

# Comparison of Methods To Enhance Separation Characteristics in Nanofiltration

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This manuscript explores the possibilities of obtaining a more efficient separation of dissolved organic compounds from an aqueous matrix using nonconventional process designs in nanofiltration. It was attempted to transform the typical sigmoid rejection curve (representing the rejection of a solute as a function of molecular size) into a step function with a sharp rejection increase at the molecular weight cutoff. This was first done using a membrane stack that contained up to three membranes in the same module without any spacers between them. Experimental results with four commercially available nanofiltration membranes showed that the rejection of various test solutes was slightly higher, but the sharpness of the rejection curve was not improved. The results were compared with an in-series filtration using up to three membrane passages. The rejection curves were steeper in this case, but the overall molecular weight cutoff shifted toward very low molecular weights, which was not intended, and the overall permeate recovery of this system was too low. However, good results were obtained by recycling each retentate stream to the feed of the previous filtration step. Mass balances showed that the steepness of the rejection curves was greatly improved.

## Introduction

Membrane processes—pressure-driven processes, in particular—have a proven performance for separating organics, salts, microorganisms, nutrients, biomass, and even micropollutants from wastewater.<sup>1–6</sup> However, the application of membrane technology in practice is still rather limited, although some innovations are slowly finding their way into large-scale applications.<sup>7,8</sup> The main reason for the hesitation to implement membrane processes on large scale, despite membranes being less expensive, safer, and more efficient, are related to (a) the fear of performance losses due to fouling and the consequent need for cleaning and membrane replacement,<sup>9–12</sup> and (b) problems related to concentrate disposal.<sup>13,14</sup> There is a strong relationship between the use of membranes in wastewater treatment and in desalination,<sup>15</sup> where similar problems occur. Nevertheless, membranes currently are the dominant technology in desalination. The objective in desalination is to remove all organic solutes and most of the inorganics. This can be achieved using reverse osmosis membranes, which allow one to reduce the salinity to ca. 500 ppm. The price for this is a relatively high energy cost, at least in comparison with other membrane processes; in comparison with thermal processes, huge energy savings can be obtained using reverse osmosis. In view of the global challenge of finding sustainable energy sources, it is clear that thermal processes should be avoided and replaced by other systems,<sup>16</sup> and it can be assumed that all remaining thermal desalination units will be gradually replaced by reverse osmosis units. A similar trend toward the application of membrane processes can be expected in wastewater treatment, although this should be attributed to the materials intensity rather than to the energy intensity. Membrane processes are clean processes in the sense that they generally do not require chemicals; they

do not produce sludge or require the input of additional liquids or solids. This makes them intrinsically more attractive than conventional techniques, on the condition that the difficulties previously mentioned can be overcome. An additional requirement is that they should operate at lower costs than what is generally accepted in desalination, except for special applications where reverse osmosis can be justified. In other cases, a membrane process must be found that delivers the desired permeate quality within a reasonable range. Taking the typical sigmoid rejection curves (rejection as a function of molecular size) for pressure-driven membranes into account, representing a far-from-perfect discrimination between different solutes on the basis of molecular size, this is certainly not evident. In addition, the chemical functionality of dissolved solutes may also have a role in the removal.<sup>17–20</sup> Most, if not all, commercial membranes remove solutes only gradually over a wide size range and deliver a permeate that contains a range of compounds with molecules larger as well as smaller than the claimed pore size; it can hardly be predicted which solute will be retained and which will not. Model calculations for the rejection of uncharged compounds invariably indicate a slow increase of rejection as a function of solute size.<sup>21–23</sup> This is illustrated by the use of the “molecular weight cutoff” (MWC), the molecular weight of a solute rejected for 90%: a good separation can only be achieved between a solute at least an order of magnitude larger than the MWC and a molecule at least an order of magnitude smaller than the MWC. The large area below the rejection curve between a “low” rejection (~10%) and an “almost complete” rejection (~90%) is not functional and leads to ill-defined products (permeate as well as concentrate). In most cases, it is not possible to use both the permeate and the concentrate, because the process is optimized in terms of one fraction, leaving the other fraction with an undefined quality.

