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Performance of Organic Solvent Nanofiltration Membrane for Purification of Omega-3

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Abstract

In this master thesis, the performance of organic solvent nanofiltration membrane for purification of Omega-3 is studied. The aim was to contribute to the improvement of the production process of a Norwegian company producing fish oil by implementing membrane technology to enhance the product quality. The behavior of two types of membranes, polymeric and ceramic, was tested in order to determine which gives an optimal range of rejection of phospholipids and triglycerides contained in the roe extract obtained from the production process.

The nanofiltration separation pressure-driven process was applied, using cross-flow filtration regime at two different pressures of 5 bar and 40 bar. The organic solvent used for defining the membrane performance during the experiment was based on three components, ethanol, water and dry matter from roe extract, in different weight percentages. The membrane rejection was analyzed using the samples taken during the filtration experiments by applying ultra high performance liquid chromatography (UHPLC) technique, as the fraction of material removed from the permeate stream.

The approaches of presenting the results are focused on the rejection percentage, standard deviation and distribution, as well as their interconnection and correlation with other parameters, such as specific flux, time, feed condition. It was observed that DuraMem 200 obtained the highest rejection for each feed type, and the rejection level was decreasing together with the molecular weight cut off (MWCO) of the membrane. The standard deviation of the rejection did not follow any defined trend, and it varied with the weight change of ethanol/ water in the feed. The results analysis showed that the specific flux decreased in time, as well as that the lower flux was related with the higher rejection percentage. By comparing the rejection results for the tests with the two different pressures, it was observed that the increase in pressure which is applied onto the membrane did not affect the level of rejection.

The conclusions based on the obtained results for the membrane performance give directions and contribute to the selection of the right membrane type and operating conditions, which offer the opportunity to transform a batch process into semi-continuous batch process that can be more easily automated and can be more compact and productive than a traditional system used in the abovementioned fish oil production company.

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List of Acronyms

AMTA	–	American Membrane Technology Association
BHT	–	Butylated Hydroxy - Toluene
CM	–	Ceramic Membrane
Da	–	Daltons
DHA	–	Docosahexaenoic Acid
EPA	–	Eicosapentaenoic Acid
EtOH	–	Ethanol
FAs	–	Fatty Acids
FAO	–	Food and Agriculture Organization
HPLC	–	High Performance Liquid Chromatography
LC	–	Long Chain
LMH	–	$L/(m^2 \cdot h)$
MF	–	Microfiltration
MW	–	Molecular Weight
MWCO	–	Molecular Weight Cut Off
NF	–	Nanofiltration
NOM	–	Natural Organic Material
OSN	–	Organic Solvent Nanofiltration
PC	–	Polycarbonates
PE	–	Polyethylene
PFA	–	Perfluoroalkoxy

PLs	–	Phospholipids
PP	–	Polypropylene
PUFAs	–	Polyunsaturated Fatty Acids
PVC	–	Polyvinyl Chloride
PVDF	–	Polyvinylidene Fluoride
RO	–	Reverse Osmosis
SD	–	Standard Deviation
TAC	–	Total Allowable Catch
TAG	–	Triacylglycerol
TGs	–	Triglycerides
THM	–	Trihalomethanes
UF	–	Ultrafiltration
UHPLC	–	Ultra High Performance Liquid Chromatography
UK	–	United Kingdom
US	–	United States

1. Introduction

The fish oil production is important branch in the Norwegian seafood industry, which represents one of Norway's largest export industries after oil and gas (SjømatNorge, n.d.). In 2005, Norway produced 30 000 tones of fish oil, which was around 3% of the world production (Norwegian Seafood Federation, 2010). The fish oil produced from herring roe, intended for human consumption contains the necessary omega-3 fatty acids which could contribute to improvement of human health. Nevertheless, the variability of the herring roe composition limits the possibilities of predicting the nutritional quality of the roe causing its sub-optimized use for different food applications. Tools for predicting the fish or fish roe quality have been limited in order to minimize its deterioration during processing, transport and marketing (Jónsson, Hafsteinsson, Klonowski & Gunnlaugsson, 2007).

The typical fish oil production process includes drying as one of the steps, which can be money and energy consuming. This type of production can have negative impacts on the quality of the product because less valuable material or environmental pollutants are extracted with the product. In order to enhance the fish oil quality membrane nanofiltration technology is applied in this study for omega-3 purification from organic solvent. The organic solvent used is based on ethanol, mixed with water and dry matter. The dry matter is a fish component, which is contained in the emulsion derived from the fish oil production process from herring roe.

Membrane technology for molecular separations in aqueous solutions has been possible since the end of the 20th century. The membranes' filtration technique has been widely used in numerous industrial applications. The main fields of application are: waste water treatment, desalination, and food and beverage industry. The organic solvent nanofiltration (OSN) has emerged during the last decade presenting a new area of membrane science, with potential for application across chemical-related industry sectors (Evonik, n.d., a). The application areas of this filtration technique are found in chemical industries, pharmaceuticals, and processing industries for natural products and oils (Sulzer, n.d.).

Research goals and objectives

The reason for choosing to work on this problem, which involves enrichment of fish oil using membrane technology, was the great challenge for a young scientist and the interest for

laboratory work applicable for real processes, enhanced by the limited data published in this field.

The main goal of this study was to improve the production process of a Norwegian company producing fish oil from immature herring roe using membrane filtration technology. Nanofiltration membranes were selected, in order to obtain highest rejection of phospholipids (PLs) and triglycerides (TGs) and improve process recovery. Both polymeric and ceramic membranes were investigated for their performance in separation of phospholipids in organic solvent.

The objective was to select membranes resistant to organic solvent which would meet the requirements for implementation in a technological process and which will enhance the product quality through obtaining the desired rejection. Indirectly, the process upgrade would have additional benefits on the consumers' health through delivering enriched and quality-increased fish oil.

2. Membrane Filtration

Membranes have the ability to control the permeation rate of a chemical species passing through. In separation applications, which will be used in this master thesis, the aim is to allow one component of a mixture to permeate the membrane freely, while the permeation of other components is hindered (Baker, 2004). Membrane filtration has gained great importance in the industrial technology and is used in a broad range of applications.

2.1. Historical development of membranes

The elements of modern membrane science had been developed in 1960, but membranes were used in small and specialized industrial applications. There were four main problems that prohibited their widespread use as a separation process, which were: unreliable, slow, unselective, and expensive. During the last 30 years, solutions to each of these problems have been found (Baker, 2004). Different manufacture processes were developed for making high performance membranes and nowadays membrane-based separation processes are common to find. By 1980, microfiltration, ultrafiltration, nanofiltration and reverse osmosis were all established processes with application in industries worldwide.

2.2. Types of Membranes

The membrane is defined as a barrier used to separate two phases (Mulder, 1996). The structure of the membrane can be symmetric or asymmetric, homogenous or heterogeneous, solid or liquid. The active layer of the membrane can carry a positive or negative charge quantified by the zeta potential.

Based on their structure synthetic membranes can be divided to isotropic and anisotropic membranes, as shown in Figure 1. The structure and composition throughout isotropic membranes is uniform. These membranes can be dense or porous. The anisotropic, also known as asymmetric membranes are composed of a number of layers with different structures and permeabilities. The interface of the membrane can be molecularly homogeneous or physically or chemically heterogeneous. The homogeneous membrane is completely uniform in composition and structure, and the heterogeneous contains holes or pores of finite dimensions or consists of some form of layered structure (Baker, 2004).

According to the materials used to produce membranes they can be ceramic, polymeric and metal membranes. Membranes are typically made from polymeric materials, which are the

point of interest in this work. Ceramic and metal membranes are also available and they can be either isotropic or anisotropic. They are more costly than other types, but they can withstand very high temperatures contributing to their use in many industrial processes (Furukawa & Burton, 1997).

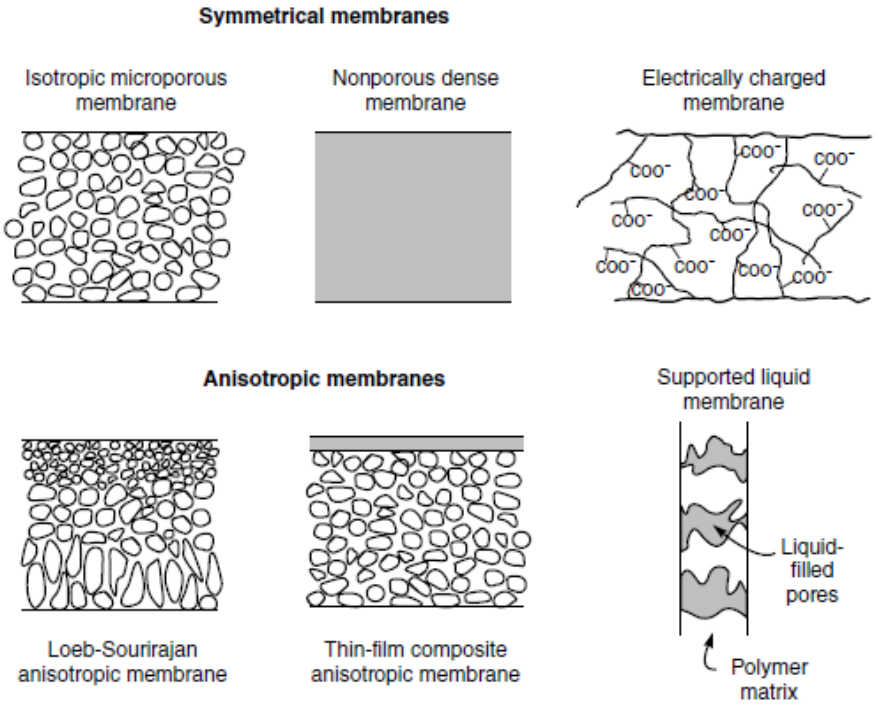


Figure 1: Types of membranes according to the structure (Baker, 2004)

Polymeric and Ceramic Membranes

The increased interests for membranes lead to formation and manufacture of membranes composed of polymer and ceramic materials.

The properties of the polymeric membranes vary depending on the type of polymer used in the manufacture process. According to Singh (2015), polymers are substances formed by linking one or more species of atoms or grouping of atoms by covalent bonds. The chemical and physical properties of the synthesized polymer depend on the method and type of linking the simple molecules that convert into macromolecular structures. Polymer properties depend on several factors such as length and conformation of polymer chain, cross-linking of chains, polar interactions and size and type of attached side groups (Singh, 2015). Modifying polymers can improve membrane selectivity and increase the range of properties important for separations.

Polymers such as polyethylene (PE), polyvinyl chloride (PVC) and polypropylene (PP) are commonly used for membrane manufacturing. The advantage is that they are less costly than fluoropolymers, such as polyvinylidene fluoride (PVDF) and perfluoroalkoxy (PFA), but their performance at high temperature conditions is less satisfactory.

The main type of ceramics currently in use for the manufacturing of filtration membranes consists of refractory oxides: alumina (α-Al_2O_3 and γ-Al_2O_3), zirconia (ZrO_2) or titania (TiO_2), as well as, cordierite, mullite, silicon nitride, silica and borosilicate glasses which are suitable materials for inorganic membrane production (Pabby, Rizvi & Requena, 2015; Soria, 1995). Ceramic membranes have good thermal, mechanical and chemical stability (Mulder, 1996) and the advantage compared to the polymeric membranes is their capacity to withstand harsh operating conditions in terms of pressure, pH and temperature. Ceramic membranes can be operated with liquid or gaseous media and they can be produced with different geometries: flat, tubular, multichannel or monolithic. They are more resistant to cleaning chemicals and have longer lifespan. The typical life of most polymer membranes varies from one to two years for hydrophilic membranes and three to five years for hydrophobic membranes, and ceramic membranes can withstand up to 10 years. However, there are some disadvantages referring to ceramic membranes, such as their brittleness and their cost, which makes them much more expensive than polymeric membranes (Hsieh, 1996).

Ceramic membranes are particularly suitable for biotechnology, food and pharmaceutical applications where repeated steam sterilization is required and their cleaning with aggressive solutions.

2.3. Membrane filtration operation

This section provides background information about types of membrane processes, the filtration regime through the membrane and the flow through porous media. Related formulas for the specific flux and rejection are presented, as well as description of the nanofiltration process.

2.3.1. Flow through porous membrane

The membranes can be described as a series of cylindrical capillary pores of diameter *d* by a simple model of liquid flow passing through the membrane. The liquid flow *(Q)* through a pore is given by Poiseuille's law with the following equation (Baker, 2004):

$$\boldsymbol{Q = \frac{\pi d^4}{128 \mu l} \cdot \Delta p} \qquad \textbf{(1)}$$

Where:

Δp – differential pressure across the pore [kg/m·s^2]

μ - liquid viscosity [kg/s·m] and

l - the pore length [m]

The flux (J) of the membrane is defined as the flow through the membrane (Q) divided by the surface area (A). The related formula is given below:

$$\boldsymbol{J = \frac{Q}{A}} \qquad \textbf{(2)}$$

J - flux through membrane [m/s or L/m^2·h]

Q – flow rate [L/h]

A – membrane area [m^2]

Flux is normalized for pressure by calculating specific flux, which is the flux at a standard temperature provided by the following formula:

$$\boldsymbol{J_{sp} = \frac{J_s}{\Delta p}} \qquad \textbf{(3)}$$

J_{sp} - specific flux at standard temperature [m^2·s/kg ≡ LMH]

J_s - flux at standard temperature [m/s]

Δp – differential pressure across membrane [kg/m·s^2]

The flux divided by the transmembrane pressure has the unit [LMH/bar]. The transmembrane pressure is the differential pressure between the feed and permeate sides of a membrane (Crittenden, Trussell, Hand, Howe & Tchobanoglous, 2012).

The rejection is expressed as the fraction of material removed from the permeate stream, and it is presented using the following formula (Singh, 2015):

$$\boldsymbol{R = \left(1 - \frac{C_p}{C_f}\right) * 100} \quad \textbf{(4)}$$

R – rejection [%]

C_p – permeate concentration [mg/L]

C_f – feed water concentration [mg/L]

2.3.2. Membrane processes

Transport through the membrane takes place when a driving force is applied to the components in the feed. In most common membrane processes the driving force is a pressure difference or a concentration (activity) difference across the membrane (Mulder, 1996). Membrane processes are continuous steady-state operations consisting of three streams: feed, product (permeate) and reject (retentate) as defined by Singh (2015). There are four types of membrane separation processes. According to the order of decreasing permeability they are divided as follows:

- Microfiltration,
- Ultrafiltration,
- Nanofiltration and
- Reverse osmosis.

Microfiltration, ultrafiltration and nanofilration are conceptually similar processes, but the difference in pore diameter produces differences in the way the membranes are used.

These membrane processes are pressure-driven processes, where hydraulic pressure is used to force water molecules through the membranes. Impurities are concentrated in the feed water which after they are retained by the membrane, becomes the reject water or concentrate stream. The water that passes through the membrane is recovered as pure water or product (Furukawa & Burton, 1997).

The relative size, the parameters of each type of pressure-driven membrane filtration is presented in Table 1.

Table 1: General parameters of pressure-driven membrane processes (Munla, 2013)

	MF	UF	NF	RO
Permeability ($l/h.m^2.bar$)	> 1,000	10 – 1000	1.5 – 3.0	0.05 – 1.5
Applied Pressure	0.1 – 2 bar 10 – 100 kPa	0.1 – 5 bar 10 – 500 kPa	3 – 20 bar 300 – 1000 kPa	5 – 120 bar 5000–120000 kPa
Pore size (nm) or MWCO	100 – 10000	2 – 100 > 1000 Da	0.5 – 2 200 – 400 Da	< 0.5 50 – 200 Da
Rejection/ Application	Particle/turbidity, bacteria, algae, protozoa	Small colloids, macromolecule, viruses	Dissolved organic matter, multivalent ions (softening)	Monovalent ions (desalination)
Separation Mechanism	Sieving/size exclusion	Sieving/size exclusion	Sieving, charge effects	Differences in solubility or diffusivity

There are several advantages and disadvantages of membrane separation processes. The advantages include smaller footprint (the membrane equipment requires 90 to 95% less space than conventional plants), reduction of labor requirements due to automation, removal of natural organic matter, inorganic matter, bacteria and viruses. As in every process also in this there are disadvantages to be considered, which are: greater electricity consumption by high pressure systems, pretreatment step for fouling prevention, decline of flux rate with time and variable recovery rate lower (Furukawa & Burton, 1997).

Nanofiltration

NF is closely related to RO, and is sometimes called "loose RO" (Schäfer, Fane & Waite, 2005). The average pore size of NF membranes is 2 nm or less, the driving force is pressure in the range from 5 to 20 bar (Mulder, 1996). The molecular weight cut off of nanofiltration membranes varies between 200 and 500 Da (Mohammad et al., 2015).

NF applications include water softening, removal of multivalent ions from brine solutions, cleaning up of contaminated groundwater, effluents treatment containing oils and heavy metals, color removal from pulp and paper waste water, salt rejection and organics removal at offshore oil platforms, food processing, yeast production, cheese whey production, pharmaceuticals, and removing trace amounts of organic and carcinogenic molecules from drinking water sources (Singh, 2015).

2.3.3. Filtration regimes

There are two filtration strategies that influence the filtration regime. Those are:

- Dead-end filtration and
- Cross-flow filtration.

The way of operation of the dead-end filtration is based on forcing the entire fluid flow through the membrane under pressure. All solids accumulate on the membrane during the filtration cycle and they are removed during the backwash cycle. This requires increase of the pressure needed to maintain the required flow. After some time the membrane must be replaced. This type of filtration regime is given in Figure 2. The other filtration regime is the cross-flow filtration, given in Figure 3. In this situation two streams are produced; one is clean particle-free permeate and the other concentrated retentate containing the particles (Baker, 2004; Crittenden et al., 2012).

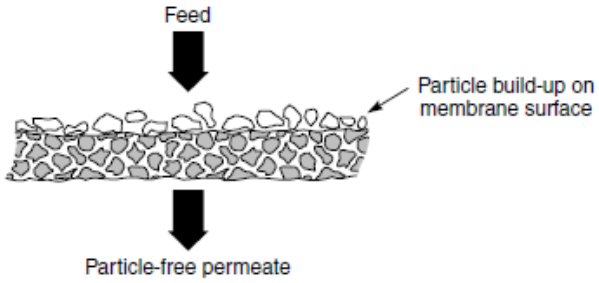


Figure 2: Schematic representation of dead-end filtration (Baker, 2004)

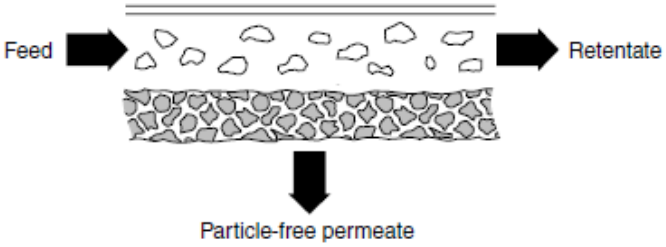


Figure 3: Schematic representation of cross-flow filtration (Baker, 2004)

Comparison of the advantages and disadvantages of dead-end filtration and cross-flow filtration is given in Table 2.

Table 2: Comparison of advantages and disadvantages of dead-end and cross-flow filtration (Baker, 2004)

Dead-end filtration	Cross-flow filtration
Low capital cost	High capital cost
High operating cost – membrane must be replaced after each use and disposal can be a problem	Operating costs modest – membranes have extended lifetimes if regularly cleaned
Operation is simple	Operation is complex – filters require regular cleaning
Best suited to dilute solution. Membrane costs increase with particle concentrations in the feed solution	Best suited to high solid content solution. Costs are relatively independent of feed solution particle concentrations
Representative applications: Sterile filtration Clarification/ sterilization of beer and wine	Representative applications: Continuous culture/ cell recycle; filtration of oilfield produced water

Cross-flow filtration has higher capital cost than dead-end filtration, but lower operating costs. The equipment required for cross-flow filtration is more complex. The operation in the dead-end filtration is simpler because this type of filtration is preferred to be used for cleaner and simpler purposes, as sterilization of water. Contrary, if the water has a high particle content, cross-flow filtration is preferred.

2.4. Membrane fouling and cleaning

Usually, the permeate flow decreases with time when operating with fluids. The major reason of such loss of productivity is the fouling phenomenon. The membrane fouling is defined by Koros, Ma and Shimidzu (1996) as the "process resulting in the loss of performance of a membrane due to deposition of suspended or dissolved substances on its external surface, or within its pores". This is a complex phenomenon, which influences the lifespan of a membrane, increases maintenance, cleaning costs and energy demand.

Fouling is characterized by the mechanism, by whether it can be removed (reversible or irreversible), and by the material causing it (particles, biofouling, and natural organic matter). There are three mechanism of membrane fouling, which include pore blocking, pore constriction and cake formation (Crittenden et al., 2012). The visualization of these phenomena is given in Figure 4.

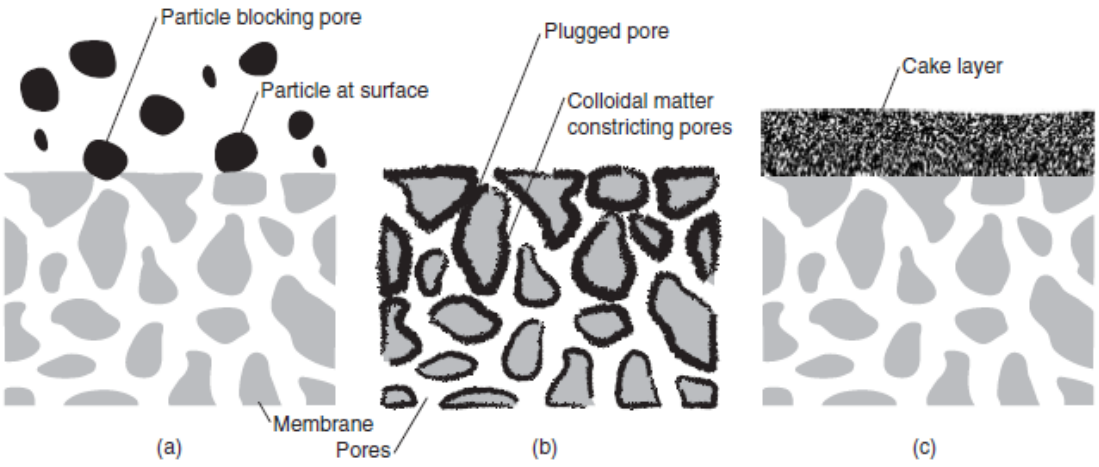


Figure 4: Fouling mechanisms in membrane filtration: (a) Pore blocking, (b) pore constriction and (c) cake layer formation (Crittenden et al., 2012)

Pore blocking occurs when the entrance to a pore is completely sealed by a particle, example (a) on Figure 4. Pore constriction happens due to the reduction of the void volume within a membrane as a result of materials adsorption within the pores, case (b). Cake formation

occurs when particles are too large to enter the pores and they collect on the membrane surface in a porous mat, case (c).

Fouling is characterized as reversible and irreversible. Permanent flux loss is called irreversible fouling. It depends from the water source and quality, as well as the type of membrane used. The loss of flux that happens during each filter run and can be recovered during backwashing is called hydraulically reversible fouling (Crittenden et al., 2012).

Four general types of fouling can be identified:

- Organic fouling is generated by the deposition or adsorption of dissolved organic materials on the membrane surface, such as proteins or polysaccharides (Jarusutthirak, Amy & Croué, 2002; Agenson & Urase, 2007).
- Scaling occurs due to oversaturation of soluble salty molecules, such as calcium and barium sulfates, calcium carbonate and silica scales, when polarization concentration causes their precipitation on the membrane surface (Schäfer et al., 2004).
- Colloidal fouling is generated from the accumulation of particles and colloidal matter, such as aggregated proteins, NOM, or inorganic colloids such as clay minerals, colloidal silica, metal oxides (Fe, Al and Mg), precipitated salts, suspended matter and organic colloids. The rate of colloidal fouling is higher when the permeate flux is higher (Singh, 2015).
- Biofouling is generated by the growth of microorganisms with accumulation of extracellular materials on the membrane surface (Mohammad et al., 2015). During filtration, microorganisms attach to the membrane surface, and once attached they can excrete extracellular material that causes additional fouling (Crittenden et al., 2012).

In order to maintain the overall process performances cleaning of the membranes must be done. There are two main types of cleaning: chemical and physical cleaning. Chemical cleaning includes solubilization, hydrolysis, enzymatic hydrolysis, saponification, chelation, or variation of pH. Whereas, physical cleaning implies backpulsation and backflush, gas bubbling, ultrasounds or application of electric fields. In order to prolong the lifespan of the membranes and reverse the fouling phenomenon, the cleaning methods can be combined (Singh, 2015). An efficient and robust filtration process is ensured by determination of the cleaning requirements and frequency of cleaning.

2.5. Industrial application of membranes

In industrial processes, the separation of components by membrane filtration can be realized without phase transfer or heat treatment. Consequently, the components in the mixture are less likely to suffer thermal degradation and this can be advantageous to some applications.

There is a wide range of applications that currently take advantage of membranes. Brief description of the membranes' applications in the industries is given below.

Food and beverage

The use of membrane technology in the food industry provides several advantages such as: ease of sterilization and cleaning, food safety, and environmental friendliness. It simplifies the process flow by avoiding more complex steps that cause chemical stress for the products and contribute to the production of high quality foods (Cuperus & Nijhuis, 1993).

Potable water

The water sources used for production of potable water may vary from site to site and in quality. That is the reason why the water industry has embraced membrane technology. This industry utilizes the membranes because of their barrier properties to exclude bacteria and microorganisms. The same technique is applied with the soft drink manufacturers, who need safe clean water, free of microorganisms, which is treated with membranes at a number of soft drink facilities (Singh, 2015; Mancinelli & Hallé, 2015).

Beer and wine production

The beer production industries are very precise and strict about the consistency and quality of the water used for the beer manufacture. Here also membrane facilities have taken their place, because of the ability to treat water sources to acceptable ionic content including hardness and alkalinity. In addition, other applications of the membranes in this industry are for: continuous beer stabilization for improvement of the brewery operating efficiency, and continuous clarification and final filtration of the beer. Membranes have been used for clarification of wine and avoidance of filter aids (AMTA, 2014).

Fruit juice production

Fruit juice manufacturers apply the membrane technology in a number of ways. Concentration of natural juices is achieved from the concentrated juice retained on the membrane from the water passing through, which is then used in the production. Because there is no heat applied,

no degradation of the complex juice sugars and flavor components happens. Color can be controlled and enhanced during the concentration step. Produced juice can be purified and clarified by removal of fine particles from juice using membranes (AMTA, 2014).

Dairy applications

Membrane filtration is a valuable part in the manufacture of dairy ingredients (Hu & Dickson, 2015). Its applications can be divided into three categories: applications to milk, applications to whey and other applications. In the applications to milk and whey is to make it more concentrated in order to produce condensed milk or provide concentrated milk.

3. Fish oil

Over a quarter of wild fish that are caught are unappetizing for direct human consumption, including small boney and oily fish such as anchovy, capelin, horse mackerel, sand eel, menhaden and pilchard. The fishing of the abovementioned species is done under controlled quota set by government agencies and is based on stock assessments, also called total allowable catch (TAC). In most of the countries which produce fish oil these limits are effectively policed by government agencies. The fish oil production from 2012 to March 2016 is shown in Figure 5. The main producing countries are Peru, Chile, Denmark, Norway and Island.

