

PROSIMPLUS APPLICATION EXAMPLE

PERVAPORATION

EXAMPLE PURPOSE

This example illustrates two pervaporation processes: the first case considers solvent pervaporation using a spiral-wound membrane module, while the second case describes ethanol dehydration using a plate-and-frame membrane module.

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CORRESPONDING PROSIMPLUS FILE	<i>PSPS_EX_EN-Solvent-Pervaporation.pmp3</i> <i>PSPS_EX_EN-Ethanol-Pervaporation.pmp3</i>
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Energy

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1. PROCESS MODELING

1.1. Process description

Pervaporation (PV) is a membrane-based separation process used for the fractionation of liquid mixtures, particularly when conventional distillation becomes inefficient or impractical. It is especially suitable for azeotropic systems, close-boiling mixtures, the removal of species present at low concentrations, and thermally sensitive compounds [ONG16].

In pervaporation, the liquid feed mixture enters the PV unit and the permeating compound passes through a dense membrane [LUI18]. Depending on the separation objective, the membrane material can be lyophilic or lyophobic. The process may operate in a single stage or multi-stage and the retentate may be recycled to the PV unit to enhance separation efficiency if required.

Transport in pervaporation is governed by the solution-diffusion mechanism, which describes mass transfer through dense membranes. The driving force for separation is a chemical potential difference between the feed and permeate sides, commonly expressed as a difference in partial vapor pressure of the permeating species. This driving force is typically established by applying a vacuum or introducing a sweep gas on the permeate side, thereby reducing the permeate-side partial pressure of the transferred compounds [LEE89].

The solution-diffusion mechanism consists of three successive steps:

1. Sorption (dissolution) of components from the liquid feed into the membrane material,
2. Diffusion through the membrane matrix driven by the chemical potential difference,
3. Desorption from the membrane into the permeate side, where the components emerge in the vapor phase.

The permeating species exit the membrane in the vapor phase and are typically condensed downstream. The following figure illustrates the pervaporation process and the associated transport mechanisms.

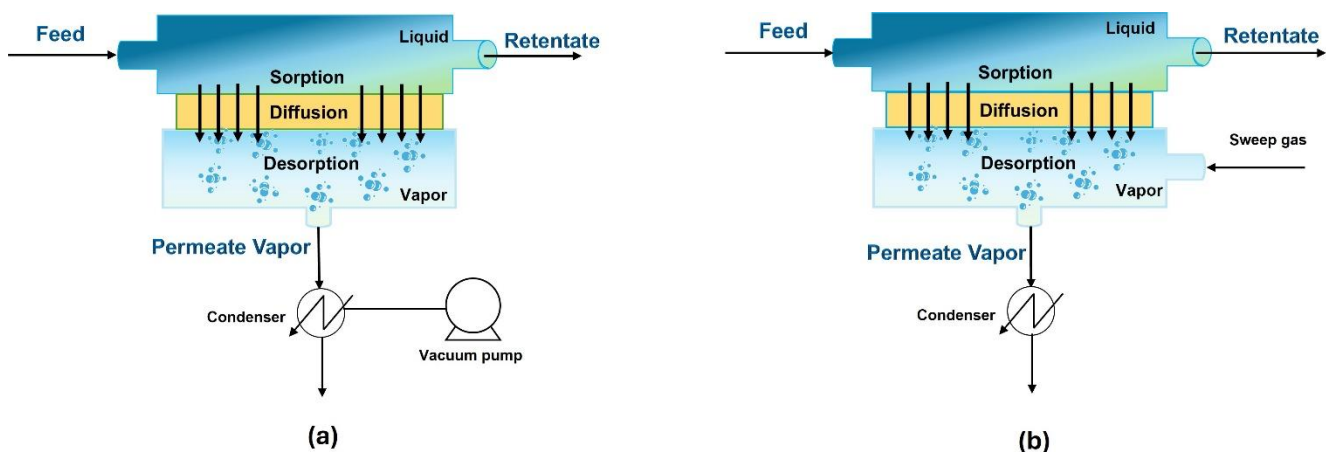


Figure 1. Schematic of the cross-flow pervaporation process. The liquid feed first dissolves into the membrane surface, then diffuses through the membrane, and finally desorbs as vapor on the permeate side under (a) vacuum or (b) sweep gas conditions.

1.2. Applications of pervaporation

The application of pervaporation depends mainly on the separation target and the required selectivity. In industrial practice, the most common use cases are:

- Solvent dehydration and azeotrope breaking: this is the most established industrial application. Typical examples include ethanol dehydration and drying of alcohol or other polar solvents [BOL11], [SOM05], [URT07]. In many plants, the membrane unit is installed downstream of a distillation column to reach low residual water content.
- Recovery of organics from aqueous streams: applied for selective recovery of valuable organics from dilute feeds, such as fermentation products (e.g., butanol, ABE¹ mixtures) and aroma/flavor compounds [CAI16].
- VOC removal and solvent recovery: Suitable for treating streams containing dilute volatile organic compounds (VOCs) [BAC15].
- Organic-organic separations: applied to systems that are difficult to separate by conventional methods, such as ethanol/ETBE and methanol/MTBE mixtures, as well as selected petrochemical applications including removal of sulfur-containing compounds [MIT12], [JAI18], [MIS21].
- Desalination of high-salinity water: Pervaporation can be used to produce fresh water from saline streams [WU18]. Water is transferred in vapor form through a hydrophobic membrane, while dissolved salts and other non-volatile species are retained in the retentate side.

Pervaporation is often applied as a downstream separation unit or combined with distillation in hybrid configurations to achieve the required product purity.

In this work, two representative examples of pervaporation applications are considered:

- Solvent dehydration using a spiral-wound membrane module [DIM19]
- Ethanol dehydration using a plate-and-frame membrane module [YAV19]

In pervaporation processes, membrane permeance cannot be considered a constant parameter, as it changes during operation due to variations in feed composition and temperature. In this simulation, a correlation based on an Arrhenius-type equation is proposed to describe permeance as a function of both composition and temperature simultaneously. Case studies were then carried out to evaluate membrane performance by varying the initial feed water concentration, membrane surface area and feed temperature in order to identify suitable operating conditions and assess their impact on separation performance.

¹ Acetone-Butanol-Butanol solvent mixture produced via fermentation

2. SOLVENT PERVAPORATION

2.1. Process flowsheet

The process flowsheet of the pervaporation unit is presented in Figure 2. The hydrated solvent feed enters the PV unit, where water selectively permeates through the hydrophilic membrane and is collected on the permeate side, while the dehydrated solvent exits the unit as the retentate stream.