This paper investigates different possibilities to improve the separation characteristics in pressure-driven membrane processes, nanofiltration in particular. The objective is to transform the sigmoid rejection curve for typical commercial nanofiltration membranes into a step function with a sharp increase at one specific molecular size. This would allow to obtain a sharp

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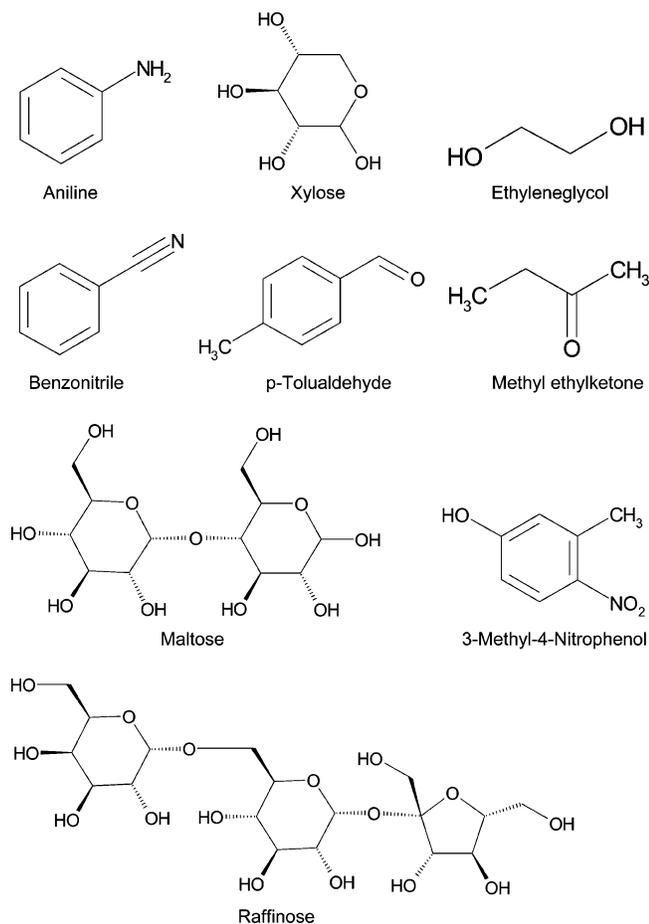


Figure 1. Structure of the solutes used in the experiments.

separation between dissolved solutes with different molecular weights. The approach used in this paper is to use different membranes in a single module and to use a sequence of filtration steps with and without recycle streams.

## Methods and Materials

**Experiments.** Filtration experiments were conducted using a commercial nanofiltration unit on a laboratory scale (Amfilter, Test Rig PSS1TZ). Flat sheet membranes were used with a diameter of 0.09 m. The active surface of the membrane is 0.0059 m<sup>2</sup>; a tortuous flow pattern was used inside the module, resulting in a rectangular flow channel of 293 mm length and a width of 10 mm that was in contact with the membrane (hydraulic diameter = 4.2 mm). Filtration was always performed until a steady state value was obtained (usually 2 h). All experiments were performed at 25°C and an operating pressure of 8 bar. The cross-flow velocity of the feed was 6 m/s. Membranes used in the experiments are N30F (Nadir), NF270 (Dow/Filmtec), and Desal 51 HL and Desal 5 DL (GE Osmonics). N30F is a permanently hydrophilic polyethersulfone membrane with an MWC of ~580 and a water contact angle of 88°. NF270 is a polyamide membrane with an estimated MWC of 180, according to the manufacturer, and a water contact angle of 27°. Desal 51 HL is a cross-linked aromatic polyamide with an MWC of 300 and a water contact angle of 47°. Desal 5 DL is also a cross-linked aromatic polyamide with an MWC of 250 and a water contact angle of 44°.

A series of nine organic test molecules was used to this purpose (methyl ethyl ketone, aniline, benzonitrile, *p*-tolualdehyde, xylose, 3-methyl-4-nitrophenol, tetraethylene glycol,

Table 1. Solutes Used (in Aqueous Solution) in Filtration Experiments, with Their Molecular Weight, log  $K_{ow}$  Value, and Determination Method

solute	molecular weight, MW	determination method	log $K_{ow}$
methyl ethyl ketone	72	gas chromatography	0.26
aniline	93	UV spectrophotometry	0.94
benzonitrile	103	UV spectrophotometry	1.54
<i>p</i> -tolualdehyde	120	UV spectrophotometry	2.26
xylose	150	Dubois et al. <sup>21</sup>	-1.98
3-methyl-4-nitrophenol	153	UV spectrophotometry	2.45
tetra ethylene glycol	194	gas chromatography	-2.02
maltose	360	Dubois et al. <sup>21</sup>	-5.03
raffinose	504	Dubois et al. <sup>21</sup>	-6.76

