Diploma Thesis

Decolourisation of Starch Hydrolysates using Ultrafiltration

carried out for the purpose of obtaining the degree of Master of Science submitted at TU Wien, Faculty of Mechanical and Industrial Engineering, by

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Vienna, February 2021

I confirm, that going to press of this thesis needs the confirmation of the examination committee.

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Abstract

Most glucose syrups, gained from starch hydrolysis are heavily coloured, which is an unavoidable side process of the production. Therefore different technologies of removing these coloured molecules, such as ion exchangers, have been developed. However, in the last years there is a trend towards membrane separation modules.
In this thesis, several polymer membranes were tested for their ability to remove colour from starch hydrolosates. Therefore, test investigating their separation abilities at different temperatures and pH values were conducted. Membranes with a large molecular weight cut off (100 kDa) were able to achieve a colour rejection of up to 50%, whereas membranes with 5 kDa led to colour rejections of up to 60%.
Furthermore, the different trials and operating conditions were compared to find an optimal compromise of a high colour rejection and a low sugar rejection. Therefore, the sugar composition of the feed, permeate, and retentate is analysed. Additionally, calculations implementing the membrane process are made, investigating the membrane area, as well as the sugar loss. Alongside multi-stage membrane solutions and alternative methods of colour removal, like treatment by hydrogen peroxide or activated carbon, are discussed.

Kurzfassung

Bei der Herstellung von Glucosesirupe entstehen häufig neben den gewollten Zuckern auch eine Reihe ungewollter Nebenprodukte. Diese zeichnen sich vor allem durch ihre intensive Farbe aus. Deshalb sind verschiedene Technologien zur Entfernung dieser Verbindungen entstanden, wie der Ionentausch. In den letzten Jahren werden allerdings Membranverfahren immer attraktiver für diese Aufgaben. In der folgenden Arbeit wurden einige Polymermembranen auf ihre Fähigkeit, diese farbigen Moleküle zu entfernen, getestet. Dazu wurden Versuche bei verschiedenen Temperaturen und pH Werten durchgeführt, und die Trenneigenschaften zu untersuchen. Mit größeren Membranen (100 kDa) konnten bis zu 50% der Farbe entfernt werden, wohingegen engere Membranen (5 kDa) zu einem Farbrückhalt von bis zu 60% führten. Zudem wurden die Ergebnisse der verschiedenen Membranen und Bedingungen miteinander verglichen um ein Optimum von hohem Farbrückhalt und geringem Zuckerrückhalt zu finden. Ergänzend wurden Berechnungen angestellt, welche die nötige Membranfläche und den Zuckerverlust eines solchen Membranprozess in einer realen Anlage untersuchen. Neben den Berechnungen eines einstufigen Prozesses, wird auch ein zweistufiger Prozess, sowie alternative Verfahren zur Farbreduktion, mittels Aktivkohle und Wasserstoffperoxid, erforscht.

Contents

Abbreviations

A ..... Absorption

C_S ..... Sugar Concentration

DE .....Dextrose Equivalent

DP ..... Degree of Polymerisation

HMF ..... Hydroxymethylfurfural

HPLC ..... High Pressure Liquid Chromatography

J ..... Flux

MWCO ..... Molecular Weight Cut Off

PES ..... Polyethersulfone

PS ..... Polysulfone

PVDF ..... Polyvinylidene Flouride

R_C ..... Colour Rejection

RCA ..... Regenerated Cellulcose Acetate

R_S ..... Sugar Rejection

SEC ..... Size Exclusion Chromatography

ρ ..... Density

Chapter 1

Introduction

1.1 Sugar

Sugar is one of the most produced high purity chemicals worldwide [1].Traditionally, it is gained from the sugar beet, mostly in Europe and North America, or the sugar cane, in southern countries, such as Brazil or Australia. In 2016, the production of sugar beets in Austria reached 3.5 million tons [2]. For the European Union the production of pure sugar in 2014 added up to 17 million tons [3] and the worldwide annual production of sugar in 2020/21 is forecast to be 188 million tons, which is an 22 million tons increase to 2019/20 [4].
The average sugar beet contains about 17% sugar, water, and pulp [2], whereas the sugar cane reaches between 12% and 16% sugar content [5]. However, due to the ever rising demand of sugar, alternative production methods are explored, such as the hydrolysis of starch.

The uses of sugar are almost limitless and reach from direct consumption to a building block for bio-fuel or bio-plastics. However, the main use for sugar is in the food industry. There are several different kinds of sugar, depending on their length and their building blocks. The most common classifications is due to the number of building blocks. This divides the sugars into monosaccharides, consisting of a single building block, such as glucose or fructose, and disaccharides, consisting of two molecules connected by a glycosidic bond, such as sucrose and lactose. Longer chains of carbohydrates, such as oligosaccharides and polysaccharides, are typically not refereed to as sugars anymore. The most commonly used sugar is sucrose, which is also refereed to as table sugar. This structure consists of one glucose and one fructose molecule linked through a β -1,4-glycosidic bond. The chemical structure, as shown in figure 1.1, leads to a chemical formula of $C_{12}H_{22}O_{11}$ and a molecular weight of 342 $g\,mol^{-1}$, however in starch hydrolysates maltose is a more common disaccharide.

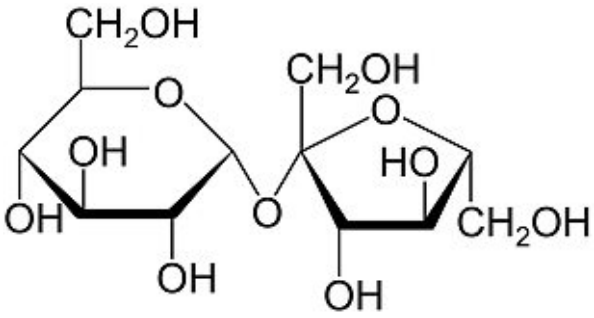


Figure 1.1: A sucrose molecule

1.2 Membrane Technology

Membrane technology is used widespread across many different industries. Especially in the last decades it is often a energy and cost-saving alternative to traditional processes.

Membrane Technology describes the separation of two or more components using a membrane as the separating medium. A membrane process consists of several components, such as the feed, which is the solution containing all the components, the permeate, which only contain the solvent, as well as smaller molecules, which are able to pass the membrane, the retentate, which consists of the molecules, which are not able to pass the membrane, and the membrane its self. As shown in figure 1.2, a membrane process divides the feed into two streams with a different composition.

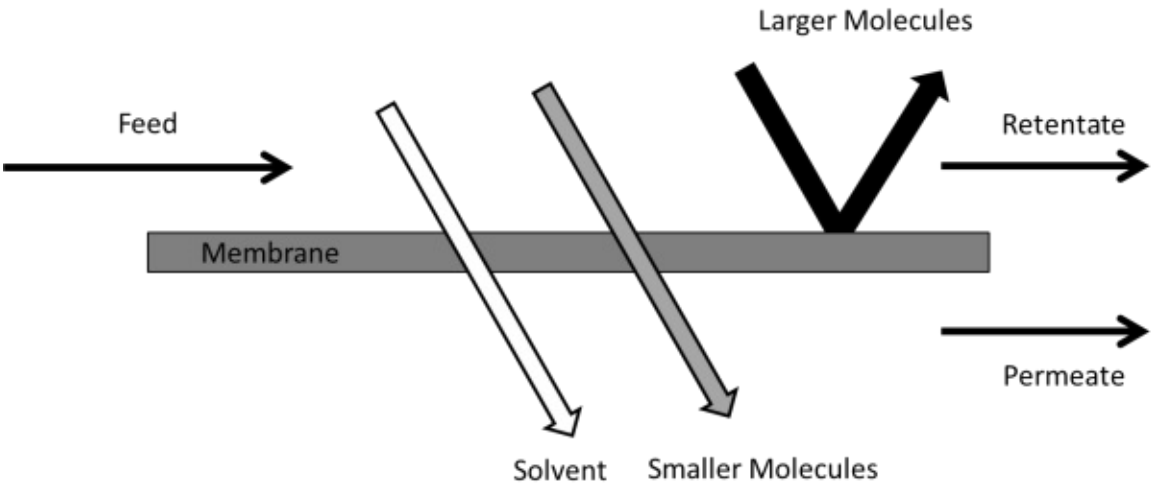


Figure 1.2: Schematic model of a membrane process, adapted form [6]

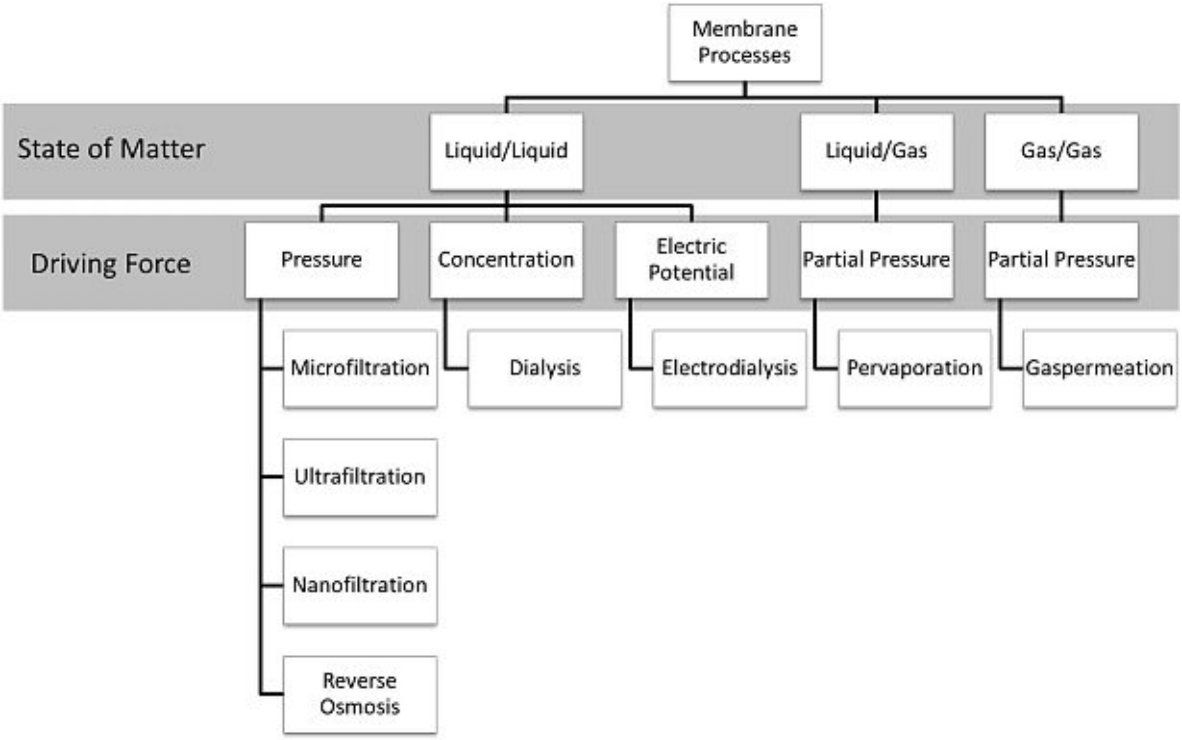


Figure 1.3: Overview of membrane processes, adapted from [6]

The driving force of a membrane process can be a concentration, electric potential, or temperature gradient; however, most of the time a pressure is applied at the membrane's retentate side to create a driving force. Additionally, the state of matter of the permeate and retentate can vary. Today a multitude of different membrane processes are available, which are shown in figure 1.3.

Table 1.1: Classification of pressure driven membrane processes

	Pore Size [nm]	Δp [bar]
Microfiltration	100-10 000	<5
Ultrafiltration	10-100	5-10
Nanofiltration	1-10	10-20
Reverse Osmosis	0.1-1	20-200

The pressure driven membrane processes have divided into several categories regarding the pores' size, and the pressure needed to produce enought driving force. The main classes are shown in table 1.1.

The composition of the membranes, as well as their structure, also varies. The most commonly used membranes include ceramic and organic compound membranes. These membranes are again divided into porous and non-porous as well as symmetrical and asymmetrical membranes. Asymmetrical membranes often have an active and a structural layer, which is necessary in order to support the membrane's pressure.

For the industrial application of membranes a variety of different modules are used. The modules are optimized to withstand the pressure applied and pack an immense amount of membrane area in the smallest volume possible. The most common modules include, tubular, hollow fiber, flatsheet and spiral wound modules.

1.2.1 Tubular Membrane Modules

Tubular membrane modules use tubes with a diameter of about 5-25 mm, on the inside of this tubes there is an active membrane layer, whereas on the outside there is the structural layer. During the membrane process feed flows through the inside of the tube. The smaller molecules, which are able to pass the membrane are pushed through the membrane to the outside of the tube, whereas the retentate is kept inside the tube. Tubular membranes are easy to maintain and insensitive against clogging; however, take up a lot of space and have little membrane area in relation to the volume flow. These modules are mainly used in microfiltration, ultrafiltration and pervaporation.

1.2.2 Hollow Fiber Membrane Modules

Hollow fiber membranes are built similar to tubular membranes, however the Feed can pass from the inside of the capillary through the membrane to the outside or outside to the inside of the tube. Because of to the smaller diameter of <5 mm there is not always a supporting layer needed, however if there is one it depends on the configuration on the outside or the inside of the active membrane layer. Due to the smaller tubes of hollow fiber membrane it is possible to put more surface area into smaller spaces. However, there is the threat of clogging the tubes. Additionally, a laminar flow inside the tube produces a smaller mass exchange with the membrane surface when the feed is flowing inside the tube. These modules are often used in reverse osmosis, gas permeation and dialysis.

1.2.3 Flatsheet Membrane Modules

Flatsheet membranes use flat pieces of membrane which are passed through by the permeate. Because they are not tube shaped, they need an additional casing in order to guide the feed to the membrane. Flatsheet membranes are easy to exchange, and not likely to foul; however, there is much additional sealing required for the casing. These modules are commonly used in microfiltration, ultrafiltration, reverse osmosis, pervaporation, and electro-dialysis.

1.2.4 Spiral Wound Membrane Modules

Spiral Wound membrane Modules use flat sheets of membrane with spacers and permeate drainage layers in between, wound around a tube. The feed flows in the spacer layer, where the smaller molecules pass through the membranes to the permeate drainage collected in the central tube. The larger molecules stay in the spacer layer and exit the module again. Spiral wound membranes have a high membrane area at a small space and a high mass transfer. However the modules are complex to clean and the flat membranes can not be changed individually. These membranes are commonly used in higher pressure applications, such as nanofiltration and reverse osmosis.

1.3 Membrane Technology in Sugar Refinement

For production of high purity sugar, some refining processes necessary, such as removing impurities, which alter the colour and the taste. The refining processes are very energy consuming; therefore, there is a high demand for alternative systems. In the last decades there was a shift towards membrane technology to clean the sugar syrups. Sources, such as [1], [7], [8], [9], [10] and [11] have tested different membranes for their ability to remove colour from various sugar syrups. However most of the sugar juices used in these tests were obtained directly from sugar cane or sugar beet, rather than from starch hydrolysis.

Table 1.2: Membranes used by sources

Membrane Material	**MWCO**	**Colour Rejection [%]**	**Source**
PES	5-100 kDa	35-83	[7]
PES	10 kDa	87	[8]
PES	5 nm	96	[10]
PES	20 kDa	-	[9]
Flouro Polymer	20 kDa	68	[8]
PS	20-100 kDa	70-86	[8]
PS	10-300 kDa	-	[1]
PS	25-100 kDa	48-72	[11]
modified PS	20	-	[9]
Cellulose Triacetate	5-20 kDa	-	[1]
Regenerated Cellulose	1-30 kDa	-	[1]
PVDF	30 -50 nm	69	[10]
Mineral (CARBOSEP)	15-50 kDa	60-90	[7]
Ceramic (Membralox)	20 nm, 1-5 kDa	39	[12]

Table 1.2 shows an overview of the previously tested membranes in connection to colour removal from sugar solutions.[7] tested polyethersulfone membranes with a molecular weight cut off (MWCO) between 5 kDa and 100 kDa and received a colour reduction from more than 80% (5 kDa) to about 40% (100 kDa). Additionally, the investigated mineral membranes between 15 kDa and 50 kDa achieved a colour reduction of up to 90%. Other sources, such as [8] used polysulphone membranes, flouro polymer, and polyethersulphone membranes, with a MWCO between 20 kDa and 100 kDa for polysulphone, 10 kDa for polyethersulphone and 20 kDa for the flouro polymer membrane and received a colour reduction of 86.5% (PES), 85.6% to 70% (PS) and 68% (FP). However the sugar concentration reduced from about 13.6 °Brix in the feed to between 7.8 °Brix (PES, 10 kDa) to 8.4 °Brix (PS, 100 kDa) in the permeate. For applications in the sugar industry this would mean to much sugar loss, and thus it would not be economical.[1] used cellulose triacetate membrane with a MWCO between 5 kDa and 20 kDa, polysulfone membranes with a MWCO between 10 kDa and 300 kDa, as well as regenerated cellulose membranes with a MWCO between 1 kDa and 30 kDa. Of the by [1] tested membranes a regenerated cellulose membrane with a MWCO of 10 kDa showed the best results regarding high flux and purity of the permeate. However, the results do not measure the colour rejection directly, but an increase in purity, therefore there are no results for colour rejection shown in table 1.2. [10] used tubular polyvinylidene flouride membranes with pore sizes between 30 nm and 50 nm and spiral wound polyethersulphone membranes with a pore size of 5 nm. Additionally, to the ultrafiltration [10] used a spiral wound polyamide nanofiltration stage with a pore size of 0.5 nm after the ultrafiltration to concentrate the solution. The spiral wound membrane achieved a colour rejection of more than 95% at a sugar rejection of 10.6%. When additionally adding the nanofiltration stage the colour rejection rises to 96.5%. With the tubular membrane modules a colour rejection of about 69% was achieved. [11] used polysolphone membranes with a MWCO of 25 kDa and 100 kDa, which resulted in a colour rejection of up to 68.4%. [12] achieved a decolorisation of about 50% when using ceramic membranes with molecular weight cut-offs of 20 nm, 5 kDa and 1 kDa and [9] used 20 kDa PES and modified PS membranes, however the colour rejection is not stated.

From these previously conducted tests, obtained from literature, an overview of the available membranes and their colour removal properties is gained. The membranes to investigate are chosen on this basis, including a variety of different materials, molecular weight cut offs, and charges. In the following thesis, these membranes are investigated on their ability to separate coloured molecules from sugar. In order to test the membranes trials with different process parameters, such as temperature and pH value are executed.

Chapter 2

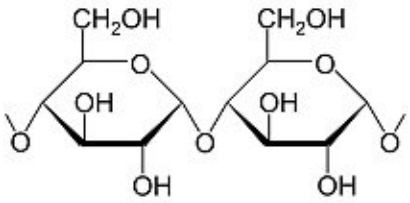
Materials and Methods

2.1 Glucose Syrup

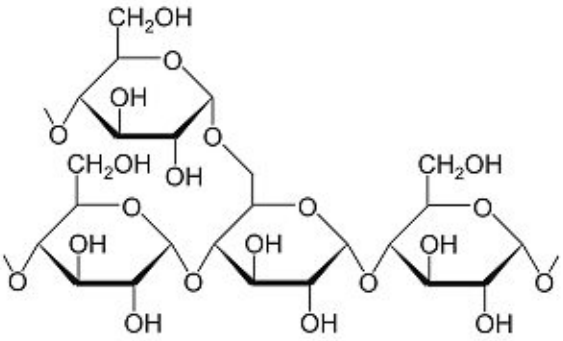
The sugar solutions were obtained from maize and wheat starch by an enzymatic hydrolysis process. Agrana provided the glucose syrup at a concentration of about 70°brix, which was diluted for the experiments using deionized water to about 30°brix.

2.1.1 Starch

Starch is the starting material to the hydrolysates, used in all further tests. It can be found in lots of plants, such as potatoes and rice; however, the starch used for the hydrolysis process came from wheat and maize. The content of starch varies between different plants. Starch consist of the linear molecule amylose (figure 2.1a), which is α-1,4- connected glucose units, and amylopectin (figure 2.1b), which has additional α- 1,6-connected glucose units attached to the linear chain, and consequently produces more complex structures. Additional to their microscopical structures, starch also forms granulates on a macromolecular scale. Additionally, to the varying starch content, the ratio of amylose to amylopectin is varying, when using different sources. Therefore, the choice of starch plant has an significant influence on the final substrate.



(a) Amylose



(b) Amylopektin

Figure 2.1: Starch molecules

2.1.2 Production of the Glucose Syrup

As previously mentioned, was the syrup obtained via hydrolysis of starch. The process of hydrolysis can be carried out using pH driven or enzymatic methods. The syrup used in the following tests was produced using the enzymatic hydrolysis process.

2.1.2.1 Hydrolysis using Enzymes

The following test's hydrolysates were obtained from enzymatic hydrolysis of starch from wheat and maize. To receive smaller molecules from the large starch chains, a slurry of water and starch, with about 30 % dry matter, was produced. For the reduction of the glucose chains the enzyme α-Amylase was used. Due to the properties of the α-Amylase a temperature of 95°C and a pH of about 6 to 6.5 [13] was adjusted.

In order to set the pH at the desired value, NaOH was added to the slurry. At this temperature gelatinization starts, which introduces coloured molecules into the slurry. Additionally the α-Amylase started to reduce the large glucose chains to smaller molecules called maltodextrins. To cut the large starch molecules into smaller ones, the α-Amylase attacked the α-1,4-glycosidic bonds. In figure 2.1 the α-1,4-glycosidic bond is the oxygen bond between the glucose building blocks. The process resulted in a fast decline in the gelatinized starch slurry's viscosity, due to the reduction of the length of the molecules [13]. This reaction to shorter molecules also contributes to the formation of coloured components. Also, glucoamylase was added, which removed α-glucose from larger chains. This process led to further forming of coloured components.

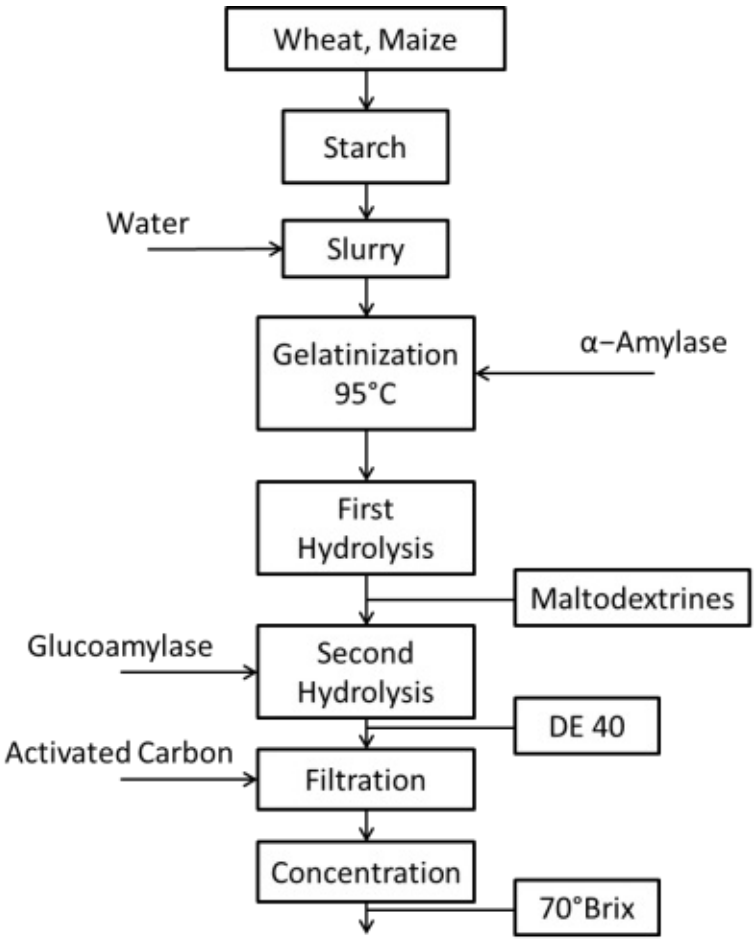


Figure 2.2: The starch hydrolysation process, adapted from [13]

With a combination of the two hydrolysis-processes a syrup with predetermined sugar composition could be achieved. After a Dextrose Equivalent (DE) of 40 was achieved, the solution was filtered using activated carbon and concentrated to 70°Brix.

