, crystallization…). The energy efficiency concept can however be addressed through different methodologies, potentially leading to different, if not opposite conclusions. A critical analysis of the energy efficiency concept for membrane separations is proposed. Starting from the most usual minimum work of separation definition, alternative expressions of this key concept are developed in order to better reflect the different types of separation situations encountered for practical purposes (solute purification and/or recovery, process selectivity). In a second step, the real work of separation of a given process, classically evaluated through modern Process Systems Engineering computations, including thermodynamic modelling and irreversible processes computations, is discussed. The interest of the entropy dissipation function, obtained from Irreversible Processes Thermodynamics (IPT) approach is then presented. The methodology is applied to the separation of a perfect gas mixture (air) and non ideal (seawater) liquid mixture. The local entropy dissipation rate offers the opportunity to analyze the impact of fluid distribution in membrane modules, possibly leading to improved designs through the entropy equipartition theory. The largely unexplored possibilities of IPT to provide a predictive evaluation of the overall energy efficiency of a separation process, based on a diffusional mass transfer mechanism, is finally illustrated.

1. Introduction

Membrane processes are considered as a key technology for sustainable processes and green engineering developments [1,2]. Continuous operation (no regeneration step, simple process), low footprint (large specific surface area of modules, intensified process) and environmental friendly operations (no chemical reactions involved, no secondary product, no waste generation) are often mentioned as major advantages [3,4]. The high energy efficiency of membrane operations, mostly due to the fact that they do not require a phase change, is however often considered as one of their most attractive characteristics [5]. Advanced membrane materials properties and optimized process designs are expected to offer outstanding opportunities for energy efficient separations [1,6].

Energy efficiency is indeed one of the key performance characteristics of a separation process which often governs its selection or rejection for a given application [3,4]. The recent evolution of the energy framework strengthens the importance of this criteria and the rigorous comparison of the energy efficiency among different separation processes is a key issue. The situation is however complex, due to the fact

that energy is a multifaceted concept [7]; additionally, a series of methodologies can be applied for energy efficiency analysis. Different evaluation criteria such as Dirac function [8], second law efficiency and exergy analysis [9], finite time thermodynamics [10,11] and irreversible processes thermodynamics [12] have been proposed for energy efficiency evaluation purposes. King states that separation processes based on an energy separation agent offer better energy efficiency compared to mass separating agent or membranes processes [2]. Table 1 summarizes the general comparative performances of different families of separation processes from the energy efficiency point of view. It can be seen that rate governed processes, such as membrane separations, are expected to be less energy efficient.

Several publications more recently addressed the analysis of the separation energy efficiency concept for comparison purposes [14] or for adsorption processes [15]. The specific case of membrane separations has not been however investigated in detail. To our knowledge, a critical synthesis of the different concepts and approaches, with a particular emphasis on membrane processes specificities, is lacking. This is however an essential prerequisite for sake of comparison to other separation equilibrium based separation processes (e.g.

List of symbols

A	Membrane surface area (m^2)
E	Overall energy requirement (W + Q) (J)
J*	Transmembrane diffusion flux ($mol.m^{-2}.s^{-1}$)
N	Transmembrane overall flux ($mol.m^{-2}.s^{-1}$)
p	Pressure (Pa)
p'	Total upstream pressure (Pa)
p"	Total downstream pressure (Pa)
P	Permeability ($mol.m^{-1}.s^{-1}$)
Q	Heat (J)
Q	Flow rate ($mol.s^{-1}$)
R	Perfect gas constant ($J.mol^{-1}.K^{-1}$)
R	Recovery ratio (-)
T	Temperature (K)
X	Mole fraction (-)
W	Work (J)

W_{MIN}	Minimum work of separation, Eq. (1) ($J.mol^{-1}$)
W'_{MIN}	Minimum specific work of purification, Eq. (3) ($J.mol^{-1}$)
W''_{MIN}	Minimum work of selective separation, Eq. (4) ($J.mol^{-1}$)
W_{REAL}	Real work of separation, Eq. (2) ($J.mol^{-1}$)
W_{IRR}	Irreversible work of separation, Eq. (2) ($J.mol^{-1}$)
Y	Mole fraction in the permeate (-)
z	Membrane thickness (m)

Greek symbols

α*	Membrane separation ideal selectivity (-)
β	Process selectivity (-)
η	Energy efficiency (-)
θ	Module stage cut (-)
φ	Heat flux (W)
σ	Entropy dissipation rate ($J.s^{-1}.K^{-1}$)
ψ	Pressure ratio (p"/p')

Table 1

Comparison of energy efficiency performances for different types of separation processes, according to King [4]. α stands for separation selectivity, corresponding to relative volatility for distillation, phase partition selectivity for mass separation agent and membrane ideal selectivity (i.e. ratio of compounds permeability) for rate governed processes.