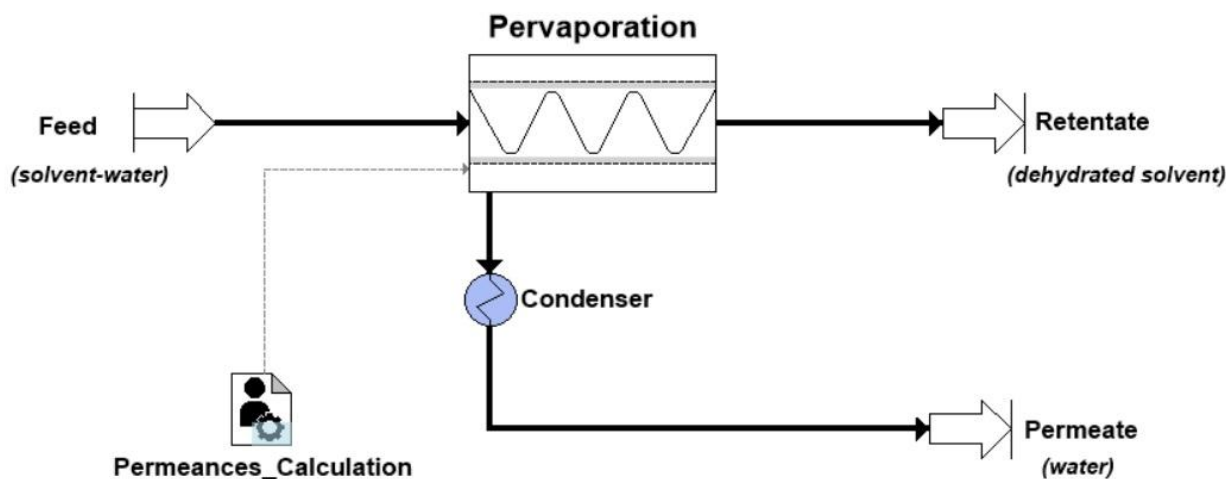


Figure 2. Simulation flowsheet of solvent pervaporation unit

2.2. Components

The compounds used in the simulation are listed below:

Component	Chemical formula	CAS number
Dipropylene glycol monomethyl ether (DPM)	$C_7H_{16}O_3$	34590-94-8
Dipropylene glycol n-butyl ether (DPnB)	$C_{10}H_{22}O_3$	29911-28-2
Propylene glycol n-butyl ether (PnB)	$C_7H_{16}O_2$	5131-66-8
Water	H_2O	7732-18-5

(*)CAS Registry Numbers® are the intellectual property of the American Chemical Society and are used by ProSim SA with the express permission of ACS. CAS Registry Numbers® have not been verified by ACS and may be inaccurate

2.3. Thermodynamic model

The PSRK (Predictive Soave-Redlich-Kwong) thermodynamic model was selected for this simulation to describe phase equilibrium in the system. This model combines the Soave-Redlich-Kwong equation of state with UNIFAC group contribution methods, enabling reliable representation of non-ideal liquid mixtures.

2.4. Operating conditions

- **Process feed**

		Hydrated solvent
Mass fraction	Water	0.1
	DPM	0.54
	DPnB	0.27
	PnB	0.09
Mass flowrate (kg/h)		100
Temperature (°C)		70
Pressure (atm)		1

- **Spiral wound membrane filter**

The spiral-wound membrane module operates in cross-flow mode, which is commonly used to mitigate concentration polarization by maintaining sufficient feed velocity along the membrane surface. The unit operation temperature is assumed to be equal to the feed temperature, neglecting temperature drop across the unit. The permeate-side pressure, controlled by a vacuum pump, is set to 10 mbar.

Configuration	
Flow direction	Cross-flow
Length (m)	1
Membrane area (m ²)	25
General	
Temperate of retentate side increment ($T_{ret} + \Delta T$)	0
Permeate side pressure (mbar)	10

Permeation		
Definition type		Permeabilities of solvent and a reference compound (selectivities)
Water permeability type		Constant molar permeance
Water permeance (kmol.m ⁻² .h ⁻¹ .atm ⁻¹)		0.2
Selectivities	DPM	0
	DPnB	0
	PnB	0

Default parameters are used for other parameters.

The permeance of water through the PERVAP 4510 membrane is reported by Dimitrov [DIM19]. The organic solvents are completely retained by the membrane [DIM19], while only water is in the permeate. In this example, water is considered as both the solvent and the reference component, and only water is allowed to permeate through the membrane, while the organic solvent is assumed to be completely retained. Therefore, the selectivity of the organic solvent relative to water is set to zero. In this work, the water permeance is described using a correlation that accounts for both temperature and feed composition, as discussed in the following section.

- **Windows script: Permeances_Calculation**

In the windows script, experimental flux data were converted to molar permeance using the solution-diffusion model before implementing the correlation in the "Permeances_Calculation" script.

The influence of operating temperature on pervaporation transport is commonly described by an Arrhenius-type relationship [FAR23]:

$$J = J_0 \exp \left(- \frac{E_i}{RT} \right) \quad (1)$$

Where

J : Permeate flux (kmol.m⁻².h⁻¹)

J_0 : Pre-exponential factor

E_i : Activation energy (J.mol⁻¹)

R : Gas constant (J.mol⁻¹.K⁻¹)

T : Temperature (K)

J_0 and E_i depend on the composition of water.

The molar permeance obtained from experimental flux data was substituted into this relationship to derive a permeance correlation as a function of temperature and feed water mass fraction:

$$\mathcal{P}(T, x) = \exp \left(A(x) - \frac{E(x)}{RT} \right) \quad (2)$$

Where

$\mathcal{P}$: Molar permeance ($\text{kmol.m}^{-2}.\text{h}^{-1}.\text{atm}^{-1}$)

x : Water mass fraction in the feed

The functions $A(x)$ and $E(x)$ are composition-dependent parameters that can be represented using linear or higher-order polynomial expressions depending on the system. For implementation in the windows script model, the correlation was expressed in the following polynomial form:

$$\mathcal{P} = \exp(c_0 + c_1 x + c_2 x^2 + c_3 x^3 + c_4 \frac{1}{T} + c_5 \frac{x}{T^2} + c_6 \frac{x^2}{T} + c_7 \frac{x^3}{T}) \quad (3)$$

c_0 - c_7 : Regression coefficients obtained using multiple linear regression

Within the "Permeances_Calculation" script, the feed water mass fraction and temperature are retrieved from the input stream of the pervaporation unit. The corresponding water molar permeance is then calculated using the above correlation and assigned to the pervaporation module as the specified molar permeance for water.