Additionally to the hydrolysis using enzymes there is an alternative method using hydrochloric acid or sulfuric acid [14]. However, this way of producing sugar from larger molecules is not as common as the enzymatic reaction and is, therefore, not discussed any further.

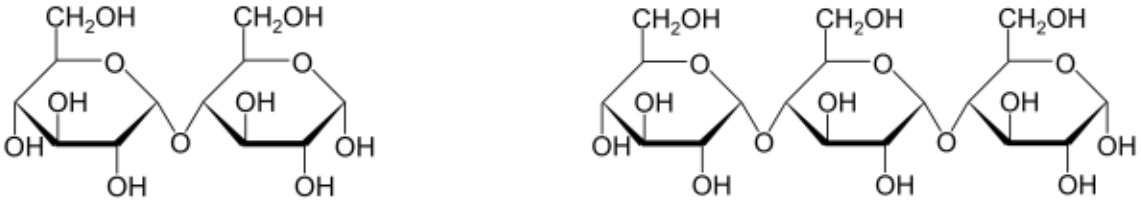
2.1.3 Composition of the Glucose Syrup

In table 2.1 are some of the components in the sugar syrup shown. The analysis of the components was made by Agrana using ionchromatography for anions and inductively coupled plasma optical emission spectrometry for kations. The carbohydrate spectrum was gathered by using a high performance liquid chromatography. As shown, there were some ions present in the solution, which led to the later on measured conductivity. However, there was a vast change in pH between the two different batches used in the tests. This suggests a different ion composition in the two glucose syrups. The carbohydrate spectrum shows that there is a rather high concentration of larger molecules, and fewer shorter molecules, such as maltose and glucose. Two shorter maltodextrine molecules, maltose (figure 2.3a) and maltotriose (figure 2.3b), are shown below. However, this analytic methods do not give an insight in the structure of the coloured molecules.

Table 2.1: Composition of the glucose syrup

		Dry Matter [%]	72
Ion Analysis	**Anion**	**Chloride Ion** [% i. DM]	0.03
		Sulfate Ion [% i. DM]	0.11
		Phosphate Ion [% i. DM]	0.01
		Citrate Ion [% i. DM]	<0.01
	Kation	**Nitrate Ion** [% i. DM]	<0.01
		Na [% i. DM]	0.0906
		K [% i. DM]	0.0156
		Ca [% i. DM]	0.0044
		Mg [% i. DM]	0.0038
Carbohydrate Spectrum		**DP4+** [% i. DM]	46.92
		Maltotriose [% i. DM]	22.32
		Maltose [% i. DM]	19.1
		Glucose [% i. DM]	9.55
		HMF [$mg\,kg^{-1}$]	15.94

Even though the browning process is desirable in lots of applications of carbohydrates, the sugar solutions gained from the starch hydrolysis should contain little to no coloured molecules for the further processing. There are several ways the brownish colour is introduced into the substrates, which can be either the Maillard reaction or a Caramelisation process [13]. At both of the reactions carbohydrates are the starting point of the reaction, which conclude in coloured molecules.



(a) Maltose

(b) Maltotriose

Figure 2.3: Maltodextrines

2.1.3.1 Maillard Reaction

During the Maillard reaction reducing sugars react with amino groups in proteins or free amino acids. Generally all of the glucose syrups contain aldehyde or ketone groups, necessary for the reaction. However, there is a relation between the sugar syrup's composition and the likeliness of the Maillard reaction, where a higher Dextrose Equivalent leads to a more reactive syrup. There are also differences in the reactivity of the amino groups, where amino acids are more reactive than larger proteins. As stated previously, the starch for the hydrolysis process is taken from starchy plants. Because there are impurities when separating the starch from the remaining plant, there are always some proteins and amino acids in the starch slurry. The number of proteins in the slurry depends on the kind of source material used. Maize or wheat present a rather high protein concentration, whereas potatoes or tapioca typically have a lower amount of proteins [13]. The Maillard Reaction is faster when the syrup is kept at higher temperatures, which is often necessary to prevent crystallization or reduce the syrup's viscosity for easier handling. [13]. Additionally, the pH has a enormous impact on the reaction's speed, where at acidic levels, the reaction is slowed down [13].

2.1.3.2 Caramelisation

Whereas the Maillard reaction occurs at lower temperatures, and only in the presence of amino groups, the caramelisation is bound only to high temperatures, but is favoured by an alkaline environment. Similar to the Maillard reaction, caramelisation is a process that occurs more at higher Dextrose Equivalents and is dependent on the types of sugar in the syrup. The different types of sugar start their caramelisation processes at different temperature, where Fructose starts at about 70°C and larger sugars are stable beyond this temperature. Caramelisation is mainly a problem if there are spots in a process which exceed this temperature. [13].

Both the Maillard reaction and the caramelisation process lead to various compounds, including some aroma substances, which are desired in several food applications, and brown compounds. These molecules vary in molecular weight, and their structure is little known [15].

2.1.4 Fermentation

Additionally, to the threat of browning there is also the probability of fermentation when handling sugar solutions at room temperature. For the subsequent trials, a fermentation during the solution's storage and the samples would falsify the results. Consequently, it is of great importance to reduce the tendency to ferment. The Dextrose Equivalent is used as an indicator of fermentability, where a higher DE leads to a larger possibility of fermentation [13]. However, the DE is not a manipulable variable in the following applications. Therefore, other influencing factors must be changed in order to encourage the least amount of fermentation. The concentration of the solution plays a rather large role in the growth speed of microorganisms. At higher sugar concentrations the water activity decreases, which leads to reduced cell division and thus slower growth [16]. The concentration of the raw glucose syrup is about 70°Brix, which is enough to slow the growth of microorganisms. However, it is diluted to a concentration of about 30°Brix to run trials, making it vulnerable to microorganism's growth. Thereupon, the samples were stored at temperatures below 0°C, when not used, to keep the syrup composition stable for longer times.

2.2 Membranes

There were different membranes tested regarding their construction material, molecular weight cut off, and charge. All the tested membranes are flat-sheet membranes made out of organic materials.
The membranes were obtained from different companies such as Koch Separation Solutions (M183, M180, P707, K328), Microdyn Nadir (UH050, UH020), and Alfa Laval (RC10PE).

Table 2.2: Membranes

Manufacturer	**Type**	**Membrane Material**	**MWCO [kDa]**
Koch Separation Solutions	M183	positively charged PVFD	100
	M180	neutrally charged PVDF	100
	P707	negatively charged PVDF	120
	K328	PES	5
Microdyn Nadir	UH050	hydrophilic PES	50
	UH020	hydrophilic PES	20
Alfa Laval	RC10PE	Regenerated Cellulose Acetate	10

As shown in table 2.2, the membranes range from a MWCO of 5 kDa to 120 kDa. Their composition materials are polyvinylidene flouride (PVDF), polyethersulfone (PES) and Regenerated cellulose acetate (RCA). Additionally to their composition the membrane vary in charge, such a positively, neutrally, and negatively charged PVDF membrane as well as their wetting properties.

2.3 Experimental Setup

The setup is a small scale membrane testing station, consisting of a membrane module for flat sheets, a pump, and two valves to dial in the desired pressure.

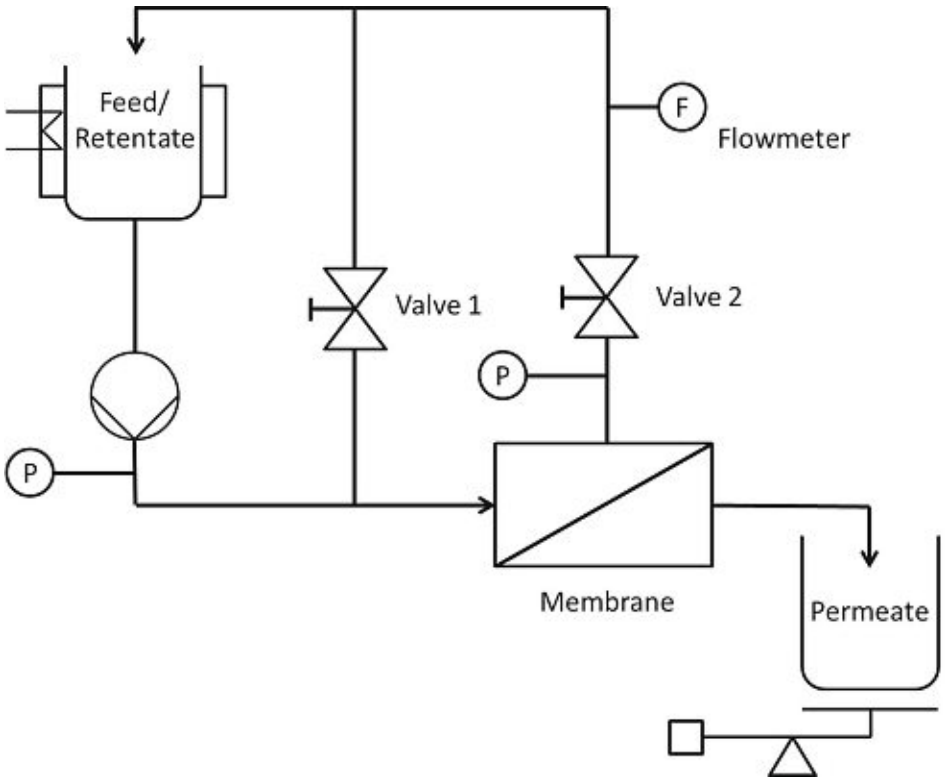


Figure 2.4: Flowsheet of the experimental setup

The plunger pump (Cat Pumps 231) is supplied from the 2-liter feed tank and pushes the bypass and the membrane's fluid. At the bypass, there is a valve in order to close it and make the fluid go only to the membrane. After the membrane, there is another valve at the retentate side to control the retentate pressure. The bypass, as well as the retentate from the membrane get recycled to the feed tank. After the pump and between the membrane and the valve at the retentate side, there is a pressure gauge to monitor the pressure. Additionally, the tank is equipped with a heating unit (VWR) to heat the liquid to the desired temperature. The equipment can be operated at a temperature ranging from 5°C to 70°C and a pressure up to 64 bar. However, in the following ultrafiltration applications the pressure does not exceed 6 bar. In figures 2.4 a flowsheet and in figure 2.5 a picture of the experimental setup is shown.

Figure 2.5: Picture of the experimental setup

2.4 Procedure

The membranes were first characterized with water in order to determine the pure water flux. After the tests with water, the sugar syrup was used. In these tests, the pH value and the temperature are varied to study their influences on the flux and decolouration. After the test with sugar syrup were finished the membrane is cleaned and again tested with water to determine any flux drop, related to fouling, compared to the first test using water.

There were several different test conducted using different membranes and different parameters. The modified parameters included temperature and pH values.

2.4.1 Test using Water

After the new membrane was installed into the apparatus, deionised water is poured into the empty tank. The valves 1 and 2, as seen in figure 2.4, were opened all the way. With this configuration, the solution is using mainly the bypass. After starting the pump valve 1 was closed to let the solution flow only to the membrane. Then the retentate pressure was dialled in using valve 2. The permeate was collected in a beaker, and the transmembrane flux was measured using a scale and a stopwatch. The tests using water were performed multiple times, measuring the mass of the transmembrane flow in a two-minute interval. After the test's conclusion, the pressure was released by opening valve 2 and shutting off the pump. All the tests using deionised water were carried out at room temperature.

2.4.2 Tests of the Polyvinylidene Flouride Membrane, 100 kDa, and Positive Charge

In the test using the sugar solutions, the procedure was similar to the tests using water. However, before, the test started, the mass poured into the equipment was weighted, and the permeate tube was unscrewed to empty the remaining water. To homogenise the solution before the start of the measurements the water, which was remaining in the equipment, was mixed in by returning the tube from the permeate side of the membrane to the feed tank and closing valve 1. However, due to the low pressure of the retentate side, there was almost no transmembrane flux. Then the heater was activated in order to heat the solution to the desired temperature. A small sample of the feed was then extracted in order to test its absorption and sugar concentration. The experiment started, after the desired temperature was reached, and the pressure was dialled in. Therefore, the permeate tube was moved from the feed tank to the permeate collecting beaker. The beaker's weight change was measured every 10 minutes and the pH, conductivity, and index of refraction was measured every 20 to 30 minutes. After the test, the permeate and retentate were weighted, and both were tested for their absorption. Following the test using the sugar solution, the membrane was again tested with deionised water to see the decrease of permeability of the membrane. The water permeability measured before and after the test was used to compensate the received data for the flux during the tests. So it can be compared to the following tests, even though the same membrane was used for multiple tests. The test was concluded once about 1200 ml of permeate were collected.

The first test used the natural pH of the solution and an operating temperature of 30°C, the second used natural pH again, but 50°C. For the third test using this membrane, the pH was increased to 9.5 using NaOH, and the temperature was again dialled in at 50°C. The final test was equal to the previous one, but the pH value was lowered to 3.3 using citric acid.

2.4.3 Tests of the Polyvinylidene Flouride Membrane, 100 kDa and Neutral Charge

The test using the second membrane were equal to the previous ones, however the test using a lower pH level was carried out by reusing the permeate and retentate from the test using a high pH. Therefore, the previous day's solutions were remixed again to get the feed solution, and citric acid was added to lower the pH. However, since there was some loss of solution in the equipment, the feed's concentration was only 20.9°Brix. Because there was less solution to start with, the experiment was concluded after receiving about 800 ml of permeate.

2.4.4 Tests of the Polyethersufone Membrane, 5 kDa

The tests were carried out equally to the previous ones, however, because the membrane had a much lower MWCO, the tests were concluded, when the retentate reached a concentration of more than 29°Brix. Due to a shortage of new sugar solution, it was reused for several experiments. Therefore, after the first test, the equipment was not cleaned with water to keep the residual sugar, which could not be removed, in the equipment. Thus, there was no permeability test, using water, performed between the first two experiments. By this strategy, there was almost no loss of sugar solution. After the second test using 50°C and natural pH, the equipment was emptied and cleaned. Furthermore, a test determining the permeability using water was executed. The third test using this membrane at a pH of 2.95, was conducted with the previous test's retentate and permeate solution. However, because the equipment was emptied and cleaned there is some new, high concentrated sugar solution added to gain a similar sugar concentration as the previous tests. For this test, there was again citric acid added in order to lower the pH. After the third test, the procedure was equal to the procedure after the first one, where the equipment was not cleaned in order to keep the residual sugar solution. The final test using this membrane was conducted using a pH of 9.64. This test was again completed when the solution's level in the feed tank reaches a minimum amount of liquid to supply the pump with enough fluid. After the tests were concluded, samples of the permeate and retentate were taken and tested for their colour.

2.4.5 Tests using the Polyvinylidene Flouride Membrane, 120 kDa and Negative Charge

The test using this membrane were conducted equally to the previous ones. The first test used a temperature of 50°C and the natural pH. After finishing this trial, there were samples collected from the permeate and the retentate. Then the permeate was returned to the feed tank, and the temperature was increased to 50°C, at which the second test was carried out. In the end, the permeate and retentate were removed from the equipment, and the apparatus was cleaned using water. For the third test, previous trials permeate and retentate were again combined to create the feed. Additionally, there was some fresh syrup added to get a similar sugar concentration in the feed. Adding a new solution was necessary, because the equipment was cleaned, and the equipment's residual water further diluted the solution. After this, the citric acid was added, until the pH value was lowered to about 3.1. The solution was added to the equipment and heated to 50°C. After finishing, the permeate and retentate were again removed from the equipment for sampling. For the last test, using a pH of 10.15, NaOH was added to the combined permeate and retentate of the previous test. The remaining trial was carried out equally to the previous ones at a temperature of 50°C. After finishing, a sample was taken from both the permeate and retentate and tested for their colour.

2.4.6 Tests using the Hydrophilic Polyethersulphone Membrane, 50 kDa

The tests using the hydrophilic polyethersulphone membrane were conducted equally to the previous ones, by reusing the substrate for all of the tests. First, the solution was tested using the natural pH value and 30°C. After the conclusion of the test a sample of the permeate and the retentate was taken, and the permeate is filled back into the feed tank. Then the heater was set to 50°C, and the second test was started. After finishing the second test, a sample was again taken from the permeate and retentate for further analysis. The rest of the permeate and retentate was then removed from the equipment and stored in a freezer. The equipment was, however, not cleaned in order to receive comparable results with further tests. For the third test, there was again the retentate and permeate combined and used as a feed. Additionally, there was NaOH added to raise the pH value to 9.73. After the desired pH value was reached, the solution was poured into the feed tank and heated to 50°C. There were again samples taken from the final permeate and retentate. For the final test, the retentate was added to the permeate, and the pH value was lowered to about 3 using citric acid. Then the solution was poured into the feed tank, and the last trial using this membrane was started.

2.4.7 Tests using the Hydrophilic Polyethersulphone Membrane, 20 kDa

The test using 30°C and 50°C at the natural pH were conducted equally to the previous tests. The solution for the first test was prepared fresh and was then reused for the subsequent tests. The pH value was again lowered using citric acid, and a test was conducted using 50°C. For the last trial, the NaOH was added and executed equally to the previous ones at 50 °C. After the test, there were samples taken from the permeate as well as the retentate.

2.4.8 Tests using the Regenerated Cellulose Acetate Membrane, 10 kDa

The tests using the regenerate cellulose acetate membrane were conducted equally to the first trials in chapter 2.4.2, using new solution each time and performing water permeability test in between all the trials. However, the new solution's natural pH was about 5.5, whereas the natural pH of the solution used in the previous test was roughly 4.4.

2.4.9 Cleaning of the Membranes

The equipment was cleaned after some tests using the sugar solution to restore some of the permeability lost during the test. For the cleaning process 50% NaOH was mixed with water in order to receive a solution with pH 11. This solution was than poured into the tank and the pump turned on. The retentate pressure was then increased in order to produce some permeate flow. After some flow, the pump was shut down and the equipment was flushed several times using water.

2.5 Additional Tests

2.5.1 Test using Permeate of the Neutrally Charged Membrane as Feed for the Positively Charged Membrane

In order to test the effects of a two-stage process compared to the single-stage processes, the permeates of the tests using the neutrally charged PVDF at a natural pH and 30 °C as well as 50°C (3.2.1 and 3.2.2) were used as the feed for a second membrane filtration using the positively charged PVDF membrane. The test was again conducted using the natural pH and 30°C. As shown in figure 2.6, a reduction in concentration was happening between the first and the second stage, because the equipment was opened and cleaned, and therefore, the remaining water diluted the feed when poured back into the feed tank. This decreased the feed sugar concentration to about 22.7°Brix.

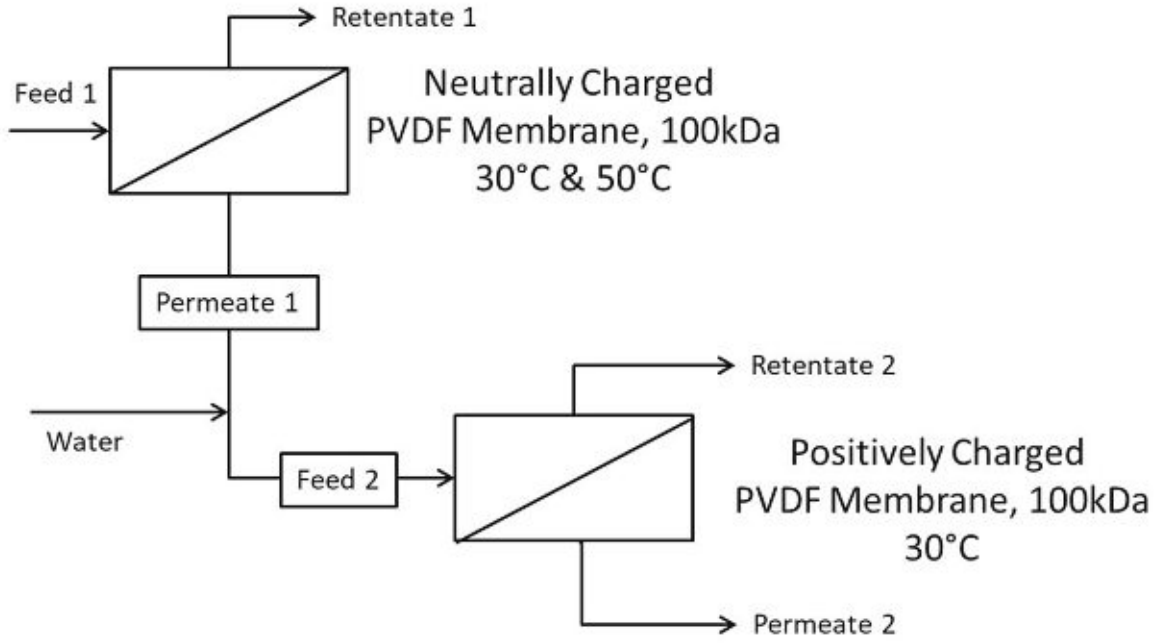


Figure 2.6: Flowsheet of the two-stage process

2.5.2 Effects of the pH Value on the Colour, using Different Alkaline and Acid

Test were concluded using different alkaline, to see the influence of the pH on the colour. Therefore, a solution with a concentration of 26.8°Brix was produced using the sugar syrup and deionised water. Then a sample of the diluted solution was taken and tested for colour. The remaining solution was divided into several parts. One portion's pH was slowly increased using calcium hydroxide, taking several samples at different pH values and testing them for colour. The test used a pH range from the natural pH of about 5.4 to a pH value of about 10. This test was repeated using sodium hydroxide, as well as magnesium oxide. After using alkaline to increase the pH value, the pH value was decreased using citric acid. There were again samples taken at different pH values, ranging from the natural pH to about 3. Additionally to the colour, the sugar concentration of all the samples was measured.

2.5.3 Effects of Activated Carbon and H_2O_2 on the Colour

To investigate additional methods of colour removal, tests using activated carbon and H_2O_2 were conducted.

For the tests using activated carbon, three different types were used. The first one was Donau Carbon Supersorbon C IV spezial, the second one Chemviron Carbon Ammonosorb, and the third one Chemviron Carbon Envirocarb AP4-60. All of the activated carbons were pelletised and therefore needed pulverization into a more delicate powder.
For all the activated carbons the same three tests were carried out, where 0.1g, 0.5g and 1g of the carbon was mixed with a solution diluted to 29.9°Brix. After one and a half hours, the solution was separated from the activated carbon using a centrifuge at 3500 rpm for 16 minutes and measured for colour and sugar, using the photometer and refractometer. However, because the centrifuge could only hold 8 samples the test using 0.5 g Chemviron Carbon Envirocarb AP4-60 was not analysed.