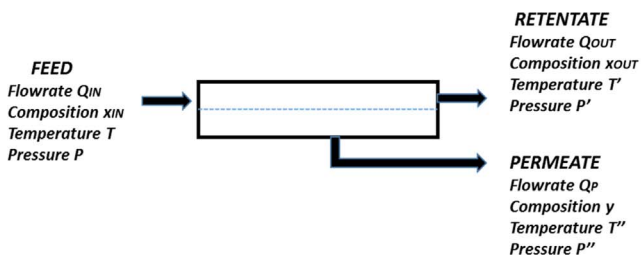| Process type	Examples	Characteristics	Net work consumption W for a difficult separation (α close to 1)
Energy separation agent	Distillation crystallization, condensation	Potentially reversible	W ~ $1/(\alpha-1)$
Mass separation agent	Absorption, extractive distillation, liquid-liquid extraction, adsorption	Partially reversible	W ~ $1/(\alpha-1)$
Rate governed processes	Membrane processes, gaseous diffusion, electrophoresis	Irreversible	W ~ $1/(\alpha-1)^2$

distillation, absorption, adsorption).

This study intends to provide a critical analysis of the evaluation of the energy efficiency of membrane processes. In a first step, the concept of minimum work of separation is critically analyzed and alternative expressions, offering a more realistic evaluation of this term, are proposed. In a second step the evaluation of the real work of separation through Process Systems Engineering simulations is compared to the Irreversible Processes Thermodynamics (IPT) approach (i.e. entropy dissipation rate function). The largely unexplored potential of the IPT approach is then discussed. More specifically, the unique possibility to offer a predictive evaluation of the energy efficiency of a given mixture membrane separation, through key molecular characteristics, is finally illustrated.

2. General framework

2.1. Membrane separation processes

For any separation process design or selection, the starting point obviously is the clear and detailed definition of the problem specifications, with the feed flowrate, temperature, pressure and feed mixture composition as necessary input data (Fig. 1). Homogeneous gaseous or liquid feed mixtures will be considered afterwards. The separation of multiphase mixtures (suspensions, emulsions) through mechanical operations (including in some cases membrane separations) will not be treated here. Depending on the application context, the objective of the separation process (output data) can be:

i) a target compound purity (y or x_{out}) as the only objective, either on the retentate (e.g. nitrogen production from air) or permeate side (e.g. drinking water from seawater, oxygen enriched air)

ii) a combined target compound purity (y or x_{out}) and recovery R (e.g. hydrogen purification; post combustion carbon capture; solute treatment by organic solvent nanofiltration)

iii) an overall process selectivity (x_{out} and y), such as natural gas treatment (minimum CO_2 retentate content and minimum methane losses on the permeate side) or hydrocarbon separations (e.g. propylene/ propane separation) for petrochemicals. This context is similar to classical distillation separation framework where a maximum ratio of the distillate / boiler compositions is wanted.

2.2. Energy efficiency evaluation methodologies

The energy efficiency concept aims to evaluate the ratio between

FEED
Flowrate Q_{IN}
Composition x_{IN}
Temperature T
Pressure P

RETENTATE
Flowrate Q_{OUT}
Composition x_{OUT}
Temperature T'
Pressure P'

PERMEATE
Flowrate Q_P
Composition y
Temperature T''
Pressure P''

SPECIFICATIONS

1. Purity (x_{OUT} or y)

2. Recovery (R)

3. Selectivity (y and x_{OUT})

Fig. 1. A generic flow diagram of a membrane separation process. The homogeneous gaseous or liquid mixture to be treated is specified, together to the inlet conditions (feed flowrate, composition, temperature and pressure). Separation performances for a given membrane material, module design and operating conditions are expressed through the flowrate, composition, temperature and pressure of the two outlet streams (retentate on high pressure side and permeate on low pressure side). The process included in the rectangular box can be single stage, multistaged or hybrid. A binary feed mixture is most often postulated in a first step for sake of simplicity; in that case, specifications 2 and 3 are equivalent in that a target compound recovery ratio R corresponds to a target outlet composition (e.g. if R = $Q_P.y/(Q_{IN}.x_{IN})$, x_{OUT} is imposed). More complex architectures, with multiple outlet streams can be proposed for multicomponent feed mixtures and multipurpose separation targets.