



Process intensification with selected membrane processes



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ABSTRACT

Membrane devices, membrane processes and membrane-based conventional chemical engineering processes have achieved extraordinary levels of process intensification (PI). Generally membrane-based devices require smaller equipment to achieve a given device production rate. Further they can eliminate dispersion-based operation and achieve extraordinary selectivity. Exploiting the compartmentalization of two regions on two sides of the membrane, membrane devices can combine two processes carried out on two sides of the membrane in one membrane device. Selected membrane processes and their applications are briefly reviewed here in terms of the level of PI achieved. The membrane processes selected are generally recently commercialized or being commercialized or have great potential for commercialization: membrane bioreactor; membrane gas–liquid contacting; membrane solvent extraction; forward osmosis; pressure-retarded osmosis; membrane distillation; membrane distillation bioreactor. Examples of potential process intensification by membrane processes are briefly illustrated for processing of lignocellulose to biofuels in a biorefinery and for produced water treatment.

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1. Introduction

Process intensification is of increasing interest in chemical process and allied industries. Several definitions of process intensification exist in the literature. One of the earliest uses of the phrase “process intensification” appeared in an article by Ramshaw [69] which described a rotating packed bed for gas–liquid contacting (HIGEE) for which US patents were issued 1981 and 1982. Ramshaw [69] identified process intensification (PI) as a strategy for reducing the plant size for a given production goal; correspondingly PI also stands for reducing equipment size for a given performance level in terms of the mass transfer rate or the reaction rate per unit equipment volume. Stankiewicz and Moulijn [87] defined PI to be any chemical engineering development that leads to a “substantially smaller, cleaner and more energy-efficient technology”. However they specified that PI should concern only engineering methods and equipment and not a new chemical route or a new catalyst. Therefore goals like atom economy and green chemistry are not part of PI. Sustainability may still be included as long as a cleaner and more energy-

efficient technology has been achieved since those two are integral parts of sustainability.

If one considers the expositions of PI provided by Stankiewicz and Moulijn [87] or that by Charpentier [9], it becomes clear that their focus is on intensifying traditional processes and devices employed in chemical engineering especially reaction engineering. Membranes appear in their lists only through membrane reactors, membrane contactors, membrane absorption, membrane distillation and membrane crystallizer. In their papers, there is no mention of membrane processes such as reverse osmosis for which there does not appear to be any analog in classical chemical engineering processes and devices. Yet reverse osmosis (RO), for example RO desalination, should qualify as an example of extraordinary process intensification compared to the classical methods of thermal desalination since there is no new chemical route or a new catalyst.

In reverse osmosis, the notion of classical filtration has merely been taken to its logical conclusion in terms of an increasingly smaller particle size being separated ultimately ending with molecular level filtration. In fact one of the terms originally proposed for what goes under the title “reverse osmosis” was “hyperfiltration” [86] since in actual reverse osmosis one cannot reverse the salt leakage that inevitably takes place during osmosis. Therefore the conventional membrane processes of nanofiltration, ultrafiltration and microfiltration are also logical candidates for PI along with hyperfiltration.

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In the PI framework by Stankiewicz and Moulijn [87], any development in the process equipment or method that contributes to dramatic improvements in manufacturing and processing qualifies as process intensification. It must ultimately result in a cheaper and more sustainable technology. There are two specific types of developments. The first type of development leads to dramatic improvements in processing by eliminating or bypassing limitations of conventional chemical engineering equipment. A second type of development consists of a combination of two or more processes or functions in one device or process resulting in PI. It is expected that such developments will lead to smaller/compact devices for the same production goal.

Illustration of the first type of development leading to dramatic improvements in processing by eliminating or bypassing limitations of conventional equipment is provided by the processes of membrane absorption, membrane extraction and membrane stripping. The devices for such processes are broadly identified as membrane contactors. In conventional devices based on phase dispersion, there are limits of phase flow rate ratios beyond which there would be flooding. By having immobilized gas–liquid interface [20,89] or liquid–liquid interface [43] at the pore mouths of porous hydrophobic membranes, the limitation on the flow rate ratio of the two phases is removed as long as appropriate pressure conditions are maintained and breakthrough pressure difference between the phases is avoided. There are a number of other basic advantages in such membrane contactors: a much enhanced surface area per unit equipment volume leading to highly compact devices when hollow fiber membranes are used; modular equipment for easy scale up or scale down; nondispersive operation eliminating foaming and weeping in gas–liquid systems, prevention of emulsification in liquid–liquid systems; elimination of the need for coalescence in liquid–liquid contacting processes; no need for density difference in liquid–liquid systems.