Table 2.3: Mixing ratios of the H_2O_2 tests

H_2O_2 [ml]	**Sugar Solution [ml]**	**Mixing Ratio**
5	5	1:1
5	10	1:2
5	15	1:3
5	20	1:4
5	25	1:5
1	10	1:10

For the tests using H_2O_2, a 30% H_2O_2 solution was added to the same sugar solution with 29.9°Brix. The tests used different mixing ratios shown in table 2.3. After the samples were mixed, they were tested for their colour and sugar concentration using the photometer and refractometer.

2.6 Analytic Methods

For characterising the solutions before and after the membrane treatment, there were several analytic methods used.

2.6.1 Analysis of the Colour Using a Photometer

The colour of the feed, the permeate and retentate were examined using a photometer (Shimadzu). Therefore, the photometer was calibrated using deionized water, then the absorption of the samples was tested at a wavelength ranging from 300 to 600 nm. The wavelength used in order to determine the colour was 420 nm, which conforms to ICUMSA Analytic Methods [17], however, the pH of the tested samples was not adjusted. In order to calculate the colour from the absorption and the sugar concentration

$$Colour = \frac{A \cdot 100000}{C_S \cdot \rho \cdot 5} \tag{2.1}$$

was used.
The colour rejection was then calculated using

$$R_C = 1 - \frac{Colour_P}{Colour_R}. \tag{2.2}$$

2.6.2 Analysis of the Concentration using a Refractometer

The index of refraction was measured using a refractometer at a temperature of 20°C. This index suggests the concentration of sugar in the solution. However, since the solution consists of other sugars besides sucrose the concentration could only be estimated. Nonetheless, this was method used for the calculation of the sugar concentration.
The sugar rejection was calculated using

$$R_S = 1 - \frac{C_{S,P}}{C_{S,R}}. \tag{2.3}$$

2.6.3 Analysis of the pH

The feed, permeate, and retentate's pH values were measured periodically using a WTW ADA S7/IDS pH electrode.

2.6.4 Analysis of the Conductivity

The solutions' conductivity was measured equally to the pH using a WTW Tetracon 925 conductivity cell.

Both the pH electrode and the conductivity cell signals were processed in a multiparameter meter WTW Multi 3430.

2.6.5 Analysis of Hydroxymethylfurfural using High-Performance Liquid Chromatography

High-performance liquid chromatography tests were conducted to determine the solutions' HMF concentration before and after the membrane process. Therefore, the samples were filtered using a 2 µm syringe filter and later diluted 1:1 with deionised water. The HPLC's analysis was made for a selection of samples, which produce the best results in terms of colour and sugar rejection. The tested samples contained the feed, the final permeate, and the retentate of the membrane trial.

2.6.6 Analysis of the Sugar Composition using Size Exclusion Chromatography (SEC)

To get an insight on the sugar composition of the solution before and after the ultrafiltration process, a size exclusion chromatography was conducted. The SEC was made by Agrana, using samples gained from various trials at the TU Wien.

Chapter 3

Results and Discussion

In this chapter, the measured data is presented and discussed. Additionally, the results of different membranes as well as different operating conditions are compared to one another. Furthermore, considerations are made on which membrane and operating conditions are the most suitable for the given task. Therefore, different approaches of judging the trials are investigated.
Figure 3.1 shows the retentate and permeate of the test using the positively charged PVDF membrane of 100 kDa at 50°C and a pH of 9.5.

Figure 3.1: Retentate and permeate of test 3.1.3

3.1 Polyvinylidene Flouride Membrane, 100 kDa and Positive Charge

3.1.1 Natural pH and 30°C

The test with a positively charged polyvinylidene flouride membrane with a molecular weight cut-off of 100 kDa, resulted, as shown in table 3.1, in a colour rejection of about 52%. The sugar rejection at the start was about 8% but raised to 16% during the test, resulting in a mean sugar rejection of about 12.6%. , the flux, as it can be seen in figure 3.3 was relatively low, starting at about 70 $\mathrm{kg\,m^{-2}\,s^{-1}}$, but dropping to about 45 $\mathrm{kg\,m^{-2}\,s^{-1}}$ after about 20 minutes and declining slowly to about 29 $\mathrm{kg\,m^{-2}\,s^{-1}}$ after 250 minutes. The results, shown in figure 3.3, are compensated with the water permeability after the test. Monitoring the water permeability was necessary because the membranes are used for several tests and the permeability changes. Additionally, the test showed heavy fouling during the sugar ultrafiltration process. The water permeability of the membrane after the test was about 54% of the starting permeability but could be restored to 69% using NaOH solution. That leads to the assumption, that the membrane was clogged with larger molecules, which decreased the ability of the sugar solution to pass at a higher rate. In figure 3.34, it is also apparent that this membrane ties molecules inside the membrane.

3.1.2 Natural pH and 50°C

A higher temperature led to a higher transmembrane flux, due to the lower viscosity. However, the higher flux resulted in a worse colour rejection of about 23.7%, which is significantly lower than the results received at a temperature of 30°C. Similar to the colour rejection the sugar rejection also dropped with the increased temperature to about 4.5%, which is almost a third of the test using 30°C.

3.1.3 Higher pH and 50°C

The colour of the feed solution, after adding the NaOH, was significantly darker than the previous tests. The darker colour may be due to some reactions happening with the added NaOH or reactions happening due to the higher pH. Another possibility is that the heat released due to the dissolving of the 50% NaOH produces areas in which the temperature rises above the caramelisation temperature, leading to the feed's stronger colouration. The flux was higher than at 50°C and natural pH. The colour rejection was about 47.1%; however, the permeate was still heavily coloured, because the feed was rather dark. The sugar rejection of the test was with 7.3% still a lot lower than with 30°C. Additionally the permeability for water improved during the test, which leads to the assumption that the high pH cleaned the membrane from blocking molecules of the previous experiments.

3.1.4 Lower pH and 50°C

The flux through the membrane was a little bit lower than the previous test with the increased pH, however still higher than the test using the natural pH and 30°C. The colour rejection was with roughly 20.5% the lowest of the tests carried out with this membrane. The sugar rejection was still relatively low with about 6.9%; however, due to the lousy colour rejection, the conditions using a lower pH are not suitable for this process.

Table 3.1: Results of the positively charged PVDF membrane, 100 kDa

Temperature [°C]	**pH**	**Colour Rejection [%]**	**Sugar Rejection [%]**
30	4.29	52.0	12.6
50	4.42	23.7	4.5
50	9.50	47.1	7.3
50	3.31	20.5	6.9

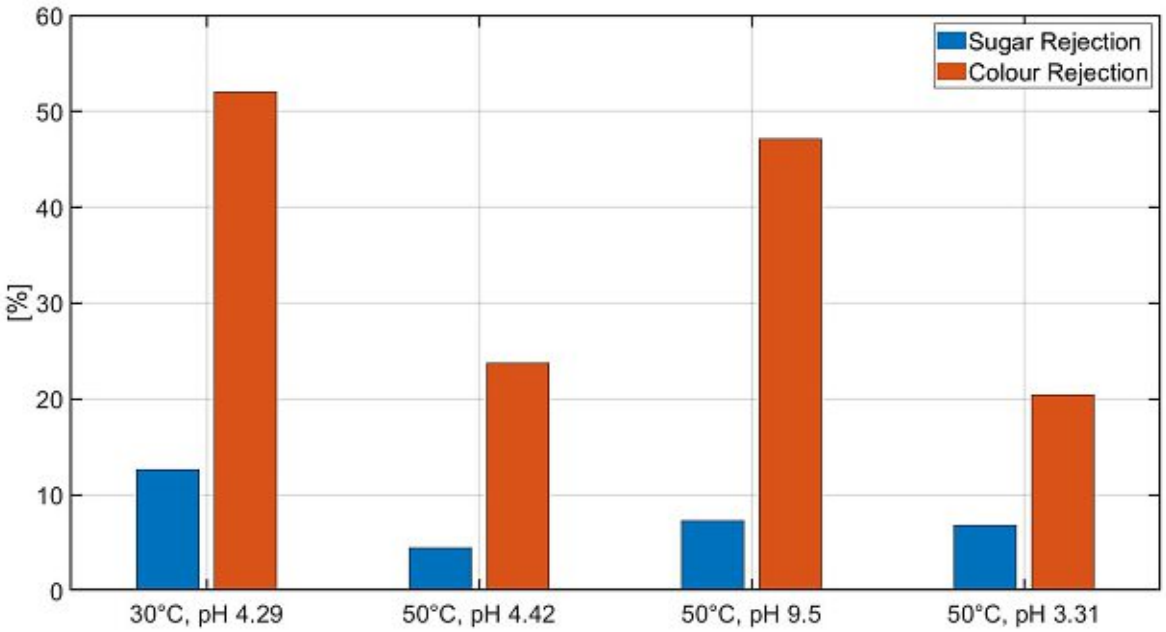


Figure 3.2: Results of the positively charged PVDF membrane, 100 kDa

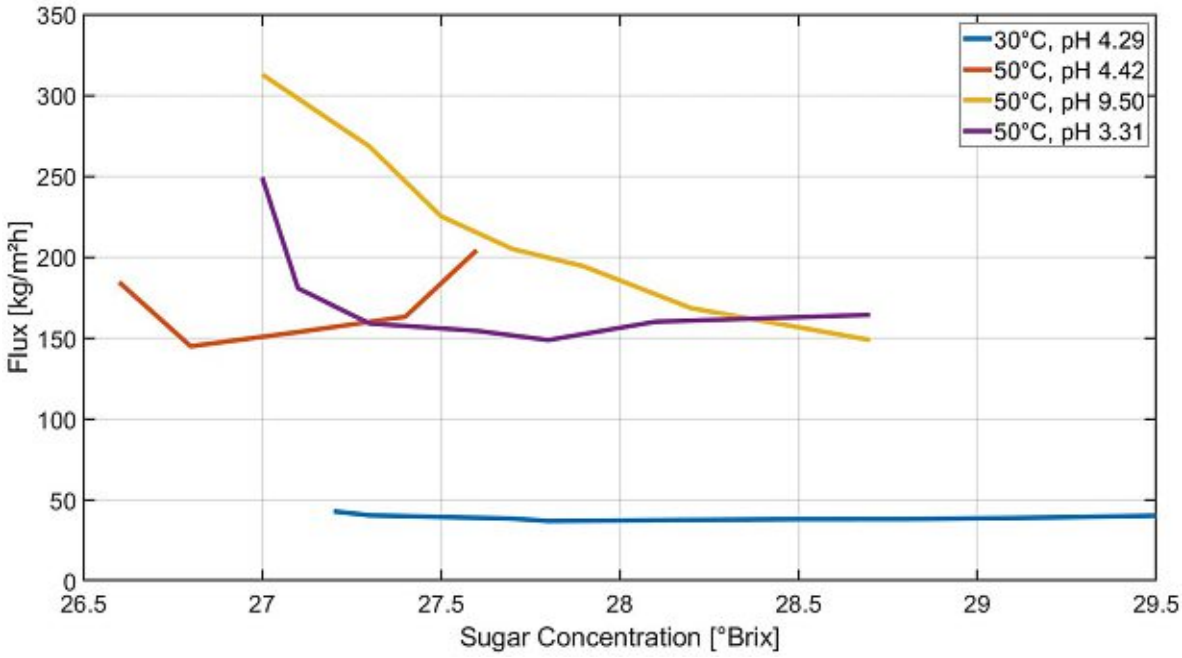


Figure 3.3: Flux of the positively charged PVDF membrane, 100 kDa

The data obtained from the first tests using the Polyvinylidene Flouride Membrane lead to the assumption that lower temperature are better conditions to remove the sugar solution's colour. However, there is very little flux at lower temperatures, which would lead to large membrane areas to receive a enough flow for the industrial application. Furthermore, the colour and sugar rejection behave somewhat similar, therefore the better the colour rejection, the higher the loss of sugar in the permeate. The changing of the pH does not bring significant benefits, even though the colour rejection of the test using a higher pH is quite good the colour of the permeate is still darker than the solution before adding the NaOH.

3.2 Polyvinylidene Flouride Membrane, 100 kDa and Neutral Charge

3.2.1 Natural pH and 30°C

Using the natural pH of the sugar solution and a temperature of 30°C resulted in a colour rejection of about 38.8% and a sugar rejection of about 0.94%. Even though the sugar rejection was relatively low, the colour rejection was not as good as the positively charged PVDF membrane. Differently from the previous test, the retentate was slightly cloudy.

3.2.2 Natural pH and 50°C

The higher temperature led, like the previous membrane, to a higher flux and a lower rejection of sugar and colour. The colour rejection was 30.7% and the sugar rejection 0.75%. Therefore, there was almost no sugar loss; however, the decolourisation was also a lot lower than desired. The relative decrease of the sugar and colour rejection at the increased temperature is identical at about 21%. However, the absolute decrease of the colour rejection is a lot higher than of the sugar rejection.

3.2.3 Higher pH and 50°C

The feed's colour was like the last tests with a higher pH, a lot darker than the previous test. This resulted in a colour rejection of about 32.0% and a sugar rejection of about 2.1%. However, since the feed was coloured heavily, the permeate is still rather darkly coloured.

3.2.4 Lower pH and 50°C

The feed used in this test was obtained from mixing the permeate and retentate of the previous test, resulting in less feed with a lower sugar concentration; therefore, the flux shown in figure 3.5 is compensated using the feed concentration, in order to make it comparable to the other tests. However, the feed colour was a lot lighter after adjusting the pH to lower values than previous test. Because of the reuse of the solution, there was NaOH as well as citric acid added. This resulted in a higher concentration of ions in the solution, seen in the higher conductivity. The tests resulted in a colour rejection of 34.7% and a sugar rejection, similar to the test using the higher pH, of about 2.1%.

During all the tests using the neutrally charged Polyvinylidene Flouride membrane, there was no cleaning using NaOH.

Table 3.2: Results of the neutrally charged PVDF membrane, 100 kDa

Temperature [°C]	pH	Colour Rejection [%]	Sugar Rejection [%]
30	4.37	38.9	0.94
50	4.48	30.7	0.75
50	9.34	32.0	2.1
50	3.15	34.7	2.1

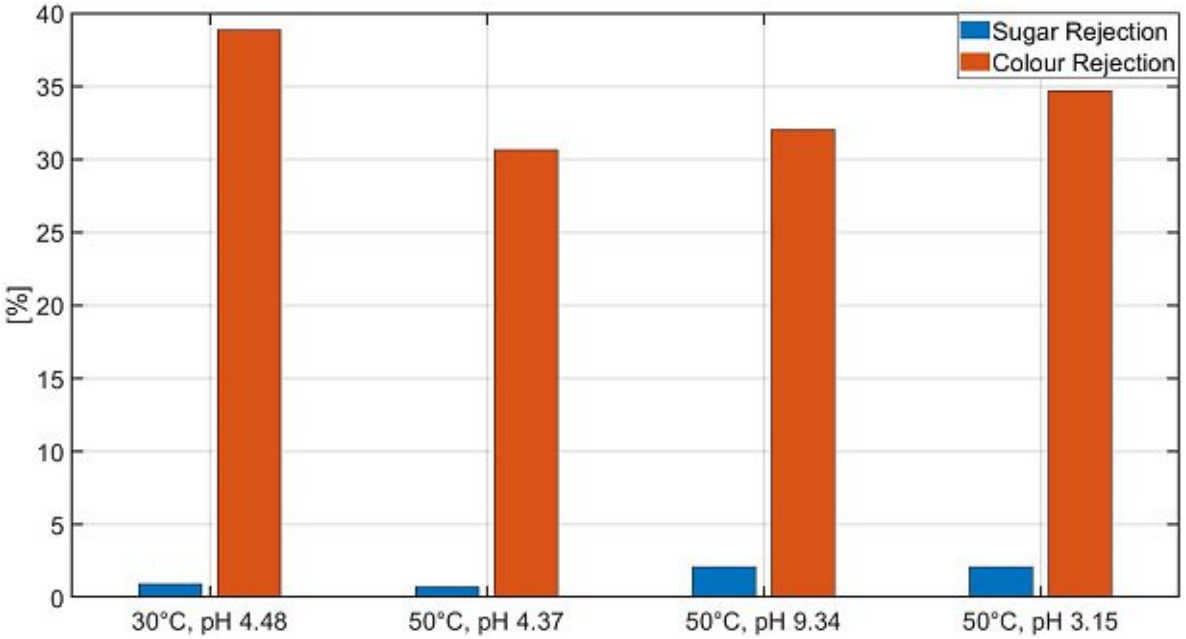


Figure 3.4: Results of the neutrally charged PVDF membrane, 100 kDa

The trials using the neutrally charged PVDF membrane vary heavily form the results of the positively charged PVDF membrane of the same MWCO, to that effect that the sugar rejection and the colour rejection was a lot lower. There was also an increase in sugar rejection when adding either citric acid or sodium hydroxide of more than 100%, whereas there was only a relatively small increase in colour rejection.

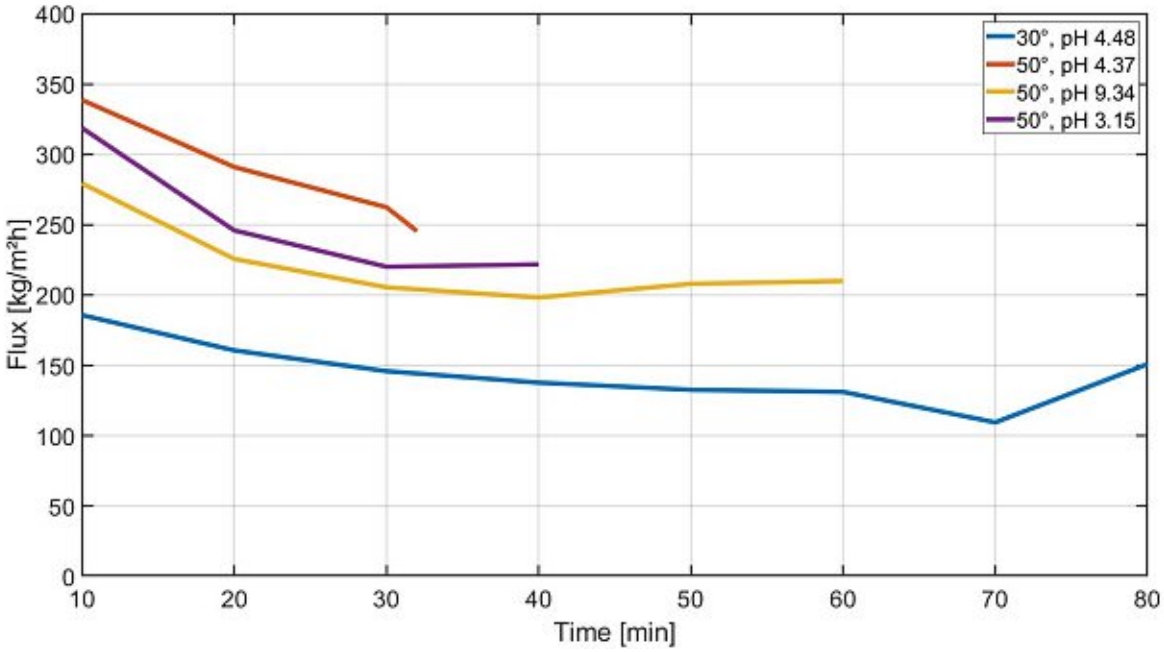


Figure 3.5: Flux of the neutrally charged PVDF membrane, 100 kDa,

Because there was very little sugar rejection while using the neutrally charged PVDF membrane, it is impossible to plot the flux over the retentate refraction index. Therefore, the flux is plotted over time. However, because the concentration of sugar at the test using a lower pH was significantly lower than the other ones, the results, seen in figure 3.5, are compensated using the sugar concentration of the feed. It is notable that unlike most other membranes, the test using an increased pH did not produce the highest flux, but the tests at natural and low pH result in a higher flux.

3.3 Polyethersulfone Membrane, 5 kDa

3.3.1 Natural pH and 30°C

From the beginning, the test's flux was very low, since the MWCO was much lower than the previous membranes. The lower MWCO makes is harder for any larger molecules, such as sucrose, to pass and the membrane blocks quickly. Additionally, the permeate's refraction index suggests a low concentration of sugar in the permeate and led to an average sugar rejection of 27.2%. Even though the colour rejection was with 53.8% relatively high, the sugar rejection and low flux make this membrane not suitable in order to separate the sugar's colour.

3.3.2 Natural pH and 50°C

As expected, the flux was higher than the test using 30°C, however, still rather low compared to the previous membranes. Even though the membrane is with a MWCO of 5 kDa rather fine-spun the colour rejection was with 34.7% still smaller than previous membranes. Furthermore, due to the lower MWCO, the sugar rejection was with 23.3%, similarly high as the previous test.

3.3.3 Lower pH and 50°C

The results of the test using a pH value of about 3 can be seen in table 3.3. The colour rejection was lower than the sugar rejection; consequently, more coloured molecules could pass the membrane than sugars. The high sugar loss for the low improvement in colour makes this membrane and operating conditions unsuitable for decolourising the sugar solution. The higher rejection for sugar than colour also directs to the assumption, that there are coloured molecules which are smaller than some sugars in the solution. Therefore, the complete decolouration of the solution can not be achieved with one single membrane process, but instead needs at least one nanofiltration stage, where the sugar is kept in the retentate, whereas smaller molecules pass the membrane.

3.3.4 Higher pH and 50°C

The introduction of NaOH in order to increase the pH resulted in a spike of the conductivity. As mentioned in previous tests (Chapter 3.2.4) the conductivity increases when both NaOH and Citric acid are added to the solution. However, when NaOH is added to the feed, recycled from a test suing a lower pH, the conductivity jumps to levels almost three times the value of the previous tests. Due to these findings, further test using acid and base are conducted in chapter 3.10.5. The permeate's conductivity was with 2260 $\mu S\,cm^{-1}$ a lot lower than the retentate's conductivity, which was 3490 $\mu S\,cm^{-1}$. This results suggest, that there was a rejection of ions happening. However, this was the only test using the PES membrane with a MWCO of 5 kDa that resulted in a high colour rejection, of 63.2%, and a sugar rejection similar to previous membranes, of 12.9%.