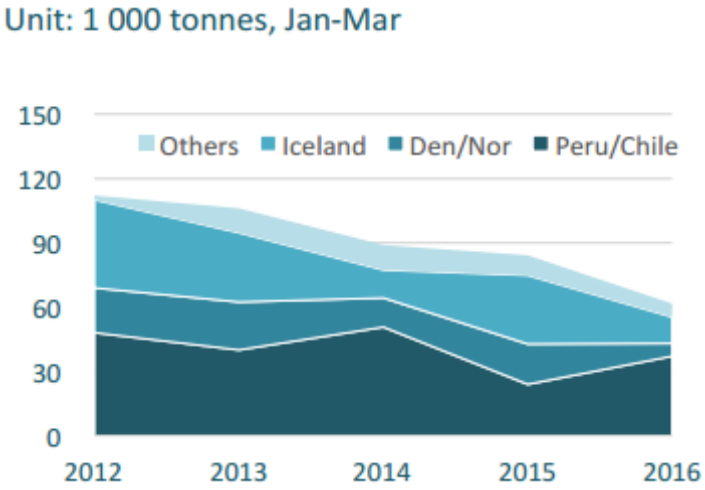


Figure 5: Major producers of fish oil in the world (FAO, 2016)

The fish meal and fish oil production process include several steps as described in Figure 6. The cut fish and by-products are directed to a cooker, where they are cooked for around 10 – 20 min using steam at temperature of approximately 90 – 95 °C. Then the heated material is transferred to a screw press where separation of solid and liquid phase is done. The press liquor is squeezed from the press cake. Afterwards, the press liquor is separated into three different phases: water, oil and solid. Fine suspended solids are removed using a decanter, whereas the oil and water separation is done via centrifugation. The other part of the process is the press cake, which is dried to reduce the moisture content using direct or indirect dryers. The fish meal is produced after the two last steps of cooling and grinding (Carvajal, n.d.).

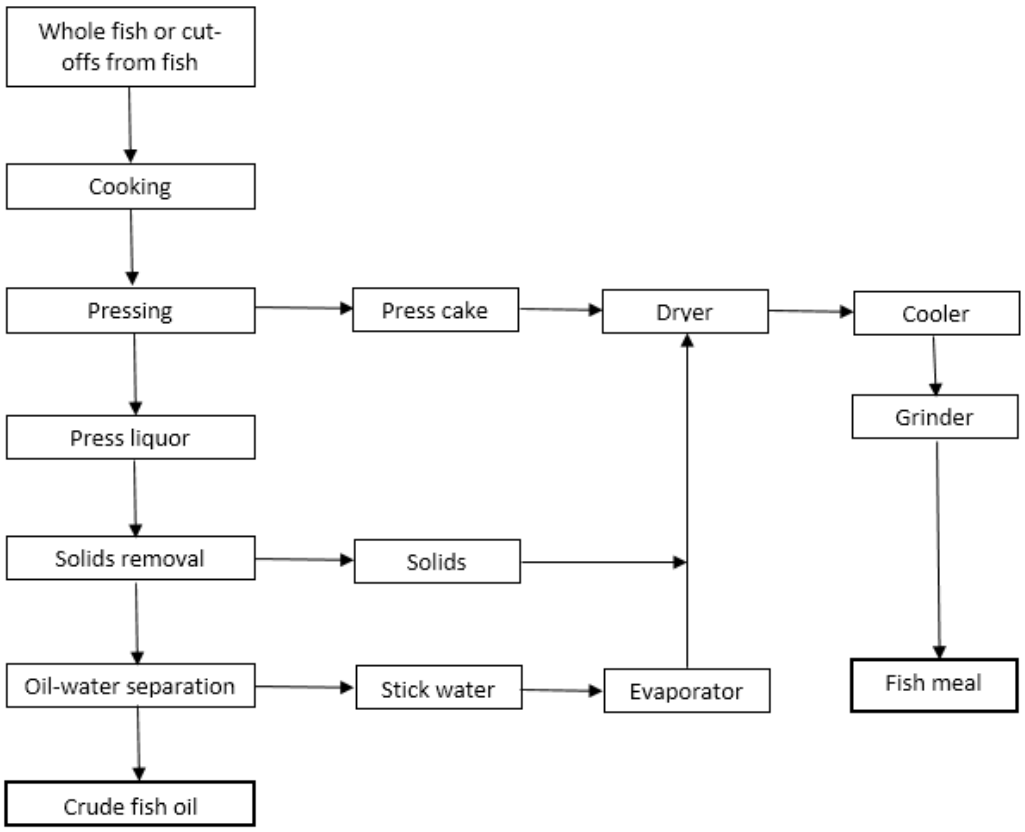


Figure 6: Flow diagram of fish meal and fish oil production (Carvajal, n.d.)

Fish oils are liquid at room temperature but generally solidify below 10 – 15 °C. The composition of fish oil depends from the type of fish. The European fish species such as capelin, herring, sand eel and sprat contain between 18 and 25% LC omega-3 fatty acids. These fish are called pelagic, which means that they swim and shoal in the upper layers of the sea. They store oil in the body rather than the liver and most of them are inedible for human (Pike & Jackson, 2010). The principal fatty acids contained in different types of fish are given in Table 3.

Table 3: Principal fatty acids in different fishes (Pike & Jackson, 2010)

	Capelin	Norway Pout	Mackerel	Sardine/ pilchard	Horse mackerel	Anchovy
Myristic 14:0	7	6	8	8	8	9
Palmitic 16:0	10	13	14	18	18	19
Palmitoleic 16:1	10	5	7	10	8	9
Oleic 18:1	14	14	13	13	11	13
Eicosenoic 20:1	17	11	12	4	5	5
Cetoleic 22:1	14	12	15	3	8	2
LC Omega-3s						
EPA 20:5	8	8	7	18	13	17
DHA 22:6	6	13	8	9	10	9

Contrary to the pelagic are demersal fish who store oil in the liver are live closer to the bottom of the sea. Those are cod and halibut which have a low content of LC omega-3s (15 to 20%).

During storage the tendency should be to eliminate the contact of the fish oil with air, pro-oxidant metals, especially those high in iron and copper. Also is it preferable to be treated with an antioxidant, such as butylated hydroxy-toluene (BHT).

It is very important to produce fish oil from fresh fish, because as fish spoils, enzymes split the oil into its component fatty acids. Ideally, free fatty-acid content should be below 2%, and there should be little oxidation (Pike & Jackson, 2010). It is also essential to keep the fish at a temperature between -1 and 0 °C and to keep the periods of fishing short. In this way the improvement of the quality of the raw fish reduces pollutant load of the wastewater and of the odor emissions, and forms a basis for an increased production of special fish products (Drivsholm & Nielsen, 1993).

3.1. General information about Omega – 3

Lipids are important nutrients that store, use and transport the energy through the human body (Drevon, 2009). The marine omega-3 phospholipids (n-3 PLs) contain n-3 long-chain PUFAs derived from marine organisms, as explained by Burri, Hoem, Banni and Berge (2012). These PLs differ from the PLs derived from vegetable sources, because they do not contain long-chain n-3 PUFAs.

In nature, n-3 FAs can be found as PLs or TGs or, due to a partial hydrolysis, in the free form. Visually the TGs and PLs structures can be seen in Figure 7. The TGs consist of three FAs esterified to a glycerol backbone, whereas PLs usually have two FAs esterified to a glycerol backbone together with a phosphorous group. This phosphorous group is linked to a headgroup which can consist of ethanolamine, inositol, choline, serine or glycerol. TGs are hydrophobic, whereas PLs are hydrophilic because of the polar headgroup. The physical-chemical properties of the two lipid groups are different and only PLs are able to form liposomes and micelles (Burri et al., 2012).

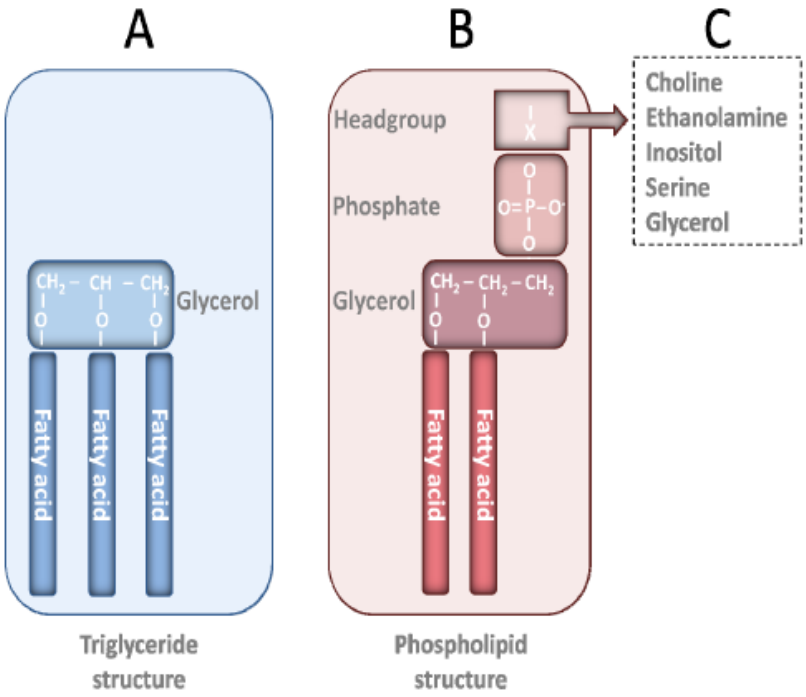


Figure 7: Triglyceride and phospholipid structures (Burri et al., 2012)

According to Calder (2013) the term omega - 3 (also notated as ω-3 or n-3) is a structural descriptor for a family of polyunsaturated fatty acids (PUFA). All omega-3 fatty acids have a double bond (C=C) at the third carbon atom from the end of the carbon chain. The fatty acids have two ends, the carboxylic (-COOH) end and the methyl (-CH3) end. The omega-3 fatty acids have systematic and common names as shown in Table 4. They are also referred to by a shorthand nomenclature that denotes the number of carbon atoms in the chain, the number of double bonds and the position of the first double bond relative to the methyl carbon (Calder, 2013).

Table 4: Omega – 3 polyunsaturated fatty acid family (Calder, 2013)

Systematic name	Common name	Shorthand nomenclature
all-cis-9,12, 15-Octadecatrienoic acid	α-Linolenic acid	18:3 ω-3
all-cis-6,9,12, 15-Octadecatetraenoic acid	Stearidonic acid	18:4 ω-3
all-cis-8,11,14, 17-Eicosatetraenoic acid	Eicosatetraenoic acid	20:4 ω-3
all-cis-5,8,11,14, 17-Eicosapentaenoic acid	Eicosapentaenoic acid	20:5 ω-3
all-cis-7,10,13,16, 19-Docosapentaenoic acid	Docosapentaenoic acid; also clupanodonic acid	22:5 ω-3
all-cis-4,7,10,13,16, 19-Docosahexaenoic acid	Docosahexaenoic acid	22:6 ω-3

The simplest omega – 3 fatty acid is the α-linolenic acid, whereas more complex are eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA). The chemical structures of EPA and DHA are given in Figure 8 and Figure 9.

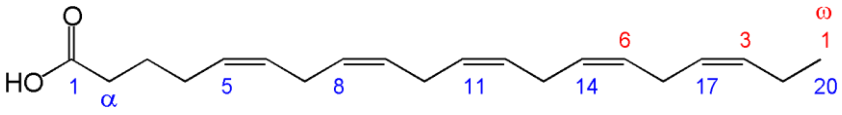


Figure 8: Chemical structure of EPA (Kapoor & Patil, 2011)

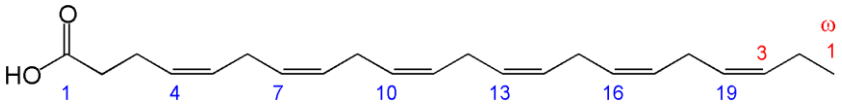


Figure 9: Chemical structure of DHA (Kapoor & Patil, 2011)

3.2. Sources of Omega-3

The main source of Omega-3 is seafood. Different types of fish contain different amounts of fatty acids and different ratios of EPA to DHA. The amounts and ratios of EPA and DHA differ based on the metabolic characteristics of the fish and their diet, as well as the water temperature and season (Calder, 2013). EPA and DHA can be obtained as extracts from the roe of cold-water fatty fish. The fattier the fish is, the more EPA and DHA it will contain. Also, significant amounts of very long-chain omega-3 fatty acids are obtained from fatty fish such as mackerel, herring, trout, salmon, eel, sardines, anchovies, as well as from krill oil, fish oil, tuna oil and cod liver oil (Drevon, 2009). The sources important for providing the necessary fatty acids are given below.

Fish

As mentioned, the main dietary source of EPA and DHA is fish, containing between 1 – 1.5% PLs and 10 – 15% TGs (Hjaltason & Haraldsson, 2006). These amounts refer to cold-water oily fish like salmon, anchovy, sardine, herring, or mackerel, where up to one third of the EPA and DHA content might exist in the form of PLs.

Fish roe

The word ‘roe’ stands for the eggs and the ovaries full of seafood eggs. Fish roe is a by-product of the fish industry used for human consumption. Fish roe is a rich source of n-3 PUFAs in PL form, containing between 38 – 75% lipids in the form of PLs from salmon, herring, flying fish and pollock. Salmon has the highest total lipid content, where 56% of the lipids are in TG form, whereas the other roes have values below 20%. More than 30% of the

total FAs are eicosapentaenoic acid, with ratio 20 EPA:5 n-3 or docosahexaenoic acid, with ratio 22 DHA:6 n-3 (Burri et al., 2012).

Krill oil

Krill oil is an important source of marine PLs and it has become increasingly popular as a food supplement during the last decade. Krill oil is extracted from the shrimp-like zooplankton - Antarctic crustacean krill (lat. *Euphausia superba*). It contains high amounts of EPA and DHA in the PL-bound n-3 PUFAs. The PLs content in the oil extracted from krill is typically around 40% (Burri et al., 2012). In fish oils from different species the EPA plus DHA range is from 11% in herring oil to 26% in anchovy oil (Pike & Jackson, 2010).

3.3. Recommended intake of Omega-3 and health benefits

The modern diet is deficient in omega-3 fatty acids and has become overloaded with pro-inflammatory omega-6 fatty acids. This heavy imbalance is thought to lead to an overall inflammatory state that might contribute to several diseases.

The daily recommended intake of LC omega-3s is in the range of 0.25 to 0.5 g per person per day. Several authorities such as the UK Government and US Heart Association have recommended people to eat fish twice a week, including oily fish, to provide 3 g weekly of LC omega-3s (Pike & Jackson, 2010).

The consumption of LC omega-3s either in fish (wild and farmed) or in encapsulated fish oil helps maintain general human health. PLs contribute as building blocks for cell membranes in almost all known living beings by playing an important role in cellular structure and function. They also have a valuable part in the formation of lipoproteins, which transport lipids to tissues through the blood stream. The omega-3s contribute to ameliorating inflammatory disorders such as asthma, eczema, psoriasis and Crohn's disease. Cardioprotective effects of n-3 long-chain polyunsaturated fatty acids (PUFAs), in particular eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA), have been attributed to reduction in fasting triacylglycerol (TAG), anti-inflammatory and anti-arrhythmic effects, blood pressure lowering, improved vascular endothelial function and insulin sensitivity, and reduced thrombotic tendency (Bjørndal et al., 2014). EPA and DHA have an important function as a component of brain and nervous tissue, and in particular in the development of these organs. Dietary LC omega-3 inclusion plays important role in the last trimester of pregnancy and in infant nutrition (Pike & Jackson, 2010).

4. Materials and methods

This section describes the materials necessary for execution of the nanofiltration process and obtaining the membrane performance. The characteristics of the membranes are provided, the description of the feed preparation process and the methods used for rejection analysis.

4.1. Nanofiltration membranes

The two types of membranes used in the nanofiltration tests for defining the rejection performance of organic constituents were polymeric and ceramic. The samples were selected on the bases of MWCO range and availability on the market. The MWCO is defined by Koros et al. (1996) as the molecular weight at which 90% of the macromolecular solute is rejected by the membrane; it is measured in Daltons [Da]. The industrial experience of the membrane producers was also important because if the process was successful, it would be implemented in a real industrial scale.

4.1.1. Polymeric membranes - DuraMem

The selected polymeric membranes were produced and provided by Evonik MET Ltd. from Germany as flat sheets. More precisely, DuraMem® 200 (T1), DuraMem® 300 (T1) and DuraMem® 500 (T1) were tested for this study. The outlook of DuraMem 500 is provided in Appendix 1. These membranes are operated in a cross-flow filtration mode. These membranes have NF layer made from polyimide and have a MWCO between 200 and 500 Da corresponding to the name of the membrane. All the polymeric membranes investigated in this study were hydrophilic with a contact angle of approximately 8° when tested with water. The contact angle defines the hydrophobicity of the surface of the membrane, meaning that if the contact angle is high the surface is hydrophobic (Crittenden et al., 2012). The chemical structure of the polyamide is given in Figure 10 below.

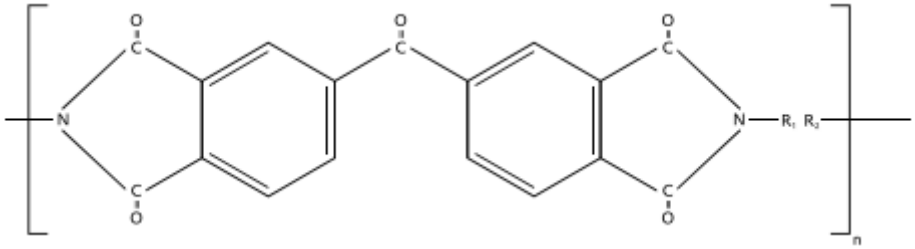


Figure 10: Chemical structure of polyamide (Evonik, n.d., b)

The recommended operation conditions are provided below:

- Recommended maximum temperature: 50 °C;
- Recommended maximum operating pressure: from 20 to 60 bar;
- Usable in: acetone, methanol, ethanol, tetrahydrofuran, isopropanol, acetonitrile, methylethylketone, ethyl acetate etc (Evonik, n.d., c).

4.1.2. Ceramic membranes - Pervatech

Five ceramic membranes produced by Pervatech in Netherlands were used for the nanofiltration experiments, including Pervatech 300-200, Pervatech 500-400, Pervatech 500D, Pervatech 700D and Silane. They were custom made for the specific tests composed of support from alpha alumina coated with a nano-filtrating layer of TiO_2 particles. The samples were delivered in form of discs with diameter of 39 mm, 2 mm thick as shown in Figure 11. The estimated MWCO of these membranes are the numerical values attached to their names. The contact angle of the ceramic membranes was also measured. Hydrophobic membranes were Silane and Pervatech 500D, whereas the rest were hydrophilic.

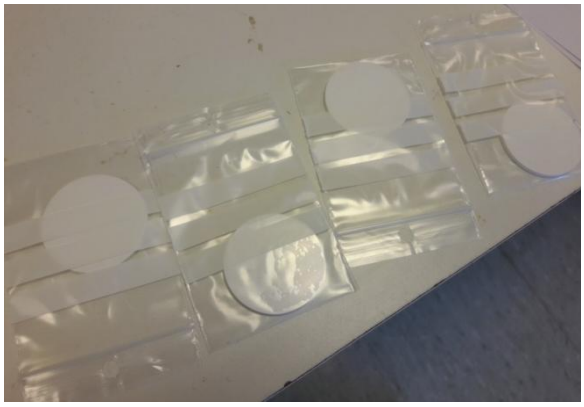


Figure 11: Visual look of four disks of the ceramic membrane Silane

4.2. Feed solution preparation

The composition of the feeds used in the experiments was based on three components expressed as percentage by weight (wt%). The balance used for weighting the compounds was a MS precision balance produced by Mettler Toledo. The feed solution was prepared using ethanol (C_2H_6O) and water (H_2O) in the following ratios, 95/ 5, 90/ 10, 85/ 15 and 80/ 20 wt% ethanol/ water, and 0.3 wt% dry matter from herring roe extract. The herring roe extract was a byproduct from the production process of a Norwegian company for production of encapsulated fish oil from immature herring roe. It was highly diverse, composed of around

30% dry matter, of which 7% were free fatty acids, 7% cholesterol; about 75% proteins; 15% fat of which 10% were PLs and the rest TGs.

For the preparation of the feed, mixing of the three components with defined weight percentages was done. This process was executed using instrument called rotary evaporator from producer Heidolph, model Hei-VAP. The picture of the equipment is given in Appendix 2.

Before starting the rotary evaporator, the necessary conditions were set manually, including:

- Mixing time: 30 – 60 min;
- Bath temperature: 40 °C;
- Rotation speed: 1300 rpm;
- Pump pressure: 350 mbar.

The role of the pump was to extract the oxygen from the tube in order the space inside the equipment to be filled with evaporating ethanol. When the mixing process was finished, the feed was cooled down using the same instrument with different working conditions input:

- Mixing time: 30 min;
- Bath temperature: /
- Rotation speed: 90 - 100 rpm;
- Pump pressure: 250 - 300 mbar.

Figure 12 presents the composition and consistency of the feed after the process of mixing.

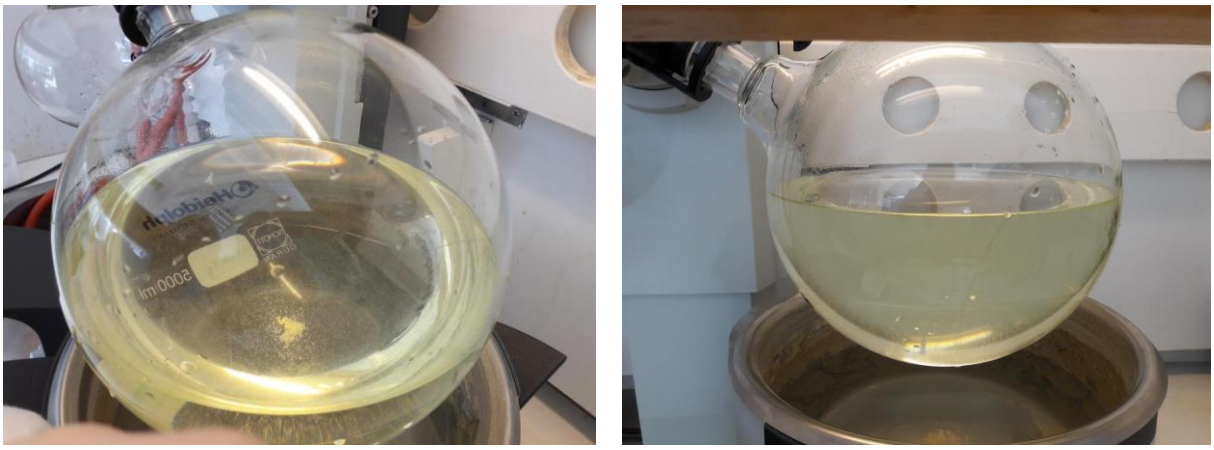


Figure 12: Composition of the feed containing 90 wt% EtOH after mixing

The next step was filtering of the feed. This action was done in order to remove greater particles contained in the dry matter from the herring roe fat extract. The possibility of clogging of the membrane because of the presence of big particles in the feed was in this way

eliminated. The filter used was a quantitative filter paper produced from Munktell with diameter of 110 mm and pore size of 1 µm. The choice of the filter was done based on the data for the size of the particles present in the feed. Thus the particles with size lower than the pore size of the filter were eliminated. Prior to this step, the herring roe fat extract was once filtered before using it as component in the feed. The filtration was done under reduced pressure through a filter with pore size 1µm.

The feed exposure to the atmosphere was limited, because the PLs contained in the roe extract were sensitive to light and oxygen. Having this in consideration, the time of exposure of the feed outside from dry and cold place during the preparation of the batch was reduced to the necessary minimum. The feed was flushed with gaseous nitrogen after each use for ensuring longer lifetime and stability. The amount of the feed prepared was based on the vessel used and usually its weight was around 2 kg. The feed was stored in amber bottle to reduce exposition to light.

4.3. Filtration experiments

The filtration experiments are described in this part of the thesis, including several steps, such as membrane preparation and pre-conditioning, together with the sampling process that is covered in detail as the focus of this point.

4.3.1. Preparation of membranes and conditioning

Before beginning with filtration experiments where the membrane's rejection would be tested, it was required to prepare and precondition the polymeric membrane for the experiment. The conditioning time is defined by Koros et al. (1996) as a "process carried out on a membrane after the completion of its preparation and prior to its use in a separation application". The polymeric membranes were provided as flat sheets, but the effective filtrating surface according to the filtration cell was around 12.5 cm^2. Considering this, it was necessary to prepare the membrane coupon by cutting the flat sheet using scalpel. The process of cutting is shown in Figure 13.

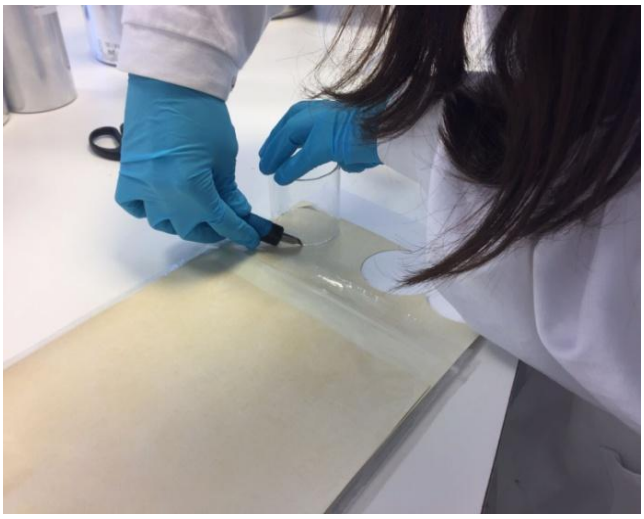


Figure 13: Preparation of polymeric membrane DuraMem 500 – cutting process

The storage of the membrane was inside the feed whose composition was same as the one used for the test. It was important to preserve the membrane wet in order not to provoke its deformation and damage. Also, in this way the possibility of membrane saturation and clogging during the pressurized test was reduced to minimal value. The feed used for pre-conditioning of the membrane was prepared based on the vessel used and its weight was around 150 g. A vessel used for pre-conditioning is showed in Figure 14. The abovementioned preparation steps were omitted for the ceramic membranes from two reasons. First, because they were custom made by Pervatech with size compatible to the filtration cell; and second, because they were not influenced by the organic molecules present in the feed due to the different properties from the polymeric membranes and did not require conditioning.

Figure 14: Pre-conditioning of three flat sheets from DuraMem 500 in feed with 90 wt% EtOH

4.3.2. Nanofiltration equipment and sampling plan

The bench-scale equipment used for membrane filtration experiments operated at constant pressure, using two types of cells. The schematic of the equipment is given in Figure 15. The overall picture of the equipment parts and their interconnections used for executing the NF process is given in Figure 16.

