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## Solvent recycle with imperfect membranes: A semi-continuous workaround for diafiltration

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## ABSTRACT

For separation of a two-component mixture, a three-stage organic solvent nanofiltration (OSN) process is presented which comprises of a two-stage membrane cascade for separation with a third membrane stage added for integrated solvent recovery, i.e. solvent recycling. The two-stage cascade allows for increased separation selectivity whilst the integrated solvent recovery stage mitigates the otherwise large solvent consumption of the purification. This work explores the effect of washing the solvent recovery unit at intervals in order to attain high product purities with imperfect solvent recovery membranes possessing less than 100% rejection of the impurity. This operation attains a purity of 98.7% through semi-continuous operation with two washes of the solvent recovery stage, even when imperfect membranes are used in a closed-loop set-up. This contrasts favourably with the 83.0% maximum purity achievable in a similar set-up with a single continuous run. The process achieves slightly lower (−0.7%) yield of around 98.2% compared to a continuously operated process without solvent recovery but consumes approx. 85% less solvent (theoretical analysis suggests up to 96% reduction is possible). 9 different membranes, both commercial (GMT, Novamem, SolSep) and in-house fabricated, are screened and tested on a separation challenge associated with the synthesis of macrocycles – amongst the membrane materials are polyimide (PI), polybenzimidazole (PBI) and, polyetheretherketone (PEEK).

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## 1. Introduction

## 1.1. Application of solvent recovery

Organic solvent nanofiltration (OSN) is a maturing membrane-based separation technology that allows discrimination between molecules in the range of approx. 100–2000 Da in organic media. Some OSN membranes can also operate in harsh pH and temperature environments. The key features and application areas of organic solvent nanofiltration have been reviewed [1–3].

Diafiltration is a technique commonly employed for separating two-, or multi-component mixtures using membranes, where at least one impurity is continuously washed out of a system through a selective membrane as shown in Fig. 1, with the product(s) being retained in the system. In order to keep the volume of the system constant when washing impurities through the membrane,

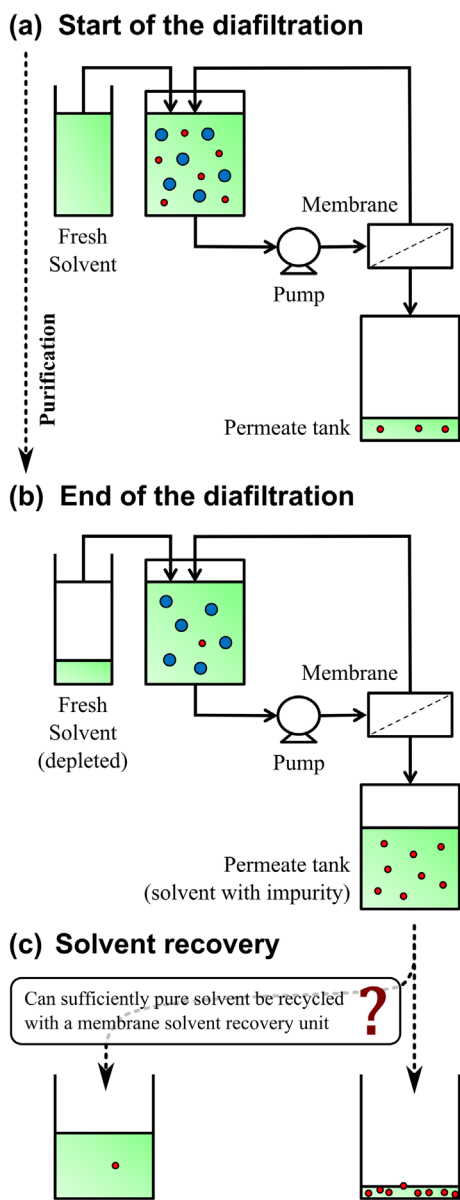
diafiltrations require the replenishment of many multiples of the system volume with fresh solvent.

Solvent throughput and disposal is typically the largest operational cost in diafiltration, and solvent recovery (SR) is therefore an increasingly sought-after concept to make processes sustainable and economically viable. Several authors have worked towards implementing some form of solvent recovery in OSN applications, with common techniques including non-selective adsorption [4,5], hybrid distillation [6] or hybrid chromatography [7] processes, as well as recovery solely through low-molecular cut-off membranes [4,8–11]. In most of these implementations, membrane-based solvent recovery promises advantages over traditional methods of solvent recovery with regards to energy consumption and sustainability.