Table 3.3: Results of the PES membrane, 5 kDa

Temperature [°C]	**pH**	**Colour Rejection [%]**	**Sugar Rejection [%]**
30	4.45	53.8	27.2
50	4.45	34.7	23.3
50	9.64	63.2	12.9
50	2.95	22.4	28.4

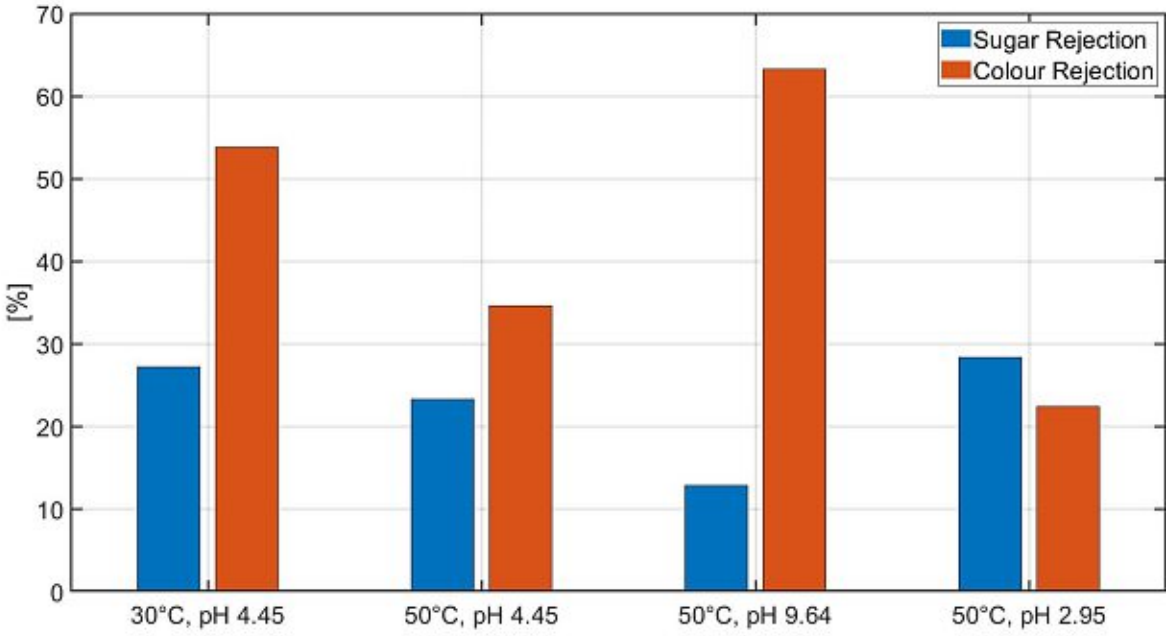


Figure 3.6: Results of the PES membrane, 5 kDa

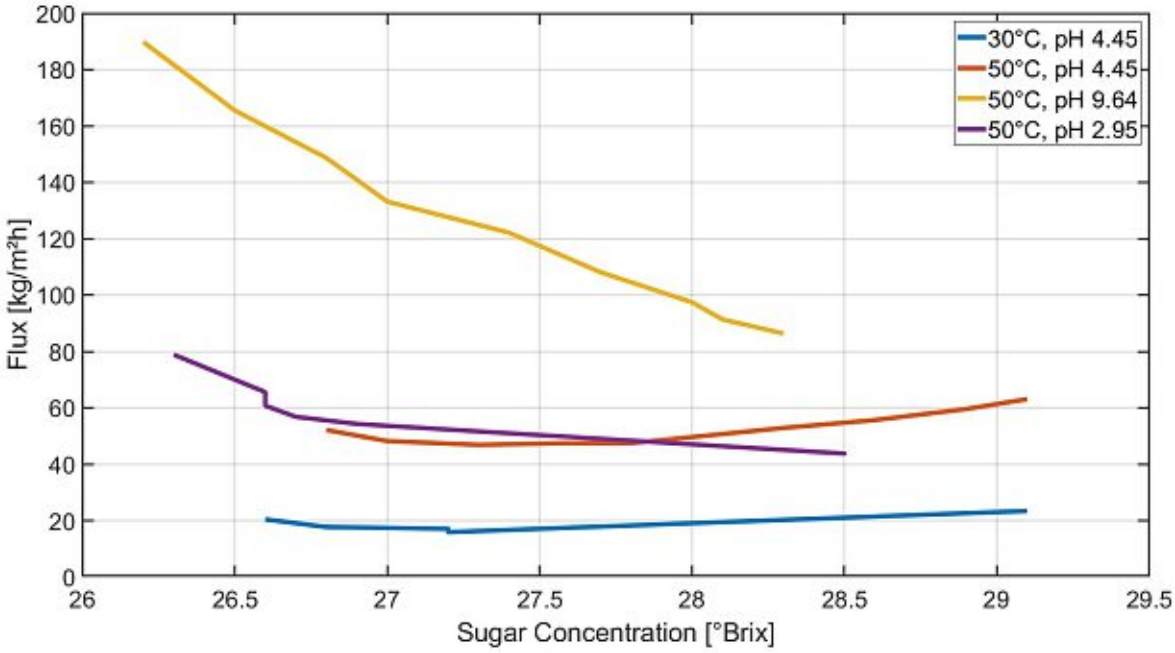


Figure 3.7: Flux of the PES membrane, 5 kDa

The test results using the polyethersulfone membrane with a MWCO of 5 kDa, as seen in table 3.3, lead to the conclusion that this kind of membrane is not suitable for the separation of sugar and colour. However, it can be used to concentrate the sugar solution after a decolouration process to reduce the product's water content.

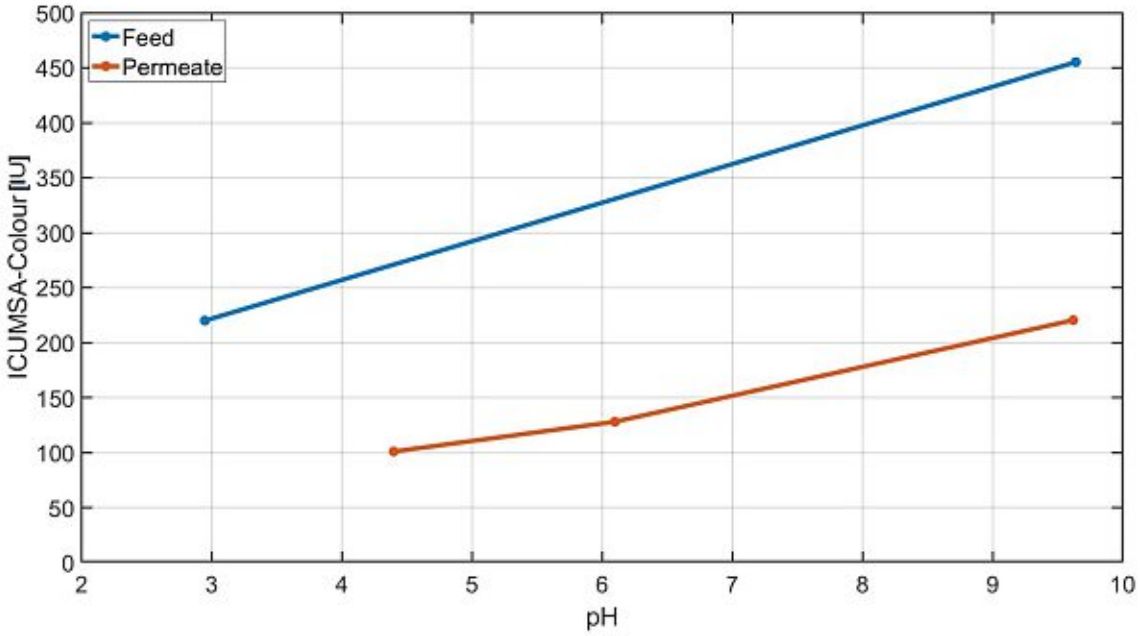


Figure 3.8: Colour over pH of feed and permeate

Like the previous test using a higher pH value, the feed colour changed drastically when the NaOH is first added. Therefore, the permeate with a high colour rejection was still heavily coloured. However, when the Citric acid was added to the permeate, the colour reduced significantly, as seen in figure 3.8. Because of this trend, the colour of the permeate after lowering the pH to 4.4 was compared to the feed before adding the NaOH, which resulted in a colour rejection of about 54.1%. Therefore, an ultrafiltration using a higher pH value and later lowering the pH again results in a relatively good colour rejection. However, the downside to this kind of process is that it is rather chemical-intensive. Additionally, there are many ions added to the product, which can also alter the properties or need to be removed again.

3.4 Polyvinylidene Flouride Membrane, 120 kDa and Negative Charge

3.4.1 Natural pH and 30°C

As can be seen in 3.4, the test using 30°C and the natural pH value of the solution resulted in a colour rejection of 22% and a sugar rejection of 3.5%. Therefore, there was a relatively little colour removal when using this membrane, compared to the previously tested membranes, however not much sugar loss.

3.4.2 Natural pH and 50°C

As expected with a higher temperature, the sugar rejection was lower; however, the colour rejection using 50 °C was higher than at 30 °C. This means that the increased temperature led to overall better results, which is a very counter-intuitive result regarding the previous tests, where the higher feed temperature generally led to a lower viscosity and made it therefore easier for molecules to pass.

3.4.3 Lower pH and 50°C

The addition of citric acid to the solution led to a lower colour rejection of about 25.4% and a higher sugar rejection of 25.4%; therefore, the effects of the lower pH are both not desirable.

3.4.4 Higher pH and 50°C

The test using a higher pH value led to very similar results as the test using natural pH. However, because the colour changed to a much darker colour when adding the sodium hydroxide, the colour rejection results cannot be considered. The final permeate received from the test still had a much darker colour than the feed before increasing the pH. Therefore, adding sodium hydroxide does not improve the membrane's performance, but introduces further impurities into the product.

Table 3.4: Results of the negatively charged PVDF membrane, 120 kDa

Temperature [°C]	**pH**	**Colour Rejection** [%]	**Sugar Rejection** [%]
30	4.49	22.0	3.6
50	4.42	35.7	2.0
50	10.15	35.5	2.4
50	3.16	25.4	2.7

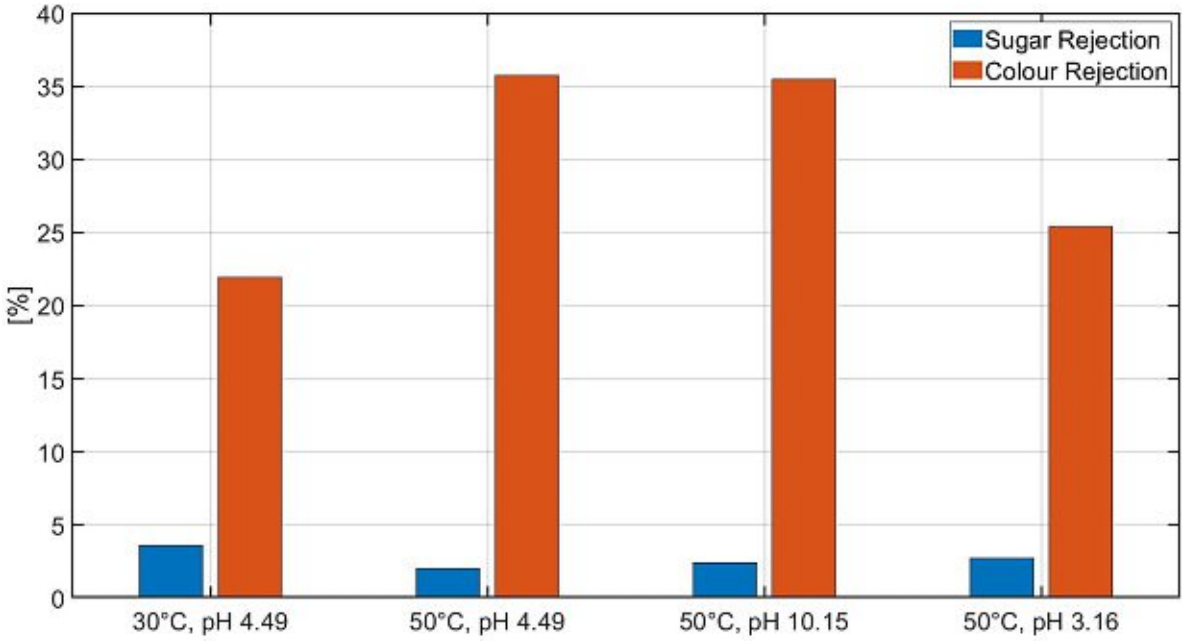


Figure 3.9: Results of the negatively charged PVDF membrane, 120 kDa

The colour rejection of this membrane was similar to previous membranes, whereas the sugar rejection was relatively low. However, with the neutrally charged PVDF membrane from chapter 3.2, a higher colour rejection with a lower sugar rejection can be produced. Therefore, the 120 kDa negative charged PVDF membrane did not yield improvements.

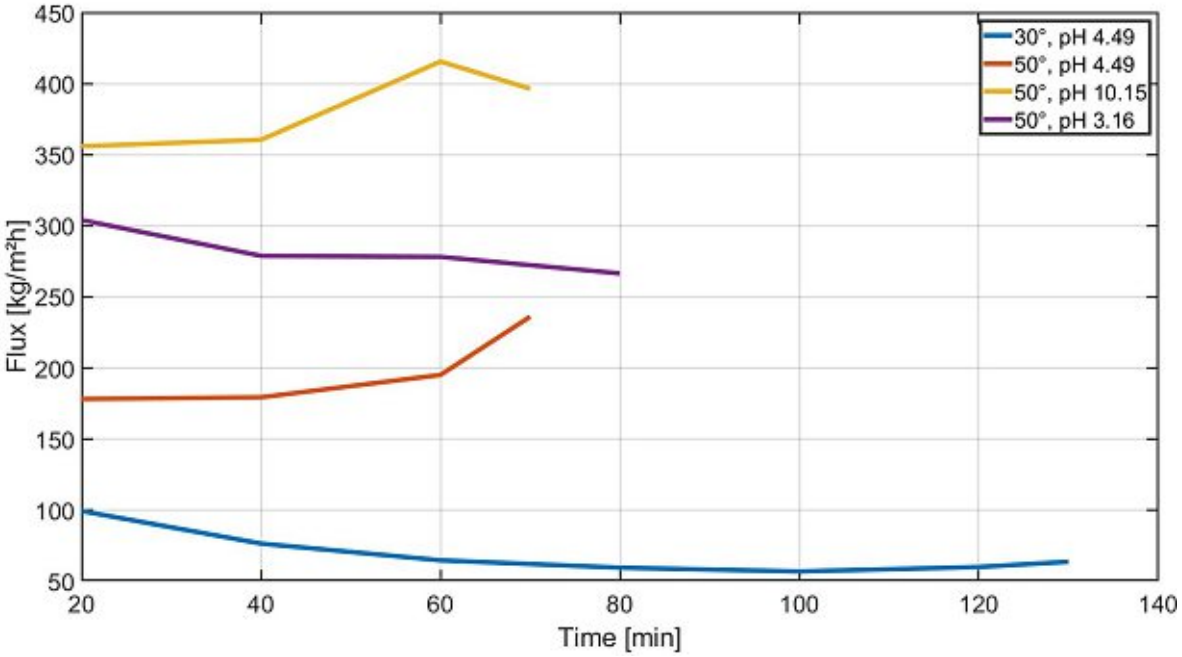


Figure 3.10: Flux of the negatively charged PVDF membrane, 120 kDa

The fluxes of the negatively charged 120 kDa PVDF membrane, displayed in figure 3.10, show a similar picture to the previous tested membrane, where the test using 30°C resulted in the lowest flux. The test using 50°C at natural pH produced a higher flux, however, still a lot lower than the test using a lowered pH. The highest flux was achieved, equally to most previous tests, at an increased pH and higher temperature.

3.5 Hydophilic Polyethersulphone Membrane, 50 kDa

3.5.1 Natural pH and 30°C

The results of the tests using this membrane are shown in table 3.5 and figure 3.11. Testing the hydrophilic PES membrane at a temperature of 30°C and a pH of 4.43, led to a colour rejection of 43.8% and a sugar rejection of about 10.8%. Even though the colour rejection was rather good, there was a lot of sugar loss due to the membrane's low MWCO.

3.5.2 Natural pH and 50°C

The sugar and the colour rejection of the test using a high temperature was lower. However, the sugar rejection of about 7% was still a lot too high to result in an economical process. The increased temperature produced the predictable result, that both colour and sugar rejection decreased.

3.5.3 Higher pH and 50°C

The increase of the pH value using NaOH led to an increased colour rejection and a decrease in sugar rejection. Both developments favour the cause; however, the problem with the darker coloured solution when adding NaOH still occurred and made the colour rejection results questionable.

3.5.4 Lower pH and 50°C

The lower pH value decreased the colour rejection and increased the sugar rejection compared to the test using the natural pH value. Therefore, the lowered pH behaved precisely opposite to the increased pH tests. Both of the developments are not desirable effects, which lead to the conclusion that the test using a lower pH is not practical for colour removal.

Table 3.5: Results of the hydrophilic PES membrane, 50 kDa

Temperature [°C]	**pH**	**Colour Rejection** [%]	**Sugar Rejection** [%]
30	4.43	43.8	10.8
50	4.42	34.0	7.0
50	9.73	39.5	4.4
50	3.08	17.7	11.0

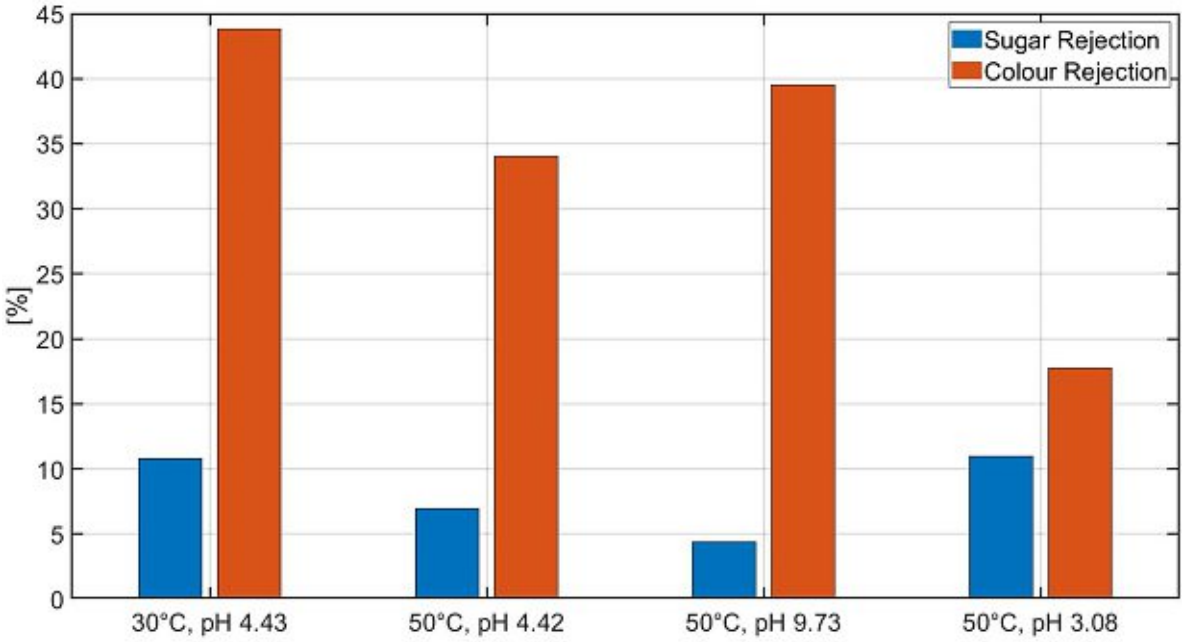


Figure 3.11: Results of the hydrophilic PES membrane, 50 kDa

To sum up, the hydrophilic PES membrane with a MWCO of 50 kDa produced a relatively high colour rejection. However, the sugar rejection was larger than desired and would result in a not economical process if implemented into an industrial process.

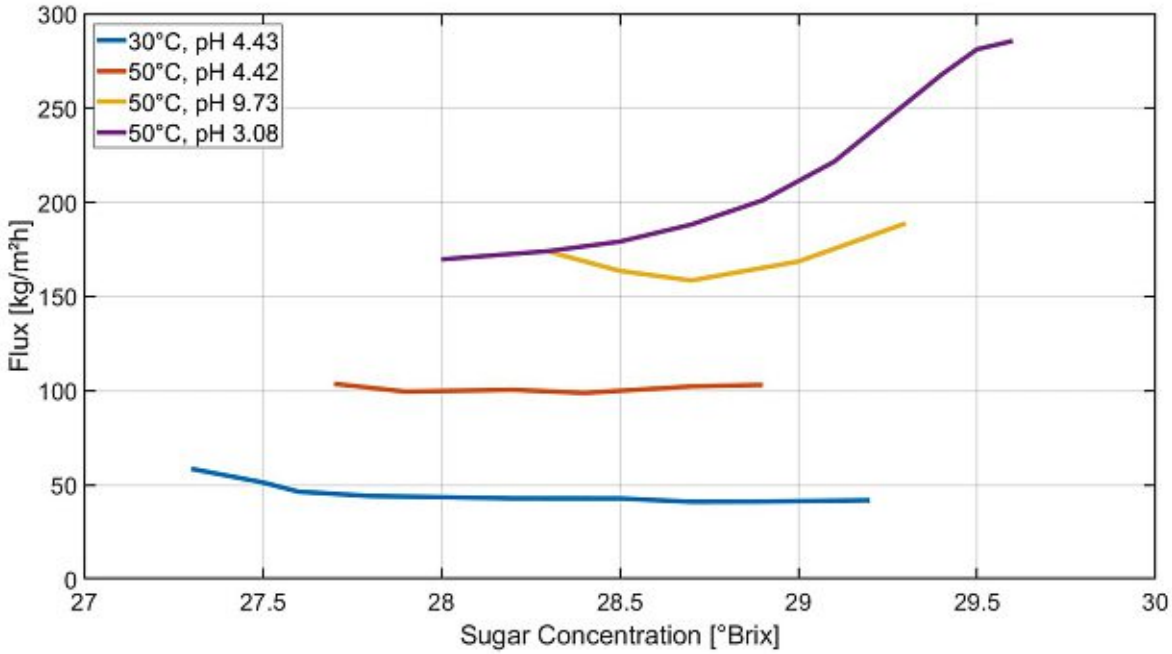


Figure 3.12: Flux of the hydrophilic PES membrane, 50 kDa

The fluxes were similar to previous tests; however, the trials using an increased and decreased pH produced somewhat similar fluxes, whereas the experiment at natural pH produced a lower value. The poorest flux was, equally to all previous tests, achieved at a lower temperature.

3.6 Hydophilic Polyethersulphone Membrane, 20 kDa

3.6.1 Natural pH and 30°C

The test results using the hydrophilic PES membrane with a MWCO of 20 kDa, a temperature of 30 °C, and the natural pH can be seen in table 3.6. The colour rejection was with about 47.3% only a slightly higher than the hydrophilic PES membrane with a MWCO of 50 kDa (Table 3.5). However, the sugar rejection was with 20.9% almost twice as high. The sugar rejection is excessive and would not be suitable for such a process. Furthermore, there can be a similar colour rejection achieved, when using the positively charged PVDF membrane of 100 kDa, with significantly lower sugar rejection (Chapter 3.1).

3.6.2 Natural pH and 50°C

As expected, there was a decrease in colour and sugar rejection when the temperature was increased to 50°C. However, the sugar rejection was with 15.5% still a lot higher than at the previous test.

3.6.3 Lower pH and 50°C

The reduction of the pH value resulted in an higher sugar rejection than at natural pH, of about 18.7%, whereas the colour rejection was lower. Similar to the previous trials, the test using a lower pH value resulted in undesirable results and is, therefore, not suitable for separating colour from sugar.

3.6.4 Higher pH and 50°C

The test using higher pH value resulted in a much lower sugar rejection of 4.6%, whereas the colour rejection was similar to the trial at 30°C. Consequently, the trial at an increased pH produced a high colour rejection, at a relatively low sugar rejection.