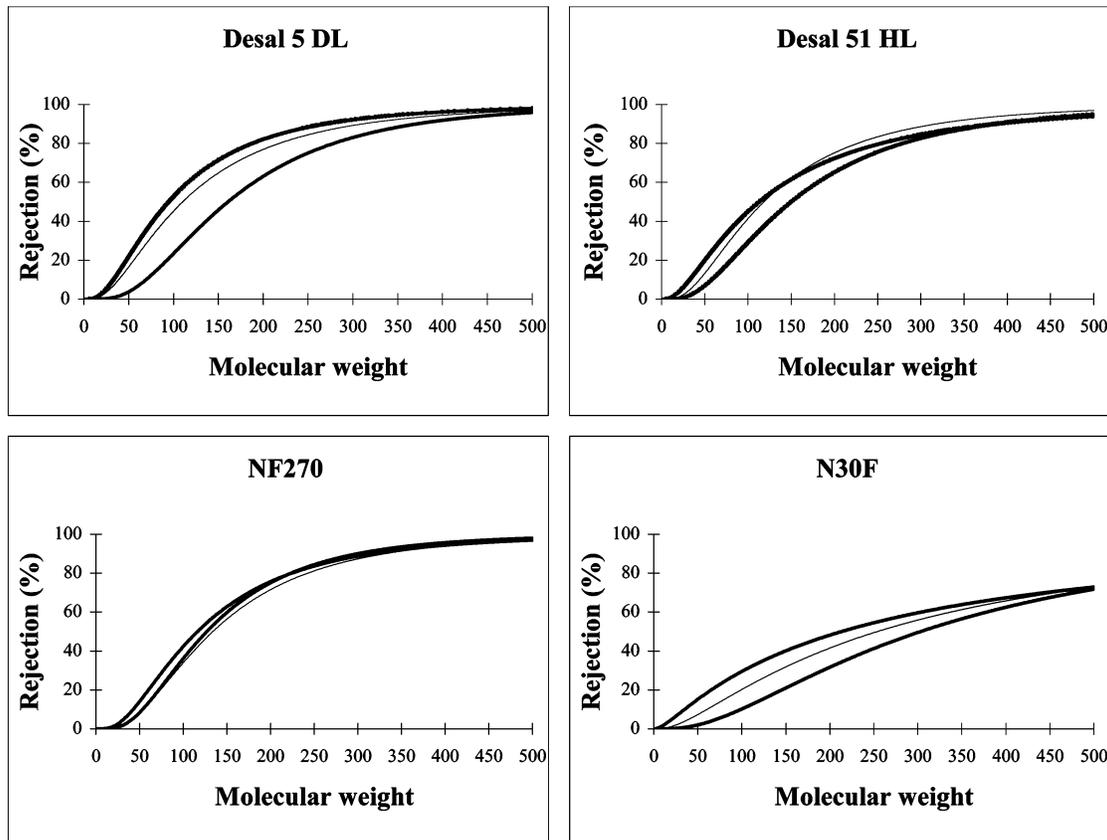
Table 2. Rejection of Three Solutes Using a Stack of up to Three Membranes (Including Desal 5 DL, Desal 51 HL, NF 270, and N30F)

	Rejection (%)		
	methyl ethyl ketone	xylose	raffinose
One Membrane			
Desal 5 DL	11	20	96
Desal 51 HL	16	61	95
NF270	20	87	98
N30F	5	20	72
Two Membranes			
Desal 5 DL	30	16	97
Desal 51 HL	26	86	97
NF270	19	86	97
N30F	13	23	73
Three Membranes			
Desal 5 DL	37	52	98
Desal 51 HL	32	81	94
NF270	27	94	97
N30F	22	42	73

maltose, raffinose). Each solute was tested separately with a feed concentration of 2 mmol/L. The molecular structure of all solutes is given in Figure 1. The molecular weight of these solutes, along with their log  $K_{ow}$  value (i.e., the logarithm of the octanol–water partition coefficient, which is a measure for the hydrophobicity of a solute) and determination method, is given in Table 1. Aniline, benzonitrile, *p*-tolualdehyde, and 3-methyl-4-nitrophenol were determined with ultraviolet (UV) spectrophotometry, using wavelengths of 228.6, 224.6, 260.6, and 230.2 nm, respectively. Xylose, maltose, and raffinose were determined by the method described by Dubois et al.,<sup>25</sup> using a reaction with phenol after the formation of furfural derivatives in sulfuric acid, which yields a colored product with a measured wavelength of 485 nm. Methyl ethyl ketone and tetraethylene glycol were determined via gas chromatography (GC), using a Hewlett–Packard model HP5890 series apparatus that was equipped with a cross-bond trifluoropropylmethyl-polysiloxane semi-capillary column with an internal diameter of 0.32 mm and a length of 30 m and a flame ionization detector (FID).