The correlation is implemented in the "Permeances_Calculation" script as follows:

```
Function OnCalculation()
  Name = "Pervaporation"
  Ind_Water = 0 'Position of water in the list of compounds

  Molar_Fractions = Project.Modules(Name).InputStream(1).MolarFractions
  Xmol_water = Molar_Fractions(Ind_Water)
  Mass_Fractions = Project.Modules(Name).InputStream(1).MassFractions
  Xmas_Water = Mass_Fractions(Ind_Water)
  T = Project.Modules(Name).InputStream(1).Temperature ' in Kelvin

  '-----
  ' Molar permeance of water(kmol.m-2.h-1.atm-1) as a function of temperature and feed water composition
  '-----

  c0 = -14.52326925
  c1 = 148.7330231
  c2 = 1725.473392
  c3 = -19027.27445
  c4 = 3603.041836
  c5 = -60217.76126
  c6 = -228766.2323
  c7 = 4801338.658

  WaterMolarPermeance = Exp (c0 + c1 * Xmas_Water + c2 * Xmas_Water ^2 + c3 * Xmas_Water^3 + c4 * 1/T
+ c5 * Xmas_Water/T + c6 * Xmas_Water^2/T + c7 * Xmas_Water^3/T)

  Module.Parameter(1) = Xmas_Water
  Module.Parameter(2) = Xmol_water
  Module.Parameter(3) = WaterMolarPermeance
  Module.Parameter(4) = T

  Project.Modules("Pervaporation").MolarPermeancesSpecified(Ind_Water+1) = WaterMolarPermeance

  'Error flag (if "True": no error reported)
  OnCalculation = True
End Function
```

- **Cooler/Heater: Condenser**

The outlet temperature of the condenser is set at the bubble-point temperature to ensure complete condensation of the vapor stream.

Name	Condenser
Constraint	Bubble-point temperature

3. RESULTS AND CONCLUSIONS

3.1. Mass and Energy Balance

		Feed	Retentate	Permeate	Condensed permeate
Temperature (°C)		70	70	70	7
Pressure (atm)		1	1	9.87×10^{-3}	9.87×10^{-3}
Mass flowrate (kg/h)		100	90.24	9.76	9.76
Mass fraction	Water	0.1	0.003	1	1
	DPM	0.54	0.598	0	0
	DPnB	0.27	0.299	0	0
	PnB	0.09	0.100	0	0

3.2. Process Performance

Stage cut (%)	9.76
Water permeance ($\text{kmol.m}^{-2}.\text{h}^{-1}.\text{atm}^{-1}$)	0.318

- Recovery ratios of solvent and water

	Recovery ratio of solvent and water (%)	
	Retentate	Permeate
Water	2.42	97.58
DPM	100	0
DPnB	100	0
PnB	100	0

3.3. Profiles

There are several operating parameters that contribute to the performance of pervaporation. These are discussed in the following sections.

3.3.1. Initial water concentration in the feed

Figure 3a presents the variation of water permeance with the initial water mass fraction in the feed at 70 °C. As the water content increases, the permeance through the membrane increases significantly. This behavior is attributed to the stronger affinity of water for the membrane material, which enhances its sorption and transport through the membrane phase. In addition, the presence of water may induce partial membrane swelling, further facilitating mass transfer.

The simulation results obtained with ProSimPlus reproduce this trend well, with predicted permeance values in good agreement with the experimental data. The same trend is reflected in Figure 3b, where the stage cut increases with increasing water content, indicating that a larger fraction of the feed is transferred to the permeate side under these conditions.

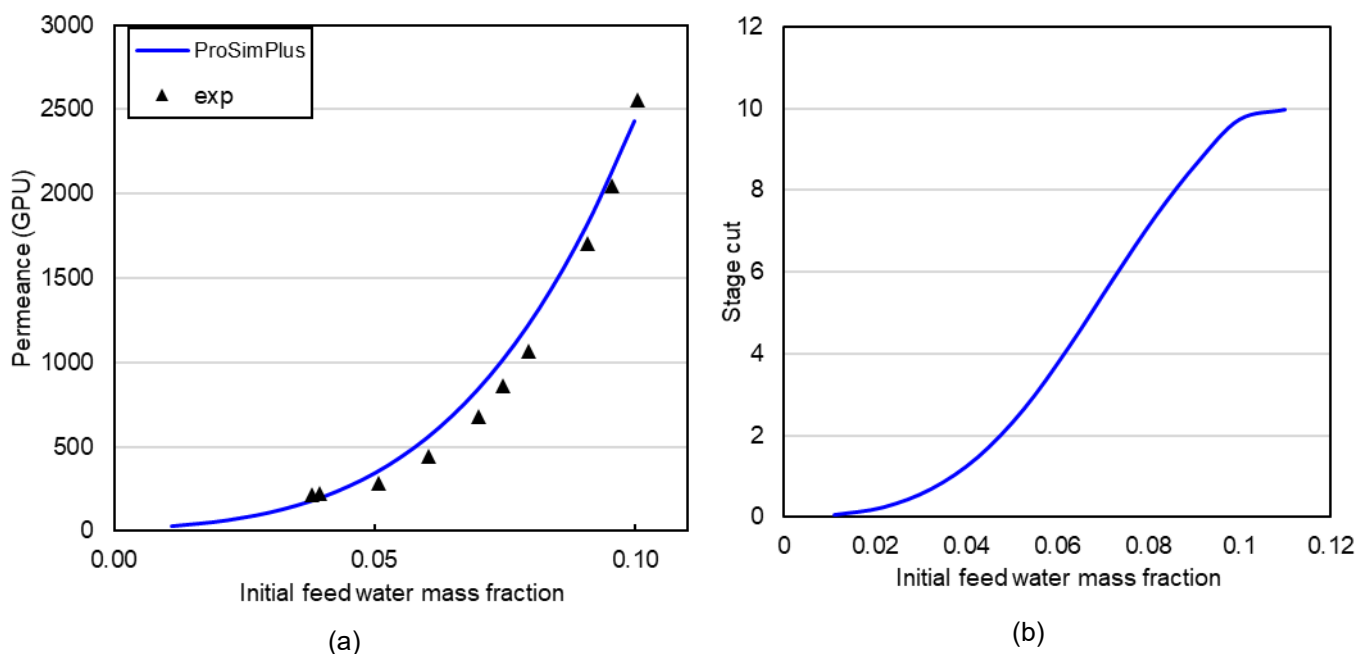


Figure 3. Effect of initial feed water mass fraction at 70 °C on (a) water permeance and (b) stage cut. ▲: experimental data ([DIM19]); solid line: ProSimPlus prediction.