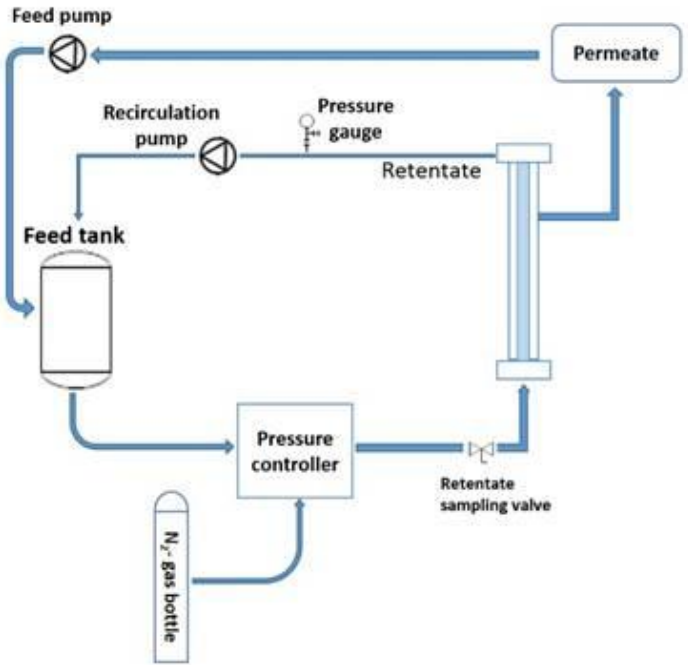


Figure 15: Schematic of the equipment used in the nanofiltration experiments

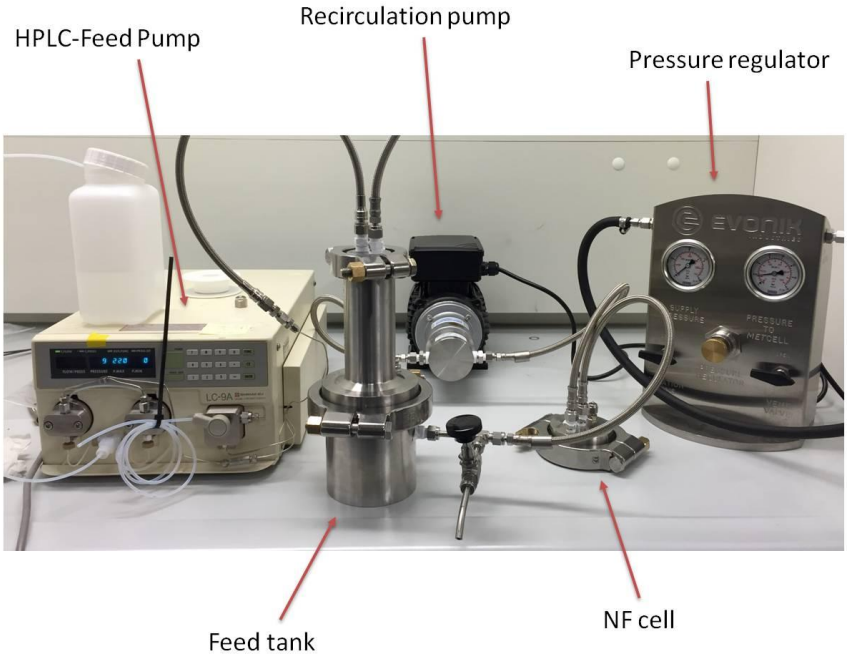


Figure 16: Bench-scale nanofiltration equipment with its constituent parts used for testing the membrane performance

One cell was used for the polymeric membranes, designed with filtrating surface of 12.5 cm^2. This cell named as 2.5" MET CrossFlow Filtration Cell is shown in Figure 17. The smooth side of the membrane disk was put on the upper side, on the center. Then the disk was pressed in order to tighten the membrane and small amount of ethanol was put in the openings of the cell.

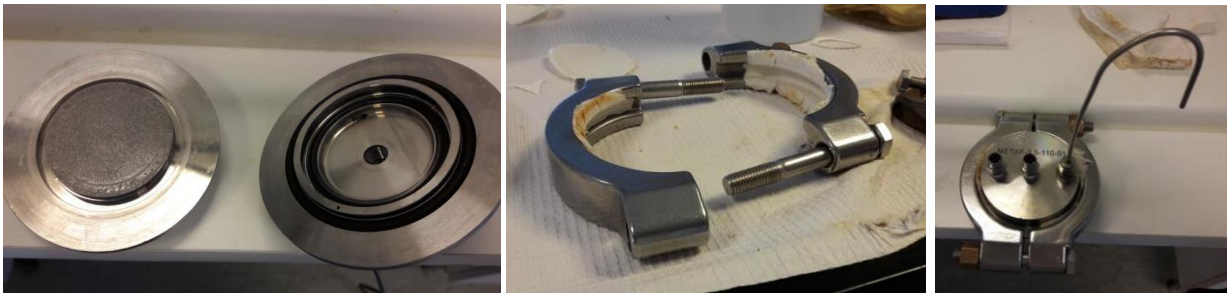


Figure 17: Filtration cell used for polymeric membranes

The other interchangeable cell used for ceramic membranes was produced by the same company that produced the membranes called Pervatech, and it is presented in Figure 18. This cell was looking different than the one for the polymeric membranes and its effective filtrating surface was 7 cm^2.

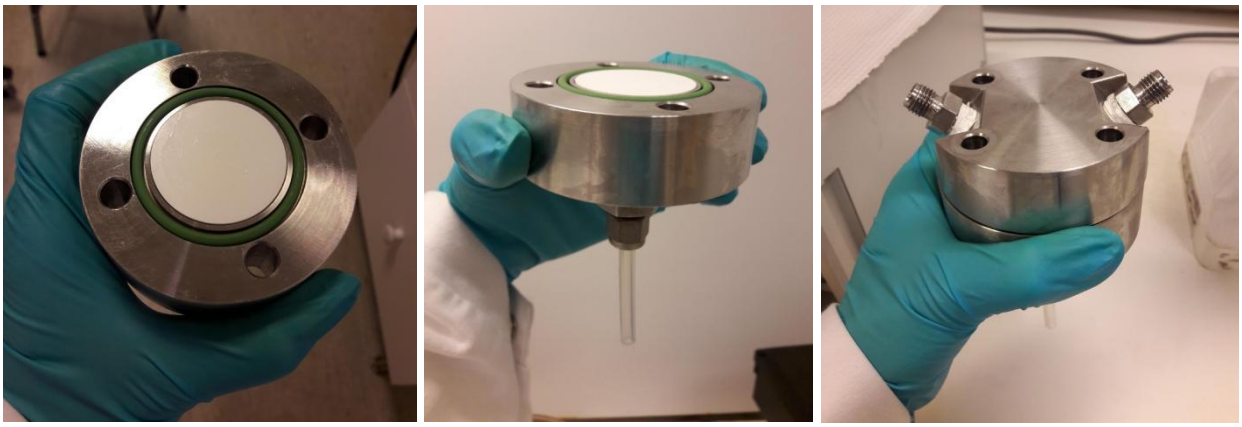


Figure 18: Filtration cell used for ceramic membranes

After placing the membrane in the cell it was necessary to fill the feed tank with the previously prepared feed. The amount of the feed was varying between 550 to 580 g depending on the EtOH concentration.

After the tank was filled with feed, a recirculation flow of 60 L/ h was induced to the system using recirculation pump. The production name of this pump is Micropump. The pressures used for testing the polymeric membranes were 5 and 40 bar which were applied manually on the pressure controller. The first experiment was done at 5 bar, but later it was increased to 40

bar in order to see how it will influence the membrane rejection having in consideration that it was in the range of recommended pressure by the producer. The ceramic membranes were tested under pressure of 5 bar because they do not withstand pressure load above 8 – 10 bars due to the characteristics. When the pressure exceeded 10 bar the ceramic membrane broke. The pressure controller was connected to the N_2 bottle under high pressure from one side and the feed tank from the other side. The permeate was injected back to the feed tank using feed pump. The available flow rate of the feed pump with technical name HPLC pump Gilson 25WTi was from 0.01 to 25 mL/ sec. The value for the flow was inserted manually on the display. In the beginning the flow rate was low, starting with around 5 mL/ sec. The reason was due to the presence of bubbles in the hose, which could cause damage to the pump. In time the bubbles were eliminated and no interaction with the pump happened.

After the conditioning time was finished the next step in the NF process was taking samples of the permeate. The sampling period is presented in Table 5, which contains the fastest and slowest sampling period from all samples taken with the polymeric membranes at two pressures and four feeds.

Table 5: Sampling period range for the polymeric membranes expressed in minutes

Membrane	**Pressure (bar)**	**Ethanol/ water composition [wt%]**			
		95 / 5	**90 / 10**	**85 / 15**	**80 / 20**
DuraMem 200	5	49 – 92	30 – 87	39 – 143	35 – 76
	40	12 – 27	19 – 21	na	14 - 31
DuraMem 300	5	43 – 67	25 – 42	24 – 45	18 – 97
	40	12 – 21	6 – 12	5 – 22	9 – 21
DuraMem 500	5	30 – 51	12 – 15	na	12 – 48
	40	12 – 22	na	na	3 - 10

na: data not available

The sampling time was not strictly defined because it was dependant from the flux through the membrane. According to theory all samples must be taken in an equilibrium situation so the time duration of the tests did not have great impact on the membrane performance.

The glass tube used for sampling the permeate was 10 ml, but the minimum required quantity was 2 ml in order to fill a vial for analysis using UHPLC instrument. After taking the sample it was appropriately labeled and placed in the UHPLC instrument or kept in the dark at 4 °C, if the rejection test was not done immediately. Three groups of samples containing four samples of the permeate and one sample of the feed are shown in Figure 19.

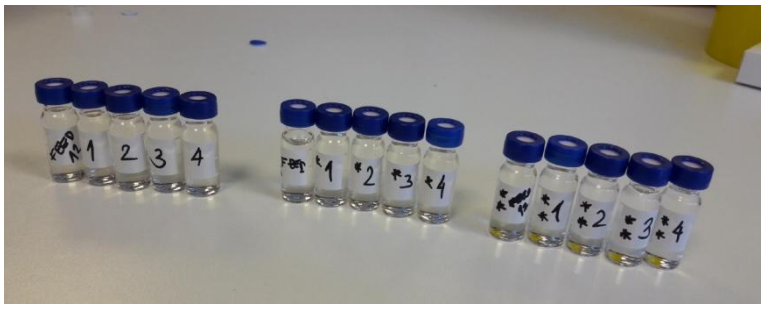


Figure 19: Samples labeled and prepared for UHPLC analysis

The number of samples taken with each membrane is given in Table 6 for the polymeric membranes and in Table 7 for the ceramic membranes. It can be seen that from one test at least three samples were taken in order to ensure accuracy of the results obtained.

Table 6: Number of tests done and number of samples (in brackets) taken with the polymeric membranes at two pressures with four feeds

Membrane	Pressure (bar)	Ethanol/ water composition [wt%]			
		95 / 5	**90 / 10**	**85 / 15**	**80 / 20**
DuraMem 200	5	3 (9)	3 (12)	3 (12)	3 (10)
	40	3 (12)	4 (16)	na	3 (10)
DuraMem 300	5	3 (12)	3 (12)	3 (12)	4 (14)
	40	3 (10)	4 (16)	3 (12)	4 (15)
DuraMem 500	5	3 (10)	3 (12)	na	3 (9)
	40	3 (12)	na	na	3 (10)

na: data not available

For a raison of time constrain, DuraMem 500 was not tested using the feed with 85 wt% EtOH at both pressures of 5 and 40 bar, nor with the feed with EtOH concentration of 90 wt% at 5 bar. Also, the other ceramic membranes with exception to Pervatech 500-400 were not

tested using the feed containing 95 wt% EtOH. The reason was because it was noticed that the EtOH concentration does not increase the level of rejection, having in consideration that the results obtained with Pervatech 500-400 using 95 wt% EtOH and 80 wt% EtOH were the same, amounting 36%. The results will be discussed in detail in the Results and Discussions section.

Table 7: Number of tests done and number of samples (in brackets) taken with the ceramic membranes at two pressures with two feeds

Membrane type	Ethanol/ water composition [wt%]	
	95 / 5	**80 / 20**
Pervatech 300-200 CM	na	2 (6)
Pervatech 500-400 CM	3 (12)	3 (12)
Pervatech 500D coated	na	1 (3)
Pervatech 700D coated	na	1 (3)
Silane	na	3 (14)

na: data not available

4.4. Analysis of membrane rejection performance

Significant parameter in membrane filtration is the size of material retained, defined also as the retention rating (Crittenden et al., 2012).

The difference in the two terms, rejection and retention was in the approach of their use. When the term rejection was used, it referred to the components removed from the feed by the membrane. Whereas, the term retention was used for defining the components retained on the membrane itself. The term used in this master thesis was rejection, and based on the rejection calculation overall conclusion about the membrane performance was obtained.

The last step in the nanofiltration process was the analysis of the previously taken samples by the UltiMate® 3000 Quaternary Analytical system produced by Thermo Scientific™ which uses Chromeleon Chromatography Data System. The instrument is presented in Figure 20.Figure 20: It works based on the UHPLC technique, which is used to analyze and separate compounds through the mass-transfer of analytes between stationary and mobile phases. This

technique utilizes a liquid mobile phase to separate the components of a mixture by forcing them to flow through a column stationary phase under high pressure. The amount of resolution of the mixture into its components depends upon the interaction between the solute components and the column stationary phase and liquid phase, which can be manipulated through different choices of both solvent and stationary phases (Bedson & Prichard, 2003).

Detailed description of the instrument's way of work, figures of its main parts, layout of the Chromeleon System and its requirements in aspect of data input are provided in the Appendix 3. Whereas, the PLs rejection results obtained by the UHPLC instrument and their analysis is presented in the next section Results and Discussions.

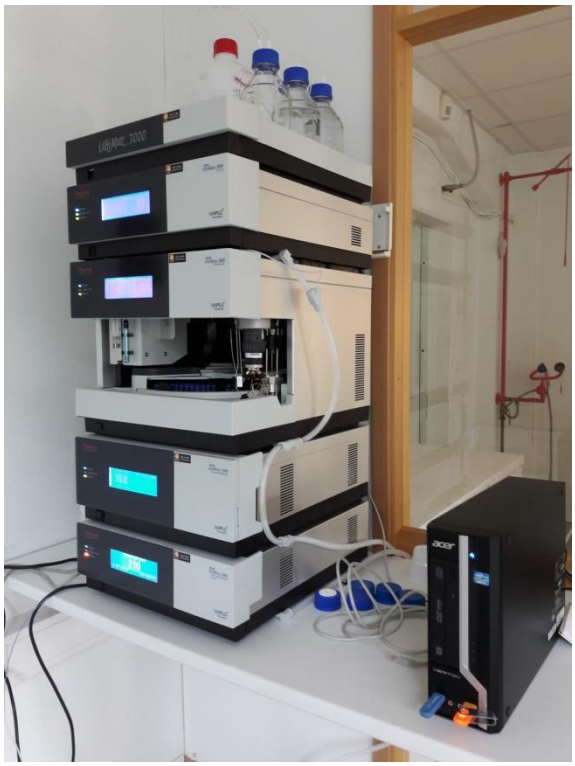


Figure 20: Outlook of the UHPLC instrument - UltiMate® 3000

4.5. Sources of error

In laboratory experiments the possibility of making an error is always present. The sources of error might be different, such as equipment imprecision, human mistake, inappropriate storage of samples for testing, improper cleaning of the equipment, outside conditions influencing the result.

Taking into consideration the variety of instruments used during the experimental procedure, the possibility of introducing an imprecision in the result was highly present. The human

mistake was probable during handling with the membranes, measuring the glass tubes and rewriting the values delivered by the balance, or defining the peak area in the UHPLC instrument. As previously mentioned, the PLs were sensitive to light and atmosphere, so longer exposure could have led to their evaporation. Cleaning of the feed tank was done after each use of the equipment, and it was mandatory to be filled to the top. If this was not the case, the hoses would not be cleaned and the residues from the previous feed used might have had influenced the results from the samples taken in the next NF process. Also, worth mentioning is that the temperature in the laboratory was not measured and therefore it presents a source of uncertainty.

5. Results and Discussions

This section presents the results of the experiments performed. It is divided into two parts according to the type of membrane, polymeric and ceramic, containing the most noticeable and influential results. The discussions focus on delivering useful conclusions for the membrane behavior and performance in organic solvent.

5.1. Phospholipids rejection

In membrane technology, the rejection of a specific component is an important parameter providing information about the membrane performance. According to the percentage of rejection of the component of interest, phospholipids, relevant conclusions can be made about the selection of membrane.

This study aimed at selecting a nanofiltration membrane which can sustain organic solvent while achieving rejection of omega-3 PLs. The rejection criterion of phospholipids was between 80% and 95%. These limits were selected based on the application of the membranes, which was retention of PLs and TGs. Rejections below 80% would not achieve the desired retention of components. While, membranes with rejection greater than 95% have high probability of retention of undesirable compounds present in the solution, such as salts and proteins.

5.1.1. Performance parameters for DuraMem membranes

The average rejection percentages and standard deviations obtained for the polymeric membranes are given in Table 8. The rejections were calculated at 5 and 40 bar pressure for the polymeric membranes DuraMem 200, 300 and 500 using four ethanol/ water mixtures. The results from all the tests are presented in previously mentioned Appendix 4 and Appendix 5.

Table 8: Phospholipids rejection obtained by DuraMem membranes at 5 and 40 bar and four ethanol/water compositions expressed in percent [%]

Membrane	Pressure (bar)	Ethanol/water composition [wt%]			
		95 / 5	**90 / 10**	**85 / 15**	**80 / 20**
DuraMem 200	5	100 ± 0	87 ± 13	84 ± 17	79 ± 4

Membrane	Pressure (bar)	Ethanol/water composition [wt%]			
		95 / 5	**90 / 10**	**85 / 15**	**80 / 20**
	40	100 ± 0	78 ± 17	na	80 ± 6
DuraMem 300	5	91 ± 2	49 ± 7	43 ± 7	63 ± 14
	40	94 ± 5	39 ± 9	40 ± 21	63 ± 10
DuraMem 500	5	76 ± 3	41 ± 4	na	40 ± 13
	40	98 ± 1	na	na	34 ± 22

na: data not available

Results show PLs rejections above 94% for the three polymeric membranes at a pressure of 40 bar and an ethanol/water composition of 95/5 wt%. Therefore, these operational conditions will not be considered for further investigation. At an ethanol/water ratio of 95/5 wt% only DuraMem 300 at 5 bar pressure achieved rejection within the criteria.

In general, for the feeds containing 90 and 85 wt% EtOH the observed rejections were higher at an operating pressure of 5 bar compared to 40 bar. However, only DuraMem 200 achieved rejection within the defined criteria at both 5 and 40 bar for this type of feed. The explanation for this observation might be related to the MWCO of the membrane DuraMem 200. It is lowest from the group, amounting in 200 Da which leads to the highest retention of all the three membranes. The results obtained with EtOH concentration of 80 wt% show that DuraMem 200 gives rejection within the set limits, at both pressures. The values are 79 ± 4% and 80 ± 6%, respectively.

If size exclusion is assumed to be the main rejection mechanism with DuraMem membranes, DuraMem 300 should have lower retention of particles compared to DuraMem 200 because its MWCO is 300 Da. This is confirmed from the results where the rejection obtained with DuraMem 300 is lower compared to the one with DuraMem 200 with the same conditions are applied. DuraMem 500 has the largest pore size at 500 Da and lowest rejection value of 41 ± 4% was observed at a pressure of 5 bar and 90 wt% ethanol.

Figure 21 and Figure 22 provide graphical representations of the rejection data. The values given are mean values obtained from all measurements done with each polymeric membrane and each type of feed.

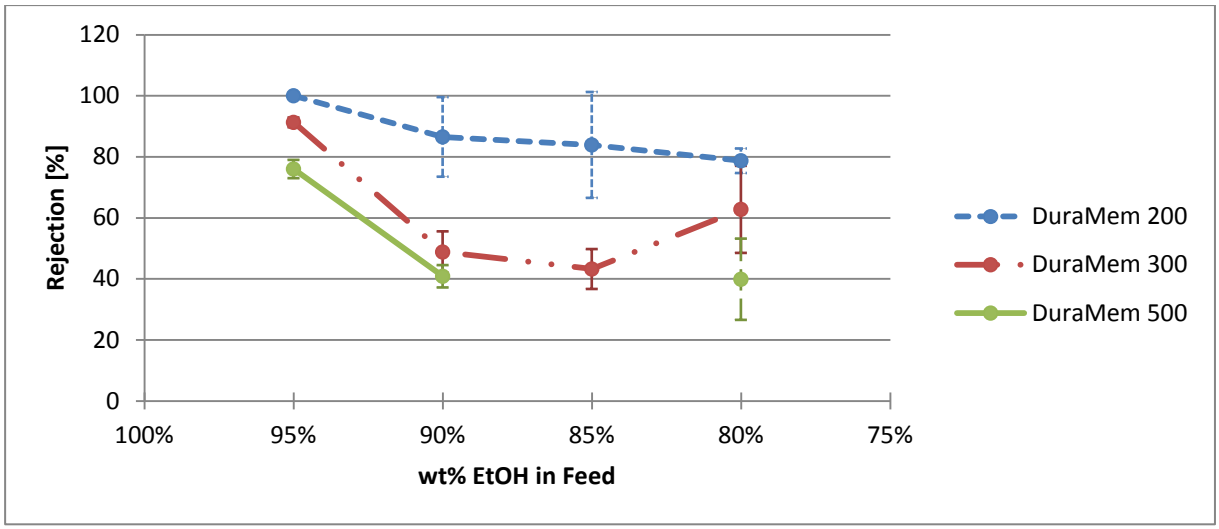


Figure 21: Rejection percentage for three types of polymeric membranes depending from the feed when 5 bar pressure was applied

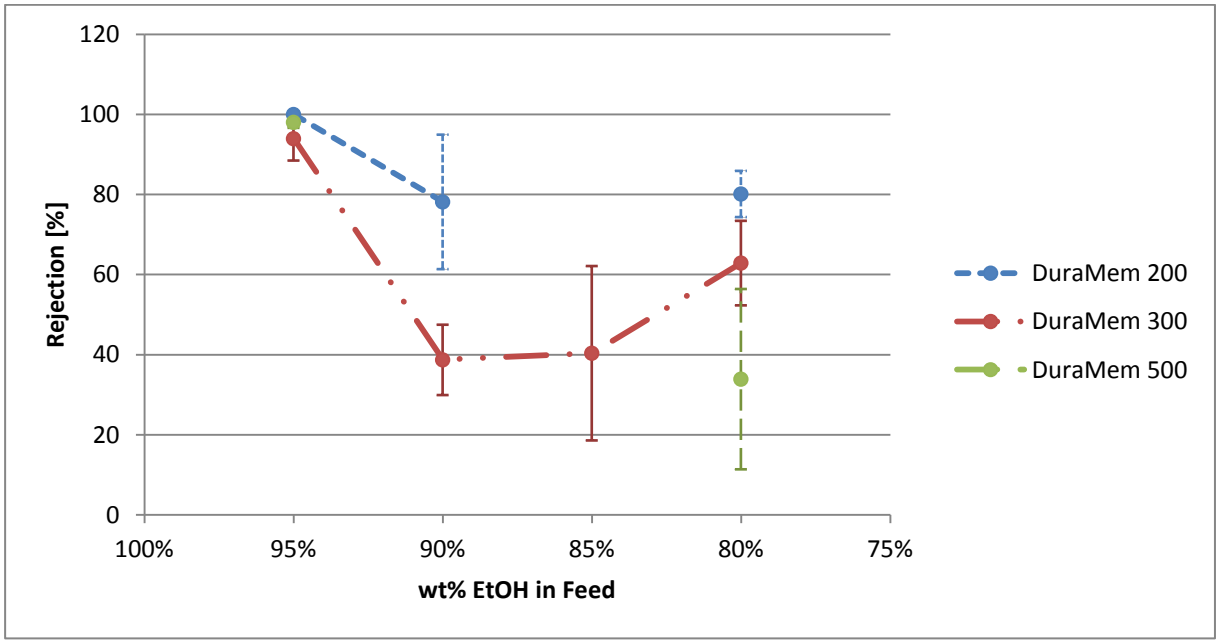


Figure 22: Rejection percentage for three types of polymeric membranes depending from the feed when 40 bar pressure was applied

DuraMem 200 follows a function close to the linear function when 5 bar pressure was used. With a decrease of the wt% of EtOH in feed the rejection is also decreasing. However, a different rejection pattern was observed when 40 bar pressure was applied. At a 95 wt% EtOH the rejection was 100% but the rejection decrease to 78% and 80% at 90 and 80 wt% EtOH respectively. The data seems to indicate stable rejection at feed composition between 90 and 80 wt% EtOH.

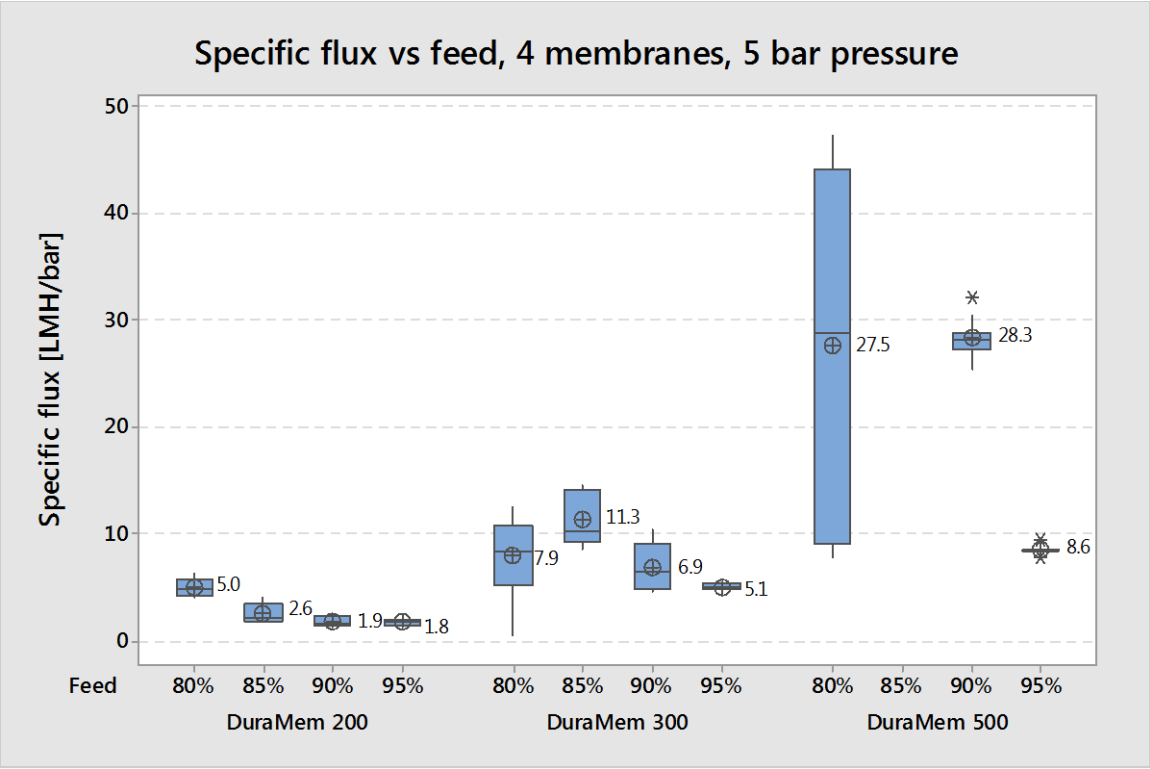
DuraMem 300 follows a similar trend as DuraMem 200 at 5 bar pressure, from a EtOH concentration of 95 to 85 wt%. Then, as the concentration of EtOH continues to decrease to 80 wt% EtOH the rejection changes the direction and starts to increase. The rejection of DuraMem 300 when 40 bar pressure was applied follows the same function as with 5 bar pressure, which is close to a parabolic function. The average rejection values for the feeds with 90 and 85 wt% EtOH at 40 bar pressure were 39 ± 9% and 40 ± 21% respectively. Whereas, when the test pressure was 5 bar the results were higher amounting 49 ± 7% and 43 ± 7%. The results within the rejection criteria are related to the feed containing 95 wt% EtOH if the SD is considered. For 5 bar pressure, the value is 91 ± 2%, whereas when 40 bar pressure was applied the rejection increased together with its SD to 94 ± 5%. These results indicate that the applied pressure during the experiment does not have an influence on the rejection results obtained with DuraMem 300.