Table 3.6: Results of the hydrophilic PES membrane, 20 kDa

Temperature [°C]	**pH**	**Colour Rejection** [%]	**Sugar Rejection** [%]
30	4.54	47.3	20.9
50	4.52	40.0	15.5
50	10.67	47.3	4.6
50	3.01	38.3	18.7

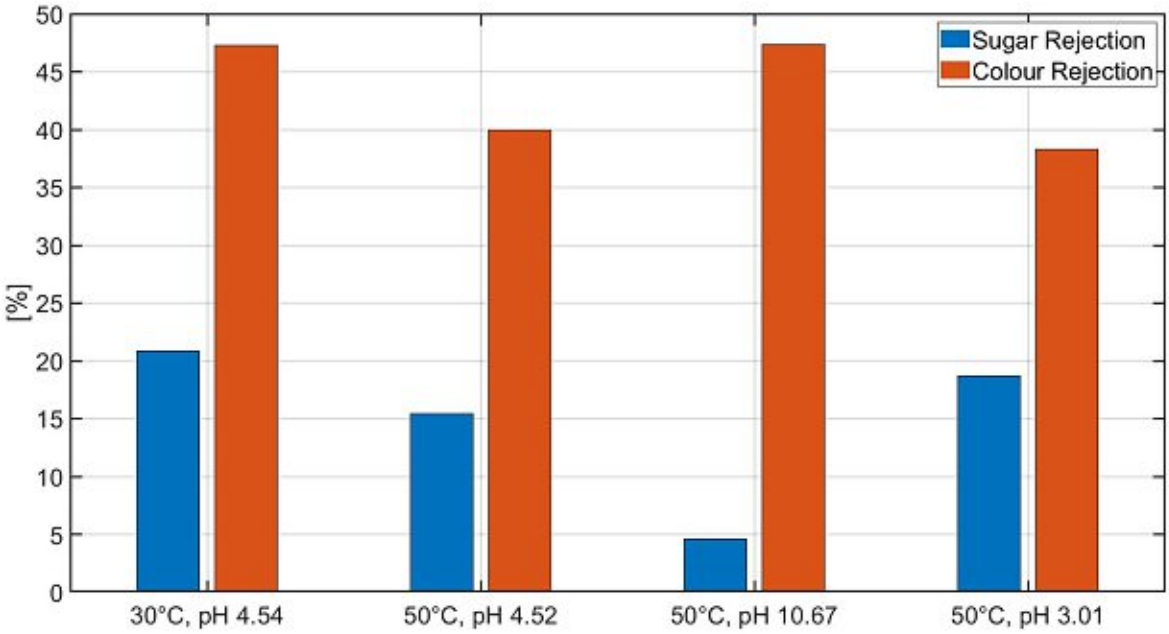


Figure 3.13: Results of the hydrophilic PES membrane, 20 kDa

To summarise, the hydrophilic PES membrane with a MWCO of 20 kDa produced permeate with only slightly better colour compared to the one with 50 kDa. However, there was a lot more sugar loss in most tests. Nonetheless, the trial at 50°C and an increased pH value resulted in a very decent colour rejection, while the sugar rejection decreased.

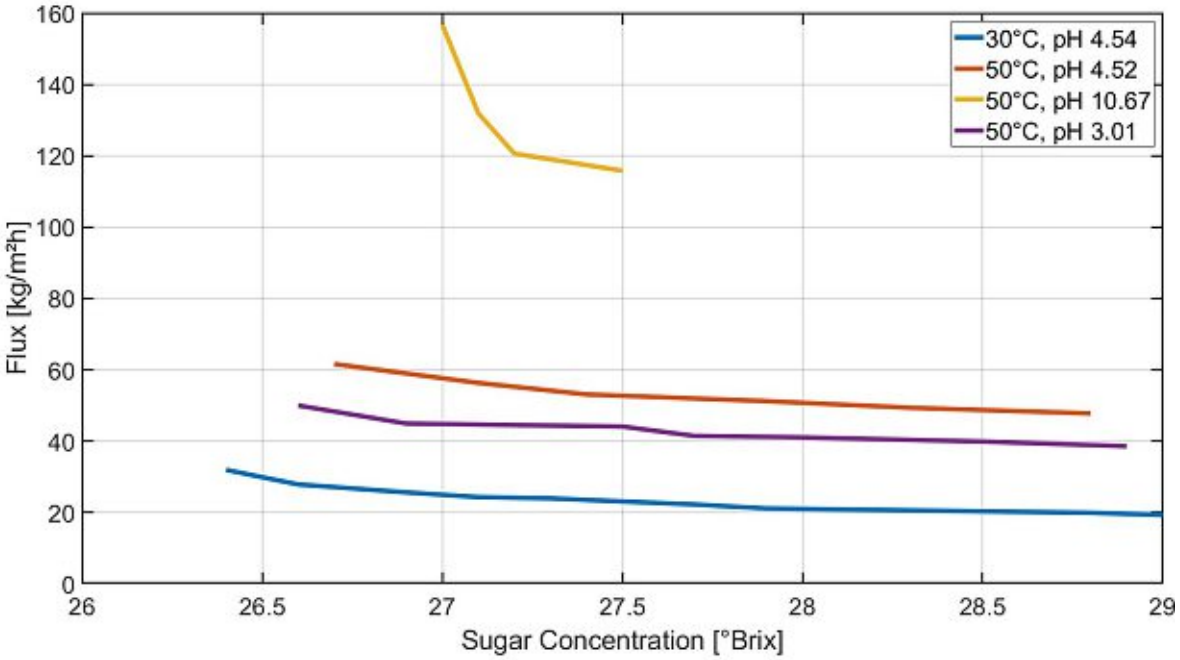


Figure 3.14: Flux of the hydrophilic PES membrane, 20 kDa

The fluxes were similar to previous trials, but at an increased pH it was a lot higher than the other ones. Generally, the this membrane's fluxes were relatively small, because the MWCO was lower than previous ones.

3.7 Regenerated Cellulose Membrane, 10 kDa

3.7.1 Natural pH and 30°C

In table 3.7 and figure 3.15, the results of this membrane are shown. At a temperature of 30°C, there was a colour rejection of about 34.7% at a sugar rejection of 9.2%. The colour rejection was decent; however, the sugar rejection was too high in order to make a economical process. Additionally, the neutrally charged PVDF membrane with a MWCO of 100 kDa produced at 30°C a higher colour rejection with less sugar rejection. Therefore, this membrane is inferior to previous tested ones when it comes to the removal of colour from sugar syrup.

3.7.2 Natural pH and 50°C

Like the other membranes, the colour and sugar rejection declined with increasing temperature. Nonetheless, the sugar rejection was still relatively high with 3.8% and a colour rejection of about 22.7% was also not in the desired level.

3.7.3 Lower pH and 50°C

When citric acid was added to lower the pH, the membrane's performance did not improve, but the colour rejection declined further to only 20.4%, and the sugar rejection rose to 5.2%. Both of the developments are not favourable for this purpose.

3.7.4 Higher pH and 50°C

At an increased pH value, the colour rejection increased slightly, but was with 25% still rather low. Moreover, the sugar rejection was with 4.9% lower than at a low pH, but higher than the trial results using 50°C and natural pH.

Table 3.7: Results of the regenerated cellulose membrane, 10 kDa

Temperature [°C]	**pH**	**Colour Rejection** [%]	**Sugar Rejection** [%]
30	5.59	34.7	9.2
50	5.44	22.7	3.8
50	9.5	25.0	4.9
50	3.24	20.4	5.2

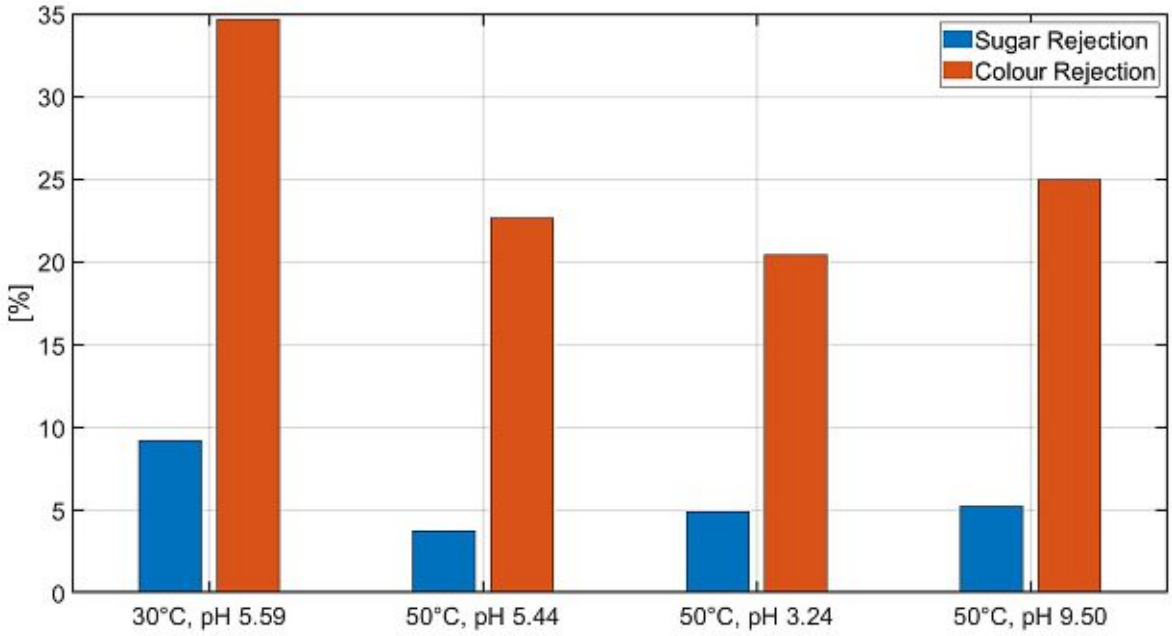


Figure 3.15: Results of the regenerated cellulose membrane, 10 kDa

The regenerated cellulose membrane produced, despite the small MWCO, a relatively moderate colour rejection, had ,however, a quite high sugar rejection. Therefore, it is not particularly suitable in order to remove coloured molecules from sugar solutions.

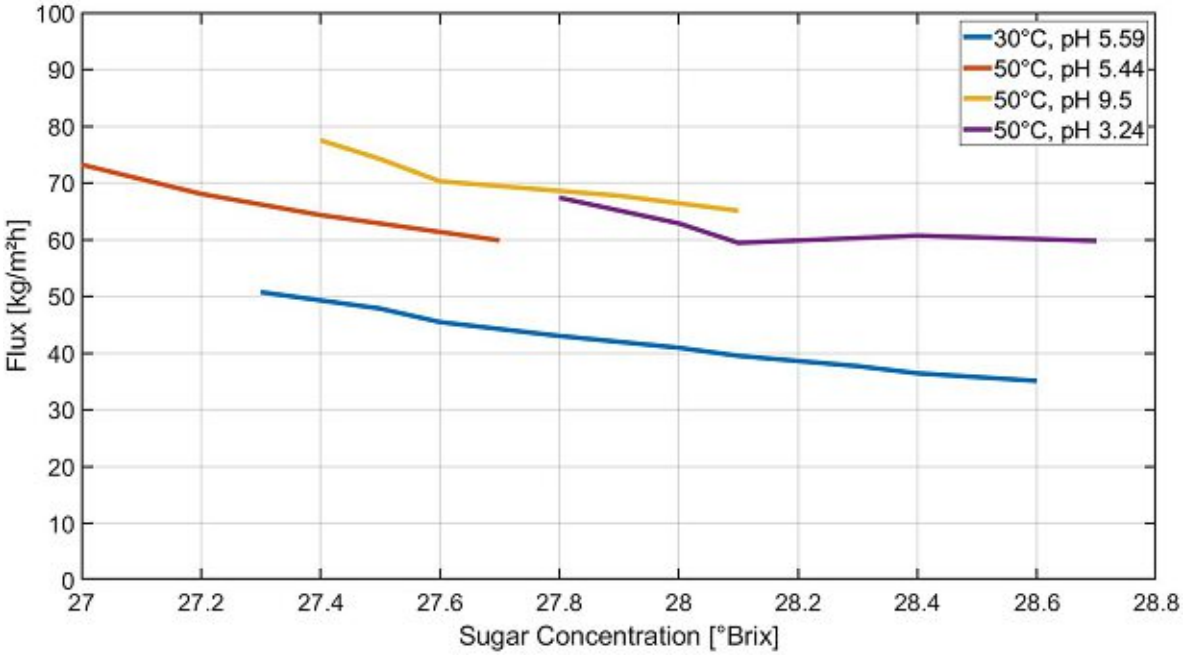


Figure 3.16: Flux of the regenerated cellulose membrane, 10 kDa

As shown in figure 3.16, the fluxes present a somewhat familiar picture, as that the highest flux was achieved at an increased pH and the lowest one at a lowered temperature. Like the other membranes, the trial's fluxes using 50°C and natural and lowered pH produced almost equal fluxes.

3.8 Comparison of the Tested Membranes

In order to find the most suitable membrane for the task, the results have to be compared.

3.8.1 Comparison of the Colour and Sugar Rejection

For comparing the different membranes, the test at the natural pH and a temperature of 50°C is considered.

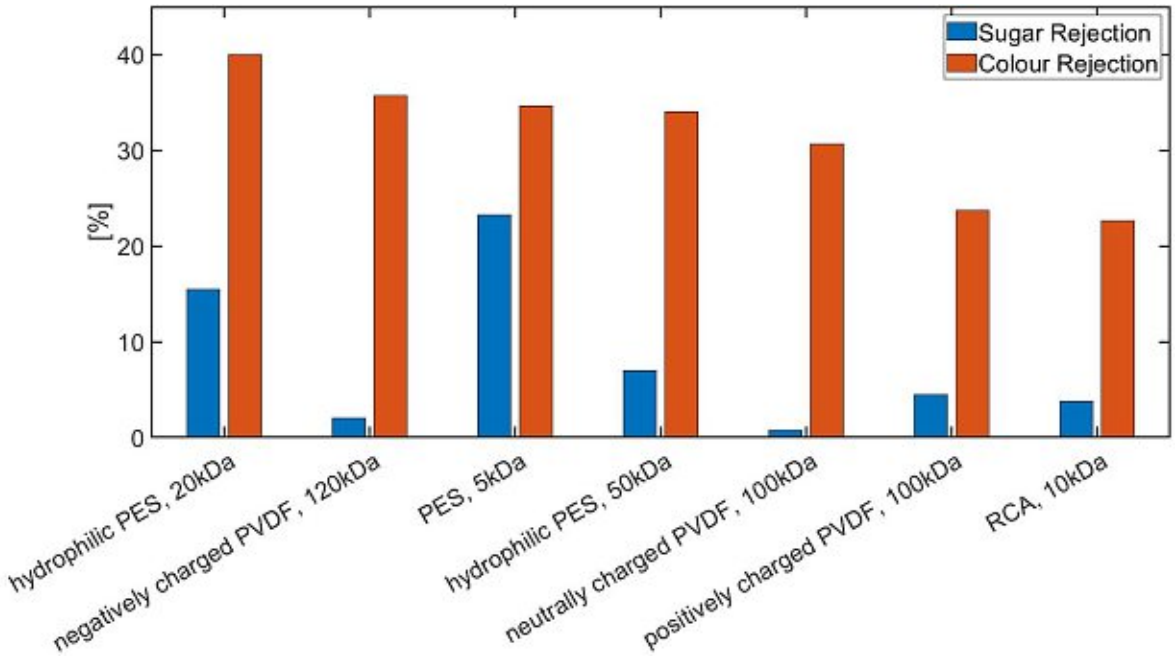


Figure 3.17: Comparison of the results of different membranes at 50°C and natural pH

In figure 3.17 it is observable that the colour rejection of the membranes range from 22% to 40%. However, there is a considerable connection between the colour rejection and the sugar rejection. If only the colour rejection is considered the best suitable membrane would be the hydrophilic PES membrane with a MWCO of 20 kDa. Nevertheless, this membrane also produced a sugar rejection of about 15%, which would lead to immense sugar losses if applied at an industrial scale. On the other hand, the negatively charged PVDF membrane (120 kDa) resulted in a sugar rejection of only about 2%, while still rejecting about 35% of the colour.

3.8.2 Comparison of the Flux

Figure 3.18 shows the fluxes of the tested membranes at 50°C and the natural pH. In order to make the results comparable, they were compensated. Therefore, the water permeability before and after the test were used to make a linear regression, which served as the compensation regression. Additionally, all the values were compensated to a concentration of 27°Brix.

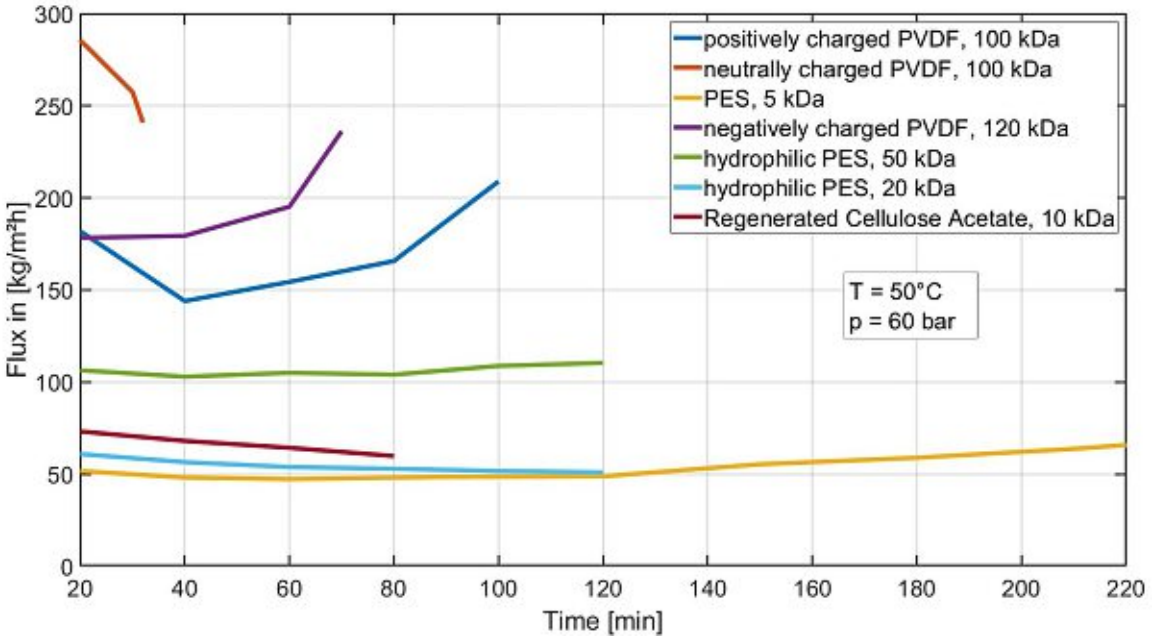


Figure 3.18: Comparison of the fluxes of different membranes at 50°C and natural pH

As seen in figure 3.18, the neutrally charged membrane with a MWCO of 100 kDa, produced the highest flux. The smallest membrane, consisting of polyethersulfone, resulted in the lowest flux. This leads to the assumption that the MWCO of the membranes have a significant influence on the flux. However, the composition is also a factor that changes the flux. Even though the regenerated cellulose acetate membrane has a MWCO of 10 kDa, it produced a higher flux than the 20 kDa hydrophilic PES membrane. Additionally, the charge affects the flux because the positively charged PVDF membrane had a much lower flux than the neutrally charged one, although they both have the same MWCO of 100 kDa.

3.9 Determination of the Most Suitable Membrane and Conditions

In order to select the membrane most suitable for this application, several data analysis methods are used. At first, tests resulting in a high sugar rejection (> 10%) or a low colour rejection (< 30%) are terminated. Therefore, several tests, not fulfilling this requirement, were dismissed. Due to all the Polyethersulfone membrane tests with a MWCO of 5 kDa having a sugar rejection of more than 10% (table 3.3) this membrane is not suitable for this purpose. From the other membranes, at least some tests satisfied both requirements.

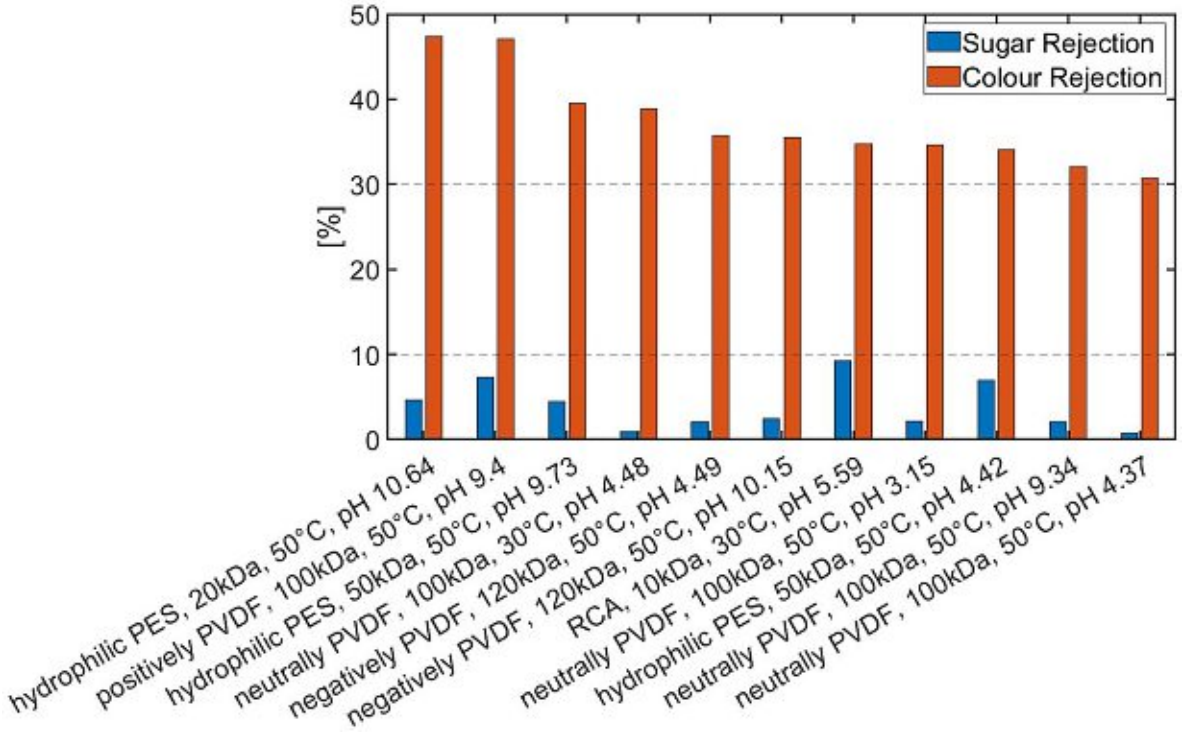


Figure 3.19: Results with >30% colour and <10% sugar rejection

When applying these requirements, the best results for removing colour from the sugar solution is received, when using the hydrophilic PES membrane of 20 kDa at a temperature of 50°C and a pH of 10.64. Additionally, it appears that higher pH values are more suitable for colour removal. The three tests resulting in the highest colour rejection use an increased pH value, whereas only one test using a decreased pH fulfils the requirements. However, there is no apparent relation between the membrane's MWCO and the colour or sugar rejection. The test using the hydrophilic PES membrane with a MWCO of 20 kDa at 50°C and an increased pH produced a colour rejection of about 47% with a sugar rejection of about 4.6%. In contrast, the same test using the positively charged PVDF membrane with a MWCO of 100 kDa produced a very similar colour rejection, but with 7.3% a much higher sugar rejection even though the MWCO is five times larger, which leads to the assumption that the composition of the membrane has a significant influence on the rejection. However, when comparing membranes of the same composition, the pore size also affects the results. When comparing the test using an increased pH and a temperature of 50°C of the hydrophilic PES membrane with 20 kDa and 50 kDa, the colour and sugar rejection decrease with increasing MWCO.