3.3.2. Membrane surface area

Figure 4a shows the dependence of the final mass fraction of the components in the dehydrated solvent (retentate) on the membrane surface area. The results were obtained from a steady-state simulation at a feed temperature of 70 °C and an initial water content of 10 wt% in the feed mixture.

Based on the required purity of the dehydrated solvent, the membrane surface area needed for a single-stage pervaporation unit can be determined. For small membrane areas, the available transfer surface is limited and only partial dehydration occurs. As the membrane area increases, the overall mass transfer capacity increases, allowing

water to permeate selectively through the membrane and significantly reducing the water concentration in the retentate. Consequently, the stage cut increases, as illustrated in *Figure 4b*, until it approaches a plateau.

However, beyond approximately 20 m², the curves become nearly constant and no significant improvement in solvent purity or stage cut is observed. This indicates that further increasing the membrane surface area does not substantially enhance dehydration performance under the selected operating conditions and would mainly result in higher capital costs.

These results highlight the importance of selecting an optimal membrane surface area during process design and scale-up to achieve the desired product specification.

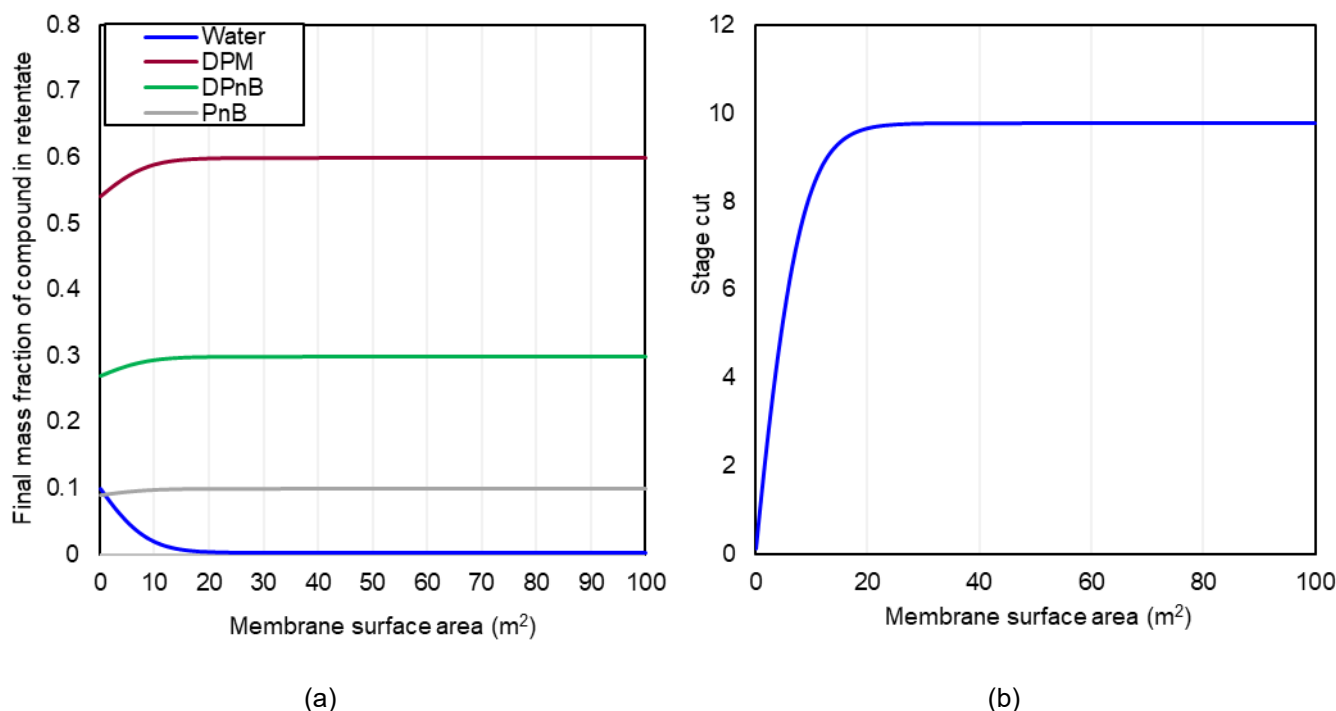


Figure 4. Effect of membrane surface area on (a) the final purity of the dehydrated solvent in the retentate and (b) the stage cut, at a feed temperature of 70 °C and an initial water content of 10 wt%.

3.3.3. Feed temperature

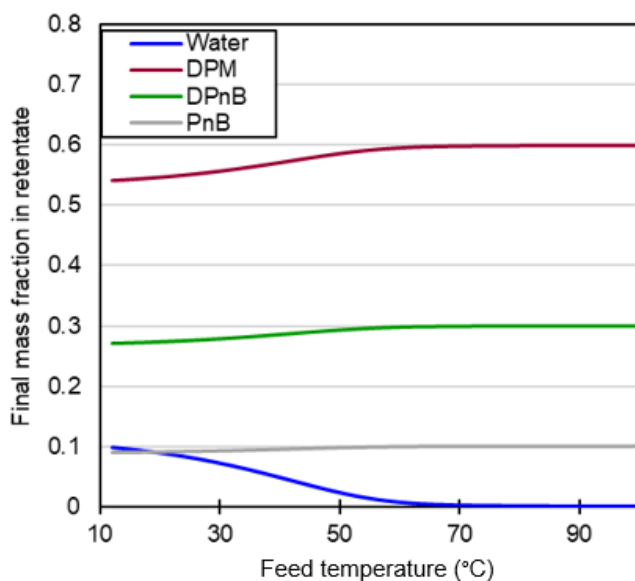
Figure 5 presents the influence of feed temperature on retentate composition, stream flowrates, water flux, and condenser duty. As shown in

Figure 5a and 5b, increasing the feed temperature improves the dehydration performance, resulting in a lower water content in the retentate stream and a higher permeate flux. The main reason for the increase in permeate flux is the increase in thermodynamic driving force. In pervaporation, mass transport is governed by the vapor pressure difference between the feed and permeate sides. Increasing the feed temperature raises the vapor pressure of water in the liquid mixture, thereby enhancing the driving force for permeation.