**Modeling.** In the modeling, it was assumed that the membranes are nanoporous and that solutes are transported by a sieving mechanism. Steric hindrance in the pores or hydrodynamic lag is not taken into account when considering the rejection; it is assumed that a molecule permeates through every pore that is larger than the diameter of the molecule. The membranes' pore size distribution is assumed to be log-normal. With these assumptions, the (maximal) rejection  $\sigma$  can be expressed as a function of the radius of the solute  $r_c$ :

$$\sigma(r_c) = \int_0^{r_c} \frac{1}{S_p \sqrt{2\pi}} \frac{1}{r} \exp\left(-\frac{(\ln(r) - \ln(\bar{r}))^2}{2S_p^2}\right) dr \quad (1)$$



**Figure 2.** Rejection curves calculated with the log-normal model using a membrane stack of up to three membranes. In each figure, the curve starting on the right-hand side is for a single membrane, the middle curve is for a stack of two membranes, and the curve starting on the left-hand side is for a stack of three membranes.

This equation is comprised of two variables,  $S_p$  and  $\bar{r}$ , where  $S_p$  is the standard deviation of the distribution (a measure for the distribution of the pore sizes) and  $\bar{r}$  is the size of a molecule retained for 50%. The radius of the solute can be expressed as a molecular width or an effective solute size as was done in previous work.<sup>22</sup> Using this approach, an empirical relation was observed between the effective diameter and the solutes' molecular weight MW (for a series of molecules with molecular weights between 100 and 600 containing most solutes used here):  $r_c = 0.0325(\text{MW})^{0.438}$ .

This finally leads to an equation expressing the (maximal) rejection as a function of molecular weight, taking the two membrane parameters into account:

$$\sigma(\text{MW}^*) = \int_0^{\text{MW}^*} \frac{1}{S_p \sqrt{2\pi}} \frac{1}{0.0325(\text{MW})^{0.438}} \times \exp\left(-\frac{[\ln(0.0325(\text{MW})^{0.438}) - \ln(0.0325(\bar{\text{MW}})^{0.438})]^2}{2S_p^2}\right) 0.0325 \cdot 0.438 \cdot (\text{MW})^{-0.562} d\text{MW} \quad (2)$$

or, in terms of MWC,

$$\sigma(\text{MW}^*) = \int_0^{\text{MW}^*} \frac{1}{S_{\text{MW}} \sqrt{2\pi}} \frac{1}{\text{MW}} \times \exp\left\{-\frac{[\ln(\text{MW}) - \ln(\text{MWC}) + 0.56S_{\text{MW}}]^2}{2S_{\text{MW}}^2}\right\} d\text{MW} \quad (3)$$

The parameters are  $S_{\text{MW}}$  and MWC.  $S_{\text{MW}}$  is a measure of the steepness of the rejection curve.

**Table 3. Modeling Results (MWC and  $S_{\text{MW}}$ )<sup>a</sup> for the Rejection Curve Using One, Two, and Three Membranes**

	One Membrane		Two Membranes		Three Membranes	
	MWC	$S_{\text{MW}}$	MWC	$S_{\text{MW}}$	MWC	$S_{\text{MW}}$
Desal 5 DL	232	0.65	174	0.81	150	0.81
Desal 51 HL	227	0.74	183	0.77	194	0.96
NF270	185	0.67	199	0.70	180	0.78
N30F	495	0.87	475	1.12	469	1.40

<sup>a</sup> MWC is the effective molecular weight cutoff, whereas  $S_{\text{MW}}$  is a measure for the sigmoidal shape of the curve with low values corresponding to a better steplike function.