3.9.1 Determination of the Most Suitable Conditions for each Membrane

As shown in figure 3.20 for the positively charged PVDF membrane with 100 kDa, the colour and sugar rejection decreased with increasing temperature. However, the colour rejection declined further with increasing temperature. This suggests that the gap between the sugar and colour rejection is larger at lower temperatures. However, due to the increased sugar loss as well as the lower flux achieved at this conditions, it is hard to determine ideal operating conditions.

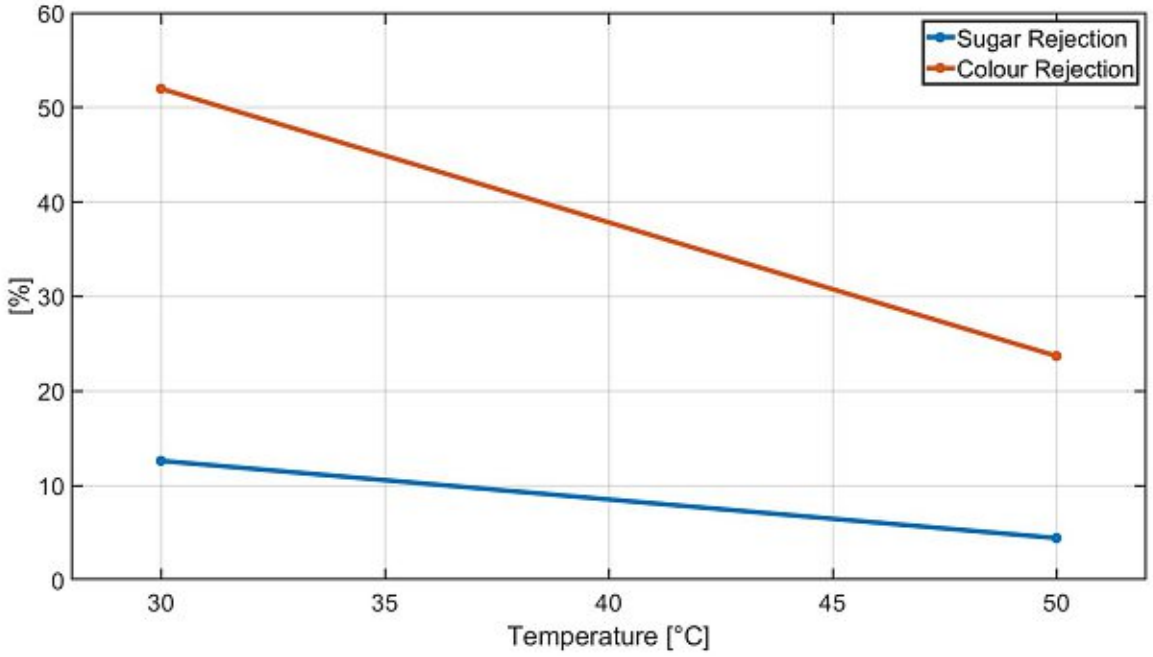


Figure 3.20: Temperature dependency of the colour and sugar rejection of the positively charged PVDF membrane, 100 kDa

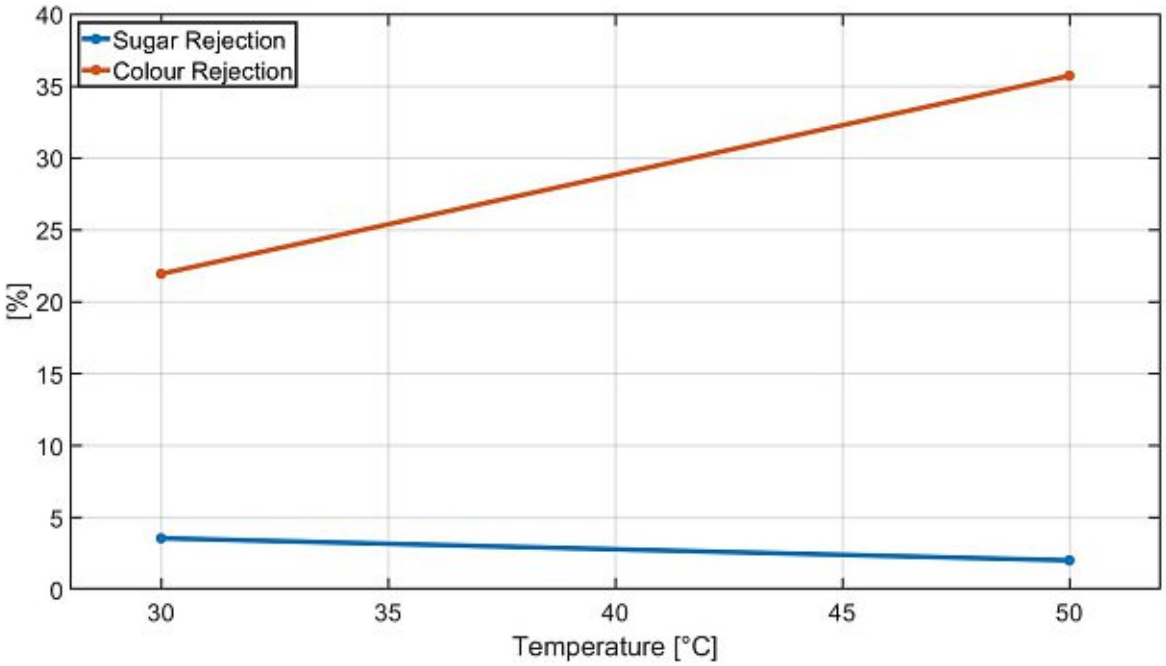


Figure 3.21: Temperature dependency of the colour and sugar rejection of the negatively charged PVDF membrane, 120 kDa

For other membranes, the behaviour was similar, except for the negatively charged PVDF membrane with 120 kDa (figure 3.21), which produced a larger colour rejection and a lower sugar rejection at higher temperatures. Therefore, the overall performance improved when increasing the temperature.

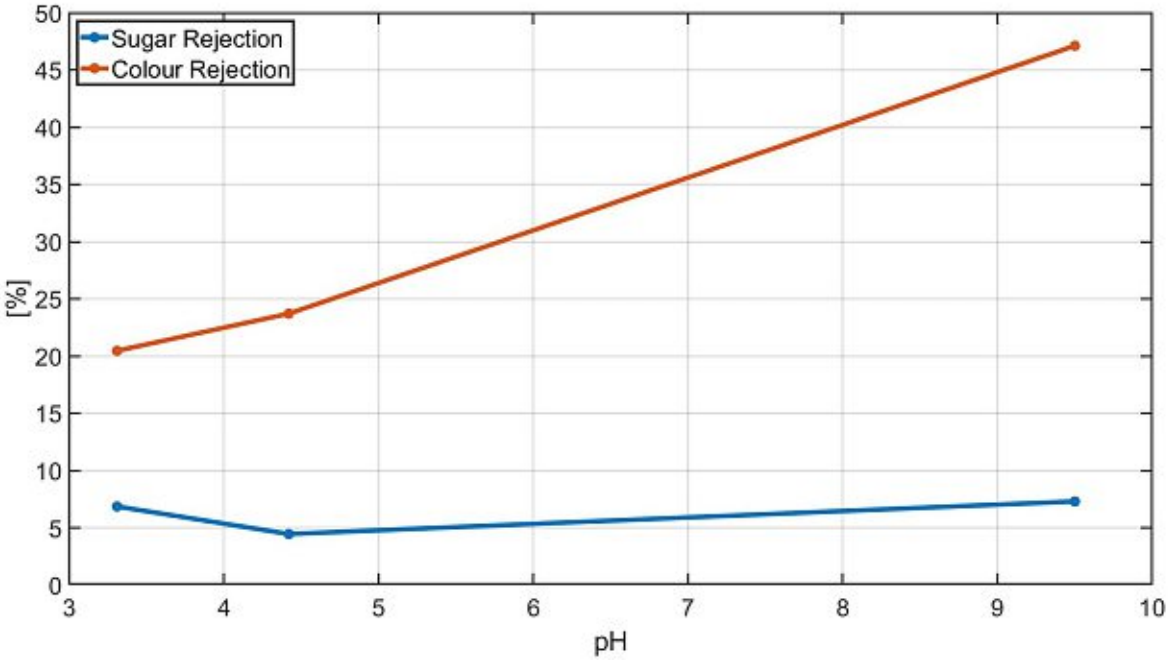


Figure 3.22: pH dependency of the colour and sugar rejection of the positively charged PVDF membrane, 100 kDa

The pH value additionally influences the results, as shown in figure 3.22. The higher the pH value, the higher the colour rejection, whereas the sugar rejection stayed almost constant. This leads to the assumption that a higher pH improves the sugar and colour separation in the sugar solution.

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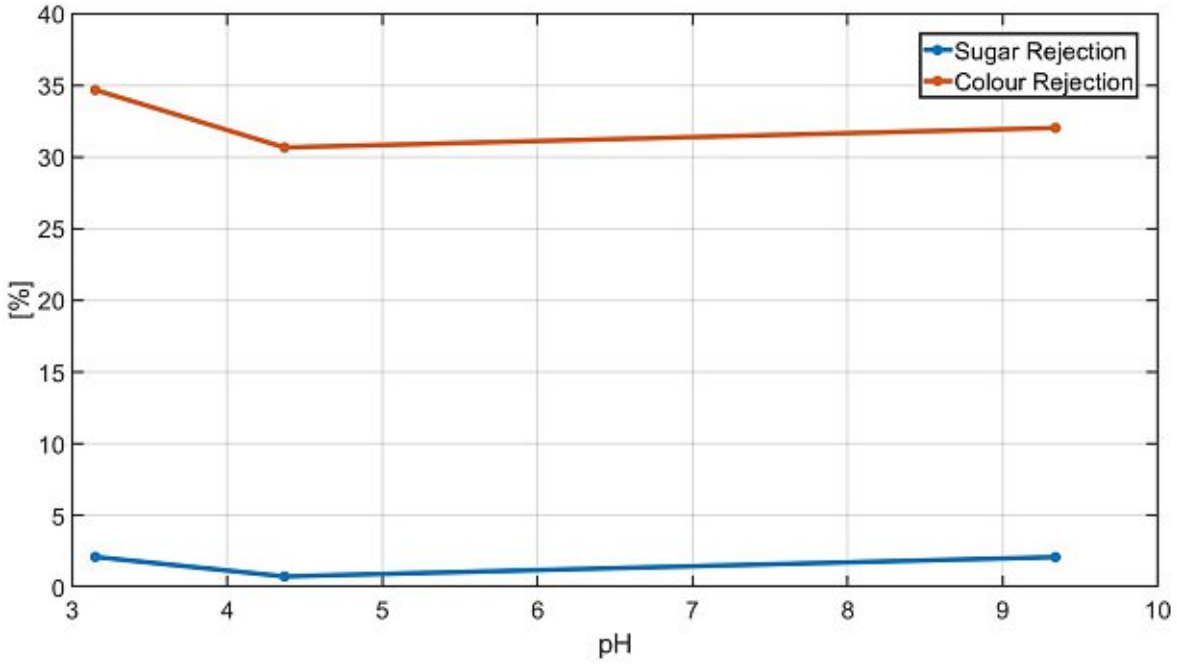

Figure 3.23: pH dependency of the colour and sugar rejection of the neutrally charged PVDF membrane, 100 kDa

The remaining membranes behaved similar to the positively charged PVDF membrane, shown in figure 3.20, however, as it can be seen in figure 3.23, the neutrally charged PVDF membrane with a MWCO of 100 kDa produced an almost constant colour rejection at different pH values. Therefore, there is no substantial improvement when changing the pH in either direction.

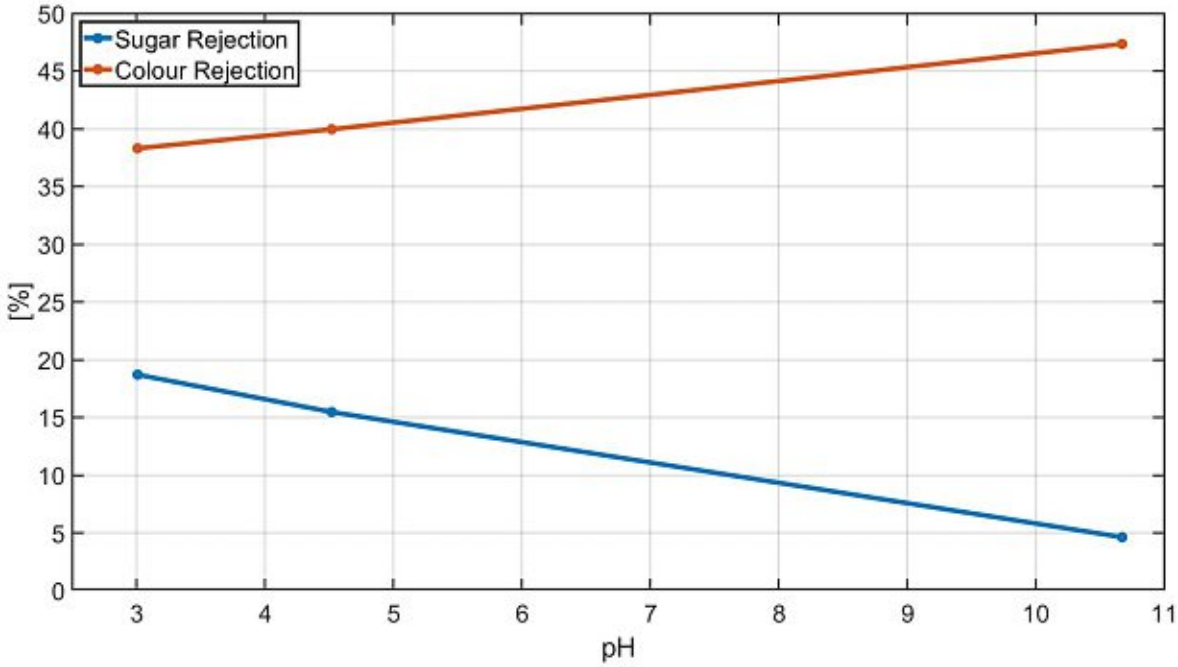

Figure 3.24: pH dependency of the colour and sugar rejection of the hydrophilic PES membrane, 20 kDa

Additionally, the PES membranes achieved a slight reduction in sugar rejection at higher temperatures. In figure 3.24, the colour and sugar rejection of the hydrophilic PES membrane with a MWCO of 20 kDa are shown. Therefore, the membrane's performance improved at an increased pH because the colour rejection increases and the sugar rejection decreases, both desirable effects.

For the determination of the ideal operating conditions for each membrane, the maximal difference the colour and sugar rejection has to be achieved. In table 3.8, the conditions, which produce the highest Δ-Rejection for each tested membrane, are shown.

Table 3.8: Highest difference between colour and sugar rejection for each membrane

Membrane Material	**MWCO [kDa]**	**Temperature [°C]**	**pH**	**Colour Rejection [%]**	**Sugar Rejection [%]**	**Δ− Rejection [%]**
PES	5	50	9.64	63.2	12.9	**50.6**
hydrophilic PES	20	50	10.67	47.3	4.6	**42.7**
positively charged PVDF	100	50	9.50	47.1	7.3	**39.8**
neutrally charged PVDF	100	30	4.48	38.9	0.94	**37.9**
hydrophilic PES	50	50	9.73	39.5	4.4	**35.1**
negatively charged PVDF	120	50	4.49	35.7	2.0	**33.7**
Regenerated Cellulose Acetate	10	30	5.59	34.7	9.2	**25.5**

When choosing these criteria to determine the most suitable membrane and conditions, the PES membrane with a MWCO of 5 kDa provided the best results. However, the membranes, which had the most considerable Δ-Rejection also had relatively high sugar rejections. The 5 kDa PES membrane produced a sugar rejection of 12.9%, which would lead to significant sugar losses if applied in an industrial scale. Therefore, it is better to define a maximum acceptable sugar rejection and find the membrane and conditions, which produced the best colour rejection, while not exceeding this predetermined value.

3.9.2 Comparing the Ideal Conditions of Each Membrane at a Maximal Acceptable Sugar Rejection

The criteria for the ideal membrane and conditions were further defined by a maximal accepted sugar rejection. When such a maximum is chosen the best colour rejection can be calculated, while not exceeding this sugar rejection. In figure 3.25, the sugar and colour rejection are shown at different temperatures and pH values. Additionally, there was a plane added at a rejection of 7%. Furthermore, there were linear functions constructed, connecting the tested data points. Therefore it is also possible that the ideal conditions consist of a temperature, pH combination, which has not been tested.

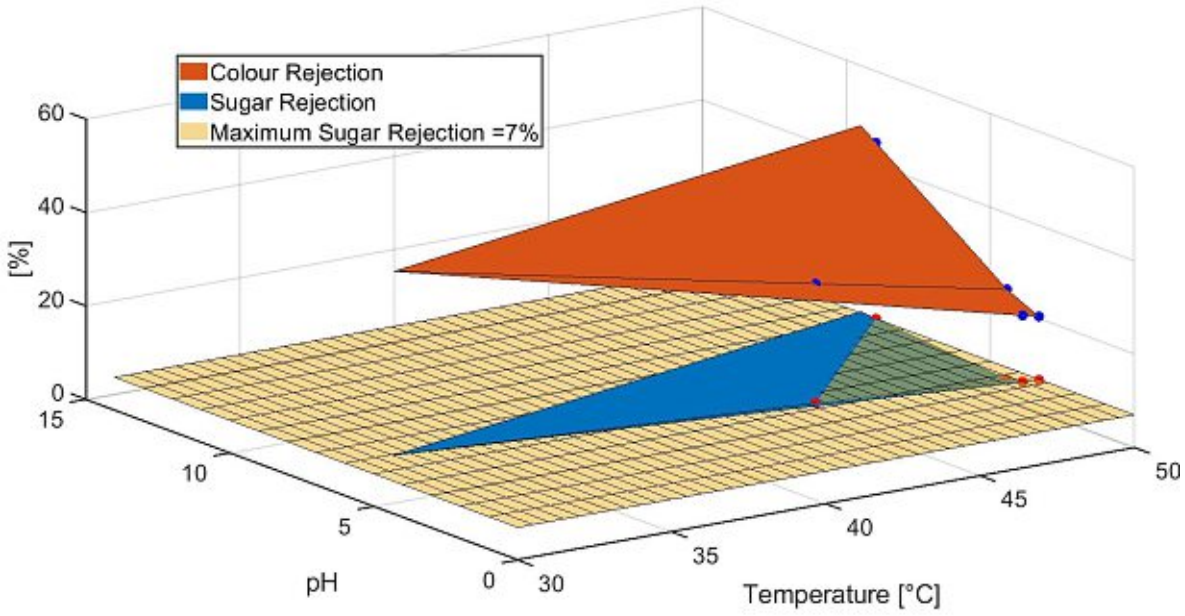

Figure 3.25: Colour and sugar rejection for a maximal sugar rejection of 7% of the positively charged PVDF membrane, 100 kDa

For the determination of the ideal operating conditions, only the values with a sugar rejection of lower than 7% were considered, which is shown by the surface below the maximum sugar rejection plane. Due to the surface structure, only few interesting points were considered, which are marked red. They were than transposed onto the colour rejection surface and marked blue. For this membrane, the considerable points are shown in table 3.9.

Table 3.9: Points of interest

Temperature [°C]	pH	Sugar Rejection [%]	Colour Rejection [%]
50.00	4.42	23.7	4.5
50.00	3.31	20.5	6.9
50.00	8.97	44.7	7.0
49.49	3.33	21.3	7.0
43.75	4.38	32.5	7.0

When considering a maximal acceptable sugar rejection of 7% the optimal conditions for this membrane are at a temperature of 50 °C and pH 8.97, which result in a colour rejection of about 44.67%.

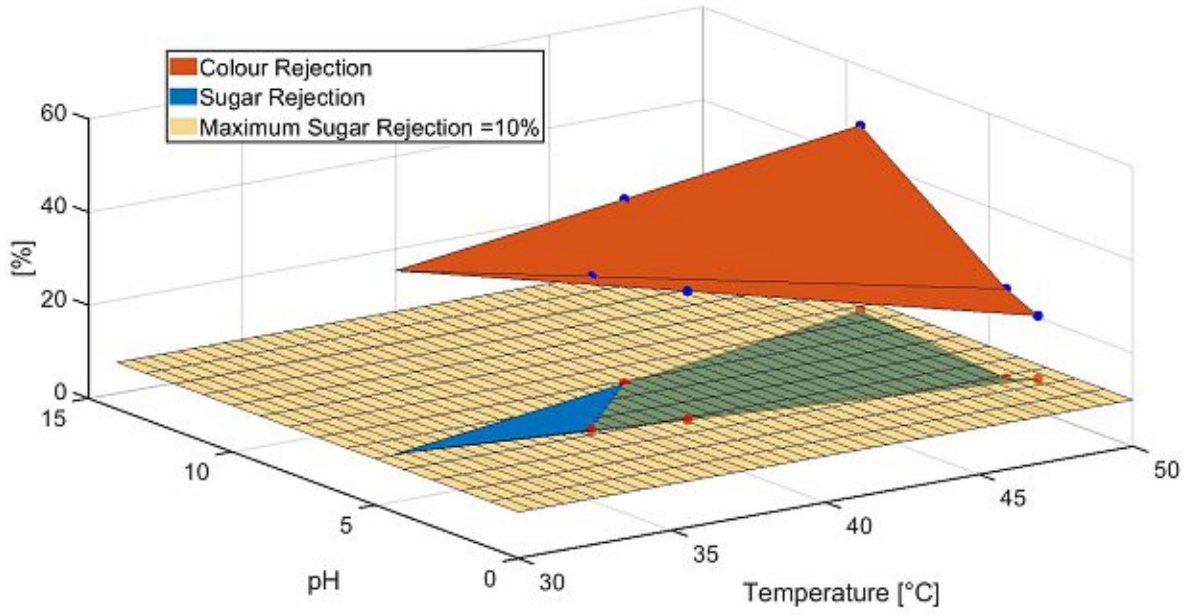

Figure 3.26: Colour and sugar rejection for a maximal sugar rejection of 10% of the positively charged PVDF membrane, 100 kDa

When the maximal acceptable sugar rejection is increased to 10% the optimal conditions change. In figure 3.26, the intersection points at a sugar rejection of 10% are shown.

Table 3.10: Most suitable membranes and conditions for different maximal sugar rejection

Maximum Sugar Rejection [%]	Membrane Material	MWCO [kDa]	Temperature [°C]	pH	Colour Rejection [%]	Sugar Rejection [%]
15	PES	5	50	9.64	63.2	12.9
10	positively charged PVDF	100	39.84	6.85	49.6	10.0
5	hydrophilic PES	20	50	10.67	47.3	4.6
1	neutrally charged PVDF	100	30	4.48	38.9	0.94

That led to optimal conditions at 39.84°C and pH 6.85, which produces a colour rejection of 49.59%, at a sugar rejection of 10%.
The best-suited conditions for all the membranes were tested using different maximal sugar rejections. The most suitable membrane and conditions for each maximal sugar rejection are shown in table 3.10.