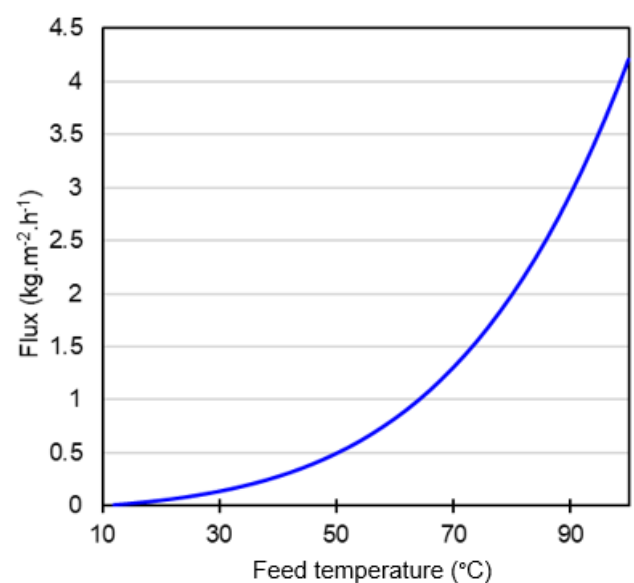
At low temperatures, pervaporation may become inefficient because the vapor pressure difference across the membrane is relatively small, resulting in limited permeation. This behavior is also reflected in

Figure 5c, where the permeate flowrate increases while the retentate flowrate decreases with increasing temperature. Around 50 °C, the process begins to operate more effectively, while temperatures close to 70 °C appear to provide near-optimal operating conditions. Beyond this point, the membrane performance approaches a plateau and further increases in temperature provide only limited improvement in separation performance.

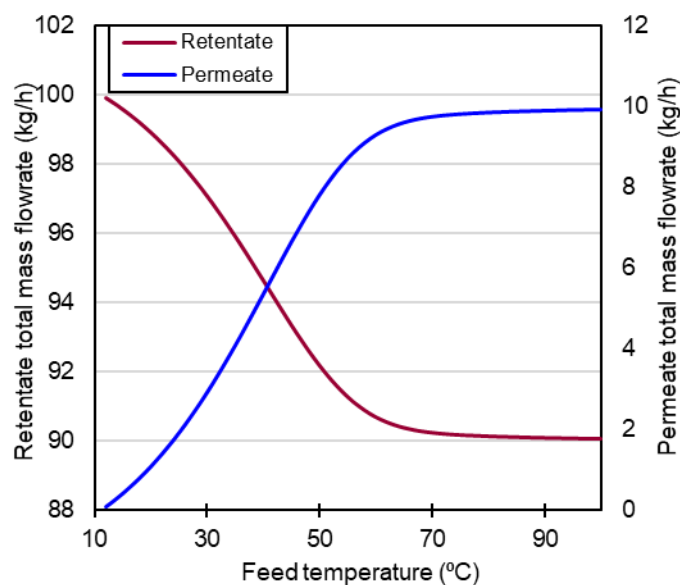
Figure 5d shows the condenser duty required to cool and condense the permeate stream at different feed temperatures. As expected, higher feed temperatures lead to a higher condenser duty because a larger amount of vapor is generated on the permeate side. Therefore, the feed temperature must be carefully selected during process design, since it affects both separation efficiency and the overall energy requirement of the pervaporation process.



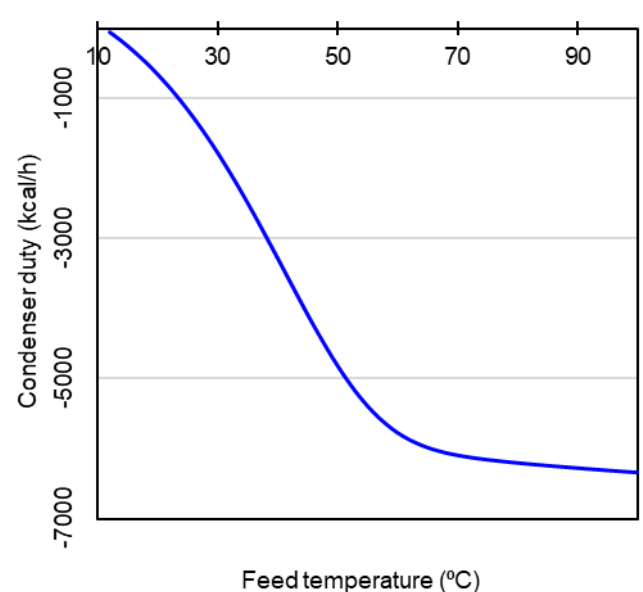
(a)



(b)



(c)



(d)

Figure 5. Effect of feed temperature on (a) final composition in the retentate, (b) water flux, (c) permeate and retentate flowrates, and (d) condenser duty. The initial feed water concentration is 10 wt%.

4. ETHANOL PERVAPORATION

4.1. Process flowsheet

The process flowsheet of the ethanol pervaporation unit is presented in Figure 6. Hydrated ethanol, containing 10% water, enters the PV unit. Due to the azeotropic behavior of the ethanol-water system, this water cannot be separated by conventional distillation. In the pervaporation unit, water selectively permeates through the hydrophilic PERVAP 4101 membrane and passes to the permeate side in the vapor phase, where it is subsequently condensed. Water with a purity of approximately 99% is collected on the permeate side. On the other hand, the dehydrated ethanol exits the unit as the retentate stream.