This model is based on the rejection of solutes due to size interactions, i.e., on a comparison of the solute's size and the pore size distribution in the membrane. The effects of differences in chemical functionality of the solutes are not taken into account. These were minimized in this study using solutes that are not too hydrophobic (see Table 1). A more-detailed discussion and an evaluation of the quality of the model can be found in a previous publication.<sup>22</sup>

## Results and Discussion

**Nanofiltration in a Membrane Stack.** A first approach to obtain a well-defined separation and a steeper rejection curve was the use of a membrane stack, i.e., a series of (identical) membranes in a single module, without any spacers between them. Thus, the separation is performed in a single step, but filtration occurs through a triple layer of membrane sheets. This method was applied successfully for the separation of proteins in ultrafiltration at low concentrations.<sup>27,28</sup> Typical (rejection) results are given in Table 2 for the nanofiltration of a solution

**Table 4. Rejection in a Filtration-in-Series without Recycle for NF270**

	Observed	Calculated	
	Rejection (%)	Rejection (%)	Rejection (%)
	one	two	three
	membrane	membranes	membranes
methyl ethyl ketone	20	36	49
aniline	0	0	0
benzotriline	7	14	20
<i>p</i> -tolualdehyde	2	4	6
xylose	87	98	99.8
3-methyl-4-nitrophenol	21	38	51
tetra ethylene glycol	82	97	99
maltose	98	99.9	99.9
raffinose	98	99.9	99.9

that contained methyl ethyl ketone (the solute with the lowest molecular weight), xylose (a solute with intermediate molecular weight), and raffinose (the solute with the largest molecular weight), using a stack of up to three identical membranes. The results for all other compounds follow the same trends. Table 2 shows that the rejection of methyl ethyl ketone increases significantly when a membrane stack is used. A similar observation was made for xylose (and other low-molecular-weight solutes; see Table 2). For raffinose, which is a compound that has already been well-rejected by a single membrane (rejections of 95%–98% were observed, except for N30F, which had a rejection of 72%), the increase seems to be insignificant.

The rejection curves calculated with the log-normal model are given in Figure 2. A visual inspection of these curves reveals that the overall difference in rejection is not dramatic. This is confirmed by the modeling results (see Table 3). The effective MWC in a membrane stack generally decreases somewhat; for Desal 5 DL, this seems to be a significant effect. However, for

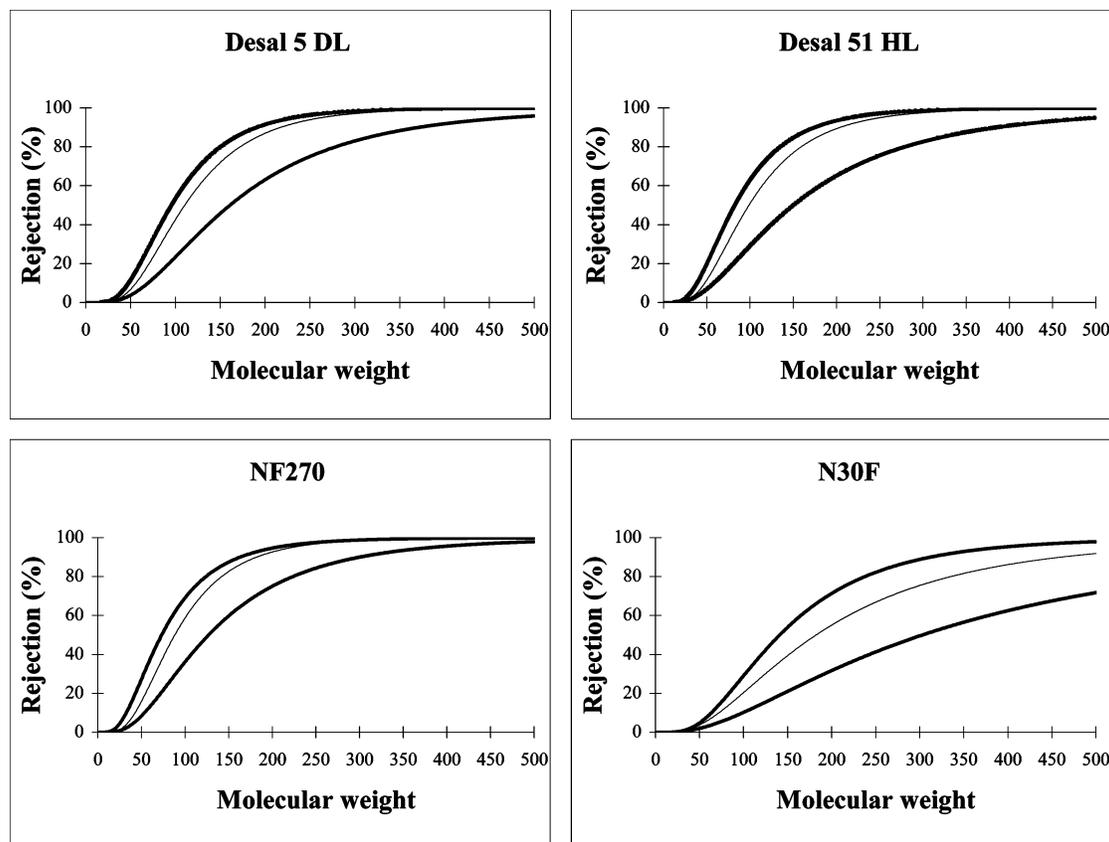
the other three membranes, the decrease of the MWC is within the experimental error, as can be derived from the irregular behavior of the MWC for NF270 when going from one membrane to three membranes. The  $S_{MW}$  parameter, which should be minimized in view of a steplike rejection curve, does not decrease as a function of the number of membranes; on the contrary, the value of  $S_{MW}$  increases. Thus, the small effect on MWC is annihilated by a negative effect on the shape of the rejection curve. This behavior can possibly be explained by an increased concentration polarization between the membrane sheets, because of the combination of relatively high concentrations and an uncontrolled flow pattern.