For DuraMem 500, with a feed composition of 95 wt%, the membrane show a rejection of 76 ± 3% and 98 ± 1% at 5 and 40 bar pressure, respectively. At 5 bar pressure the rejection decrease to approximately 40% at feed composition of 90 and 80 wt%. At a pressure of 40 bar, low rejection of 34 ± 22% was measured at a feed composition of 80 wt%.

Important observation is that the higher pressure applied led to an increase in the rejection of the polymeric membranes for the feeds containing 95 wt% EtOH. For the other feed composition, 90, 85 and 80 wt% EtOH, the rejection performance of the polymeric membranes was higher when the pressure used was 5 bar.

The SD of the tests with 5 and 40 bar pressure differs and no precise correlation can be found. In the experiments with 5 bar pressure the highest deviation is related to DuraMem 200 with the feed containing 85 wt% EtOH. Whereas in the case when 40 bar pressure was applied the deviation is highest with DuraMem 500 with 80 wt% EtOH feed. If the SD of all the three membranes is compared when 5 bar pressure was applied, it can be noticed that it increases with decreasing the amount of EtOH in feed. Exception to this is DuraMem 200 when the EtOH concentration was lowest and the SD decreased. In the tests with 40 bar pressure the SD varies from feed to feed. The large SD observed at a feed composition of 80 wt% indicates that rejection values between DuraMem 200 and 300 cannot be differentiated. Similar observation is valid for DuraMem 300 and 500 at a feed composition of 80 wt%. The might explained based on the recommended use of the membranes, which is in organic solvent which in this case is ethanol.

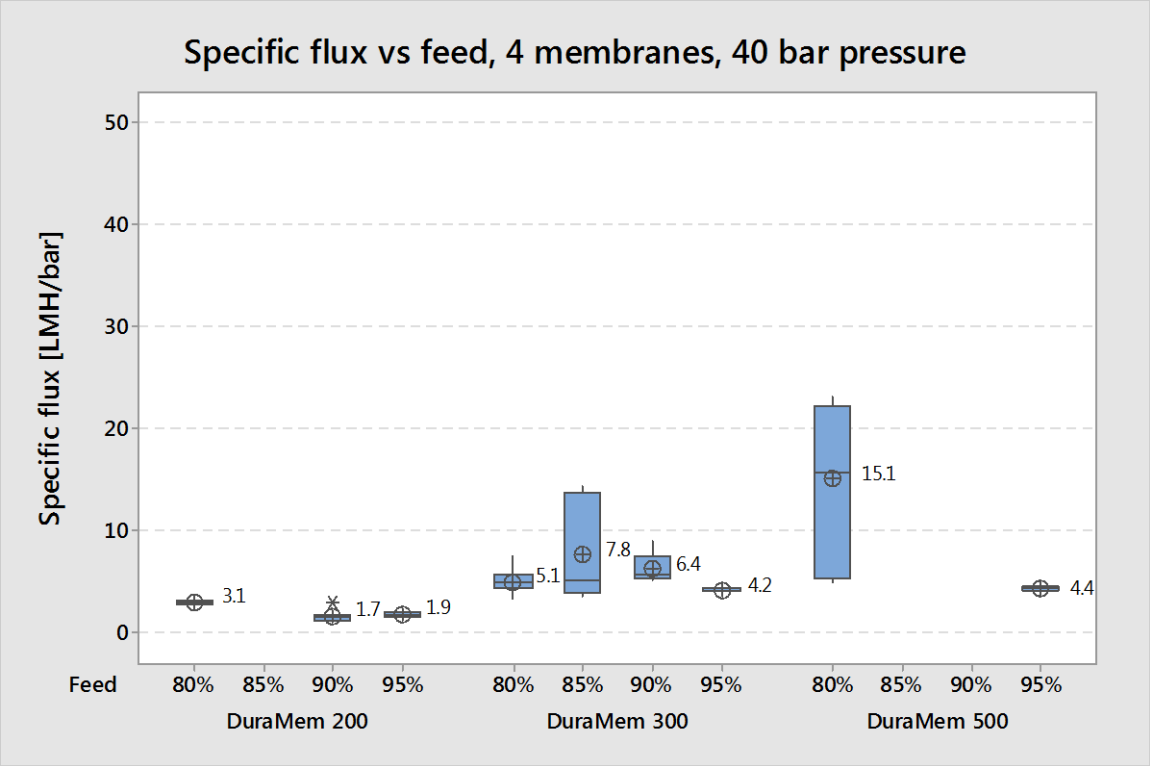
In addition to rejection, another important operational parameter for the evaluation of membrane performance is the specific flux through the membrane. Figure 23 and Figure 24 show the distribution of the average specific fluxes and their SDs depending from the type of feed for the three polymeric membranes.



* outlier symbol: an unusually large or small observation (Minitab, 2007)

Figure 23: Average specific flux distribution for the polymeric membranes with four types of feeds when 5 bar pressure was applied

By analyzing Figure 23 and Figure 24 it can be seen that the SD of the specific fluxes increased with increasing the MWCO of the membrane, for each feed respectively. Another important observation is that the average specific flux values are lower at higher pressure for all the three polymeric membranes. The lowest and most stable specific flux for each membrane was measured with 95 wt% EtOH feed. Also, in most of the cases it was noticed that when the EtOH concentration in the feed was decreasing the SD of the specific flux was increasing. This is opposite to the theory of the ethanol/ water mixture viscosity. According to Tanaka, Yamamoto, Satomi, Kubota, and Makita (1977), when the quantity of ethanol in the feed increases above 30wt% EtOH the viscosity decreases, meaning that with higher ethanol percentage the feed becomes less opposed to the relative motion between the two surfaces, in this case the feed and the membrane.



* outlier symbol: an unusually large or small observation (Minitab, 2007)

Figure 24: Average specific flux distribution for the polymeric membranes with four types of feeds when 40 bar pressure was applied

Additionally, the polymeric membrane is expected to react to the presence of organic solvent and it is expected to experience a structure modification, also called swelling in presence of ethanol. The apparent MWCO of a membrane can decrease as the swelling effect increase with an increase of ethanol percentage in the solution. This would be the explanation for the lower average specific flux during the tests when the EtOH concentration was higher with 95wt%.

If the flux distribution is analyzed by the type of membrane, it can be noticed that DuraMem 200 is the membrane with the lowest specific flux in both experiments with 5 and 40 bar. This is correlated to the MWCO which is the lowest from the group leading to the lowest specific flux. Nevertheless, this membrane offers the most stabile specific flux with the lowest SD from all the three membranes. Opposite, the membrane with highest SD of specific flux is DuraMem 500 with feed having 80 wt% EtOH in the two cases with different pressures.

Figure 23 and Figure 24 present the average specific flux values and cannot be used to analyze the timeline of the flux. Having this in consideration additional graphs will be

presented where the specific flux is given consecutively during time for each test done with specific membrane.

Figure 25 and Figure 26 present the specific flux in function of time for each test done with DuraMem 200. In Figure 25, it can be observed that the flux varies during time but when comparing the values of the first and last sample of a test it can be seen that in all cases the specific flux decreases. One exception is test 3 with feed containing 80 wt% EtOH. The specific fluxes in time for each sample from this test are 4.61, 4.39 and 5.20 LMH/bar. The specific flux of the last sample increased which might indicate damage of the membrane. Nevertheless, this cannot be confirmed because of the lack of additional samples which show the specific flux distribution afterwards.

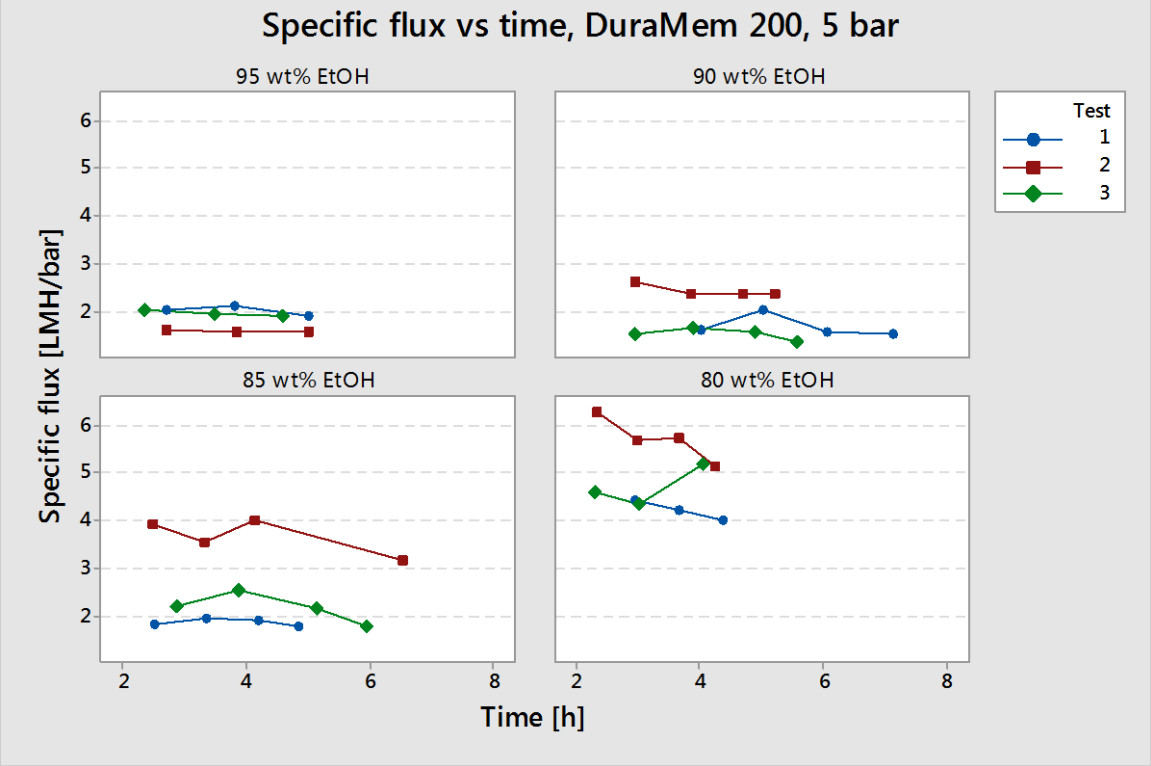


Figure 25: Specific flux behavior during time for each test done with DuraMem 200 depending on the type of feed when 5 bar pressure was applied

At a pressure of 40 bar DuraMem 200 shows a more consistent specific decrease, as shown in Figure 26. The reason might be because of higher interaction of the organic materials with the surface of the membrane due to higher pressure. Here also a deviation of the specific flux can be noticed for test number 3 when the ethanol concentration was 90 wt%. The values for the specific flux for each sample are as follows: 2.08, 2.05, 3.11 and 1.87 LMH/bar. The damage of the membrane is excluded as the reason, because the rejection returns to normal after the third sample. In this case, the high value of the specific flux might present an outlier. By

analyzing Figure 25 and Figure 26, it can be observed that the specific flux is increasing with decreasing of the amount of EtOH, which is to be expected based on the explanation provided previously.

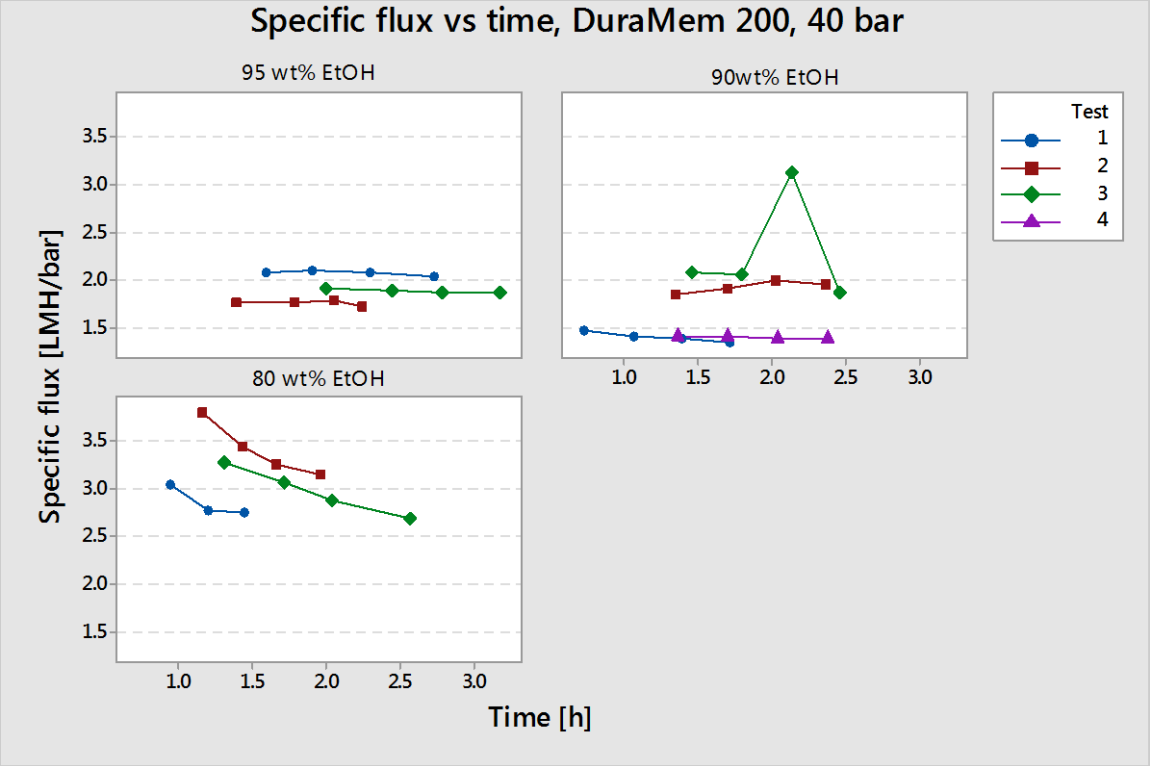


Figure 26: Specific flux behavior during time for each test done with DuraMem 200 depending on the type of feed when 40 bar pressure was applied

Figure 27 and Figure 28 show the specific flux in function of time for each sample of DuraMem 300 tested at 5 and 40 bar pressure respectively.

In tests 1 and 2 with the feed with 95 wt% ethanol when 5 bar pressure was applied a slow increase in the specific flux can be seen. Also, the same case is in test 3 with 95 wt% EtOH feed and test 1 when 85 wt% EtOH was used at 40 bar as presented in Figure 28. The rest of the tests show flux decrease during time. The reason might be because of the accumulation of materials are present in the feed on or within the membrane causing flux reduction.

In both experiments, where two different pressures were applied, the same observation can be delivered. The specific fluxes increase as the EtOH concentration decreases till the value of 85 wt%. Then, when the EtOH concentration reaches 80 wt% in the feed, the specific fluxes decrease to a value below to the one when 85 wt% EtOH feed was used. In this range of EtOH concentration other theory is applied, which says that the decrease of EtOH concentration to 20 wt% causes increase of viscosity (Tanaka et al., 1977). The increase of

viscosity causes higher opposition of the fluid to the relative motion in contact with the membrane, causing lower flux.

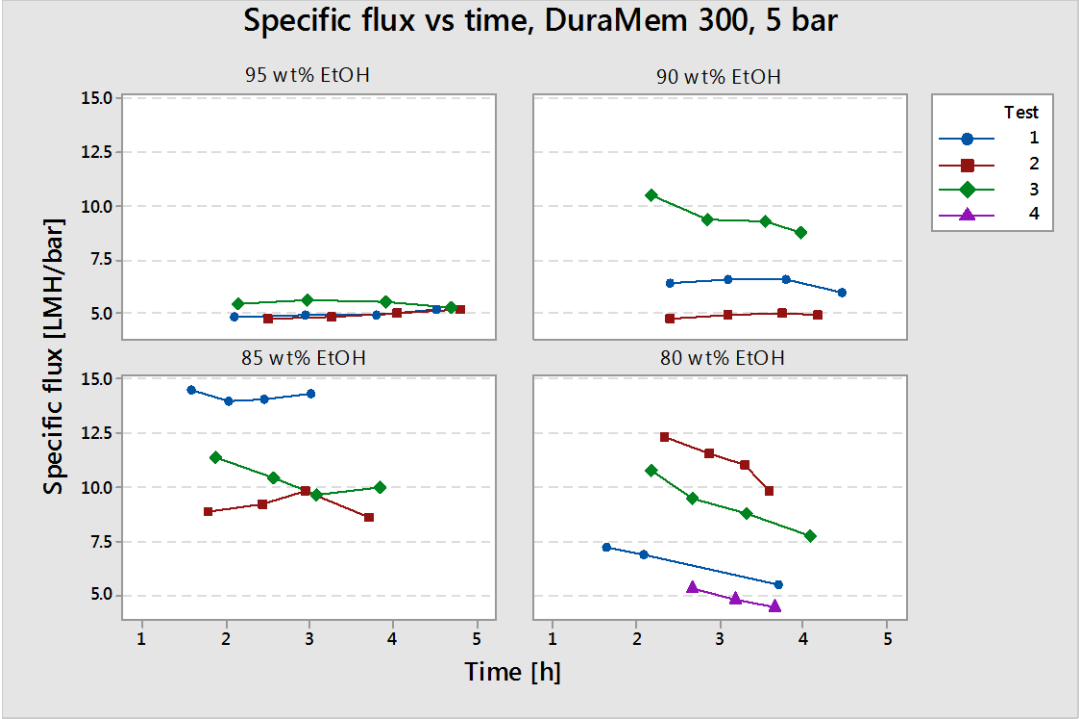


Figure 27: Specific flux behavior during time for each test done with DuraMem 300 depending on the type of feed when 5 bar pressure was applied

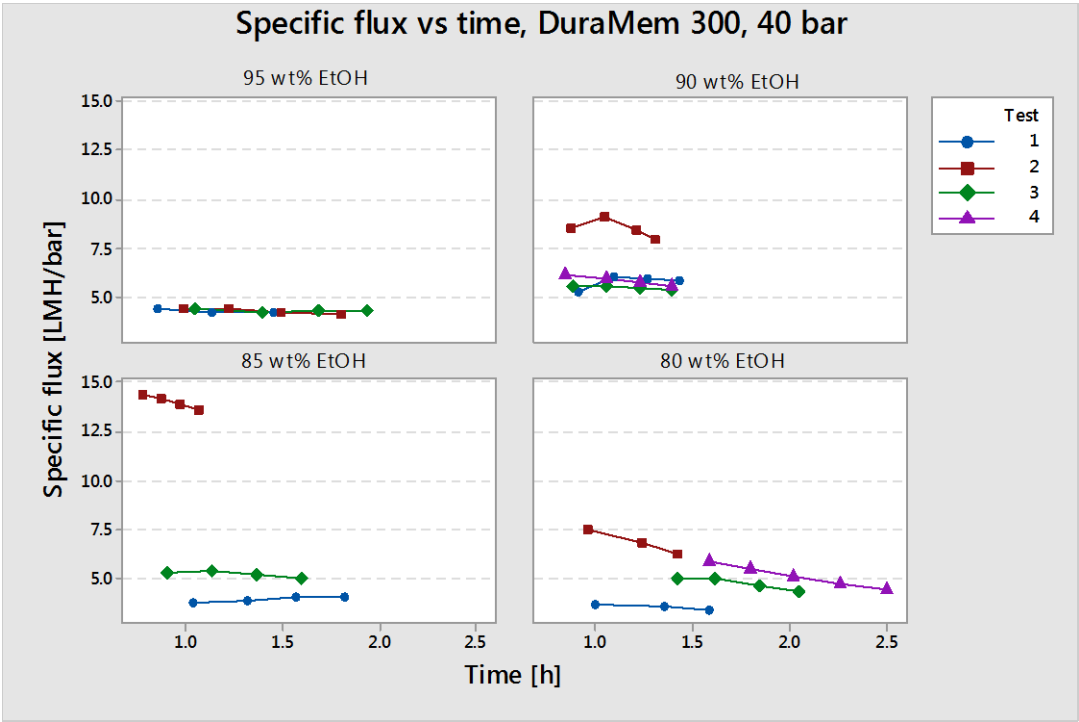


Figure 28: Specific flux behavior during time for each test done with DuraMem 300 depending on the type of feed when 40 bar pressure was applied

In the next Figure 29 and Figure 30, the same presentation of specific flux behavior during time is done, including all samples from the tests done with DuraMem 500.

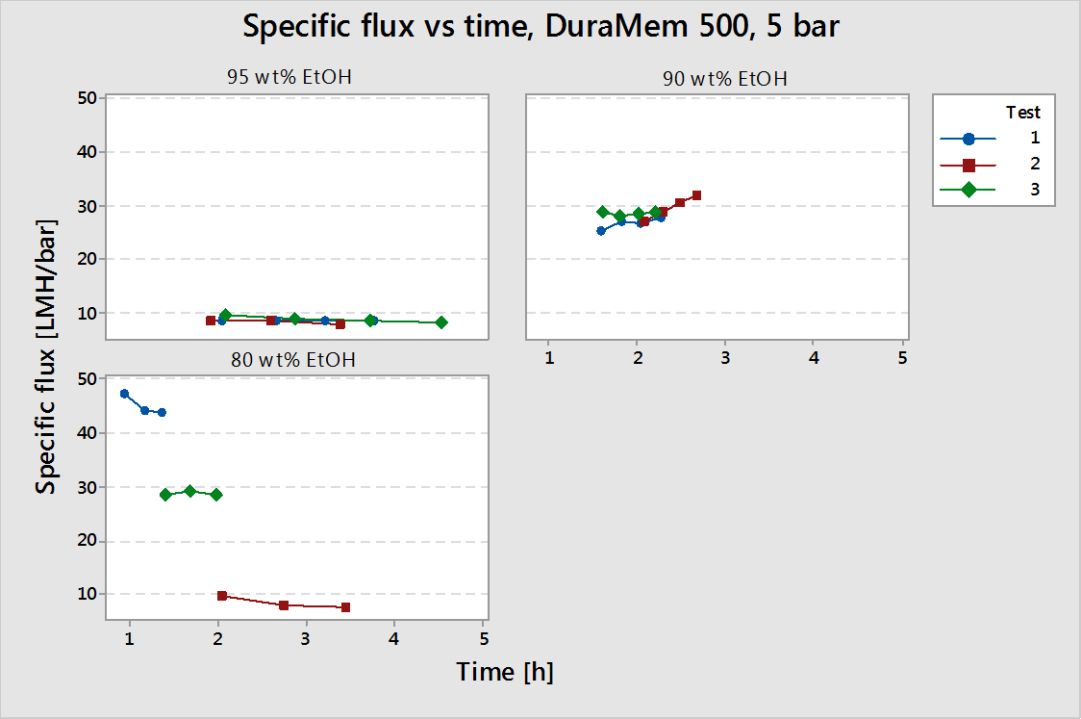


Figure 29: Specific flux behavior during time for each test done with DuraMem 500 depending on the type of feed when 5 bar pressure was applied

In the experiment when 5 bar pressure was applied each test delivers different behavior of the specific flux depending on the type of feeds. In two tests with feed containing 95 wt% EtOH the specific fluxes are decreasing, whereas in the three tests with 90 wt% EtOH in feed the specific fluxes are increasing. Latest, in the final case analyzed with feed containing 80 wt% ethanol the specific fluxes are constant. The observation from the three graphs presented when 5 bar pressure was used is that when using DuraMem 500 with different feeds the specific flux behavior is drastically different. The specific flux variations obtained with 95 and 90 wt% ethanol at 5 bar indicates that this pressure may be out of the optimal range for this membrane.

By viewing Figure 30, it can be seen that in the case when 40 bar pressure was applied during the tests, the specific fluxes decreased during time. When comparing Figure 29 and Figure 30 it can be observed that in the tests with 80 wt% EtOH feed the deviation between the tests is constant. Here again it is confirmed that the increase of pressure affects the specific flux inversely proportionally.

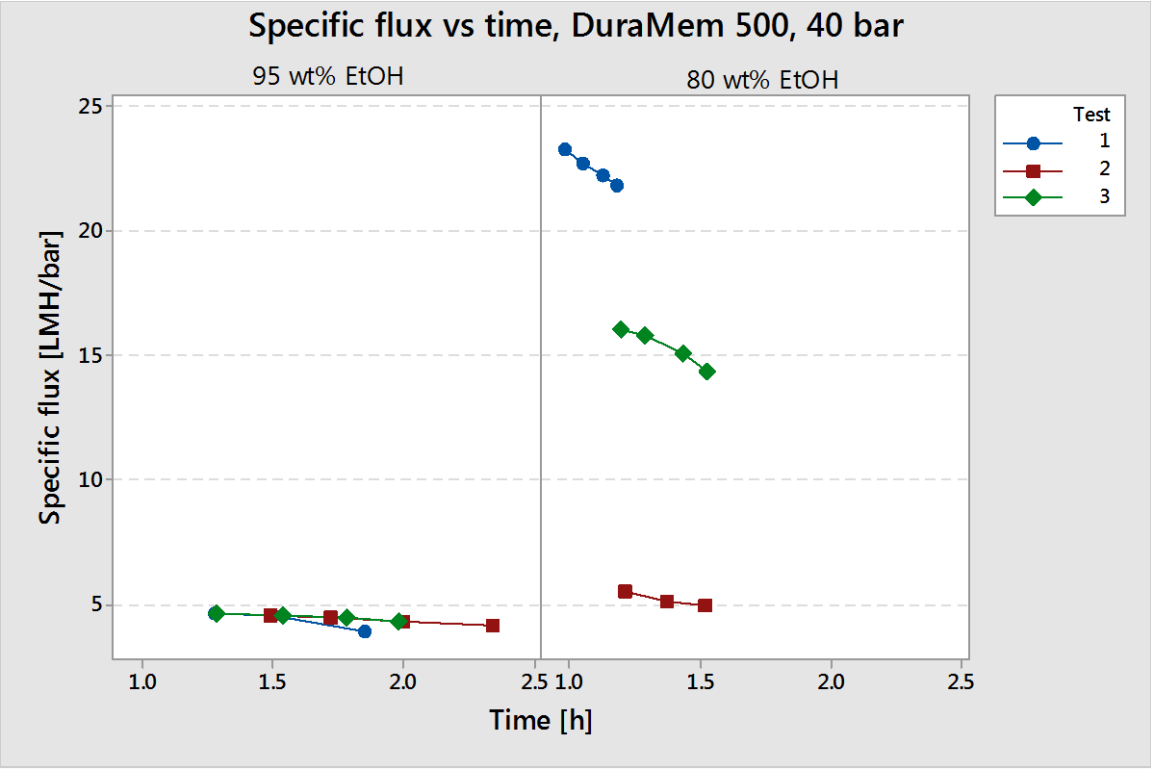


Figure 30: Specific flux behavior during time for each test done with DuraMem 500 depending on the type of feed when 40 bar pressure was applied

The range of specific flux observed during the separate tests with DuraMem 200, 300 and 500 may indicate variation in the membrane material. The coupon used for the experiment had a surface area of 12,5 cm^2 therefore a commercial module may produce a specific flux within the range observed.