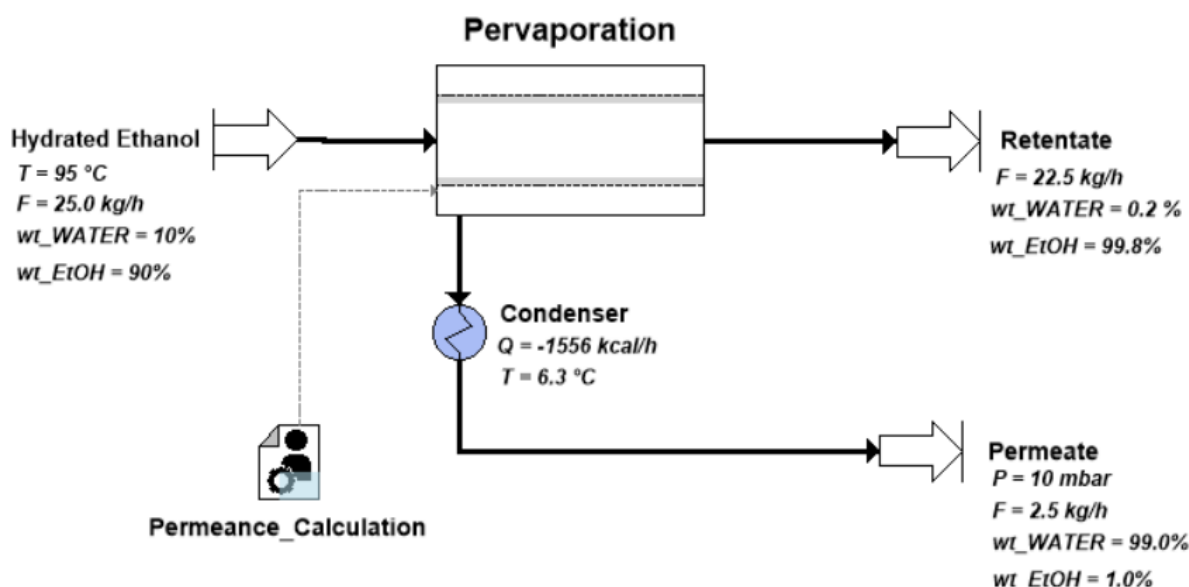


Figure 6. Simulation flowsheet of ethanol dehydration unit

4.2. Components

The compounds used in the ethanol dehydration simulation are listed below.

Component	Chemical formula	CAS number
Water	H ₂ O	7732-18-5
Ethanol	C ₂ H ₆ O	64-17-5

(*)CAS Registry Numbers® are the intellectual property of the American Chemical Society and are used by ProSim SA with the express permission of ACS. CAS Registry Numbers® have not been verified by ACS and may be inaccurate

4.3. Thermodynamic model

The NRTL model was selected to describe the vapor-liquid equilibrium of the water-ethanol system.

4.4. Operating conditions

- **Process feed**

		Hydrated solvent
Mass fraction	Water	0.1
	Ethanol	0.9
Mass flowrate (kg/h)		25
Temperature (°C)		95
Pressure (atm)		2

- **Plate and frame membrane filter**

The plate and frame membrane module operates in cross-flow mode. The unit operation temperature is assumed to be equal to the feed temperature, neglecting temperature drop across the unit. The permeate-side pressure, controlled by a vacuum pump, is set to 10 mbar.

Configuration	
Flow direction	Cross-flow
Length (m)	1
Membrane area (m ²)	10
General	
Temperate of retentate side increment (Tret + ΔT)	0
Permeate side pressure (mbar)	10
Permeation	
Definition type	Permeability of a reference compound and selectivities
Water permeability type	Constant molar permeance
Water permeance (GPU)	2000
Selectivity of ethanol/water	2×10^{-4}

Default parameters are used for other parameters.

- **Permeation**

The permeance of water through the PERVAP 4101 membrane at different feed water compositions is reported in [YAV19]. The ethanol is partially retained by the membrane, while water is in the permeate. In this example, water is considered as both the solvent and the reference component. Therefore, the selectivity of the ethanol relative to water is set to 2×10^{-4} . In this work, the water permeance is described using a correlation that accounts for both temperature and feed composition, as discussed in the following section.

- **Windows script: Permeances_Calculation**

In the windows script, the experimental flux data were first converted to molar permeance using the solution-diffusion model before implementing the correlation in the “Permeances_Calculation” script. The molar permeance obtained from experimental flux data was substituted into Equation (2) to derive a permeance correlation as a function of temperature and feed water mass fraction.

In this example, the functions $A(x)$ and $E(x)$ are composition-dependent parameters that are assumed to be linear functions of composition. Due to the linearity of the correlation for ethanol pervaporation, the regression coefficients in Equation (3) corresponding to the second and third-order terms are equal to zero.

Within the “Permeances_Calculation” script, the feed water mass fraction and temperature (K) are retrieved from the input stream of the pervaporation unit. The corresponding water molar permeance ($\text{kmo.m}^{-2}.\text{h}^{-1}.\text{atm}^{-1}$) is then calculated using the above correlation and assigned to the pervaporation module as the specified molar permeance for water.

The correlation is implemented in the "Permeances_Calculation" script as follows:

```
Function OnCalculation()
  Name = "Pervaporation"
  Ind_Water = 0 'Position of water in the list of compounds

  Molar_Fractions = Project.Modules(Name).InputStream(1).MolarFractions
  Xmol_water = Molar_Fractions(Ind_Water)
  Mass_Fractions = Project.Modules(Name).InputStream(1).MassFractions
  Xmas_Water = Mass_Fractions(Ind_Water)
  T = Project.Modules(Name).InputStream(1).Temperature ' in Kelvin

  '-----
  ' Molar permeance of water(kmol.m-2.h-1.atm-1) as a function of temperature and feed water composition
  '-----

  c0 = 0.041539414
  c1 = 39.6545137
  c2 = 0
  c3 = 0
  c4 = -907.9004284
  c5 = -12556.59855
  c6 = 0
  c7 = 0

  WaterMolarPermeance = Exp (c0 + c1 * Xmas_Water + c2 * Xmas_Water^2 + c3 * Xmas_Water^3 + c4 * 1/T +
c5 * Xmas_Water/T + c6 * Xmas_Water^2/T + c7 * Xmas_Water^3/T)

  Module.Parameter(1) = Xmas_Water
  Module.Parameter(2) = Xmol_water
  Module.Parameter(3) = WaterMolarPermeance
  Module.Parameter(4) = T

  Project.Modules("Pervaporation").MolarPermeancesSpecified(Ind_Water+1) = WaterMolarPermeance

  'Error flag (if "True": no error reported)
  OnCalculation = True
End Function
```

- **Cooler/Heater : Condenser**

The outlet temperature of the condenser is set at the bubble-point temperature to ensure complete condensation of the vapor stream.

Name	Condenser
Constraint	Bubble-point temperature

5. RESULTS AND CONCLUSIONS

5.1. Mass and Energy Balance

		Feed	Retentate	Permeate	Condensed permeate
Temperature (°C)		95	95	95	6.3
Pressure (atm)		2	2	9.87×10^{-3}	9.87×10^{-3}
Mass flowrate (kg/h)		25	22.5	2.5	2.5
Mass fraction	Water	0.1	0.002	0.99	0.99
	Ethanol	0.9	0.998	0.010	0.01

5.2. Process Performance

Stage cut (%)	9.9
Water permeance ($\text{kmol.m}^{-2}.\text{h}^{-1}.\text{atm}^{-1}$)	0.154

- Recovery ratios of ethanol and water

	Recovery ratio of solvent and water (%)	
	Retentate	Permeate
Water	1.86	98.15
Ethanol	99.89	0.11

5.3. Profiles

There are several operating parameters that contribute to the performance of pervaporation. These are discussed in the following sections.