The membrane fluxes were lower when more membranes were used (the pressure applied to obtain the same flux was higher). When two membranes were used, the fluxes were lower by a factor of <2 (1.4–1.7), depending on the membrane. When three membranes were used, the fluxes were lower by a factor of <3 (1.6–2.1). These factors were always in the following order:

$$N30F \approx \text{Desal 5 DL} < \text{Desal 51 HL} < \text{NF270}$$

Thus, the pressure increase to be applied is less than linear. However, the effect on the separation was not very significant. Therefore, it could be concluded that the approach of using a membrane stack was unsuccessful.

**Comparison: Filtration in Series without Recycle.** A comparison with a filtration in series without recycle (i.e., in an operation where two or three membrane modules are used consecutively, and where the permeate stream obtained from each module is used as the feed for the following module; the concentrate from each module is discharged) is useful to



**Figure 3.** Rejection curves calculated with the log-normal model using a multiple membrane passage without recycle. In each figure, the curve starting on the right-hand side is for a single membrane passage, the middle curve is for two membrane passages in series, and the curve starting on the left-hand side is for three membrane passages in series.

**Table 5. Modeling Results (MWC and  $S_{MW}$ )<sup>a</sup> for the Rejection Curve Using One, Two, and Three Membrane Passages**

	One Membrane		Two Membranes		Three Membranes	
	MWC	$S_{MW}$	MWC	$S_{MW}$	MWC	$S_{MW}$
Desal 5 DL	232	0.65	149	0.53	130	0.54
Desal 51 HL	227	0.74	136	0.57	115	0.58
NF270	185	0.67	122	0.56	104	0.62
N30F	495	0.87	274	0.72	200	0.62

<sup>a</sup> MWC is the effective molecular weight cut-off, whereas  $S_{MW}$  is a measure for the sigmoidal shape of the curve with low values corresponding to a better steplike function.

**Table 6. Modeling Results (MWC and  $S_{MW}$ )<sup>a</sup> for the Rejection Curve Using One, Two, and Three Membrane Passages with Recycle of the Retentate**

	One Membrane		Two Membranes		Three Membranes	
	MWC	$S_{MW}$	MWC	$S_{MW}$	MWC	$S_{MW}$
Desal 5 DL	232	0.65	151	0.49	127	0.45
Desal 51 HL	227	0.74	142	0.55	112	0.49
NF270	185	0.67	121	0.50	102	0.52
N30F	495	0.87	272	0.67	192	0.55

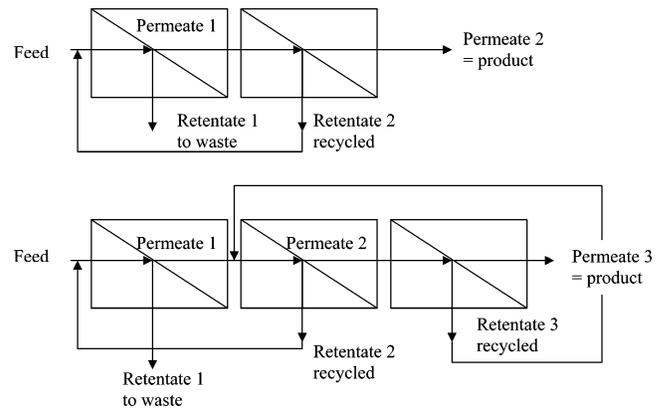
<sup>a</sup> MWC is the effective molecular weight cut-off, whereas  $S_{MW}$  is a measure for the sigmoidal shape of the curve with low values corresponding to a better steplike function.