Depending on the maximal acceptable sugar rejection, different membranes produce the best colour rejection. The higher the accepted sugar rejection, the smaller the membrane. However, the positively charged PVDF membrane produced a relatively high colour and sugar rejection. Thus, at a maximal acceptable sugar rejection of 10%, this is the most suitable membrane. For lower maximal sugar rejection, more sweeping membranes have to be used to let most of the sugar pass. However, there is a lot less colour rejection. Generally, there is a trend to higher pH values, because there is less sugar rejection, at a relatively high colour rejection. For a maximal sugar rejection of 1%, only the neutrally charged PVDF membrane is relevant, because all the other membranes produced a higher sugar rejection.

3.10 Additional Tests

3.10.1 Two-Stage Process with Neutrally and Positively Charged PVDF Membrane

The second stage alone had a colour rejection of 23.8% and a sugar rejection of 4.29%. This resulted in an overall colour rejection of 50.28% for both membrane stages and an overall sugar rejection of 5.11%. Therefore, as shown in figure 3.27, the colour rejection of this two-staged process was, similar to the single stage process of the 30°C with the positively charged polyvinylidene flouride membrane, however, the sugar rejection was much lower when using the two-staged process.

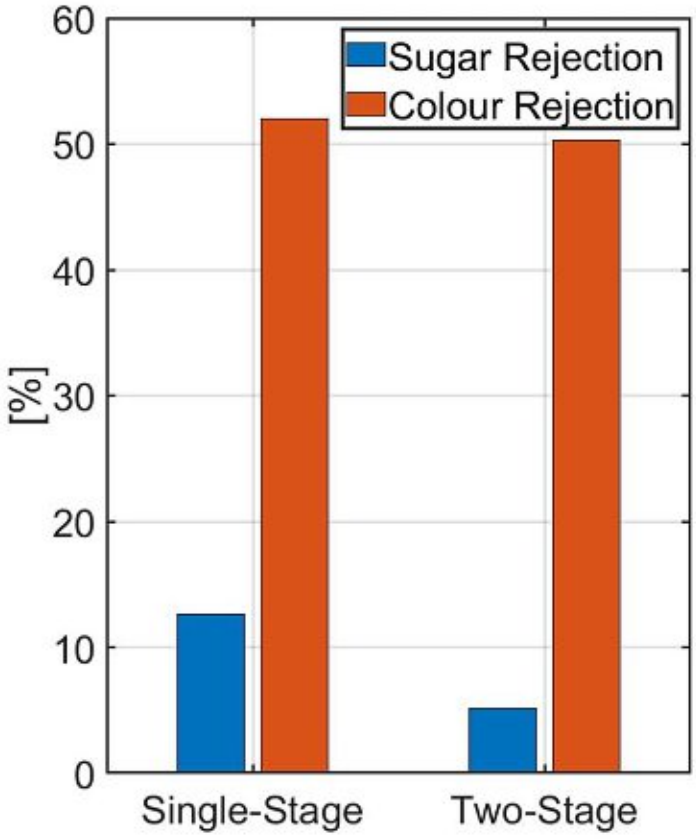

Figure 3.27: Comparison of a single-stage and a two-stage process

Table 3.11: Comparison of single-stage and two-stage process

	Single-Stage	Two-Stage
	M-180	M-180 and M-183
	30°C	30°C
Colour Rejection [%]	52.0	50.3
Sugar Rejection [%]	12.6	5.1

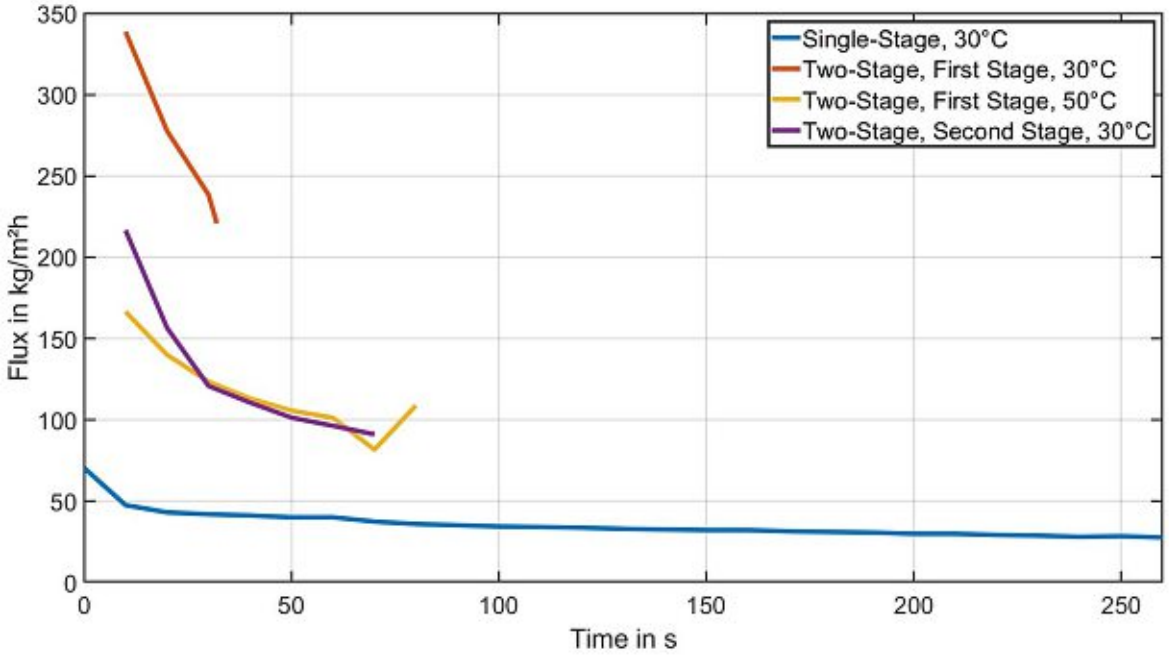

Figure 3.28: Comparison of the flux of a single-stage and a two-stage process

Additionally to the lower sugar rejection, the two-stage process' flux was a lot higher, resulting in a lower surface area. In figure 3.28 it is shown, that the first stage using the neutrally charged PVDF membrane with a MWCO of 100 kDa had a relatively high flux. Since the first stage separated larger molecules, the positively charged PVDF with a MWCO of 100 kDa did not clog as fast and produced a higher flux than the single-stage process. Because all the tests, except for the neutrally charged PVDF membrane at 50°C, used new membranes, the flux results shown in figure 3.28 were not compensated.

3.10.2 Effect of the Ultrafiltration on the Concentration of Hydroxymethylfurfural (HMF)

The high-performance liquid chromatography results show that there is very little difference in the concentration of hydroxymethylfurfural in the retentate and permeate. The molecular mass of hydroxymethylfurfural is with 126 $g\,mol^{-1}$ lower than that of sucrose with 342 $g\,mol^{-1}$. Therefore, it is expected, that there is not much rejection happening in the process. In some of the tests made, as shown in table 3.12, there was even an increase of HMF in the permeate. Suggesting that it was easier for the HMF to pass the membrane than for other molecules. Additionally, the results suggest an influence of the pH value on the concentration of HMF. The neutrally charged PVDF membrane tests with a MWCO of 100 kDa show a decrease in concentration at a lower pH. As shown in table 3.12, the concentration of HMF in the feed at a pH of 4.37 was 2.400 $mg\,L^{-1}$ then decreased to 1.592 $mg\,L^{-1}$ at a pH of 3.15. Further HPLC tests show that at higher pH values, the HMF concentration decreases as well. However, because the results fall below the equipment's detectability, they are not shown in table 3.12.

Table 3.12: Results of the HPLC analysis

Membrane Material	**MWCO [kDa]**	**Temperature [°C]**	**pH**	**HMF Feed** [$mg\,L^{-1}$]	**HMF Permeate** [$mg\,L^{-1}$]	**HMF Retentate** [$mg\,L^{-1}$]
positively charged PVDF	100	30	4.29	2.403	2.379	2.341
		50	4.42	2.228	2.332	2.342
neutrally charged PVDF	100	30	4.48	2.367	2.361	2.314
		50	4.37	2.400	2.383	2.319
		50	3.15	1.592	1.550	1.598
negatively charged PVDF	120	50	4.49	2.337	2.474	2.190
hydrophilic PES	50	50	4.42	2.357	2.424	2.417
Regerenated Cellulose Acetate	10	50	4.74	1.749	1.678	3.757

The only membrane producing a decent rejection for HMF was the regenerated cellulose membrane with a MWCO of 10 kDa. The rejection of HMF calculates to 55.3%, whereas the sugar rejection was with 6.47% a lot lower, which leads to the assumption that the regenerated cellulose acetate is selective for HMF, rather than for sugar.
These results additionally lead to the conclusion that HMF is not single-handedly responsible for the solution's colour, because some of the membranes produce a relatively high colour rejection, however almost no rejection for HMF.

3.10.3 Influence of the Ultrafiltration on the Sugar Composition

Since all tested membranes bring some sugar rejection with them, the sugar composition change was investigated. Therefore, a size exclusion chromatography was provided by Agrana. For figures 3.29, 3.30, 3.31, and 3.32 the three lines represent the feed, permeate and retentate. The peak at 180 Da represents the amount of monosaccharides, such as glucose or fructose, at about 342 Da disaccharides, such as sucrose or maltose, at about 504 Da trisaccharides, such as maltotriose, and the peak at about 666 Da tetrasaccharide, such as maltotetraose.

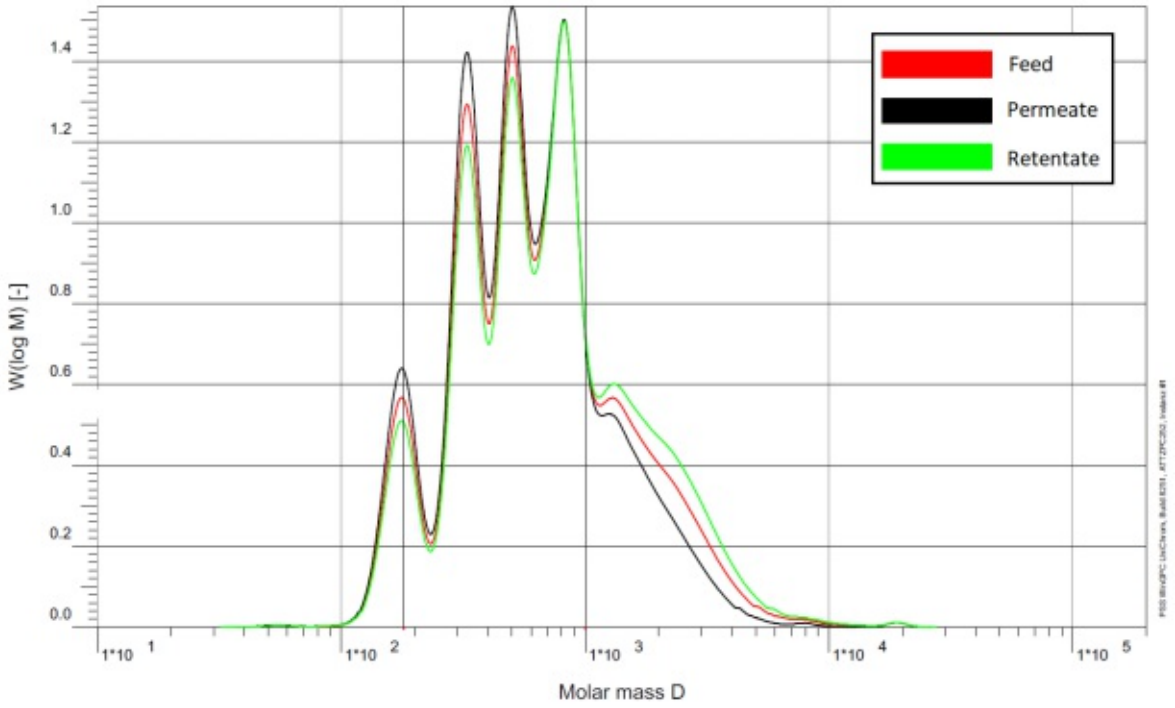

Figure 3.29: SEC results for the positively charged PVDF membrane, 100 kDa at 30°C (chapter 3.1.1), provided by Agrana

The figure 3.29 shows the composition of the feed, permeate and retentate of the tests using the positively charged, 100 kDa PVDF membrane at a temperature of 30°C and a pH of 4.29 (Chapter 3.1.1). There is a difference in molar mass distribution between the permeate and retentate. There is a higher concentration of mono-, di-, and trisaccharides, however, a smaller number of larger molecules in the permeate, whereas the retentate contains larger quantities of larger molecules.

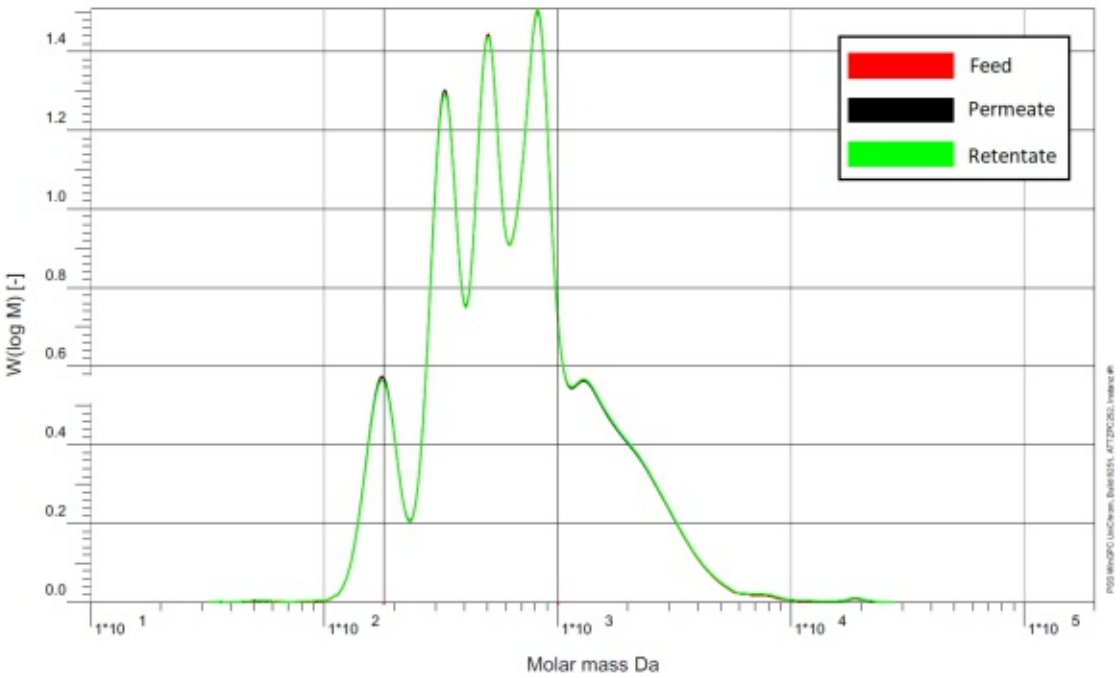

Figure 3.30: SEC results for the neutrally charged PVDF membrane, 100 kDa at 30°C (chapter 3.2.1), provided by Agrana

For the neutrally charged 100 kDa PVDF membrane at 30°C and a pH of 4.48, the compositions, as shown in figure 3.30, look differently. For this membrane, there was almost no change in the composition between the retentate and the permeate. This is also visible in the sugar rejection, which is with 0.94% much smaller than the 12.62%, which are produced by the positively charged PVDF membrane.

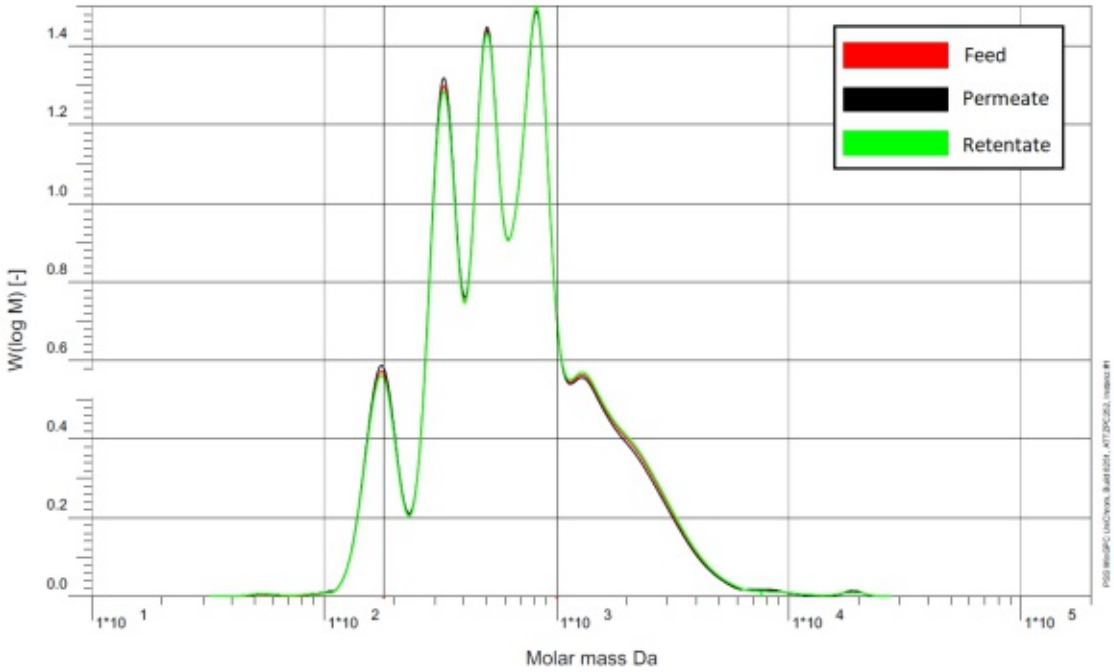

Figure 3.31: SEC results for the positively charged PVDF membrane, 100 kDa at 30°C with the Permeate as Feed (chapter 3.10.1), provided by Agrana

Even though the same membrane, as in figure 3.29, is used, when being pre-treated by a neutrally charged 100 kDa membrane, there is significantly less separation. As shown in figure 3.31, the size separation was not nearly in the extend shown in 3.29. That leads to the assumption that in order to keep the sugar composition constant throughout the membrane process, it is favourable to use a multi-stage process.

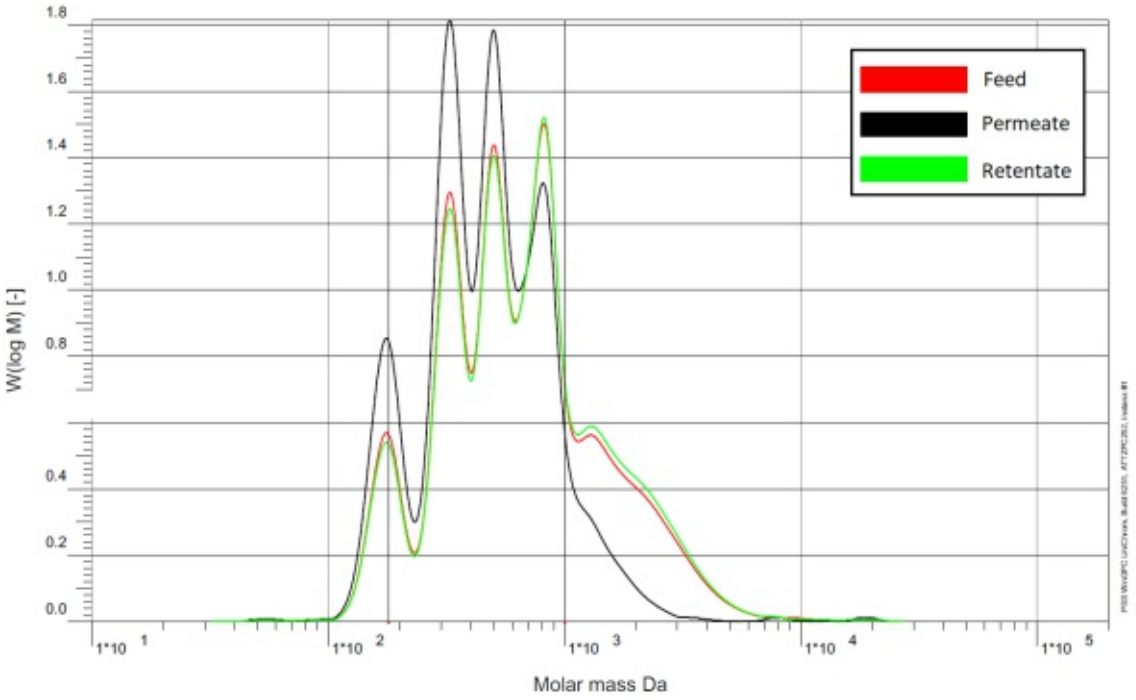

Figure 3.32: SEC results for the PES membrane, 5 kDa at 30°C (chapter 3.3.1), provided by Agrana

As predictable through the sugar rejection of 27.21%, the 5 kDa PES membrane produced a more vital separation of larger and smaller sugars. As shown in figure 3.32, there was a significant increase in smaller molecules in the permeate; however, molecules at more than 1000 Da got rejected immensely.

Generally, there is a large correlation between the sugar rejection, the molecular weight cut-off and the sugar composition. Therefore, when using tighter membranes, there is a more significant sugar rejection and a shift in concentration towards smaller molecules in the permeate, whereas membranes with larger MWCO do not influence the composition of the solution significantly.

3.10.4 Effects of the pH Value on the Colour, using Different Alkaline

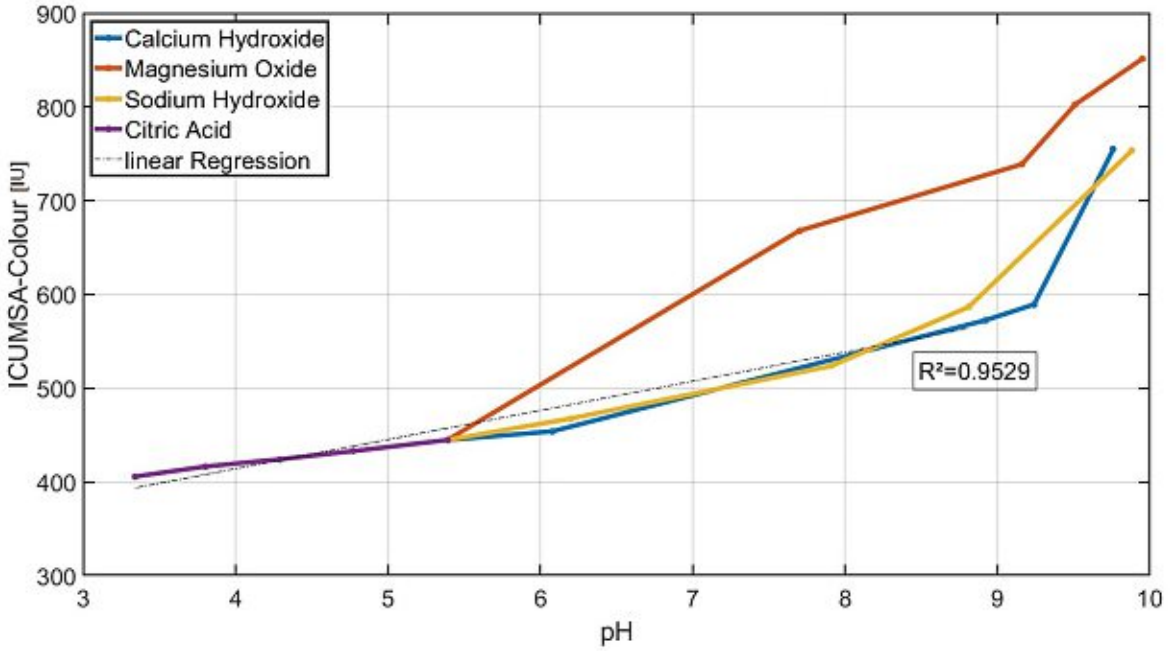

Figure 3.33: Influence of the pH value on the colour

In figure 3.33, it can be observed that there is a strong correlation between the pH value and the colour. Calcium hydroxide and sodium hydroxide had a relatively similar correlation between the pH and the colour, whereas magnesium oxide had a much stronger influence. Additionally, there is an almost linear correlation in the acidic and the alkaline range to a pH of about 9. However, there is a big jump in the colour at a pH value higher than 9.3. The linear regression, seen in figure 3.33, was made using from the acidic test results using citric acid and the alkaline test using sodium hydroxide.