5.3.1. Initial water concentration in the feed

Figure 7a presents the variation of water permeance with the initial water mass fraction in the feed at 95 °C. As the water content increases, the permeance through the membrane increases significantly. The ProSimPlus simulation reproduces this trend well, showing good agreement with the experimental data.

Figure 7b shows that, as the initial water concentration in the feed increases, the water fraction in the permeate rises initially and then reaches an almost constant value, while the ethanol fraction correspondingly decreases to a very low level.

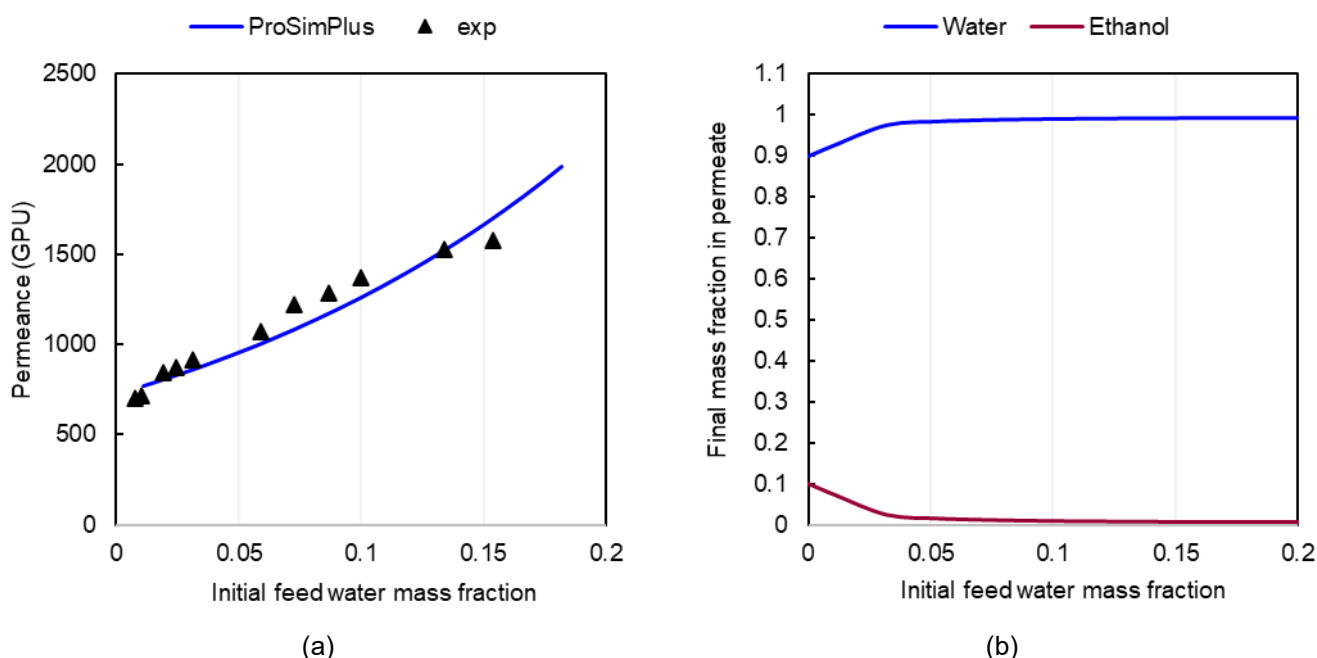


Figure 7. Effect of initial feed water mass fraction at 95 °C on (a) water permeance and (b) final purity of water in the permeate

▲ : experimental data ([YAV19]); solid line: ProSimPlus prediction.

5.3.2. Membrane surface area

Figure 8a shows the dependence of the final mass fraction of the components in the dehydrated ethanol (retentate) on the membrane surface area. The results were obtained from a steady-state simulation at a feed temperature of 95 °C and an initial water content of 10 wt% in the feed mixture.

Based on the required purity of the dehydrated ethanol, the membrane surface area needed for a single-stage pervaporation unit can be determined. For small membrane areas, the available transfer surface is limited and only partial dehydration occurs. However, considering the feed flowrate in this case, even a membrane area of 5 m² for a plate-and-frame module is sufficient. Increasing the membrane area beyond this value does not significantly improve

the stage cut (Figure 8b) or the purity of the dehydrated ethanol in the retentate and water in the permeate stream, while it would increase the operational cost.