The earliest illustrations of the second type of development are provided by membrane distillation (MD) [29,71,73] and membrane crystallization (MC) [95]. The process of membrane distillation, especially direct contact membrane distillation (DCMD), combines evaporation of a volatile species on one surface of a porous non-wetted hydrophobic membrane with its condensation in the distillate stream on the other surface of the membrane; the primary application has been removal of water from a hot brine into a cold distillate stream. In air gap membrane distillation (AGMD), the two functions take place in two distinct parts of one separation device: evaporation takes place on one surface of the porous hydrophobic membrane as the water vapor emerging from the other side of the non-wetted membrane encounters a contiguous cold surface and condenses. In membrane crystallization, the process of membrane distillation is utilized to concentrate the solution initiating crystallization.

An even earlier illustration of the second type of development is found in a supported liquid membrane (SLM) or an immobilized liquid membrane (ILM) or later in a contained liquid membrane (CLM). In a SLM, the pores of a porous/microporous membrane are filled with a liquid which is held by capillary forces. Feed liquid solution meant to undergo solvent extraction contacts one surface of the pore liquid on one side of the membrane where solvent extraction takes place. The feed liquid and the pore liquid (solvent) must be immiscible. On the other side of the membrane, back extraction of the extracted solute takes place from the pore solvent into a back extraction solvent with which the pore liquid must be immiscible. The phrase ILM is used more often when a gas mixture is separated through the liquid membrane immobilized in the pores of the support membrane. Contained liquid membrane achieves separation in a similar fashion except the liquid membrane is now placed between two separate membranes or two separate sets of porous hollow fibers.

Membrane reactors illustrate also a combination of two or more processes/functions in one device. The functions include among others combining multiple reactions on two sides of the membrane, combining reaction with separation or separation with reaction, immobilizing or segregating a catalyst along with separation and reaction. Distributed introduction of reactants through the membrane, nondispersive operation, heat exchange, functioning as an electrode etc., are additional functions implemented via membranes in a membrane reactor. The large number of functions that can be carried out in a membrane reactor for chemical, biochemical, biopharmaceutical and petrochemical manufacturing has been summarized in Sirkar et al. [84]. An early review of membrane bioreactors is available in Cheryan and Mehaia [12]. Although there are quite a few large-scale applications of membrane bioreactors in amino acid production and pharmaceutical synthesis, the largest volume application is the membrane bioreactor (MBR) for municipal water treatment. In a MBR, the suspended growth activated sludge system utilizes a microporous membrane for ultrafiltration which carries out solid/liquid separation and eliminates the secondary clarifiers.

In the next section we focus on selected individual membrane processes and membrane-based processes and provide illustrations of the process intensification achieved. When we refer to membrane-based processes ideally we refer to conventional processes in chemical engineering such as absorption, desorption, stripping, extraction, back extraction, distillation, crystallization and chemical reaction implemented using a membrane. On the other hand, any type of filtration process using a membrane is identified as a membrane process. We do not dwell much on this distinction in the next section as individual processes are considered. References to appropriate literature as well as important reviews will be added for each process. In the last section two potential examples for PI by membrane processes are provided: the process route to biofuels from lignocellulose in a biorefinery; produced water from oil exploration. Both illustrate how membrane processes can introduce considerable PI in challenging separation applications.

2. Selected membrane processes for process intensification

We first identify the membrane processes and devices of interest here. Our primary criteria for process selection are as follows. These are new membrane processes or membrane-based processes and are either commercialized or being commercialized. Some processes have been commercialized over the last decade and a half: membrane absorbers, membrane bioreactors, membrane extractors, membrane strippers. The processes of membrane distillation (MD), forward osmosis (FO) and pressure-retarded osmosis (PRO) are being commercialized.

2.1. Membrane bioreactor (MBR)

The membrane bioreactor for wastewater treatment is a classic example of a hybrid membrane process and PI. The conventional activated sludge process (CASP) involves an aerobic suspended growth bioreactor followed by a settling tank to provide a settled treated wastewater of low BOD. The settled sludge is returned to the bioreactor and a small fraction is discharged, as 'waste sludge', to maintain a constant level of mixed liquor suspended solids (MLSS), or biomass, in the reactor. In the CASP the concentration of MLSS is a compromise as higher values improve the biological removal of BOD but also lead to less efficient settling and residual SS in the treated water. A value of MLSS of about 3–5 g/L is typical.

The advent of ultrafiltration membranes in the late 1960s provided the opportunity to replace the settling tank with a more efficient solid/liquid separation. Initially the approach was to

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