The next section presents the rejection of phospholipids in function of the specific flux. In the previous section, we have seen that for the same membrane type different specific fluxes could be registered. Therefore, analyzing the rejection in function of the specific flux could provide additional information regarding the performance of the selected membrane.

Figure 31 and Figure 32 below provide the rejection distribution depending from the specific flux for DuraMem 200. The numbers on each of the points in the graphs are the sequence numbers of the samples.

By analyzing Figure 31, it can be seen that higher rejection was obtained when the specific flux was lower, with exception to the case using feed containing 85 wt% EtOH. This may be an indicator that the flux influences inversely proportional the level of rejection of the membrane. Nevertheless, this statement is still to be confirmed. Also noticed, is that when the specific flux was below 2 LMH/ bar the rejection was above 95%. From Figure 31 it can be observed that flux increase causes increase of the SD of the rejection. The SD of the rejection

with feeds containing 95 and 80 wt% EtOH is greater when 5 bar pressure was applied due to decreased compaction of the membrane compared to the case with 40 bar. This causes the membrane structure to be less homogenous producing higher performance variation. Additional indicator would be that the 5 bar pressure applied is outside the recommended operational range contributing to the unstable membrane performance.

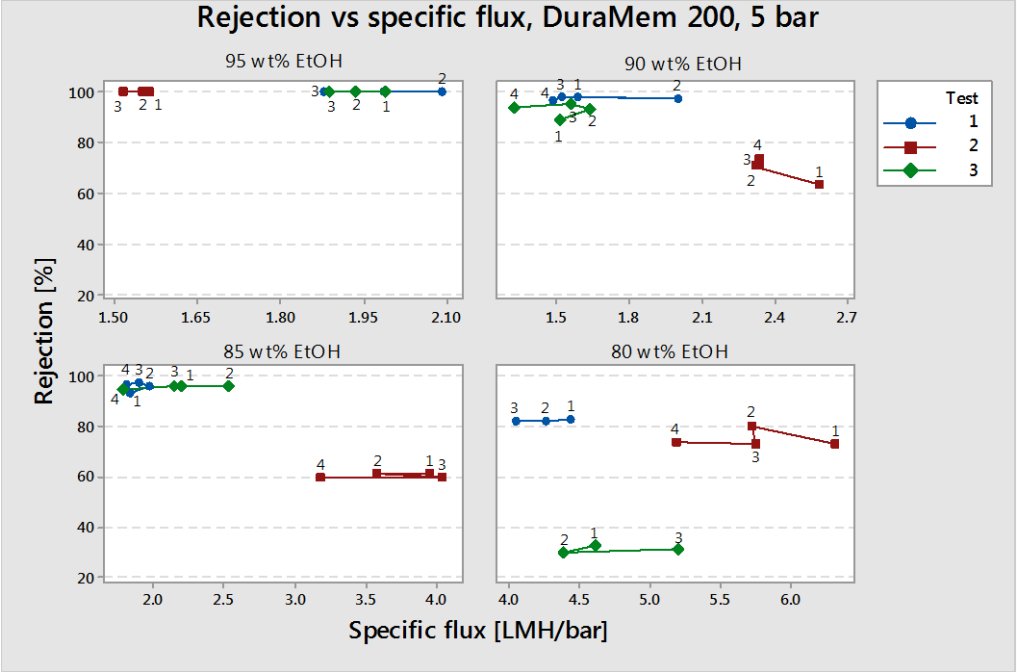


Figure 31: Dependence of the rejection from the specific flux in all the tests with DuraMem 200 when 5 bar pressure was applied

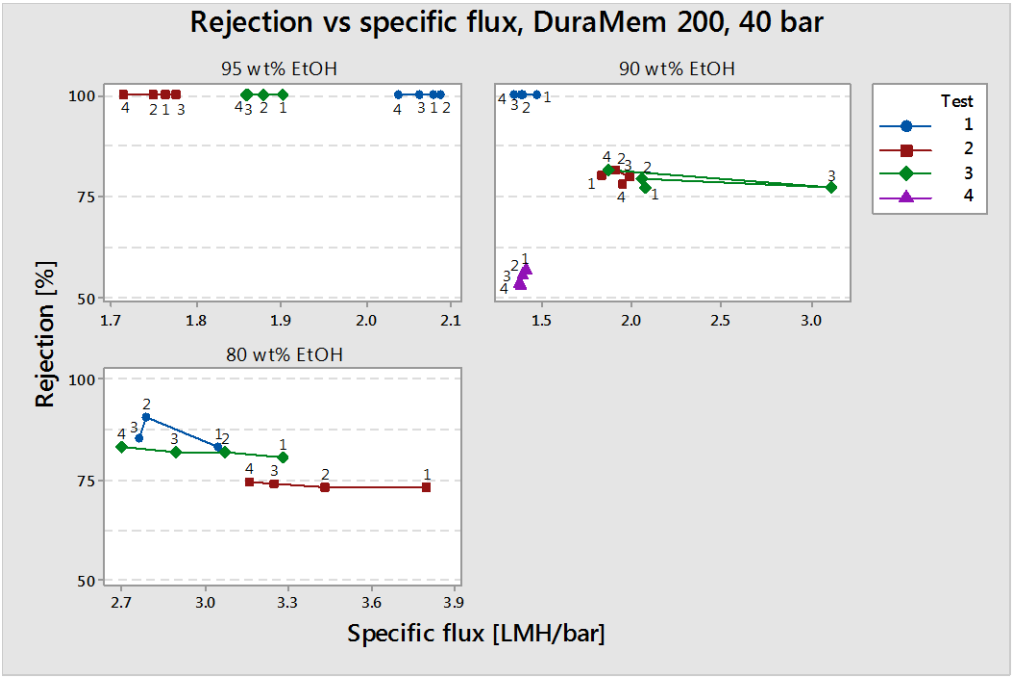


Figure 32: Dependence of the rejection from the specific flux in all the tests with DuraMem 200 when 40 bar pressure was applied

From Figure 32 it can be observed that when the specific flux does not exceed the value of 4 LMH/ bar, the rejection does not decrease below 50%. The highest rejection performance is noticed to be when the specific flux was below 2 LMH/ bar, with exception to test 4 with feed containing 90 wt% EtOH. This situation might be due to clogging of the membrane resulting in reduced performance. When the specific flux was in the range of 2 to 4 LMH/ bar the rejection was around 75%.

The rejection in function of the specific flux for DuraMem 300 at 5 and 40 bar is presented in Figure 33 and Figure 34.

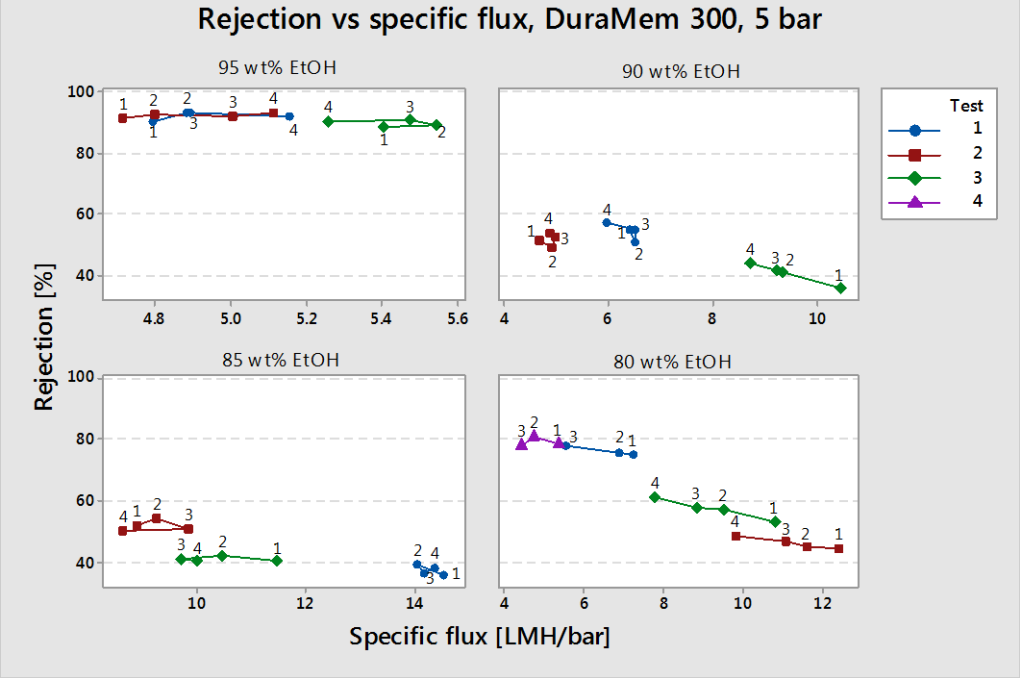


Figure 33: Dependence of the rejection from the specific flux in all the tests with DuraMem 300 when 5 bar pressure was applied

Based on Figure 33, it can be noticed that constant decrease of the specific flux was obtained when feed with 80 wt% EtOH was used, which also contributed to increase in rejection. Whereas, when the rest of the feeds were used, the specific fluxes were varying in narrow range delivering constant rejection.

The correlation that can be derived from Figure 34 is that in most of the cases the decrease of flux causes increase of the rejection. The situation with the feed with 80 wt% EtOH is similar if compared to the rejection results in the experiment where 5 bar pressure was used. The lowest rejection values were obtained by the membranes when the specific flux was high and constant, as in test 1 and 2 with the feed having 85 wt% EtOH when 5 and 40 bar was applied, respectively. From both previous graphs, it can be noticed that the highest rejection was

obtained with the lowest specific flux. Also, the highest rejection having lowest SD was obtained when the flux variation was the lowest which in both cases was with the feed having 95 wt% EtOH.

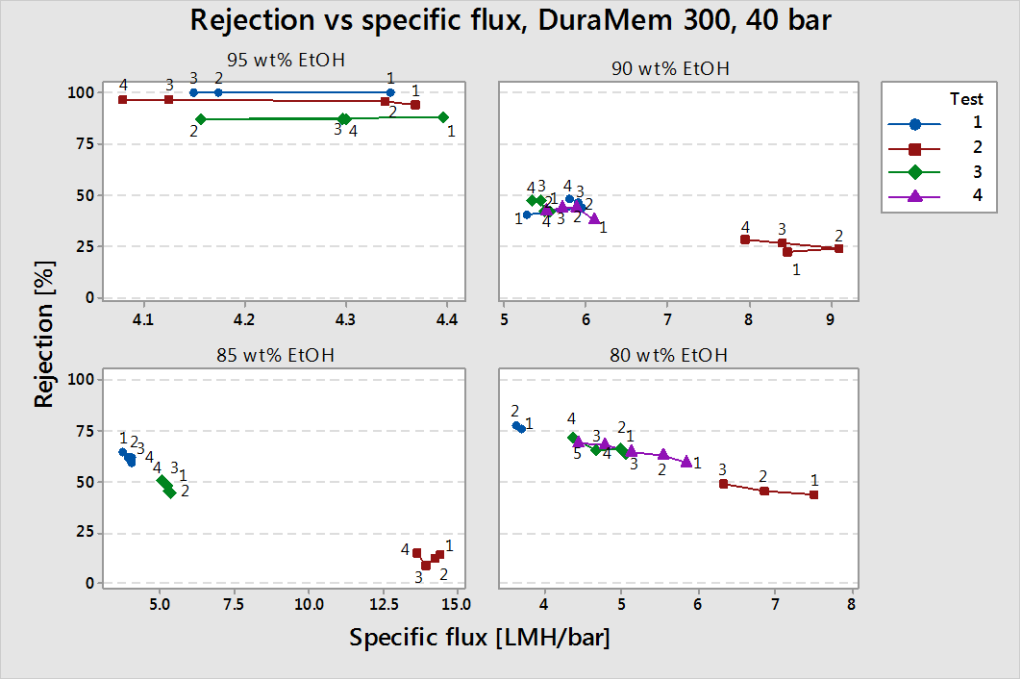


Figure 34: Dependence of the rejection from the specific flux in all the tests with DuraMem 300 when 40 bar pressure was applied

The last graphs providing the correlation between the rejection and the specific flux for DuraMem 500 are presented in Figure 35 and Figure 36.

From the two graphs it can be seen that in the case when 80 wt% EtOH feed was used the high difference between the specific fluxes in the tests did not influence the level of rejection. The average rejection when 5 bar pressure was applied was 40%, and with 40 bar was 34%. Whereas, the double decrease of the specific flux with 95 % EtOH when 40 bar pressure was applied caused increase of the rejection of around 20%.

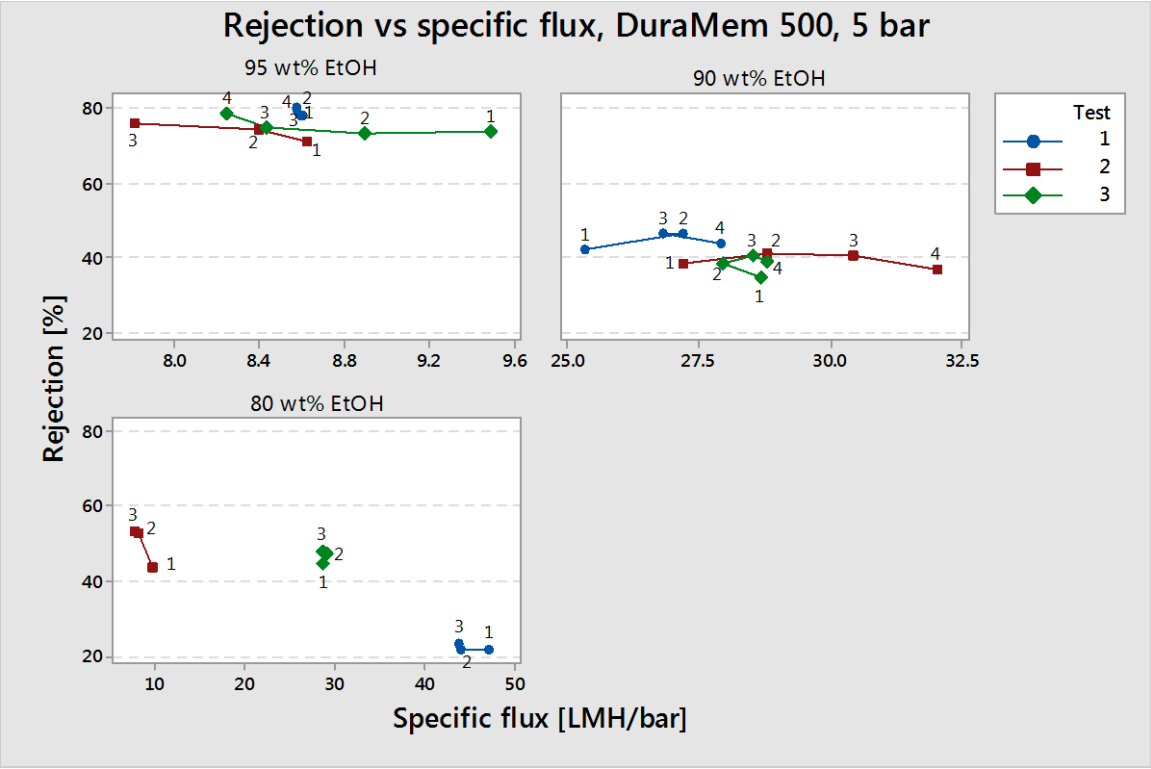


Figure 35: Dependence of the rejection from the specific flux in all the tests with DuraMem 500 when 5 bar pressure was applied

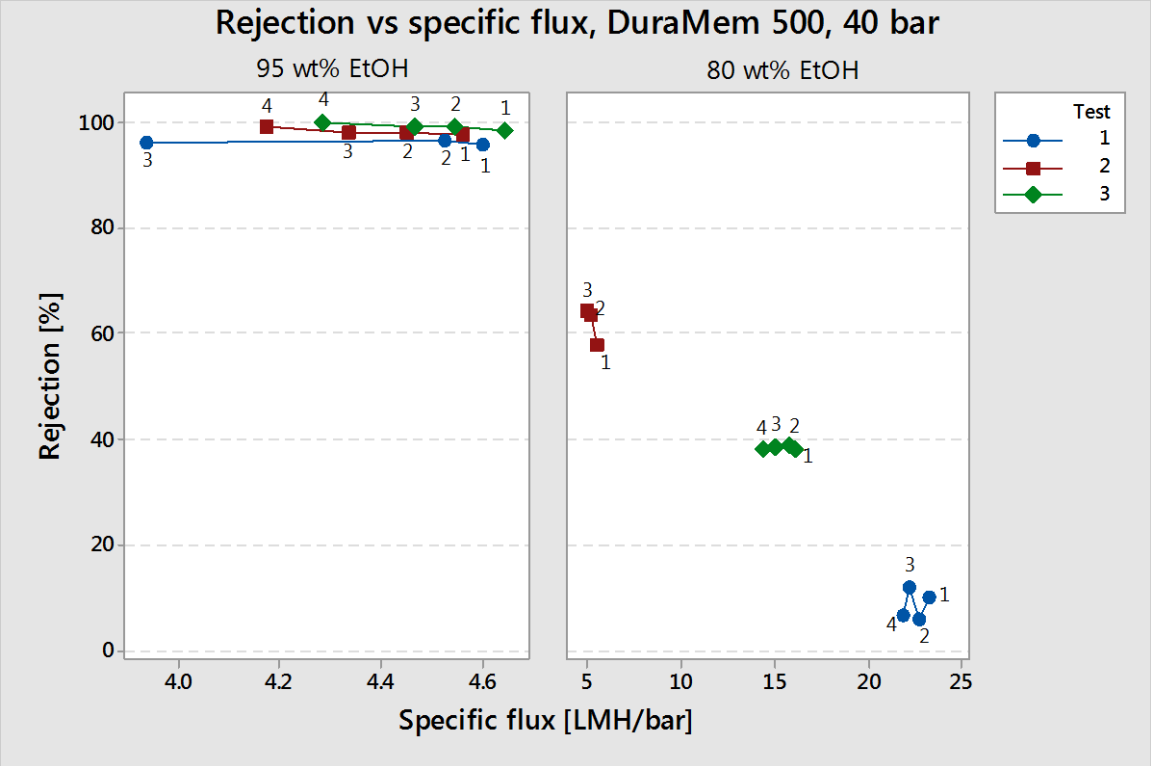


Figure 36: Dependence of the rejection from the specific flux in all the tests with DuraMem 500 when 40 bar pressure was applied

In the tests with DuraMem 300 another situation was observed, which is dependency between the rejection and the condition of the feed (cold versus warm). The feed was considered to be

cold when the experiment with the specific membrane was begun with fresh feed from fridge at 4 °C. Whereas, when the next experiment with new membrane continued to use the same feed as in the previous experiment, the feed was considered warm at approximately 20 °C. From these tests it appeared that the obtained rejection was higher when the feed was cold. This can be seen in the Figure 37 below. This phenomenon is in accordance with the theory (Tanaka et al., 1977), considering that as the temperature increases the viscosity of the ethanol/ water mixture decreases providing higher specific flux through the membrane. Based on the previous observations, when the specific flux was higher the rejection was lower. Nevertheless, this phenomenon cannot be delivered as conclusion because it was not observed for the other membranes and it should be further investigated.

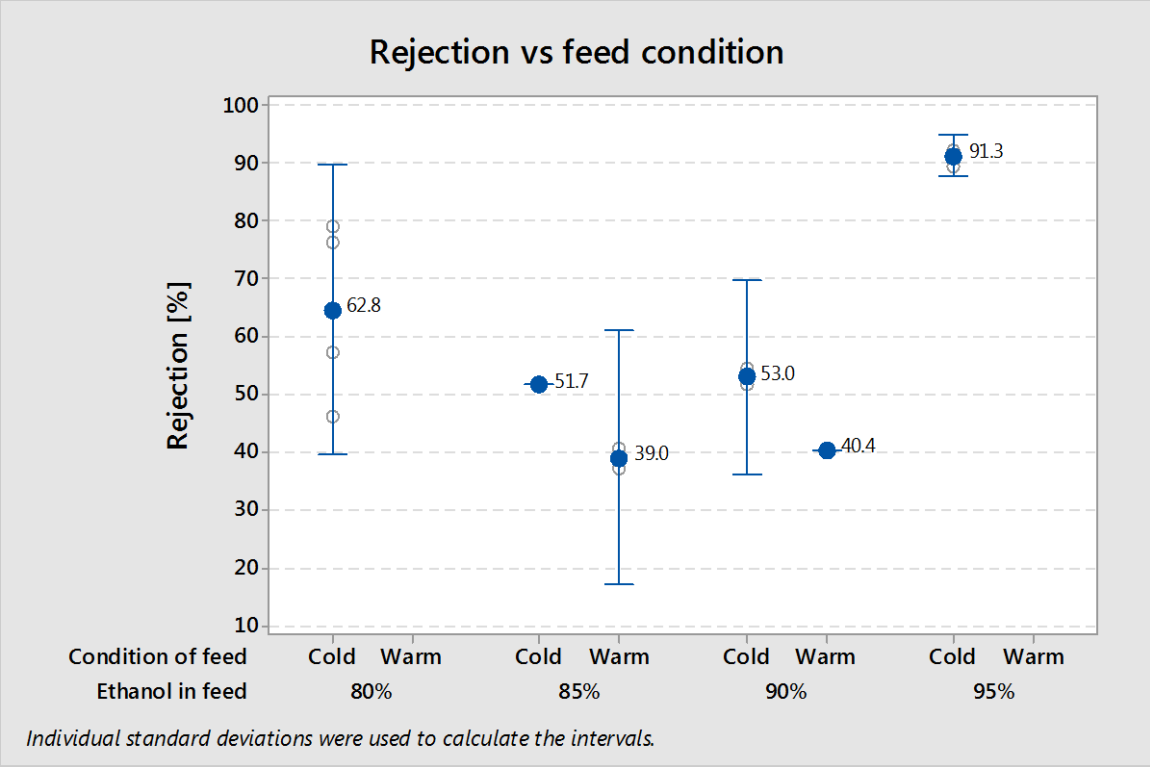


Figure 37: Rejection dependency from feed temperature in the case of DuraMem 300 when 5 bar pressure was used

Additional analysis that was done about the rejection of each membrane was its correlation with the conditioning time of the tests. Conditioning time is the time from the moment of stabilization of the membrane flux till taking the first sample. The two graphs representing the correlation between the conditioning time and rejection when 5 and 40 bar pressures were used are given in Appendix 6.

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5.1.2. Performance parameters for Pervatech membranes

In order to have wider range of choice of membranes which would meet the industrial application requirements another type of membrane was tested. This type of membrane was the ceramic membrane. In Table 9 below the rejection percentage and the SDs of the rejection for all ceramic membranes which were tested under 5 bar pressure is given. The performance parameters of all ceramic membranes are provided in Appendix 7.

Table 9: Phospholipids rejection obtained by ceramic membranes at 5 and 40 bar and four ethanol/water compositions expressed in percent [%]

Membrane type	**Ethanol/ water composition [wt%]**	
	95 / 5	**80 / 20**
Pervatech 300-200 CM	na	20 ± 5
Pervatech 500-400 CM	36 ± 5	36 ± 6
Pervatech 500D coated	na	17
Pervatech 700D coated	na	8
Silane	na	25 ± 7

na: data not available

The same representation given in Table 9 is done graphically. The rejection distribution for all ceramic membranes using the type of feed with 80 wt% EtOH is given in Figure 38. As it can be observed the average rejection values for all ceramic membranes are below 40%, which are lower from the ones prescribed in the designated criteria.

Figure 39 below represents the values for the rejection only for the Pervatech 500-400 with the two types of feeds tested, containing 80 and 95 wt% EtOH. Again, in both of the tests the rejection is below 40%.