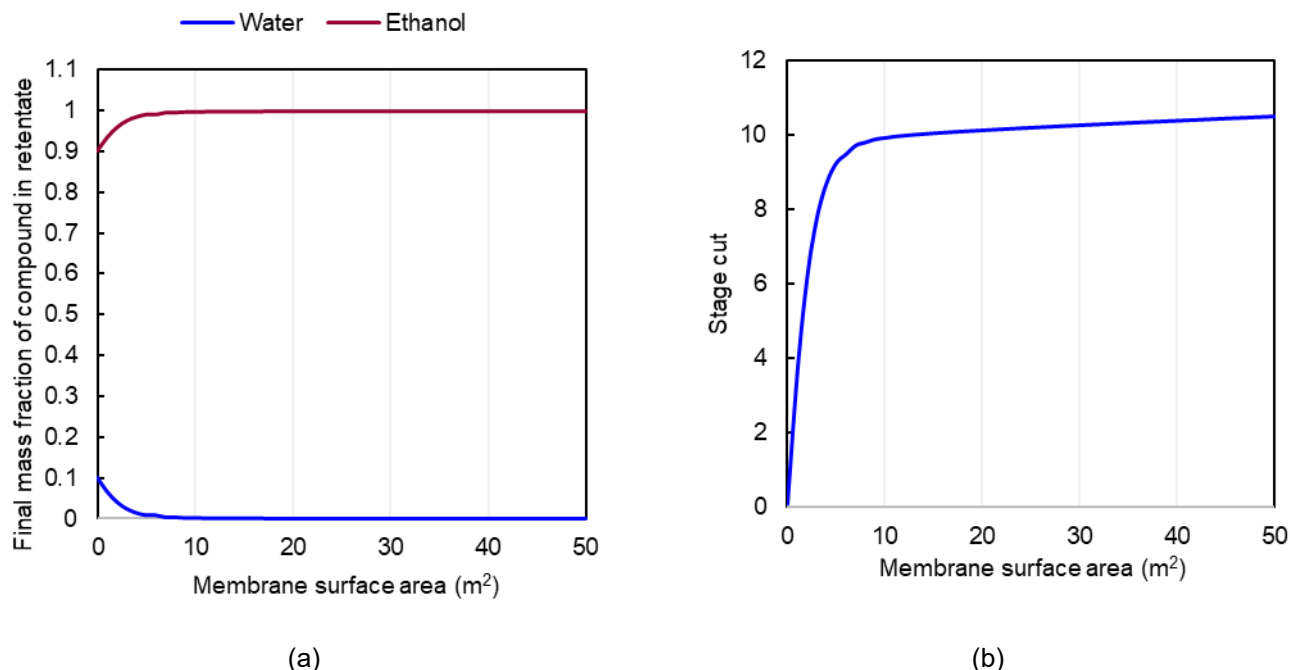


Figure 8. Effect of membrane surface area on (a) the final purity of dehydrated ethanol in the retentate and (b) the stage cut, at a feed temperature of 95 °C and an initial water content of 10 wt%.

5.3.3. Feed temperature

Figure 9 presents the influence of feed temperature on the performance of ethanol dehydration by the pervaporation process. As shown in Figure 9a, increasing the feed temperature improves the dehydration performance, resulting in higher ethanol purity in the retentate and higher water recovery in the permeate. In addition, the permeate flux increases (Figure 9b) due to the increase in the driving force, which is defined in terms of the vapor pressure difference across the membrane. At low temperatures, pervaporation may become inefficient because the vapor pressure difference across the membrane is relatively small, resulting in limited permeation.

Moreover, around 60-70 °C the process begins to operate more effectively, while temperatures close to 80 °C appear to provide near-optimal operating conditions. Beyond this point, the membrane performance approaches a plateau, and further increases in temperature provide only limited improvement in separation performance. This indicates that 80-85 °C is approximately the optimal temperature under these operating conditions.

Figure 9c illustrates the predicted molar permeance of water calculated by ProSimPlus using Equation (3). As expected for a hydrophilic membrane (PERVAP 1401), the molar permeance increases with temperature.

Figure 9d shows the condenser duty required to cool and condense the permeate stream at different feed temperatures. As expected, higher feed temperatures lead to higher condenser duty because a larger amount of vapor is generated on the permeate side.

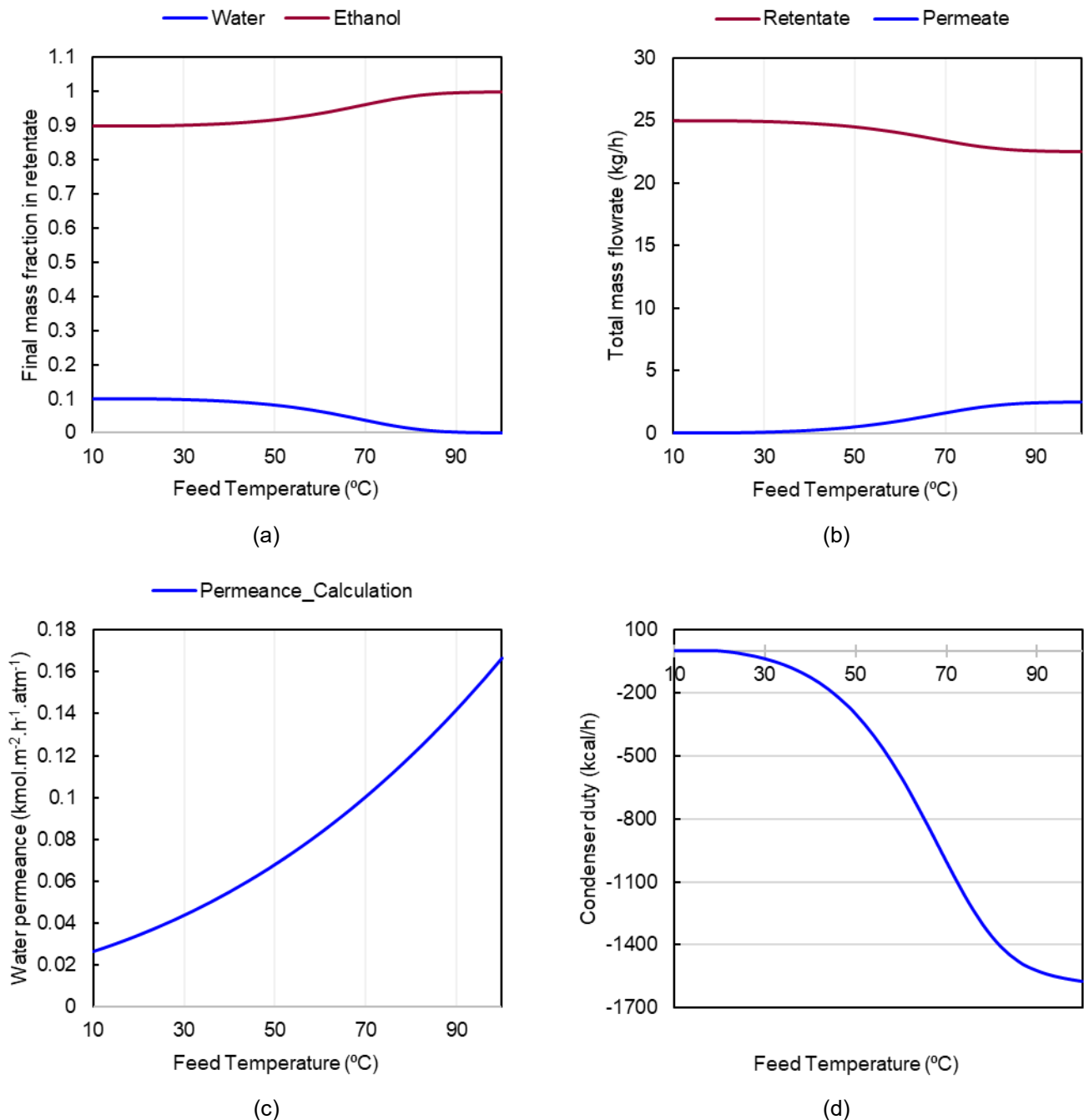


Figure 9. Effect of feed temperature on (a) final composition in the retentate, (b) permeate and retentate total mass flowrates, (c) molar permeance of water, and (d) condenser duty. The initial feed water concentration is 10 wt%.

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