The main limitation of membrane-based solvent recovery is the inability of most currently available OSN membranes to retain very small impurities in the molecular weight range below 200 Da. When rejection is not absolute (< 100%), recycled solvent from a solvent recovery unit will contain some impurity which, in a closed loop system, will recirculate back into the separation stages.

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**Fig. 1.** Conceptual depiction of (a) the start and (b) the end of a diafiltration process and (c) a key challenge in reducing the operational cost of diafiltrations – the recovery of solvent. A diafiltration process comprises at least two solutes and one solvent, here shown as large blue circles (product), small red circles (impurity) and green background coloration (solvent) respectively. Adapted from [1] under a Creative Commons CC-BY Usage Agreement. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

In a typical diafiltration, the smallest solute that needs to be retained is the main product and rejections over 95% are usually acceptable, yielding close to quantitative product recovery in a two-stage cascade operation [12]. However, in a membrane solvent recovery operation, all solutes need to be retained and rejections as high as 99% can be insufficient for effective solvent recovery in a continuous closed-loop system, if high product purities are required. As a result, where membrane-based solvent recovery was demonstrated, the solutes to be retained were usually large enough to allow close to absolute rejection in the SR stage as summarised in Table 1. There are exceptions where solvent recovery was used in settings where some permeation of solutes is permissible, usually in closed-loop configurations.

**Table 1**

Typical rejections (> 99%) for feasible membrane-based solvent recovery, and examples of applications where some solute permeation is permissible, thus allowing a lower rejection.

Species <sup>a</sup>	MW	Rejection	Solvent	Membrane	Ref.
Macrocyclic intermediate <sup>b</sup>	675	99.5–99.7%	THF	DuraMem™ 300	[8]
Triphenylmethanol	260	99.8%	MeOH	DuraMem™ 150	[9]
Active Pharmaceutical Ingredient <sup>b</sup>	≈ 600	> 99.9	IPAc	Starmem™ 122	[11]
		99.7–99.8%		Starmem™ 240	
		99.6–99.8%		Puramem™ 280	
Desmopressin	1,069	≥ 96.2% <sup>c</sup>	H <sub>2</sub> O	Inopor™ 0.9 nm TiO <sub>2</sub> ceramic	[10]
Lube oil		≈ 98% <sup>c</sup>	MEK-Toluene	Polyimide based	[13]

<sup>a</sup> In studies where more than one species is retained by the solvent recovery membrane, the rejection and corresponding molecular weight of the least rejected impurity is given.

<sup>b</sup> The exact structures were not disclosed in these publications.

<sup>c</sup> In these applications, some permeation of the product is permissible.

Examples include a recent cyclisation of a peptide in a high dilution reactor [10] and the MAX-DEWAX process for recovery of chilled solvent from lube oil filtrates after dewaxing [13]. These and further applications such as recovery of extraction solvents e.g. from vegetable oil processing have been reviewed from an industrial viewpoint [14].

In a continuous closed-loop diafiltration where the process relies solely on a membrane to perform solvent recovery, our colleagues have found complete solvent-solute separation infeasible for a very small impurity [4]. It was observed that none of the membrane rejections were sufficiently high to perform membrane purification of a solvent containing the low molecular weight 4-(dimethylamino)pyridine impurity (MW = 122.17 g mol<sup>-1</sup>) [4].

The mathematics for continuous operation in closed-loop diafiltration systems (as discussed in Section 2.5.2) dictate that high purities should not be attainable unless rejections of all species in the solvent recovery stage are close to 100% [9]. We will highlight a semi-continuous workaround for these systems that allows operating with imperfect solvent recovery membranes. This will involve periodic washing of the solvent recovery unit when the impurity is most concentrated.

## 1.2. Control of membrane cascades and implementation of solvent recovery

With the recognition that membranes with outstanding selectivity (very tight MWCO curves, approximating step-change rather than sigmoidal) may be unattainable at least in the short term, much research has focused on workarounds by improving selectivity through process operation. Membrane cascades offer potentially large gains in selectivity and they are often discussed on a purely theoretical basis [15–17] although partially experimental [18–23] and fully experimentally validated examples exist [4,12,24–27]. Much of the published work extrapolates single-stage screening results to predict and analyse the performance of multiple stages. Whilst the predictive capability for two- or three-stage cascades may actually be sufficient to warrant the conclusions drawn, there are major obstacles to industrial implementation. Beside the diminishing economic benefit of introducing additional stages [18], process control is challenging with large cascades due to the step increase in

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