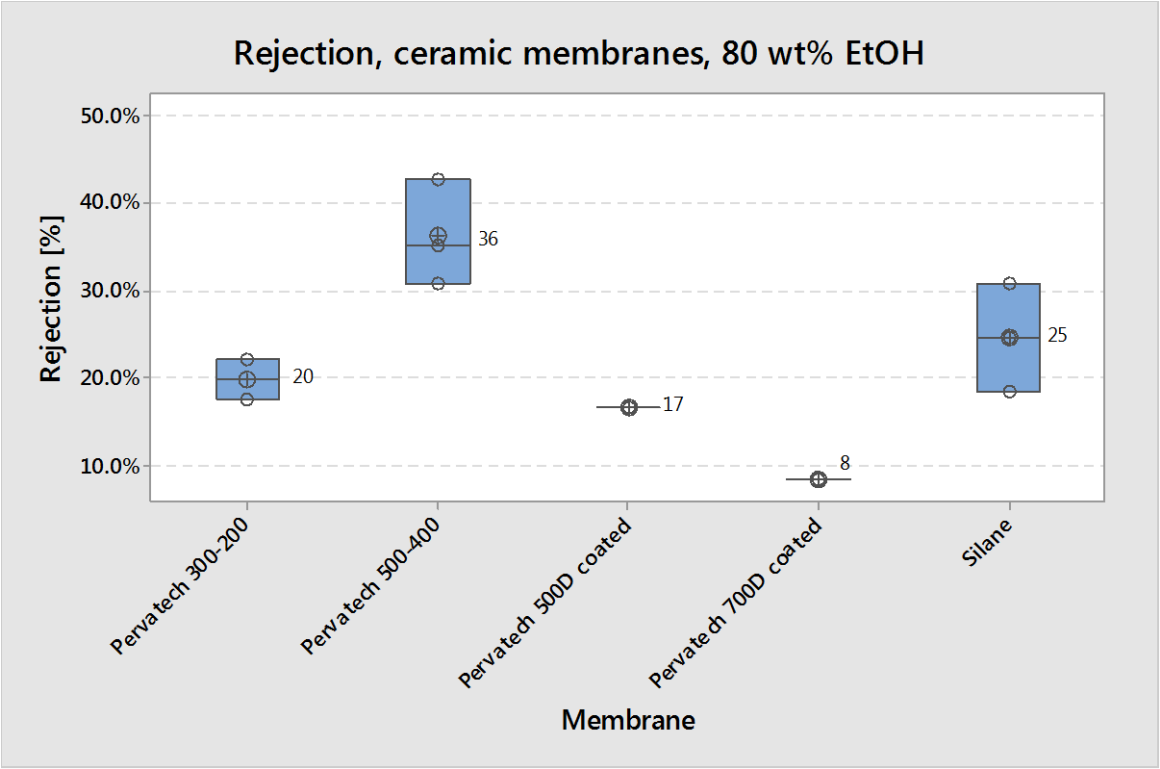

Figure 38: Rejection values for all tests done with the ceramic membranes using feed with 80 wt% EtOH

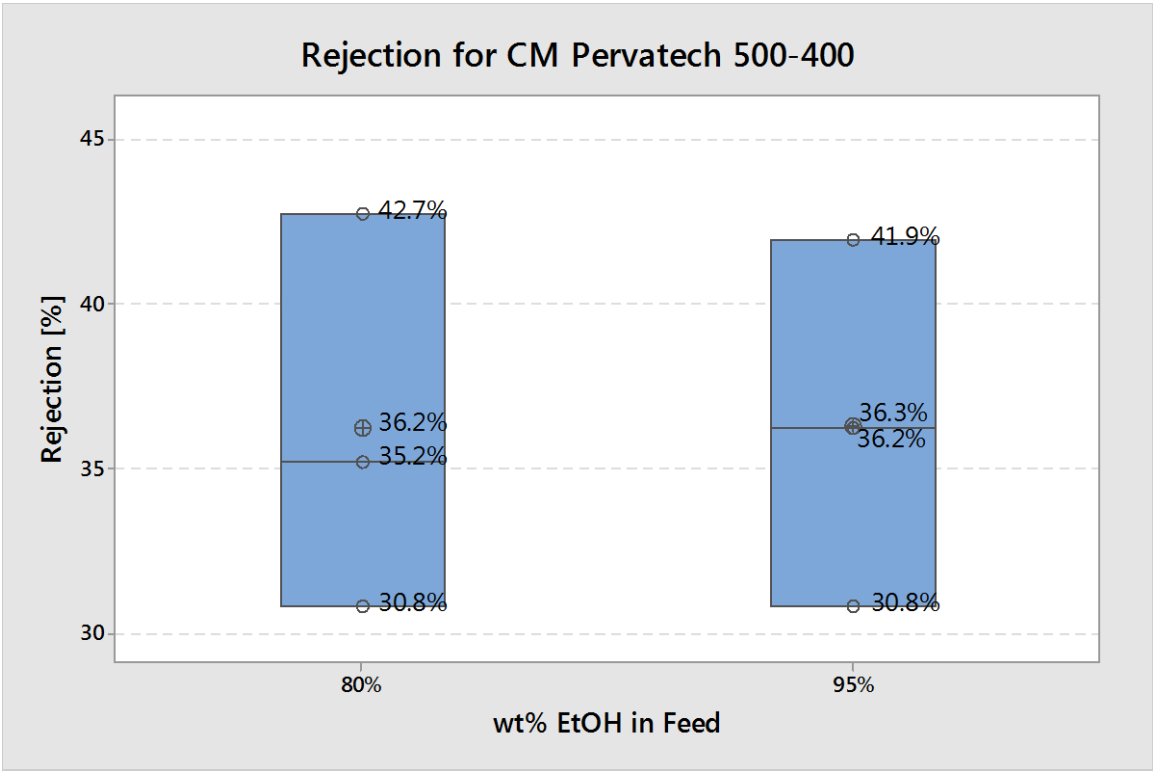

Figure 39: Rejection values for the tests done with Pervatech 500-400 using feed with 90 wt% and 80 wt% EtOH

6. Conclusions

Membranes that gave PLs rejection results in the prescribed range from 80% to 95% are DuraMem 200 and 300. These two polymeric membranes obtained the desired rejection values for different types of feed. DuraMem 500 obtained rejection outside the prescribed limits for the conditions investigated. All rejection values delivered from the tests with 5 bar pressure were below the level of 80%, as the results from the tests with 40 bar pressure with an exception of one test, which gave rejection above 95%. Accordingly, DuraMem 500 will not be considered for future investigation.

DuraMem 200 delivered the best rejections when using the feeds with EtOH concentration of 90, 85 and 80 wt%. When the EtOH concentration was 80 wt% the rejection of DuraMem 200 was 79 ± 4% and 80 ± 6% at a pressure of 5 and 40 bar respectively. With this concentration of EtOH the highest average specific flux was obtained, 5.0 LMH/ bar. With feed having 85 wt% EtOH the rejection calculated at 5 bar pressure was 84 ± 17%. The highest rejection was obtained with 5 bar with the feed with EtOH concentration of 90 wt%, amounting in 87 ± 13%. With 40 bar and same ethanol composition the rejection was 78 ± 17%. It can be noticed that at pressure of 5 bar the best rejection values are also associated with the highest standard deviation.

On the contrary, DuraMem 300 showed best rejection with the feed containing 95 wt% EtOH. For DuraMem 300 the rejection obtained with this feed was 91 ± 2% with 5 bar pressure and 94 ± 5% with 40 bar. As observed, these values are on the upper limit of the defined criteria and have the lowest SD. The average specific fluxes are 5.1 and 4.2 LMH/ bar for each pressure, respectively.

If a membrane should be chosen it would be DuraMem 300, used with feed containing 95 wt% EtOH at both 5 and 40 bar pressure because it obtained lowest SD and highest specific flux. DuraMem 200 delivered rejection with high SD with both feeds with 90 and 85 wt% EtOH. Even though the feed with 80 wt% EtOH concentration would be the best option, having in consideration the financial benefits when reducing the amount of EtOH, the rejection delivered was on the lower limit of the range, and even below considering the SD.

The general conclusions for the flux and rejection distribution are provided from the results, which were taken into consideration for the selection process of the membrane. The specific flux distribution is based on these correlations:

1. It decreases when the pressure increases;
2. It decreases during time;
3. The membrane with smallest MWCO has the lowest flux; and
4. It is stabilizes when the EtOH concentration in the feed increases.

The dependency of the rejection from the specific flux can be presented by delivering the following conclusions:

1. The rejection is increasing when the flux is decreasing;
2. The highest rejection is obtained with the lowest flux;
3. The highest rejection is obtained by the membrane with the lowest MWCO (or pore size); and
4. When the flux during the test is constant, the rejection is also constant.

The above listed findings refer to the correlation between the time, flux and rejection. Whereas, below are provided some general conclusion obtained from the summarized results.

1. The duration of the conditioning time of the experiments does not influence the level of rejection. Whereas, the conditioning time using 40 bar pressure is shorter than using 5 bar pressure.
2. The pressure applied on the membrane does not influence the level of rejection delivered by that membrane. More precisely, the increase in pressure does not increase the rejection percentage.
3. The pressure increase delivers faster membrane flux stabilization.

7. Further work

One of the steps for improvement of the membrane performance is upgrading the bench-scale equipment currently used. This could be done by small investment costs, through installing device for measuring the feed temperature and a pressure regulator. The certainty of these two parameters would contribute to the accuracy of the rejection results obtained by the specific membrane.

The rejection tests were done with intention to use the membrane in NF process for rejection of omega-3 PLs in a real fish oil production company, so the next step would be scaling of the process. This step would bring additional change which would be implementation of the spiral wound membrane due to the proportions of the equipment. This step is important in order to see if the selected membranes would meet the requirements for rejection of PLs and TGs in real scale and if the results obtained during the filtration experiments reflect the actual situation. If the answer turns out to be positive this kind of small-scale experiments could be reliable and used for pre-application of a membrane in the industrial process.

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Appendix 1: DuraMem 500 flat sheet

The packing of DuraMem 500 is provided in Figure 40, whereas the flat sheet of the specific membrane is given in Figure 41.

Figure 40: Packing for the polymeric membrane DuraMem 500

Figure 41: Flat sheet of the polymeric membrane DuraMem 500

Appendix 2: Rotary evaporator

The operating panel of the rotary evaporator is given in Figure 42, whereas the other part of the instrument is shown in Figure 43. During the experiments executed as part of the project done for this master thesis this instrument was used for mixing the feed during its preparation process, and for recycling of ethanol.

Figure 42: Operating panel of Heidolph rotary evaporator

Figure 43: Main equipment of Heidolph rotary evaporator

Appendix 3: UHPLC instrument - Principle of working

The UltiMate® 3000 Quaternary Analytical system uses Chromeleon Chromatography Data System. This instrument which is used for UHPLC analysis is composed of several parts:

- Pump,
- Autosampler,
- Column compartment and
- UV detector (lamp).

The pump is composed of 4 solvent channels with integrated four-channel degasser. The flow rate range is from 8 to 200 mL/min, the optimal pressure range is up 620 bar, but it could go up to 800 bar. The flow accuracy of the pump is ± 0.1%. The four-solvent quaternary pump is shown in Figure 44, whereas the parameters input to the Chromeleon Data System are given in Figure 45.

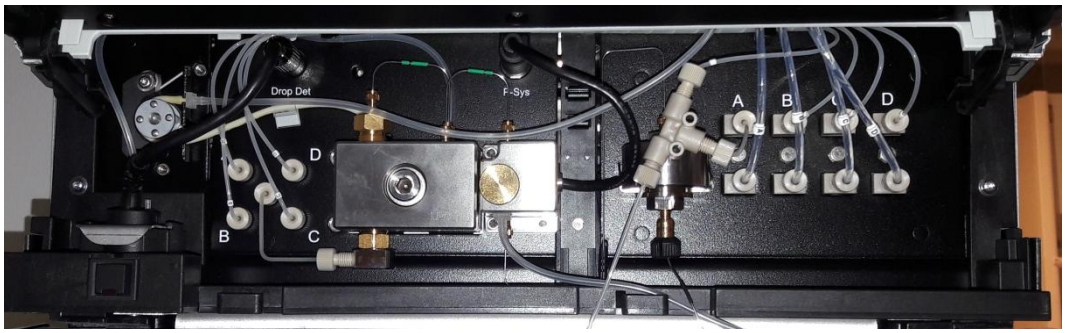

Figure 44: Pump compartment of the UHPLC instrument

In the experiments two solvents were used, ethanol with 3 lines that intake 30% solvent and water with 1 line that intake 10%. The four solvents entering the pump were mixed together to form a solution which then was pumped at pressure of 670 bar to the degasser. This solution (composed of ethanol and water) was used for normal operation of the pump, or precisely for directing and transporting the samples to the column. The degasser was used for eliminating the bubbles in the lines which might have appeared during the transport of the liquid because of high pressure.

The sampler was used for precise and accurate injections of the defined volume of liquid. The injection volume range of the sampler is from 0.01 to 100 μL, the injection volume accuracy is ± 0.5% at 50 and 90 μL. The injection cycle time is < 15s for 5 μL. In these experiments the amount of fluid injected by the sampler was 1 and 3 μL, which can be seen in Figure 46.

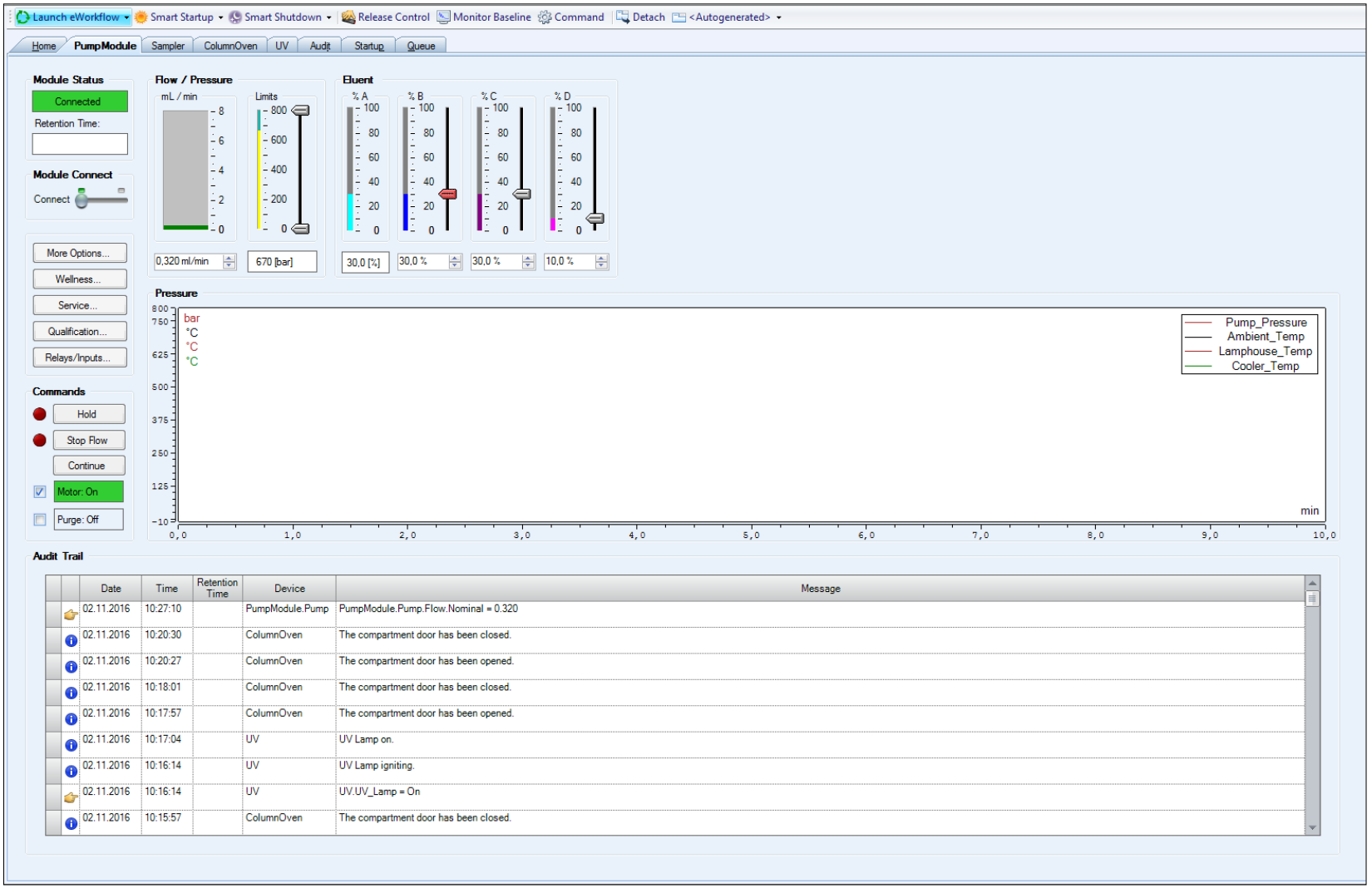

Figure 45: Parameters for the pump input in the Chromeleon Data System

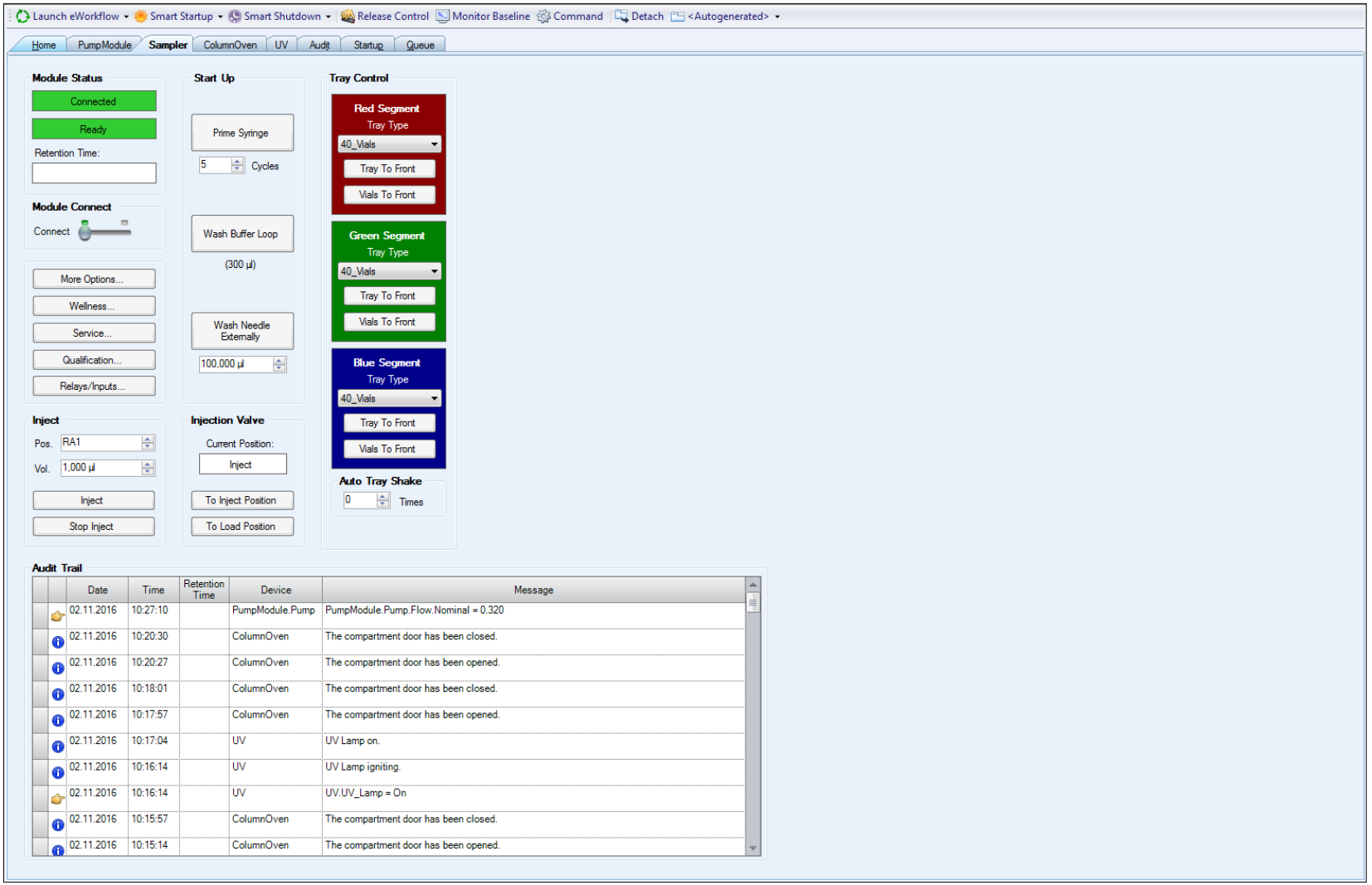

Figure 46: Parameters for the sampler input in the Chromeleon Data System

The sampler compartment is given in Figure 47.

Figure 47: Sampler compartment of the UHPLC instrument

The thermostatted column was based on silica gel packing material. The compartment contained two field-upgradable column switching valves. It used fan-based forced-air design to provide efficient cooling and heating, when changing the set temperature or when opening the front door. The column compartment is shown in Figure 48.

Figure 48: Column compartment of the UHPLC instrument

The optimal temperature range was from 5 to 80 °C, but it could obtain temperature up to 110 °C. The temperature accuracy was ± 0.5 °C. During the tests the temperature control was on and the temperature was set to 35 °C. The rest of the parameters inserted in the system are given in Figure 49.

After obtaining the set temperature, the sampler injected the defined amount of the sample to the pre-column and then to the main column. The reason for installation of the pre-column was in order to protect the main column, for example, from dirt in the sample which would lead to malfunction of the main column. The pre-column is cheaper and easier to repair. The pre-column was apolar, whereas the main column was polar and porous containing silica substrates inside the core. The polarity of the column lead to separation and binding of the organic molecules from the permeate and their detection using the UV detector. For example, the PLs are polar so they were attracted by the column and their movement was slowed down so the UV detector could easily detect them. On the other side, the fats are non polar and were not attached by the column. Nevertheless, some of the fats are bound to the PLs and the size detected would be for the both molecules, whereas some of them exist freely.

The UV detector covers a wide detection range from 190 to 800 nm. The maximum data collection rate is 100 Hz. In the experiment wavelength of 210 nm and rate of data collection of 2.5 Hz were used, as it can be seen from Figure 50.

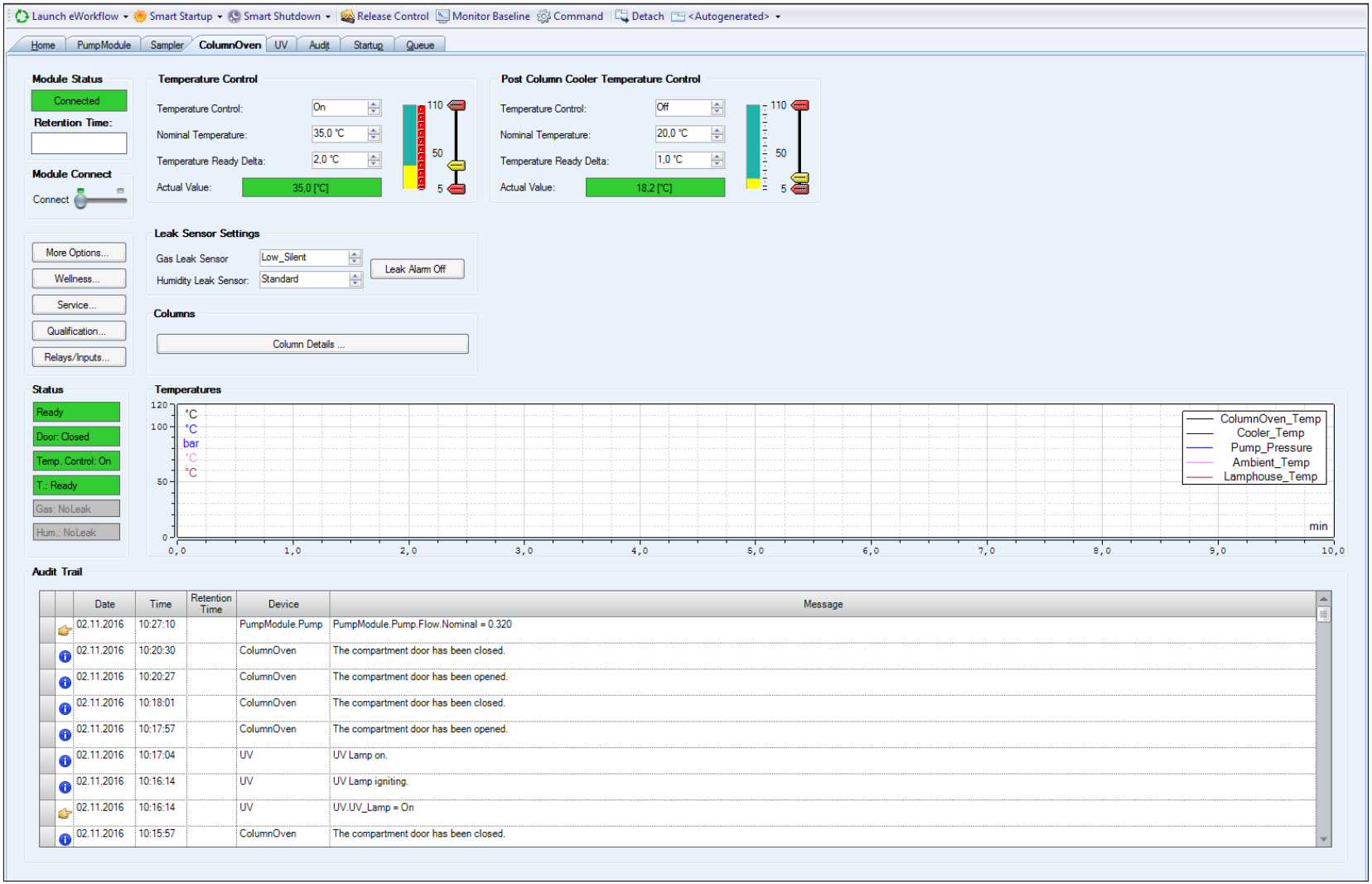

Figure 49: Parameters for the column input in the Chromeleon Data System

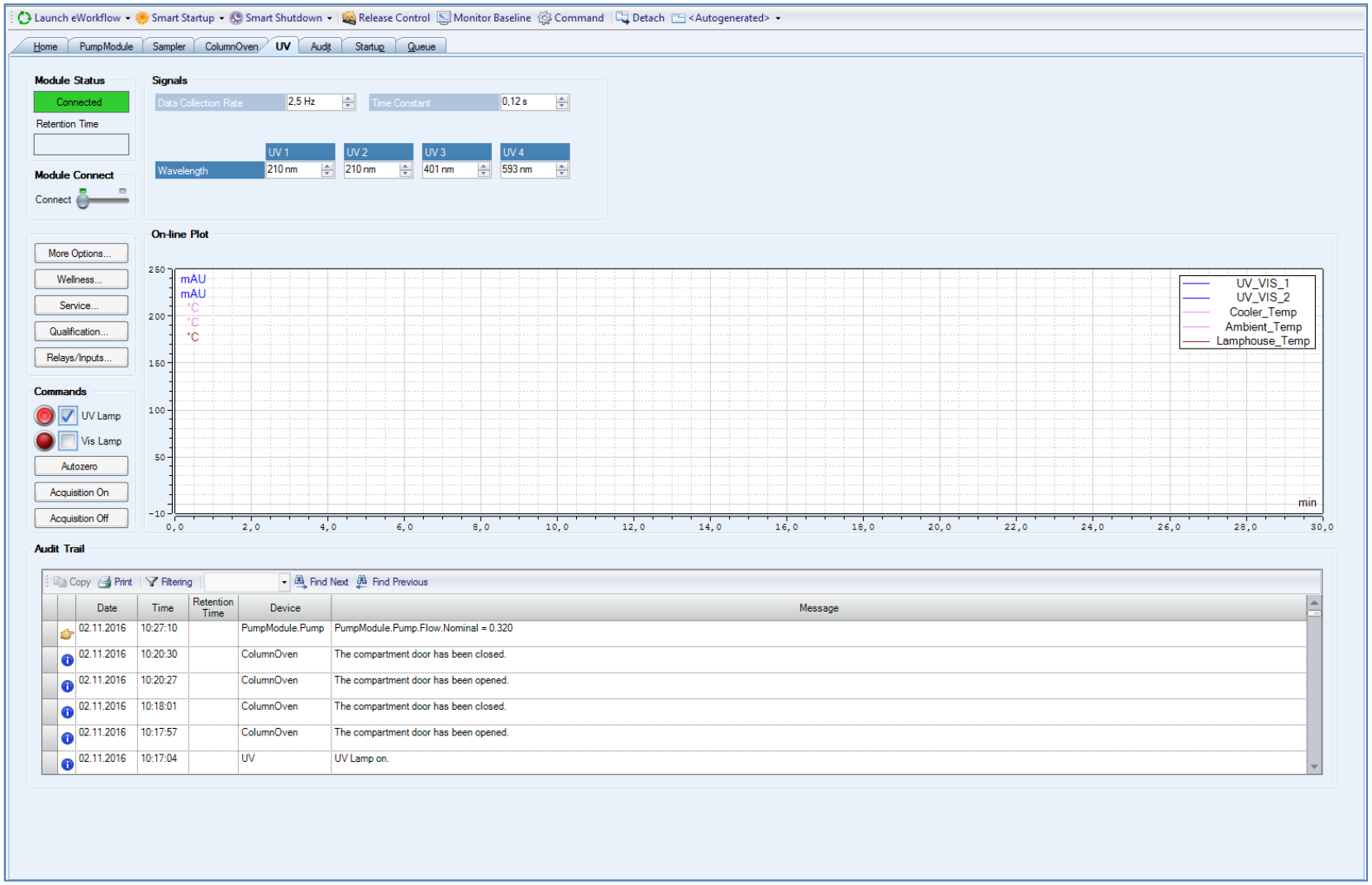

Figure 50: Parameters for the UV detector input in the Chromeleon Data System

Appendix 4: Performance parameters for polymeric membranes at 5 bar pressure

The raw data obtained during the nanofiltration experiments by DuraMem polymeric membranes when 5 bar pressure was applied is provided in Table 10.