investigate the maximal achievable rejection of various compounds. This can be done using mass balances and assuming the same intrinsic rejection in each stage. The result of this calculation is given in Table 4 for NF270 (the same observations were made for the three other membranes). The experimental results show that the rejections of more hydrophobic solutes are slightly lower, which confirms the findings from other studies;<sup>17–20</sup> however, generally, the rejections increase with molecular size, as assumed by the log-normal model. Solutes that are not or are hardly retained by a single membrane passage (e.g., aniline and *p*-tolualdehyde), have a low rejection with three membrane passages as well. However, even for solutes with a low to average rejection (e.g., methyl ethyl ketone, 3-methyl-4-nitrophenol), there is a large increase in rejection. Solutes that are well-retained by a single membrane (>80%) are almost completely removed after three membrane passages. The calculated rejection curves using the log-normal approach are given in Figure 3, for comparison with Figure 2. The (overall) MWC and  $S_{MW}$  are given in Table 5. It can be seen that the MWC values, in particular, show a dramatic decrease.

However, note that the overall permeate recovery with three membrane passages and no recycle is much lower than that in the system with a membrane stack (assuming a permeate recovery of 80% in a single module, the permeate recovery in the system with three membrane passages is only 51%). In addition, the energy requirements for this system are much higher (~3 times higher than that in a single passage, compared to the factor of 1.6–2.1 previously observed).

#### Comparison: Filtration in Series with Retentate Recycle.

An alternative process design with retentate recycle is shown in Figure 4. The general idea is that solutes that are well-retained in a single step are present in very low concentrations in the retentate of the second step; therefore, recycling decreases the feed concentration in the first step, which has a positive influence on the separation. However, recycling does not change the performance for solutes that are not well-retained, because the concentrations in each individual stream are practically the same.

**Figure 4.** Design of a filtration in series with recycle, using two and three membrane passages.

The mass balances for the system with three membrane passages are given as follows (in terms of the retentate in module  $n$  ( $R_n$ ), the permeate in module  $n$  ( $P_n$ ), and the feed  $F$ ):

$$F + R_2 = R_1 + P_1 \quad (4)$$

$$P_1 + R_3 = R_2 + P_2 \quad (5)$$

$$P_2 = P_3 + R_3 \quad (6)$$

$$\text{overall recovery} = \frac{P_3}{F} \quad (7)$$

$$\text{recovery in step 1} = \frac{P_1}{F + R_2} \quad (8)$$

$$\text{recovery in step 2} = \frac{P_2}{R_3 + P_1} \quad (9)$$

where  $P$  refers to the permeate stream in the different filtration steps and  $R$  refers to the retentate stream in the different filtration steps. Again,  $F$  is the feed stream. The overall rejection was calculated as  $\text{Rej}_{\text{overall}} (\%) = (1 - c_{p,3}/c_{f,1}) \times 100$ , where  $c_{p,3}$  and  $c_{f,1}$  were solved from