Table 10: Performance parameters of the polymeric membranes from the nanofiltration experiments at 5 bar pressure with four types of feed

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s* bar]	Specific flux [LMH/bar]	Density
95	DuraMem 300 (T1) - M255	Cold	002142	Feed 4	70.6509	69.2904	/	/	/	/	/	/	/	/	/	/
			002142	Conditioning	/	/	/	/	/	5	4869	/	/	/	/	/
				1	7.2001	6.2665	89.8	91.0	92.1	5	2721	2.1	3638	0.27	4.80	0.80705
				2	4.5112	4.9793	93.6	92.8		5	3039	3.0	4140	0.27	4.89	0.80705
				3	4.7823	5.1076	93.2	92.6		5	3087	3.8	4214	0.27	4.90	0.80705
				4	5.5832	5.9866	92.1	91.4		5	2581	4.5	3710	0.29	5.16	0.80705
		Cold	002143	Feed 4	74.7797	73.3942	/	/	/	/	/	/	/	/	/	/
			002143	Conditioning	/	/	/	/	/	5	4991	/	/	/	/	/
				1	7.0718	5.8894	90.5	92.0	92.2	5	4000	2.5	5257	0.26	4.71	0.80705
				2	4.5422	6.5203	93.9	91.1		5	2798	3.3	3743	0.27	4.80	0.80705
				3	6.7140	5.2035	91.0	92.9		5	2771	4.0	3868	0.28	5.01	0.80705
				4	6.0439	4.1979	91.9	94.3		5	2812	4.8	4010	0.29	5.12	0.80705
		Cold	002144	Conditioning	/	/	/	/	/	5	4567	/	/	/	/	/
				1	8.4103	8.9326	88.8	87.8	89.6	5	3147	2.1	4744	0.30	5.41	0.80705

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s* bar]	Specific flux [LMH/bar]	Density
				2	8.6371	7.3706	88.4	90.0		5	3043	3.0	4704	0.31	5.54	0.80705
				3	6.9391	7.2427	90.7	90.1		5	3350	3.9	5114	0.31	5.48	0.80705
				4	7.0312	7.3363	90.6	90.0		5	2785	4.7	4084	0.29	5.26	0.80705
	DuraMem 500 (T1) - M267	Cold	002145	Feed 4	74.7797	73.3942	/	/	/	/	/	/	/	/	/	/
			002145	Conditioning	/	/	/	/	/	5	5571	/	/	/	/	/
				1	16.4236	16.0783	78.0	78.1	78.9	5	1800	2.0	4317	0.48	8.60	0.80705
				2	15.3276	15.0582	79.5	79.5		5	2261	2.7	5406	0.48	8.58	0.80705
				3	15.9474	16.7141	78.7	77.2		5	1978	3.2	4742	0.48	8.60	0.80705
				4	15.5891	13.7399	79.2	81.3		5	1952	3.8	4670	0.48	8.58	0.80705
		Cold	002148	Feed 5	68.6240	69.9784	/	/	/	/	/	/	/	/	/	/
			002148	Conditioning	/	/	/	/	/	5	4205	/	/	/	/	/
				1	19.7125	20.6055	71.3	70.6	73.5	5	2712	1.9	6519	0.48	8.62	0.80705
				2	17.6169	18.3747	74.3	73.7		5	2474	2.6	5794	0.47	8.40	0.80705
				3	16.5427	17.3671	75.9	75.2		5	2802	3.4	6103	0.44	7.81	0.80705
		Cold	002149	Feed 5	68.6240	69.9784	/	/	/	/	/	/	/	/	/	/
			002149	Conditioning	/	/	/	/	/	5	4867	/	/	/	/	/
				1	18.7342	18.0710	72.7	74.2	74.9	5	2710	2.1	7169	0.53	9.49	0.80705
				2	19.1207	18.0560	72.1	74.2		5	2784	2.9	6906	0.50	8.90	0.80705
				3	18.6724	16.3024	72.8	76.7		5	3071	3.7	7220	0.47	8.43	0.80705
				4	15.9242	14.1083	76.8	79.8		5	2915	4.5	6706	0.46	8.25	0.80705

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]	Rejection [%]	Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s*bar]	Specific flux [LMH/bar]	Density	EtOH in Feed [wt%]	Type of membrane
	DuraMem 200 (T1) - M256 B	Cold	002157	Feed 7	74.6889	73.0801	/	/	/	/	/	/	/	/	/	/
			002157	Conditioning	/	/	/	/	/	5	6011	/	/	/	/	/
				1	below detection limit		100.0	100.0	100.0	5	3792	2.7	2100	0.11	1.99	0.80705
				2	below detection limit		100.0	100.0		5	3942	3.8	2300	0.12	2.09	0.80705
				3	below detection limit		100.0	100.0		5	4313	5.0	2260	0.10	1.88	0.80705
		Cold	002158	Conditioning	/	/	/	/	/	5	5585	/	/	/	/	/
				1	below detection limit		100.0	100.0	100.0	5	4228	2.7	1847	0.09	1.57	0.80705
				2	below detection limit		100.0	100.0		5	3993	3.8	1730	0.09	1.55	0.80705
				3	below detection limit		100.0	100.0		5	4280	5.0	1811	0.08	1.52	0.80705
		Cold	002159	Feed 7	78.0669	78.1342	/	/	/	/	/	/	/	/	/	/
			002159	Conditioning	/	/	/	/	/	5	5525	/	/	/	/	/
				1	below detection limit		100.0	100.0	100.0	5	2948	2.4	1635	0.11	1.99	0.80705
				2	below detection limit		100.0	100.0		5	4079	3.5	2201	0.11	1.94	0.80705
				3	below detection limit		100.0	100.0		6	4033	4.6	2549	0.11	1.89	0.80705

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]	Rejection [%]	Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s*bar]	Specific flux [LMH/bar]	Density	EtOH in Feed [wt%]	Type of membrane
90	DuraMem 500 (T1) - M267	Cold	002165	Feed 9	65.5701	63.9589	/	/	/	/	/	/	/	/	/	/
			002165	Conditioning	/	/	/	/	/	5	4859	/	/	/	/	/
				1	37.7189	37.0594	42.5	42.1	44.9	5	878	1.6	6120	1.39	25.34	0.81797
				2	34.4590	34.4924	47.4	46.1		5	774	1.8	5792	1.50	27.20	0.81797
				3	33.7710	35.2022	48.5	45.0		5	800	2.0	5903	1.48	26.82	0.81797
				4	35.8593	36.8990	45.3	42.3		5	827	2.3	6348	1.54	27.91	0.81797
		Cold	002166	Conditioning	/	/	/	/	/	5	6780	/	/	/	/	/
				1	39.5209	40.2420	39.7	37.1	39.4	5	720	2.1	5388	1.50	27.21	0.81797
				2	37.6443	38.2251	42.6	40.2		5	741	2.3	5864	1.58	28.77	0.81797
				3	38.2630	38.7095	41.6	39.5		5	708	2.5	5927	1.67	30.43	0.81797
				4	40.0934	41.3422	38.9	35.4		5	695	2.7	6123	1.76	32.03	0.81797
		Cold	002168	Conditioning	/	/	/	/	/	5	5021	/	/	/	/	/
				1	42.4098	42.2273	35.3	34.0	38.3	5	732	1.6	5771	1.58	28.66	0.81797
				2	39.8069	39.8110	39.3	37.8		5	724	1.8	5570	1.54	27.97	0.81797
				3	38.3458	38.2611	41.5	40.2		5	733	2.0	5747	1.57	28.50	0.81797
				4	39.8859	39.0284	39.2	39.0		5	727	2.2	5756	1.58	28.78	0.81797
	DuraMem 300 (T1) - M255	Cold	002170	Feed 10	64.0712	65.1240	/	/	Average	/	/	/	/	/	/	/
			002170	Conditioning	/	/	/	/	/	5	6214	/	/	/	/	/
				1	30.2175	28.2465	52.8	56.6	54.3	5	2410	2.4	4246	0.35	6.40	0.81797
				2	32.2457	31.4825	49.7	51.7		5	2495	3.1	4465	0.36	6.51	0.81797

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]	Rejection [%]	Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s*bar]	Specific flux [LMH/bar]	Density	EtOH in Feed [wt%]	Type of membrane
				3	29.3015	28.9628	54.3	55.5		5	2515	3.8	4510	0.36	6.52	0.81797
				4	27.4698	28.1045	57.1	56.8		5	2429	4.5	3990	0.33	5.97	0.81797
		Cold	002171	Feed 10	63.2296	62.9501	/	/	Average	/	/	/	/	/	/	/
			002171	Conditioning	/	/	/	/	/	5	6167	/	/	/	/	/
				1	30.7923	30.5539	51.3	51.5	51.7	5	2424	2.4	3110	0.26	4.66	0.81797
				2	32.3915	31.8358	48.8	49.4		5	2493	3.1	3362	0.27	4.90	0.81797
				3	30.7241	29.3176	51.4	53.4		5	2412	3.7	3288	0.27	4.96	0.81797
				4	29.7065	28.6177	53.0	54.5		5	1515	4.2	2031	0.27	4.87	0.81797
		Warm	002172	Feed 10	67.1286	67.1550	/	/	Average	/	/	/	/	/	/	/
			002172	Conditioning	/	/	/	/	/	4	5413	/	/	/	/	/
				1	43.4329	42.9212	35.3	36.1	40.4	4	2413	2.2	5544	0.57	10.44	0.81797
				2	40.5819	38.8170	39.5	42.2		4	2441	2.9	5003	0.51	9.31	0.81797
				3	40.0361	38.9040	40.4	42.1		4	2448	3.5	4965	0.51	9.22	0.81797
				4	38.7482	36.7008	42.3	45.3		4	1528	4.0	2933	0.48	8.72	0.81797
	DuraMem 200 (T1) - M256 B	Cold	00174	Feed 10	65.3752	63.1289	/	/	Average	/	/	/	/	/	/	/
			002174	Conditioning	/	/	/	/	/	6	10838	/	/	/	/	/
				1	1.7542	1.1314	97.3	98.2	97.4	6	3614	4.0	1893	0.09	1.59	0.81797
				2	1.6191	1.6568	97.5	97.4		4	3639	5.0	1600	0.11	2.00	0.81797
				3	1.7768	1.2669	97.3	98.0		4	3692	6.1	1233	0.08	1.52	0.81797
				4	1.9098	2.2984	97.1	96.4		5	3908	7.1	1597	0.08	1.49	0.81797
		Cold	002175	Feed 10	67.2641	67.1485	/	/	Average	/	/	/	/	/	/	/

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]	Rejection [%]	Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s*bar]	Specific flux [LMH/bar]	Density	EtOH in Feed [wt%]	Type of membrane
			002175	Conditioning	/	/	/	/	/	6	5441	/	/	/	/	/
				1	25.1523	24.3911	62.6	63.7	69.5	4.5	5120	2.9	3273	0.14	2.58	0.81797
				2	20.1662	19.5464	70.0	70.9		4.5	3278	3.8	1884	0.13	2.32	0.81797
				3	20.1470	19.1641	70.0	71.5		4.5	3071	4.7	1773	0.13	2.33	0.81797
				4	17.0878	18.4652	74.6	72.5		4.5	1816	5.2	1049	0.13	2.33	0.81797
		Warm	002178	Feed 10	62.1000	64.8217	/	/	Average	/	/	/	/	/	/	/
			002178	Conditioning	/	/	/	/	/	5	5368	/	/	/	/	/
				1	6.7798	7.6579	89.1	88.2	92.7	5	5221	2.9	2172	0.08	1.51	0.81797
				2	4.7917	3.6682	92.3	94.3		4.5	3431	3.9	1387	0.09	1.63	0.81797
				3	3.1456	3.4015	94.9	94.8		4	3613	4.9	1235	0.09	1.55	0.81797
				4	4.3194	3.3217	93.0	94.9		4.5	2407	5.6	790	0.07	1.33	0.81797
85	**DuraMem 200 (T1) - M256 B**	Cold	002179	Feed 11	65.0216	65.7631	/	/	Average	/	/	/	/	/	/	/
			002179	Conditioning	/	/	/	/	/	6	5408	/	/	/	/	/
				1	4.7502	3.8613	92.7	94.1	95.7	6	3604	2.5	2147	0.10	1.83	0.83095
				2	3.5552	2.1527	94.5	96.7		6	3062	3.4	1963	0.11	1.97	0.83095
				3	1.7734	2.1341	97.3	96.8		6	3086	4.2	1905	0.10	1.90	0.83095
				4	2.0744	2.0675	96.8	96.9		6	2355	4.9	1383	0.10	1.81	0.83095
		Warm	002180	Feed 11	68.8129	70.4827	/	/	Average	/	/	/	/	/	/	/
			002180	Conditioning	/	/	/	/	/	6	5375	/	/	/	/	/
				1	26.9163	26.9033	60.9	61.8	60.4	5.5	3547	2.5	4181	0.21	3.96	0.83095

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]	Rejection [%]	Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s*bar]	Specific flux [LMH/bar]	Density	EtOH in Feed [wt%]	Type of membrane
				2	26.5776	27.5682	61.4	60.9		5	3006	3.3	2917	0.19	3.58	0.83095
				3	27.1320	29.1468	60.6	58.6		5	2986	4.1	3272	0.22	4.05	0.83095
				4	28.4535	27.7216	58.7	60.7		6	8580	6.5	8894	0.17	3.19	0.83095
			002181	Feed 11	66.1494	65.0052	/	/	Average	/	/	/	/	/	/	/
		Cold	002181	Conditioning	/	/	/	/	/	6	6299	/	/	/	/	/
				1	3.4345	2.4008	94.8	96.3		6	4080	2.9	2915	0.12	2.20	0.83095
				2	3.0701	2.4532	95.4	96.2		5	3637	3.9	2498	0.14	2.54	0.83095
				3	2.8303	2.3991	95.7	96.3	95.5	5.5	4481	5.1	2873	0.12	2.15	0.83095
				4	3.8985	3.1790	94.1	95.1		6	2995	6.0	1739	0.10	1.79	0.83095
	DuraMem 300 (T1) - M255		002182	Feed 11	69.6860	70.6936	/	/	Average	/	/	/	/	/	/	/
		Warm	002182	Conditioning	/	/	/	/	/	6	4319	/	/	/	/	/
				1	44.4059	46.0933	36.3	34.8		5	1435	1.6	5656	0.79	14.56	0.83095
				2	40.4953	44.6980	41.9	36.8		5	1594	2.0	6073	0.76	14.07	0.83095
				3	42.8188	46.6177	38.6	34.1	37.3	5	1527	2.5	5866	0.77	14.19	0.83095
				4	44.3634	42.7641	36.3	39.5		5	1999	3.0	7792	0.78	14.40	0.83095
			002183	Feed 11	65.2671	63.7951	/	/	Average	/	/	/	/	/	/	/
		Cold	002183	Conditioning	/	/	/	/	/	5.5	4565	/	/	/	/	/
				1	31.9641	30.1492	51.0	52.7		4.5	1903	1.8	4119	0.48	8.88	0.83095
				2	30.3306	29.0714	53.5	54.4		4	2302	2.4	4616	0.50	9.26	0.83095
				3	31.6166	31.9440	51.6	49.9	51.7	4	1874	3.0	3997	0.53	9.85	0.83095
				4	31.8023	32.6960	51.3	48.7		5	2710	3.7	6330	0.47	8.63	0.83095

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]	Rejection [%]	Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s*bar]	Specific flux [LMH/bar]	Density	EtOH in Feed [wt%]	Type of membrane
80	DuraMem 300 (T1) - M255	Warm	002184	Feed 11	62.4866	60.8506	/	/	Average	/	/	/	/	/		
			002184	Conditioning	/	/	/	/	/	5.5	4891	/	/	/		
				1	38.3327	35.5849	38.7	41.5	40.7	5	1876	1.9	5824	0.62	11.47	0.83095
				2	37.9512	33.6287	39.3	44.7		5	2503	2.6	7101	0.57	10.48	0.83095
				3	37.1536	35.8349	40.5	41.1		5.5	1886	3.1	5455	0.53	9.71	0.83095
				4	36.7210	37.1572	41.2	38.9		5	2693	3.8	7299	0.54	10.01	0.83095
		Cold	002127	Feed 3	78.4870	77.9540	/	/	/	/	/	/	/	/	/	/
			002127	Conditioning	/	/	/	/	/	5	3600	/	/	/	/	/
				1	20.8436	18.4658	73.4	76.3	76.2	5	2298	1.6	4431	0.39	7.23	0.84344
				2	17.9960	19.7611	77.1	74.7		5	1614	2.1	2965	0.37	6.89	0.84344
				3	17.4080	17.2711	77.8	77.8		5	5822	3.7	8586	0.29	5.53	0.84344
		Cold	002128	Feed 3	82.2949	78.0346	/	/	/	/	/	/	/	/	/	/
			002128	Conditioning	/	/	/	/	/	5	6785	/	/	/	/	/
				1	43.4781	45.3347	47.2	41.9	46.0	5	1626	2.3	5387	0.66	12.42	0.84344
				2	45.1652	43.6797	45.1	44.0		5	1929	2.9	5964	0.62	11.59	0.84344
				3	44.6974	41.3572	45.7	47.0		5	1511	3.3	4470	0.59	11.09	0.84344
				4	41.0799	41.3050	50.1	47.1		5	1069	3.6	2802	0.52	9.83	0.84344
		Cold	002129	Feed 3	78.0346	82.2949	/	/	/	/	/	/	/	/	/	/
			002129	Conditioning	/	/	/	/	/	5	5405	/	/	/	/	/
				1	37.5634	37.5398	51.9	54.4	57.2	5.5	2392	2.2	7576	0.58	10.79	0.84344

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]	Rejection [%]	Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s*bar]	Specific flux [LMH/bar]	Density	EtOH in Feed [wt%]	Type of membrane
				2	35.1287	33.9288	55.0	58.8		5	1808	2.7	4588	0.51	9.51	0.84344
				3	33.9886	33.9979	56.4	58.7		5	2295	3.3	5407	0.47	8.83	0.84344
				4	30.8878	31.2596	60.4	62.0		5	2801	4.1	5818	0.42	7.79	0.84344
		Cold	002133	Feed 3	80.9865	80.4595	/	/	/	5	/	/	/	/	/	/
			002133	Conditioning	/	/	/	/	/	5	5349	/	/	/	/	/
				6	17.8485	16.7391	78.0	79.2		5	4237	2.7	6053	0.29	5.36	0.84344
				7	14.0828	16.5961	82.6	79.4	79.1	5	1827	3.2	2315	0.25	4.75	0.84344
				8	19.7356	16.0173	75.6	80.1		5	1716	3.6	2031	0.24	4.44	0.84344
	DuraMem 200 (T1) - M256 B	Cold	002137	Feed 3	75.4345	71.7845	/	/	/	/	/	/	/	/	/	/
			002137	Conditioning	/	/	/	/	/	5	6001	/	/	/	/	/
				1	13.5959	12.4751	82.0	82.6		5	4575	2.9	5422	0.24	4.44	0.84344
				2	14.2343	12.7284	81.1	82.3	82.0	5	2620	3.7	2976	0.23	4.26	0.84344
				3	13.2615	13.1901	82.4	81.6		5	2531	4.4	2734	0.22	4.05	0.84344
		Cold	002138	Conditioning	/	/	/	/	/	5	5815	/	/	/	/	/
				1	19.7139	20.0397	73.9	72.1		4	2514	2.3	3388	0.34	6.31	0.84344
				2	17.6295	11.7384	76.6	83.6		5	2397	3.0	3659	0.31	5.72	0.84344
				3	19.6354	19.7894	74.0	72.4	75.0	5	2445	3.7	3749	0.31	5.75	0.84344
				4	19.5564	19.0070	74.1	73.5		5	2086	4.2	2885	0.28	5.18	0.84344
		Cold	002139	Conditioning	/	/	/	/	/	5	5765	/	/	/	/	/
				1	14.0115	12.0084	81.4	83.3	81.9	5	2480	2.3	3051	0.25	4.61	0.84344

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]	Rejection [%]	Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s*bar]	Specific flux [LMH/bar]	Density	EtOH in Feed [wt%]	Type of membrane
				2	13.9450	13.1656	81.5	81.7		4.5	2577	3.0	2717	0.23	4.39	0.84344
				3	13.9401	12.7382	81.5	82.3		4.5	3680	4.0	4597	0.28	5.20	0.84344
	DuraMem 500 (T1) - M267	Cold	002141	Feed 3	70.2385	71.6643	/	/	/	/	/	/	/	/	/	/
			002141	Conditioning	/	/	/	/	/	5	2679	/	/	/	/	/
				1	54.6107	55.7428	22.2	22.2		5	738	0.9	9309	2.52	47.28	0.84344
				2	56.0036	54.8292	20.3	23.5	22.6	5	809	1.2	9517	2.35	44.10	0.84344
				3	54.6423	53.5595	22.2	25.3		5	719	1.4	8423	2.34	43.91	0.84344
		Cold	002154	Feed 6	77.1697	73.8084	/	/	/	/	/	/	/	/	/	/
			002154	Conditioning	/	/	/	/	/	5	4505	/	/	/	/	/
				1	43.0107	41.4712	44.3	43.8		5	2881	2.1	7639	0.53	9.94	0.84344
				2	36.5556	34.8746	52.6	52.7	50.1	5	2550	2.8	5659	0.44	8.32	0.84344
				3	35.1275	34.9635	54.5	52.6		5	2523	3.5	5219	0.41	7.75	0.84344
		Cold	002155	Feed 6	65.9917	68.1423	/	/	/	/	/	/	/	/	/	/
			002155	Conditioning	/	/	/	/	/	5	4051	/	/	/	/	/
				1	37.6102	36.3073	43.0	46.7		5	1031	1.4	7893	1.53	28.70	0.84344
				2	35.2405	35.1560	46.6	48.4	46.9	5	972	1.7	7561	1.56	29.16	0.84344
				3	35.5974	33.6726	46.1	50.6		5	1128	2.0	8630	1.53	28.68	0.84344

Appendix 5: Performance parameters for polymeric membranes at 40 bar pressure

The performance of the polymeric membranes during the nanofiltration measurements at 40 bar pressure is presented in Table 11 providing the corresponding parameters.

Table 11: Performance parameters of the polymeric membranes from the nanofiltration experiments at 40 bar pressure with four different types of feed

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s]	Specific flux [LMH/bar]	Density
95	DuraMem 300	Warm	002142	Feed 4	70.6509	69.2904	/	/	/	/	/	/	/	/	/	/
			002142	Conditioning	/	/	/	/	/	40	2141	/	/	/	/	/
				5	B.D.L	B.D.L	100.0	100.0	100	40	945	0.9	9157	0.24	4.34	0.80705
				6	B.D.L	B.D.L	100.0	100.0		40	1016	1.1	9460	0.23	4.17	0.80705
				7	B.D.L	B.D.L	100.0	100.0		40	1150	1.5	10643	0.23	4.15	0.80705
		Warm	002143	Feed 4	74.7797	73.3942	/	/	/	/	/	/	/	/	/	/
			002143	Conditioning	/	/	/	/	/	40	2831	/	/	/	/	/
				5	4.8556	3.3956	93.5	95.4	95.9	40	744	1.0	7252	0.24	4.37	0.80705
				6	2.6500	3.8087	96.5	94.8		40	853	1.2	8256	0.24	4.34	0.80705
				7	2.1241	2.1262	97.2	97.1		40	973	1.5	8953	0.23	4.13	0.80705
				8	2.0301	3.1680	97.3	95.7		40	1089	1.8	9907	0.23	4.08	0.80705
		Warm	002144	Conditioning	/	/	/	/	/	40	2500	/	/	/	/	/
				5	8.4093	9.5942	88.8	86.9	87.5	40	777	0.9	7622	0.25	4.40	0.80705
				6	10.6134	8.5021	85.8	88.4		40	1240	1.3	11495	0.23	4.16	0.80705
				7	10.2208	8.5146	86.3	88.4		40	1037	1.5	9941	0.24	4.30	0.80705
				8	10.0397	8.4229	86.6	88.5		40	927	1.8	8893	0.24	4.30	0.80705

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s]	Specific flux [LMH/bar]	Density
	DuraMem 500	Cold	002147	Feed 5	68.624	69.9784	/	/	/	/	/	/	/	/	/	/
			0002147	Conditioning	/	/	/	/	/	40	3737		/	/	/	/
				1	2.5946	3.3095	96.2	95.3	96.2	40	867	1.3	8898	0.26	4.60	0.80705
				2	2.4369	2.2548	96.4	96.8		40	778	1.5	7857	0.25	4.53	0.80705
				3	2.9294	2.3138	95.7	96.7		40	1293	1.9	11361	0.22	3.94	0.80705
				4	lost sample		no value			40	873	2.1	/	/	/	/
		Warm	0002149	Conditioning	/	/	/	/	/	40	4389	/	/	/	/	/
				5	1.4466	1.6896	97.9	97.6	98.2	40	969	1.5	9858	0.25	4.56	0.80705
				6	1.3686	1.2468	98.0	98.2		40	830	1.7	8239	0.25	4.45	0.80705
				7	1.3036	1.4207	98.1	98.0		40	1010	2.0	9770	0.24	4.34	0.80705
				8	0.432	0.9368	99.4	98.7		40	1226	2.3	11423	0.23	4.18	0.80705
		Cold	0002150	Conditioning	/	/	/	/	/	40	3665	/	/	/	/	/
				1	0.8488	1.1822	98.8	98.3	99.3	40	945	1.3	9790	0.26	4.64	0.80705
				2	0.6251	0.2782	99.1	99.6		40	910	1.5	9226	0.25	4.55	0.80705
				3	0.4243	0.7178	99.4	99.0		40	885	1.8	8816	0.25	4.47	0.80705
				4	B.D.L	B.D.L	100	100		40	737	2.0	7045	0.24	4.29	0.80705
	DuraMem 200	Warm	002157	Feed 6	74.6889	73.0801	/	/	/	/	/	/	/	/	/	/
			002157	Conditioning	/	/	/	/	/	40	4501	/	/	/	/	/
				4	B.D.L	B.D.L	100.0	100.0	100%	40	1241	1.6	5757	0.12	2.08	0.80705
				5	B.D.L	B.D.L	100.0	100.0		40	1119	1.9	5212	0.12	2.09	0.80705
				6	B.D.L	B.D.L	100.0	100.0		40	1416	2.3	6519	0.12	2.06	0.80705
				7	B.D.L	B.D.L	100.0	100.0		40	1539	2.7	6995	0.11	2.04	0.80705
		Warm	002158	Conditioning	/	/	/	/	/	40	4031		/	/	/	/
				4	B.D.L	B.D.L	100.0	100.0	100%	40	956	1.4	3761	0.10	1.76	0.80705