$$c_{f,1}F + c_{r,2}R_2 = c_{r,1}R_1 + c_{p,1}P_1 \quad (10)$$

$$c_{p,1}P_1 + c_{r,3}R_3 = c_{r,2}R_2 + c_{p,2}P_2 \quad (11)$$

$$c_{p,2}P_2 = c_{p,3}P_3 + c_{r,3}R_3 \quad (12)$$

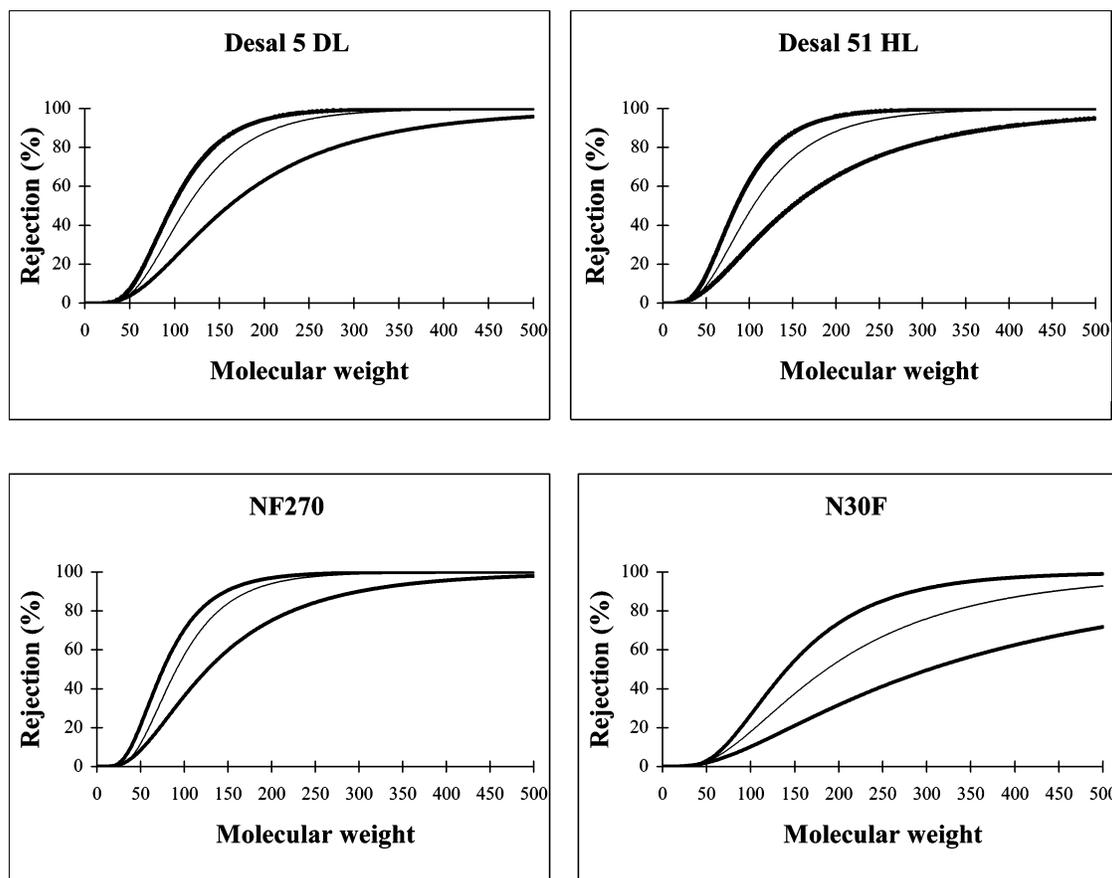
$$\text{Rej}_{\text{one step}} = 1 - \frac{c_{p,3}}{c_{p,2}} \quad (13)$$

$$\text{Rej}_{\text{one step}} = 1 - \frac{c_{p,2}}{(c_{r,3}R_3 + c_{p,1}P_1)/(R_3 + P_1)} \quad (14)$$

$$\text{Rej}_{\text{one step}} = 1 - \frac{c_{p,1}}{(c_{f,1}F_1 + c_{r,2}R_2)/(F_1 + R_2)} \quad (15)$$

where  $c_{p,n}$  is the concentration of the permeate in module  $n$ ,  $c_{r,n}$  is the concentration of the retentate in module  $n$ , and  $c_{f,n}$  is the concentration of the feed to module  $n$ .

Using a recovery of 85% in the first step, a recovery of 90% in the second step, and an overall recovery of 80%, rejections were calculated based on the experimental observations in one step. The resulting rejection curves are given in Figure 5. The shape of the curves indicates that is more similar to a step



**Figure 5.** Rejection curves calculated with the log-normal model using a multiple membrane passage with recycle of the retentate. In each figure, the curve starting on the right-hand side is for a single membrane passage, the middle curve is for two membrane passages in series, and the curve starting on the left-hand side is for three membrane passages in series.

function. This is confirmed by an analysis of the model parameters (see Table 6). The MWC is still as good as in the case without recycle streams; the more membrane passages are used, the lower the MWC. The  $S_{MW}$  factor is lower with a recycle stream, which confirms that the rejection curve is steeper in the case when no recycle stream was used.

Note that the recoveries in the different filtration steps were not optimized, so that the results could be improved even more.

## Conclusions

The use of a membrane stack, although successful for ultrafiltration in dilute solutions, was ineffective to improve the separation efficiency in nanofiltration, possibly because of the combination of high concentrations and an uncontrolled flow pattern in a semi-steady-state operation. It was shown that a filtration in series helps to obtain a steplike function, although the water recovery (permeate yield) is (too) low in this configuration. Using an intelligent design with a recycle of retentate streams, the overall MWC remains the same but the rejection curve is more similar to a step function than in the case when no retentate was recycled, with a permeate recovery identical to a single membrane operation.

A further envisaged optimization is the selection of a proper membrane that would allow to shift the MWC toward a value suitable for a given separation, which would make efficient separations possible between solutes with a relatively small difference in molecular weight and, possibly, between organic solutes and ions.

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