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s]	Specific flux [LMH/bar]	Density
				5	B.D.L	B.D.L	100.0	100.0		40	1429	1.8	5581	0.10	1.75	0.80705
				6	B.D.L	B.D.L	100.0	100.0		40	957	2.0	3795	0.10	1.78	0.80705
				7	B.D.L	B.D.L	100.0	100.0		40	713	2.2	2728	0.10	1.72	0.80705
			002159	Feed 6	78.0669	78.1342	/	/	/	/	/	/	/	/	/	/
		Warm	002159	Conditioning	/	/	/	/	/	40	5711	/	/	/	/	/
				4	B.D.L	B.D.L	100.0	100.0		40	1462	2.0	6202	0.11	1.90	0.80705
				5	B.D.L	B.D.L	100.0	100.0		40	1613	2.4	6766	0.10	1.88	0.80705
				6	B.D.L	B.D.L	100.0	100.0	100%	40	1233	2.8	5117	0.10	1.86	0.80705
				7	B.D.L	B.D.L	100.0	100.0		40	1425	3.2	5911	0.10	1.86	0.80705
			002187	Feed 12	67.1197	67.1604	/	/	/	/	/	/	/	/	/	/
			002187	Conditioning	/	/	/	/	/	40	2601	/	/	/	/	/
		Cold		1	39.9640	40.6242	40.5	39.5		40	681	0.9	7899	0.29	5.27	0.81797
				2	36.8856	38.9828	45.0	42.0		40	637	1.1	8343	0.33	5.95	0.81797
				3	36.4809	36.3028	45.6	45.9	44.2	40	617	1.3	8002	0.32	5.89	0.81797
				4	33.6618	36.5442	49.8	45.6		40	608	1.4	7746	0.32	5.79	0.81797
			002188	Feed 12	65.6586	65.4109	/	/	/	/	/	/	/	/	/	/
			002188	Conditioning	/	/	/	/	/	39	2489	/	/	/	/	/
		Warm		1	48.7079	53.5794	25.8	18.1		39	649	0.9	11790	0.47	8.47	0.81797
				2	47.2959	53.0638	28.0	18.9		38	605	1.0	11510	0.50	9.10	0.81797
				3	47.4020	49.3220	27.8	24.6	24.9	41	606	1.2	11490	0.46	8.41	0.81797
				4	45.9750	48.5965	30.0	25.7		43	348	1.3	6546	0.44	7.95	0.81797
			002189	Feed 12	63.1868	65.4784	/	/	/	/	/	/	/	/	/	/
		Cold	002189	Conditioning	/	/	/	/	/	39	2541	/	/	/	/	/
				1	38.2175	37.0911	39.5	43.4	44.4	39	639	0.9	7614	0.31	5.55	0.81797
90	**DuraMem 300 (T1) - M255**															

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s]	Specific flux [LMH/bar]	Density
				2	37.2641	37.3813	41.0	42.9		39	622	1.1	7319	0.30	5.48	0.81797
				3	33.7462	34.0764	46.6	48.0		39	602	1.2	7044	0.30	5.45	0.81797
				4	35.5300	32.8655	43.8	49.8		40	616	1.4	7227	0.29	5.33	0.81797
		Warm	002191	Feed 12	63.2348	63.3302	/	/	/	/	/	/	/	/	/	/
			002191	Conditioning	/	/	/	/	/	41	2431	/	/	/	/	/
				1	41.4954	37.6287	34.4	40.6	41.5	41	603	0.8	8280	0.33	6.09	0.81797
				2	35.1187	36.0349	44.5	43.1		41	763	1.1	10117	0.32	5.88	0.81797
				3	35.2381	36.6830	44.3	42.1		41	616	1.2	7916	0.31	5.70	0.81797
				4	37.7269	36.4678	40.3	42.4		41	602	1.4	7489	0.30	5.52	0.81797
	DuraMem 200	Warm	002193	Feed 12	66.0384	69.5888	/	/	/	/	/	/	/	/	/	/
			002193	Conditioning 1	/	/	/	/	/	45	5963	/	/	/	/	/
				Conditioning 2	/	/	/	/	/	40	1385	/	/	/	/	/
				1	B.D.L	B.D.L	100.0	100.0	100	40	1221	0.7	3955	0.08	1.47	0.81797
				2	B.D.L	B.D.L	100.0	100.0		40	1204	1.1	3696	0.08	1.39	0.81797
				3	B.D.L	B.D.L	100.0	100.0		40	1213	1.4	3703	0.08	1.39	0.81797
				4	B.D.L	B.D.L	100.0	100.0		41	1138	1.7	3450	0.07	1.34	0.81797
		Cold	002194	Feed 12	64.3714	66.0650	/	/	/	/	/	/	/	/	/	/
			002194	Conditioning	/	/	/	/	/	40	3695	/	/	/	/	/
				1	14.0717	12.0555	78.1	81.8	79.7	40	1178	1.4	4751	0.10	1.83	0.81797
				2	13.9388	10.4914	78.3	84.1		40	1234	1.7	5182	0.10	1.91	0.81797
				3	14.4512	12.2738	77.6	81.4		40	1179	2.0	5154	0.11	1.99	0.81797
				4	13.1324	15.5563	79.6	76.5		40	1217	2.4	5216	0.11	1.95	0.81797
		Warm	002195	Feed 12	64.5819	69.6207	/	/	/	/	/	/	/	/	/	/

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s]	Specific flux [LMH/bar]	Density
				Conditioning	/	/	/	/	/	40	4046	/	/	/	/	/
			002195	1	14.4480	16.3357	77.6	76.5	78.7	40	1211	1.5	5545	0.11	2.08	0.81797
				2	12.7199	15.0888	80.3	78.3		40	1214	1.8	5482	0.11	2.05	0.81797
				3	14.3927	16.5872	77.7	76.2		40	1201	2.1	8221	0.17	3.11	0.81797
				4	11.7058	12.9454	81.9	81.4		40	1185	2.5	4877	0.10	1.87	0.81797
		Warm	002197	Feed 12	66.7493	65.1108	/	/	/	/	/	/	/	/	/	/
			002197	Conditioning	/	/	/	/	/	40	3614	/	/	/	/	/
				1	29.5983	27.7180	55.7	57.4	54.3	40	1284	1.4	3996	0.08	1.41	0.81797
				2	29.7101	29.6502	55.5	54.5		40	1235	1.7	3794	0.08	1.40	0.81797
				3	31.2943	31.0335	53.1	52.3		40	1192	2.0	3627	0.08	1.38	0.81797
				4	31.2895	30.8010	53.1	52.7		40	1223	2.4	3705	0.08	1.38	0.81797
85	**DuraMem 300**	Cold	002198	Feed 13	68.4941	69.2321	/	/	/	/	/	/	/	/	/	/
			002198	Conditioning	/	/	/	/	/	40	2458	/	/	/	/	/
				1	24.9808	23.5344	63.5	66.0	61.9	40	1306	1.0	10588	0.20	3.74	0.83095
				2	27.3253	25.4711	60.1	63.2		40	990	1.3	8421	0.21	3.93	0.83095
				3	26.8031	26.2588	60.9	62.1		40	904	1.6	7922	0.22	4.05	0.83095
				4	28.1963	27.3909	58.8	60.4		40	910	1.8	8026	0.22	4.07	0.83095
		Warm	002199	Feed 13	69.2204	67.7081	/	/	/	/	/	/	/	/	/	/
			002199	Conditioning	/	/	/	/	/	40	2406	/	/	/	/	/
				1	58.7606	58.6831	15.1	13.3	12.4	40	400	0.8	12488	0.78	14.41	0.83095
				2	58.3178	61.7377	15.8	8.8		40	369	0.9	11376	0.77	14.23	0.83095
				3	63.1437	62.3870	8.8	7.9		40	340	1.0	10266	0.75	13.94	0.83095
				4	58.1044	58.6105	16.1	13.4		41	326	1.1	9886	0.74	13.66	0.83095
		Cold	002200	Feed 13	70.2499	76.9018	/	/	/	/	/		/	/	/	/

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s]	Specific flux [LMH/bar]	Density
			002200	Conditioning	/	/	/	/	/	40	2405		/	/	/	/
				1	39.0170	41.6441	44.5	45.8	46.9	40	848	0.9	9734	0.29	5.30	0.83095
				2	40.6578	41.1100	42.1	46.5		40	848	1.1	9890	0.29	5.38	0.83095
				3	38.4503	38.1822	45.3	50.3		40	829	1.4	9431	0.28	5.25	0.83095
				4	36.8298	36.0767	47.6	53.1		44	837	1.6	10114	0.27	5.07	0.83095
80	DuraMem 300	Warm	002127	Feed 3	78.4870	77.9540	/	/	/	/	/	/	/	/	/	/
			002127	Conditioning	/	/	/	/	/	40	2704	/	/	/	/	/
				4	19.2864	18.6723	75.4	76.0	76.6	40	876	1.0	6935	0.20	3.71	0.84344
				5	19.1384	16.1392	75.6	79.3		40	1287	1.4	9983	0.19	3.63	0.84344
				6	lost sample		no value			40	825	1.6	5967	0.18	3.39	0.84344
		Warm	002128	Feed 3	82.2949	78.0346	/	/	/	/	/	/	/	/	/	/
			002128	Conditioning	/	/	/	/	/	40	2887	/	/	/	/	/
				5	45.2888	45.3857	45.0	41.8	45.8	40	561	1.0	8985	0.40	7.50	0.84344
				6	43.6100	43.9098	47.0	43.7		40	1000	1.2	14611	0.37	6.85	0.84344
				7	42.7320	39.7131	48.1	49.1		40	675	1.4	9118	0.34	6.33	0.84344
		Warm	002129	Feed 3	78.0346	82.2949	/	/	/	/	/	/	/	/	/	/
			002129	Conditioning	/	/	/	/	/	40	4323	/	/	/	/	/
				5	29.3580	29.3908	62.4	64.3	66.8	40	770	1.4	8311	0.27	5.06	0.84344
				6	27.1500	26.3365	65.2	68.0		40	711	1.6	7567	0.27	4.99	0.84344
				7	27.5830	27.4369	64.7	66.7		40	829	1.8	8270	0.25	4.67	0.84344
				8	21.0380	24.4024	73.0	70.3		40	737	2.0	6857	0.23	4.36	0.84344
		Cold	002133	Feed 3	80.9865	80.4595	/	/	/	/	/	/	/	/	/	/
			002133	Conditioning	/	/	/	/	/	40	4925	/	/	/	/	/
				1	34.1694	32.2789	57.8	59.9	64.6	40	785	1.6	9803	0.31	5.85	0.84344

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s]	Specific flux [LMH/bar]	Density
				2	30.1331	29.9601	62.8	62.8		40	744	1.8	8815	0.30	5.55	0.84344
				3	28.7220	28.9516	64.5	64.0		40	815	2.0	8935	0.27	5.14	0.84344
				4	26.3424	24.7370	67.5	69.3		40	855	2.3	8739	0.26	4.79	0.84344
				5	27.2414	23.6274	66.4	70.6		40	884	2.5	8365	0.24	4.43	0.84344
	DuraMem 200	Warm	002137	Feed 3	75.4345	71.7845	/	/	/	/	/	/	/	/	/	/
			002137	Conditioning	/	/	/	/	/	40	2515	/	/	/	/	/
				4	12.9103	11.6641	82.9	83.8		40	856	0.9	5565	0.16	3.05	0.84344
				5	1.8845	11.9538	97.5	83.3	86.4	40	924	1.2	5499	0.15	2.79	0.84344
				6	9.8920	11.4309	86.9	84.1		40	901	1.4	5320	0.15	2.77	0.84344
		Warm	002138	Conditioning	/	/	/	/	/	40	2819	/	/	/	/	/
				5	19.6182	20.2680	74.0	71.8		40	1361	1.2	11038	0.20	3.80	0.84344
				6	19.4725	19.7722	74.2	72.5		40	951	1.4	6977	0.18	3.44	0.84344
				7	18.7384	19.6246	75.2	72.7	73.6	40	850	1.7	5899	0.17	3.25	0.84344
				8	18.2933	19.3715	75.7	73.0		40	1053	2.0	7102	0.17	3.16	0.84344
		Warm	002139	Conditioning	/	/	/	/	/	40	3315	/	/	/	/	/
				4	13.9963	14.3571	81.4	80.0		39.5	1385	1.3	9583	0.18	3.28	0.84344
				5	12.9286	13.4938	82.9	81.2		39.5	1494	1.7	9680	0.16	3.07	0.84344
				6	13.6338	13.0450	81.9	81.8	82.0	39.5	1168	2.0	7123	0.15	2.89	0.84344
				7	12.2961	12.2475	83.7	82.9		40	1885	2.6	10874	0.14	2.70	0.84344
	DuraMem 500	Warm	002141	Feed 3	70.2385	71.6643	/	/	/	/	/	/	/	/	/	/
			002141	Conditioning	/	/	/	/	/	40	3068	/	/	/	/	/
				5	61.9789	65.9019	11.8	8.0		40	435	1.0	21594	1.24	23.26	0.84344
				6	68.2846	65.5775	2.8	8.5	8.5	40	266	1.0	12855	1.21	22.64	0.84344
				7	61.2272	63.8306	12.8	10.9		40	258	1.1	12210	1.18	22.18	0.84344

EtOH in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Timeline [h]	Weight [mg]	Flux [mg/s]	Specific flux [LMH/bar]	Density
				8	66.1780	66.5054	5.8	7.2		40	196	1.2	9120	1.16	21.80	0.84344
			002154	Feed 6	77.1697	73.8084	/	/	/	/	/	/	/	/	/	/
			002154	Conditioning	/	/	/	/	/	40	3901	/	/	/	/	/
		Warm		4	32.7799	30.7620	57.5	58.3		40	444	1.2	5224	0.29	5.51	0.84344
				5	28.6321	26.2863	62.9	64.4	61.9	40	589	1.4	6442	0.27	5.12	0.84344
				6	27.3476	26.7671	64.6	63.7		40	516	1.5	5472	0.27	4.97	0.84344
			002155	Feed 6	65.9917	68.1423	/	/	/	/	/	/	/	/	/	/
			002155	Conditioning	/	/	/	/	/	40	3971	/	/	/	/	/
		Warm		4	40.3664	42.7292	38.8	37.3		40	329	1.2	11252	0.86	16.03	0.84344
				5	41.4557	40.4633	37.2	40.6		40	321	1.3	10798	0.84	15.76	0.84344
				6	41.9816	40.4209	36.4	40.7	38.4	40	520	1.4	16685	0.80	15.04	0.84344
				7	42.9285	40.2155	34.9	41.0		40	340	1.5	10387	0.76	14.32	0.84344

Appendix 6: Correlation between membrane rejection and conditioning time of experiment

By analyzing Figure 51 and Figure 52 it can be observed that the two graphs show different conditioning times when 5 bar and 40 bar pressures were used. At 5 bar pressure, the rejection value for the conditioning time in between half an hour to three hours was variable amounting from 30% to 100%. When 40 bar pressure was applied the conditioning time was shorter, lasting up to hour and a half. The rejection variation was in a wide range from around 10% up to 100%. Additionally, it can be seen that the change of conditioning time caused by the change of pressure did not influence the level of rejection. The graphs can be used to emphasize that there is no defined function of dependence or correlation between the two parameters.

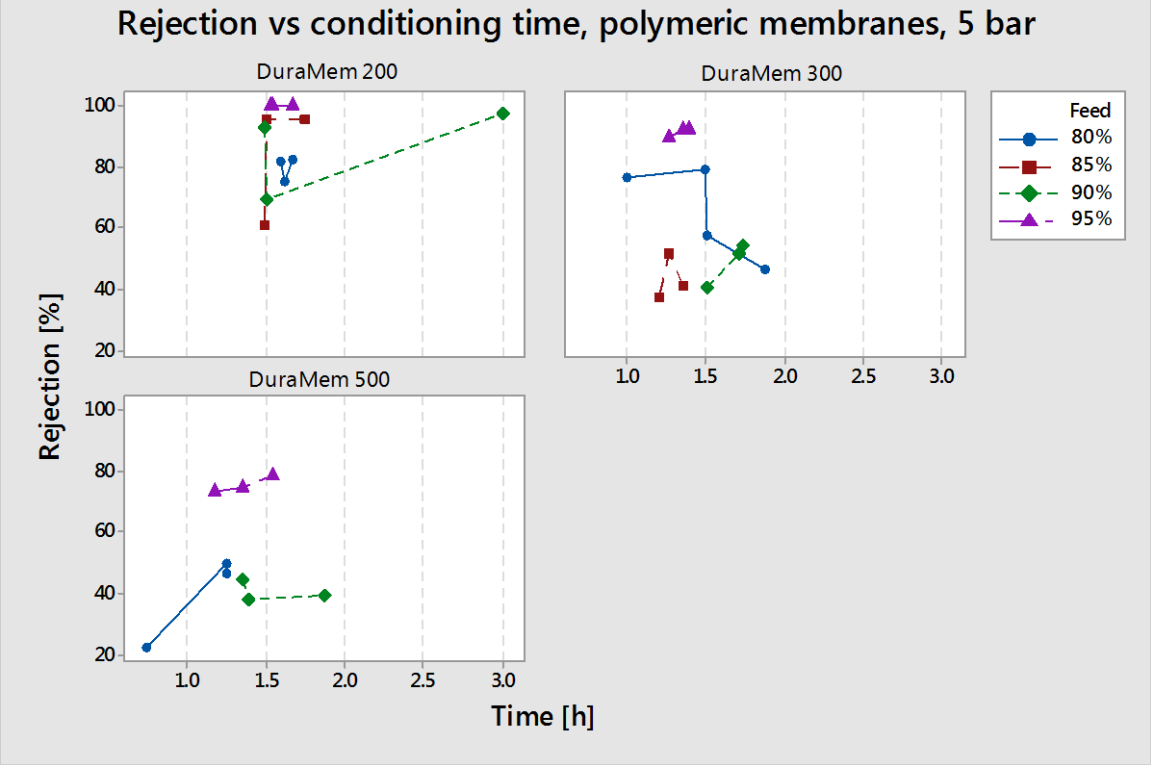

Figure 51: Correlation between rejection and conditioning time for each polymeric membrane when 5 bar pressure was used

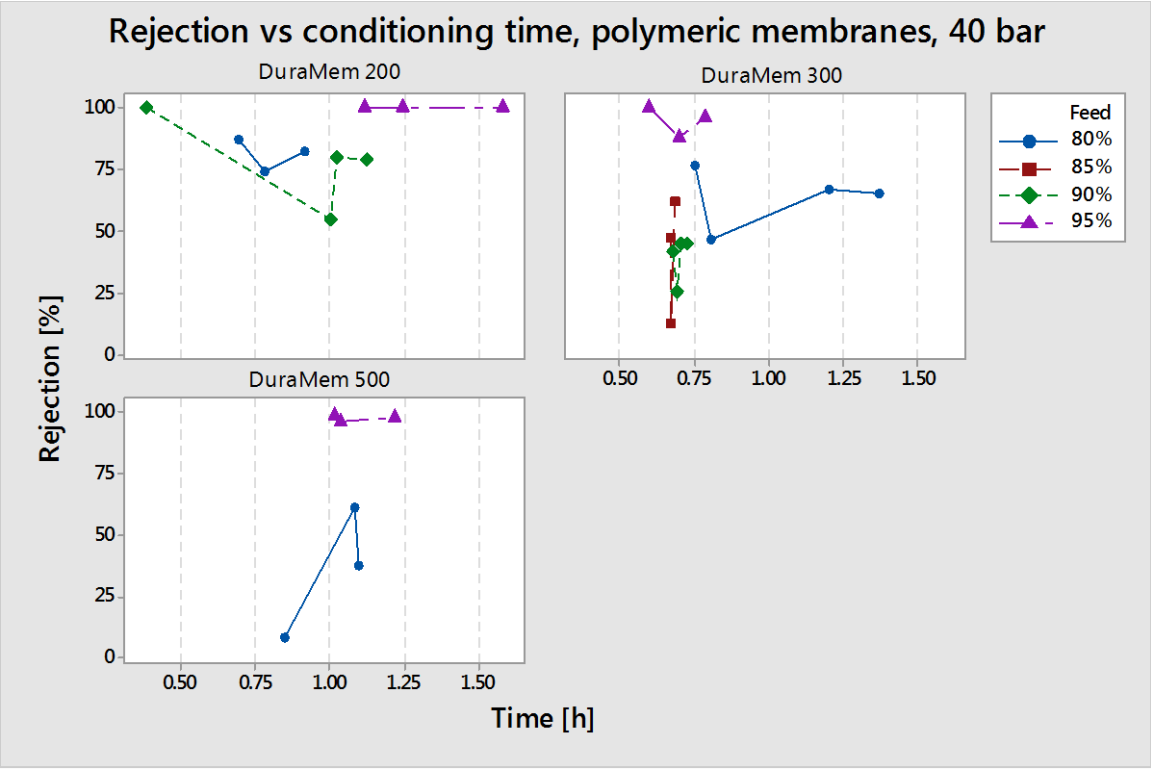

Figure 52: Correlation between rejection and conditioning time for each polymeric membrane when 40 bar pressure was used

Appendix 7: Performance parameters for ceramic membranes at 5 bar pressure

The ceramic membranes have performed the experiments according to the parameters in Table 12.

Table 12: Performance parameters of the ceramic membranes from the nanofiltration experiments at 5 bar pressure with two types of feed

Water in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Weight [mg]	Flux [mg/s]
5	Pervatech 500 - 400 CM	Cold	002161	Feed 7	77.6432	70.0490	/	/	/	/	/	/	/
			002161	Conditioning	/	/	/	/	/	5	4505	/	/
				1	47.5540	46.6587	38.8	33.4	36.2	5	2746	2172	0.16
				2	47.3682	48.4323	39.0	30.9		5	3293	2891	0.18
				3	46.7996	48.4002	39.7	30.9		5.5	4280	3635	0.15
				4	46.5280	44.4251	40.1	36.6		4	3209	1638	0.13
		Cold	002163	Feed 8	77.4978	73.8015	/	/	/	/	/	/	/
			002163	Conditioning	/	/	/	/	/	5	4625	/	/
				1	46.5778	41.9642	39.9	43.1	41.9	4	4213	3166	0.19
				2	44.5406	41.2874	42.5	44.1		5	2968	2136	0.14
				3	43.6284	43.3416	43.7	41.3		5	4227	2546	0.12
				4	44.3092	45.8737	42.8	37.8		5	7504	3507	0.09
		Cold	002164	Feed 8	69.2816	72.2350	/	/	/	/	/	/	/
			002164	Conditioning	/	/	/	/	/	5	4755	/	/
				1	51.1808	49.8122	26.1	31.0	30.8	5	4866	3018	0.12
				2	48.5884	49.3616	29.9	31.7		5	4345	2187	0.10
				3	47.8283	49.1263	31.0	32.0		5	5255	949	0.04
				4	49.3979	46.4436	28.7	35.7		5	7457	320	0.01

Water in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Weight [mg]	Flux [mg/s]
20	Pervatech 300-200 Da CM	Cold	002130	Feed 3	80.7369	89.9292	/	/	/	5	/	/	/
			002130	Conditioning	/	/	/	/	/	5	6045	/	/
				1	69.1759	66.8118	14.3	25.7	17.6	5	5770	1364	0.05
				2	70.7768	73.5514	12.3	18.2		5	6650	1803	0.05
				3	no sample		no value			5	10876	3478	0.06
		Cold	002131	Feed 3	83.7369	85.9292	/	/	/	5	/	/	/
			002131	Conditioning	/	/	/	/	/	5	3931	/	/
				1	63.3949	61.5500	24.3	28.4	22.2	5	3752	1755	0.09
				2	67.3936	69.4636	19.5	19.2		5	7899	3136	0.08
				3	68.6765	65.3896	18.0	23.9		5	10314	2547	0.05
	Pervatech 500 - 400 CM	Cold	002134	Feed 3	75.4345	71.7845	/	/	/	5	/	/	/
			002134	Conditioning	/	/	/	/	/	5	9030	/	/
				1	39.9845	42.0117	47.0	41.5	42.7	5	3280	2694	0.16
				2	44.6356	42.2167	40.8	41.2		5	3741	3058	0.16
				3	40.8385	42.0110	45.9	41.5		5	4641	2945	0.13
				4	43.5746	41.7586	42.2	41.8		5	5075	2152	0.08
		Cold	002135	Conditioning	/	/	/	/	/	5	6331	/	/
				1	48.5623	48.7852	35.6	32.0	35.2	5	6518	5298	0.16
				2	44.4208	47.1732	41.1	34.3		5	5940	4191	0.14
				3	47.7794	48.1551	36.7	32.9		5	4540	2630	0.12
				4	48.0847	48.0649	36.3	33.0		5	6421	2835	0.09
		Cold	002136	Conditioning	/	/	/	/	/	5	6090	/	/
				1	49.4350	50.4464	34.5	29.7	30.8	5	4925	4030	0.16
				2	52.2399	49.4005	30.7	31.2		5	6830	4849	0.14

Water in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Weight [mg]	Flux [mg/s]
				3	52.0397	51.1726	31.0	28.7		5	5109	2818	0.11
				4	51.2033	51.2967	32.1	28.5		5	5230	1769	0.07
	Pervatech 500D - coated	Cold	002152	Feed 6	75.8760	72.8633	/	/	/	/	/	/	/
			002152	Conditioning	/	/	/	/	/	5	8931	/	/
				4	57.3530	55.0744	24.4	24.4		5	3452	1768	0.10
				5	65.7869	62.6517	13.3	14.0	16.6	5	2769	1480	0.11
				6	67.3836	63.8485	11.2	12.4		5	3560	1856	0.10
	Pervatech 700D - coated	Cold	002153	Feed 6	73.8084	77.1697	/	/	/	/	/	/	/
			002153	Conditioning	/	/	/	/	/	5	5394	/	/
				4	70.2961	69.4452	4.8	10.0		5	3078	3197	0.21
				5	69.2828	67.6805	6.1	12.3	8.4	5	1945	2028	0.21
				6	69.9712	68.0797	5.2	11.8		5	2017	1962	0.19
	CM Silane	Cold	002203	Feed 14	65.1066	65.7403	/	/	/	/	/	/	/
			002203	Conditioning	/	/	/	/	/	5	6353	/	/
				1	46.1521	45.4083	29.1	30.9		5	3539	806	0.05
				2	52.9833	52.122	18.6	20.7	24.7	5	2741	519	0.04
										5	3041	525	0.03
										5.5	2336	369	0.03
				3	48.3485	50.6438	25.7	23.0		6	15096	2038	0.02
		Cold	002204	Feed 14	/	/	/	/	/	/	/	/	/
			002204	Conditioning	/	/	/	/	/	5.5	8683	/	/
				1	/	/	/	/	/	6	6283	no flux	no flux
		Cold	002205	Feed 14	64.1813	63.8613	/	/	/	/	/	/	/

Water in Feed [wt%]	Type of membrane	Feed condition	Journal number	Sample	Area [mAU*min]		Rejection [%]		Average rejection [%]	Pressure [bar]	Time [sec]	Weight [mg]	Flux [mg/s]
			002205	Conditioning	/	/	/	/	/	6	8223	/	/
				1	39.2155	41.1461	38.9	35.6	30.7	5	4257	1036	0.05
										5	4220	629	0.03
										5	3554	526	0.03
				2	48.8637	48.1898	23.9	24.5		5	2829	423	0.03
										6	19178	2646	0.02
		Cold	002206	Conditioning	/	/	/	/	/	6	10641	/	/
				1	51.3756	53.5164	20.0	16.2	18.1	6	3544	504	0.02
										6	3492	404	0.02
										6	3271	146	0.01
				2	/	/	/	/	/	6	6372	no flux	no flux