3.10.5 Effects of the Addition of Acid as well as Alkaline to the Solution

Due to the shortage of sugar solution at some points during the work, the pH had to be first raised and then lowered. When adding both acid and alkaline, there are many chemicals introduced into the solution, which has a significant impact on the conductivity. However, the order in which the alkaline and acid are introduced influences the conductivity significantly.

Table 3.13: Influence of different chemicals on the conductivity

Added Chemicals	**pH**	**conductivity** [$\mu S\,cm^{-1}$]
none	5.39	687
NaOH	9.70	817
Citric Acid	3.33	807
NaOH + Citric Acid	3.31	952
Citric Acid + NaOH	9.77	3720

As shown in table 3.13, the conductivity and the ions dissolved in the solution increased slightly when adding either sodium hydroxide or citric acid. When adding NaOH in order to increase the pH and then citric acid to lower it, the conductivity further increased by about 18%. However, when lowering the solution first using citric acid, a lot more NaOH had to be introduced to increase the pH to a higher level. Therefore, the conductivity rose drastically to about 3700 $\mu S\,cm^{-1}$.

3.10.6 Aging of the Membranes

After finishing the test using one membrane, it was removed from the equipment. Therefore, the membranes could be examined after they were used. In 3.34, the vast differences of the aging of the different membranes can be observed. The positively charged PVDF membrane bound lots of molecules, whereas the neutrally and negatively charged ones bound less. This leads to the assumption, that lots of molecules in the solution are firstly, charged negatively, and secondly have trouble getting inside membranes with a lower MWCO, such as the 5 kDa PES membrane and the 10 kDa regenerated cellulose acetate membrane. Additionally, the 20 kDa hydrophilic PES membrane was less heavily coloured than the same membrane with 50 kDa, supporting this assumption.

Figure 3.34: Membranes after use from left to right, M183, M180, K328, P707, UH050, UH020, RC10PE

3.10.7 Effects of Activated Carbon and H_2O_2 on the Colour

The results of the test using the activated carbon are shown in table 3.14

Table 3.14: Results of the tests using activated carbon

Activated Carbon Type	Solution [ml]	Charcoal [g]	Sugar Rejection [%]	Colour rejection [%]
Donau Carbon	10	0.1	0.33	12.0
		0.5	2.7	21.2
		1	2.0	13.1
Chemviron Ammonsorb	10	0.1	-0.33	21.9
		0.5	-0.33	39.9
		1	-0.33	38.2
Chemviron Envirocarbon	10	0.1	0.00	0.79
		1	0.67	-22.7

From the results, it is evident that some of the activated carbons work better for this application than others. The Donau Carbon produced a maximum in colour rejection of 21.2%, had, however, a sugar rejection of about 2.7%, whereas, the Chemviron Ammonsorb Carbon produced nearly 40% colour rejection with no sugar rejection. Some measurements producd a negative sugar rejection, which is not logical. Therefore, these values are zero. Additionally, it is shown that there is an optimal ratio between sugar solution and activated carbon. Then Donau Carbon and the Ammonsorb activated carbon produced their highest colour rejection when adding 0.5g. When adding more activated carbon, the colour rejection decreased again. The Envirocarbon produced a negative colour rejection, when adding 1 g to 10 ml, which implies an introduction of colour into the solution, rather than a removal. Thus, this activated carbon is not suitable for the decolourisation of sugar solution.

For the tests using the H_2O_2, the results are represented in table 3.15. However, the concentration of sugar in the solution changed when adding H_2O_2, due to the dilution. This effect had to be taken into account when calculating the sugar loss. The sugar concentration was calculated using the refraction index, but the H_2O_2 solution has a refraction index other than 1 and therefore distort the results and had to be compensated. The refraction index of the 30% H_2O_2 was taken from [18] at a temperature of 20°C and is accepted to be 1.3529. The refraction index of the sugar solution was calculated from the concentration, using the ICUMSA table. The mixing ration of the components was used in order to find the mixture's refraction index, which was then used to calculate the sugar loss.

Table 3.15: Results of the tests using H_2O_2

H_2O_2 [ml]	**Sugar Solution[ml]**	**Mixing Ratio**	**Sugar Rejection [%]**	**Colour Rejection [%]**
5	5	1:1	-2.2	45.4
5	10	1:2	-0.13	37.5
5	15	1:3	0.07	30.6
5	20	1:4	-0.30	27.9
5	25	1:5	2.0	25.9
1	10	1:10	-1.2	12.4

Even though the refraction index of the H_2O_2 was considered when calculating the sugar rejection, there are still negative values, which would increase the sugar concentration. However, these values are relatively small and are accepted to be zero, and therefore do not produce a sugar rejection.

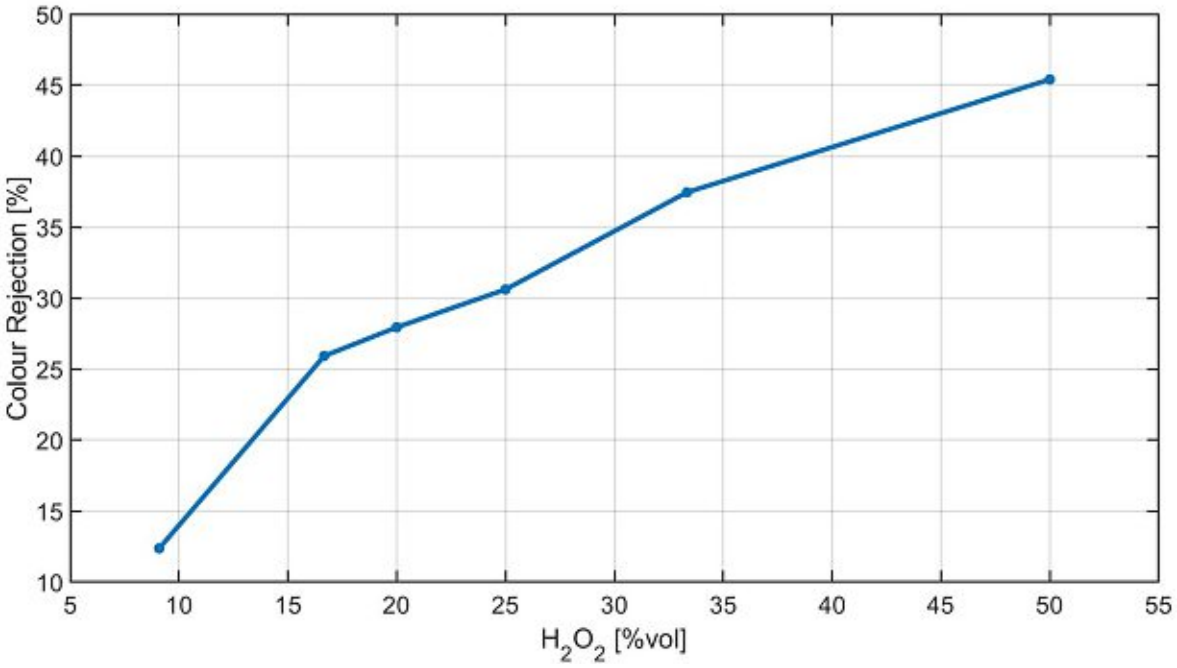

Figure 3.35: Colour rejection over volume fraction of H_2O_2

The colour rejection is dependent on the mixing ratio, as shown in figure 3.35. The higher the concentration of H_2O_2, the larger the resulting colour rejection. With a mixing ratio of 1:1, there was a colour rejection of 45.4%. Whereas, with a ratio of 1:10, there was only about 12.4% colour rejection.

3.11 Applications of the Results in an Industrial Scale

Next to the previously determined colour and sugar rejection, the membrane area is of great importance at an industrial scale. The area of membranes needed, directly correlates to the costs of the membrane processor's acquisition and operation. The membrane area needed to achieve a certain transmembrane flowrate was calculated using the mean flux measured.

$$\dot{m}_{trans} = J_{mean} \cdot A_{Membrane}. \tag{3.1}$$

Therefore, for a given flowrate, the membrane area was calculated using

$$A_{Membrane} = \frac{\dot{m}_{trans}}{J_{mean}}. \tag{3.2}$$

For further calculations a flowrate of $\dot{m}_{trans}$ =100 kg h^{-1} is assumed.

3.11.1 Comparison of the Membrane Areas

To use the tested membranes in a real process, not only the colour and sugar rejection are important, but also the cost. Therefore, the membranes chosen in figure 3.19, which achieve a colour rejection of over 30% with less than 10% sugar rejection, were compared regarding their membrane area at a transmembrane flow of 100 kg h^{-1}. In order to calculate the necessary membrane area of each membrane, equation 3.1 was used.

Table 3.16: Membrane area

#	Membrane Material	MWCO [kDa]	Temperature [°C]	pH	Colour Rejection [%]	Sugar Rejection [%]	Membrane Area [m²]
1	neutrally charged PVDF	100	50	9.34	32.0	2.1	**20.0**
2	neutrally charged PVDF	100	50	4.37	30.7	0.75	**20.3**
3	neutrally charged PVDF	100	50	3.15	34.7	2.1	**20.8**
4	neutrally charged PVDF	100	30	4.48	38.9	0.94	**21.2**
5	negatively charged PVDF	120	50	4.49	35.7	2.0	**21.5**
6	negatively charged PVDF	120	50	10.15	35.5	2.4	**21.5**
7	hydrophilic PES	50	50	9.73	39.5	4.4	**22.0**
8	hydrophilic PES	20	50	10.67	47.3	4.6	**23.3**
9	hydrophilic PES	50	50	4.42	34.0	7.0	**23.5**
10	positively charged PVFD	100	50	9.5	47.1	7.3	**24.7**
11	Regenerated Cellulose Acetate	10	30	5.59	34.7	9.2	**25.6**

Due to the different fluxes, the needed surface area in order to produce 100 $kg\,h^{-1}$ changes. Table 3.16 shows, that the membrane area correlates heavily with the MWCO, because the membrane with the least membrane area needed is one with the largest MWCO, whereas the largest membrane area is needed at a membrane with only 10 kDa. Additionally, the test producing the most extensive colour rejection, of the considered trials, needs with 23.28 m^2, a relatively large area. The experiments resulting in a lower colour rejection, like the neutrally charged PVDF of 100 kDa at 50°C and a pH of 4.37, need with 20.3 m^2 only smaller membrane areas.

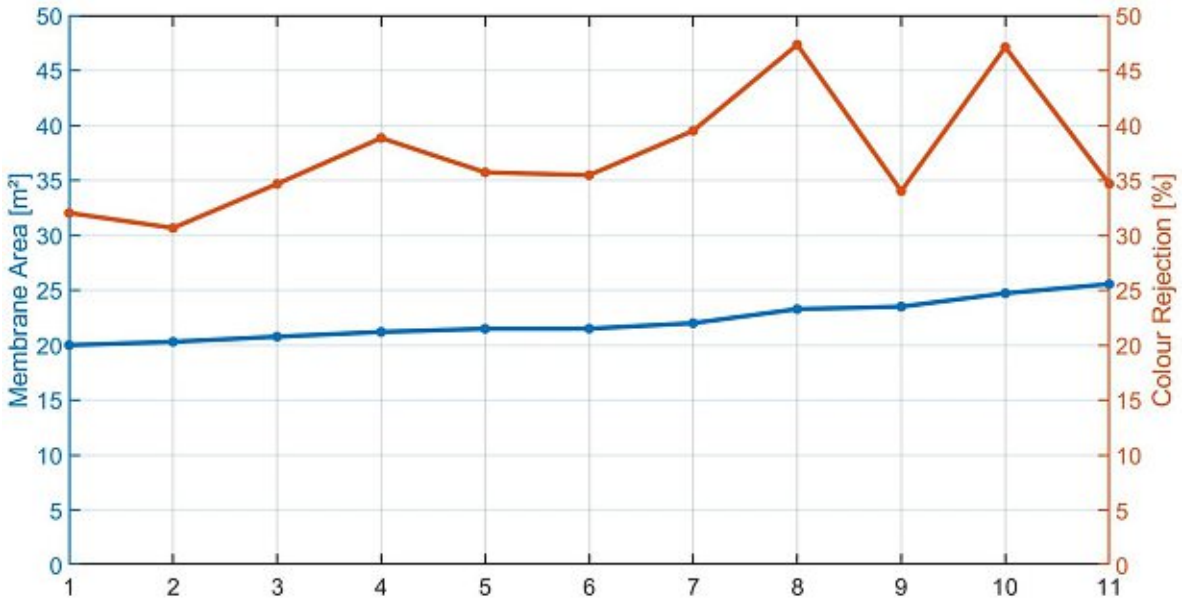

Figure 3.36: Correlation of the membrane area and colour rejection from tests in table 3.16

The plot shown in figure 3.36 illustrates the membrane area and colour rejection of the trials from table 3.16 sorted by ascending membrane area. There is a noticeable correlation between these values. However, there are also some experiments producing a lower colour rejection than expected, while needing a rather big surface area, like the tests of the regenerated cellulose acetate membrane, as well as the hydrophilic PES membrane at 50°C and natural pH.

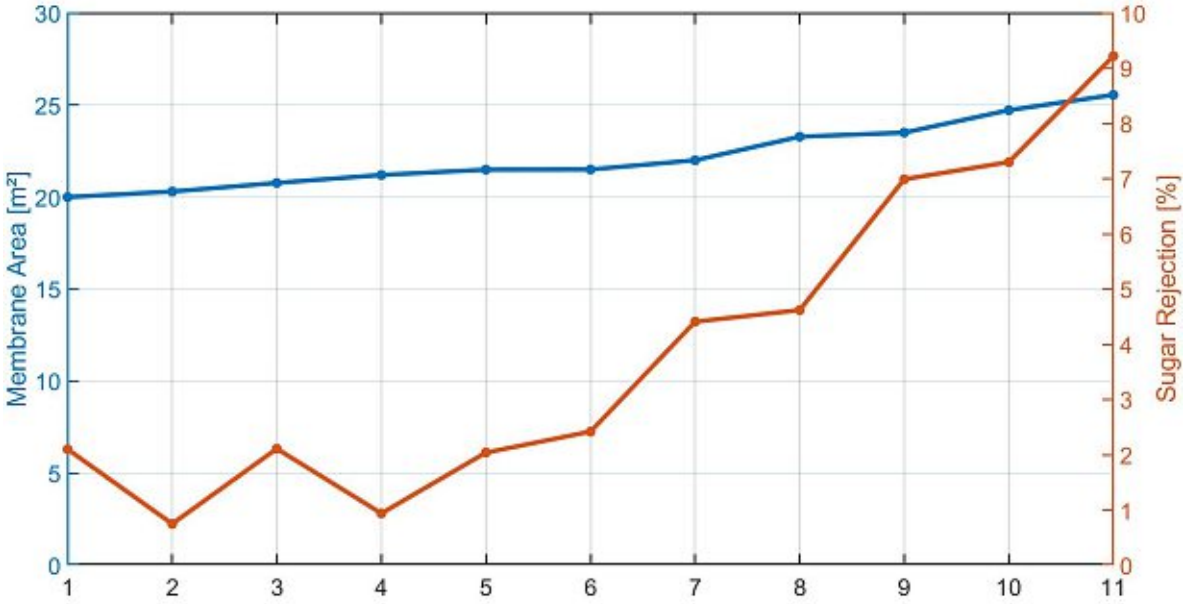

Figure 3.37: Correlation of the membrane area and sugar rejection from tests in table 3.16

When plotting the membrane area with the sugar rejection, as shown in figure 3.37, there is a much more obvious correlation. With a few exceptions, the sugar rejection follows the trend of the needed membrane area.

3.11.2 Comparison of a Single and Two-Stage Process at an Industrial Scale

In chapter 3.10.1, the colour and sugar rejection of a single-stage and a two-stage process are compared. However, to be an economical process, the membrane area and the sugar loss has to be calculated. For the single-stage process, at a transmembrane flow of $\dot{m}_{trans}$= 100 kg h^{-1}, the needed membrane area calculated, as seen in table 3.17, to 2.81 m^2. For the two-stage process the second stage's transmembrane flow was assumed to be $\dot{m}_{trans}$= 100 kg h^{-1}. Furthermore, it was assumed that the first stage produces 80% permeate and 20% retentate, which leads to a transmembrane flow of $\dot{m}_{trans}$= 125 kg h^{-1} for the first stage. Therefore, the first stage's membrane area calculated to 1.06 m^2 and the membrane area of the second stage to 0.78 m^2. The membrane area needed to implement the two-stage process is lower, even though there the permeate needs to pass two membranes compared to the single-stage process. The membrane at the single-stage process most likely clogged early on, and, therefore, the flux was relatively low, leading to a larger membrane area, whereas the pre-treatment using the neutrally charged PVDF membrane rejected larger particles and keeping the positively charged membrane unclogged for a longer time.

Table 3.17: Membrane area of the single and two-stage process

	Single-Stage	**Two-Stage**	
		First Stage	Second Stage
Flux [kg m^{-2} h^{-1}]	35.6	117.6	127.7
Membrane Area [m^2]		1.06	0.78
total Membrane Area [m^2]	**2.81**	**1.84**	

However, when looking at the two different processes' sugar loss, the two-stage process produces two separate retentate streams. In the following calculations, there is a permeate to feed ratio of 0.8 assumed. Therefore, 80% of the feed pass the membrane, whereas 20% remain in the retentate. Additionally, the permeate's sugar concentration was calculated using the measured refraction index of the permeate, whereas the sugar in the retentate was calculated using a mass balance and the total mass of sugar in the feed and permeate. For these calculations, a mass flow of 100 kg h^{-1} of feed was assumed. The first stage of the two-stage process was calculated using the data of the test carried out at 50°C. Additionally, there was a dilution happening in the two-stage process between the first and second stage.

Table 3.18: Sugar loss of the single and two-stage process

	Single-Stage	**Two-Stage**	
		First Stage	Second Stage
Feed Sugar [kg h^{-1}]	27.20	26.50	21.12
Permeate Sugar [kg h^{-1}]	19.52	21.12	16.00
Retentate Sugar [kg h^{-1}]	7.68	5.38	5.12
total Sugar Loss [kg h^{-1}]	**7.68**	**10.50**	
relative Sugar Loss [%]	**28.24**	**39.62**	

As seen in table 3.18, there is a significantly higher sugar loss in the two-stage process, because there are two retentate streams. However, in modern processes, the retentate streams are not lost but can be used in further processes or different applications.

Overall, a two-stage process is a economical alternative to a single-stage process, if the retentate streams are used in another way. Then a smaller membrane area needed, and the lower sugar rejection at almost the same colour rejection produces desirable results.

3.11.3 Comparison of a Single-Stage Process and a Two-Stage Process with Recycling

To achieve less sugar loss, compared to the calculations in chapter 3.11.2, recycling was added. Therefore, the second stage's retentate was treated using H_2O_2, as tested in chapter 3.10.7, and then recycled to the feed. That leads to only one final permeate and one retentate. The updated process is shown in figure 3.38.

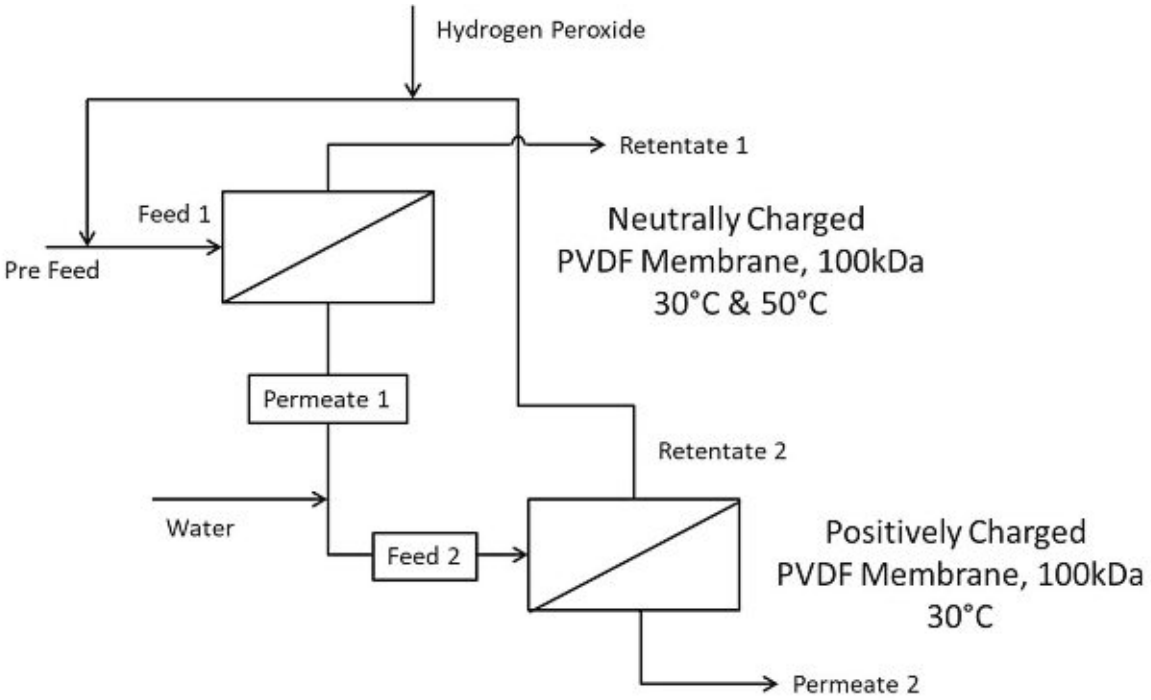

Figure 3.38: Two-stage process with recycling

To calculate the states, again a permeate to feed ratio of 0.8 for both stages was assumed. Additionally, the permeate's sugar concentration and colour was taken from the previous trials (Chapters 3.2 and 3.10.1). The added H_2O_2 was to be taken as pure water because it was assumed to react when added to the sugar solution ultimately. The mass flow of the added H_2O_2 was considered to be 33.33% of the retentate 2 mass flow, reducing the colour of the solution, according to table 3.15, to 69.3% of the original colour.
For this configuration to make sense, there have to be a few conditions fulfilled. Firstly, the membranes used in this configuration have the same molecular weight cut off. This is important because otherwise molecules that are larger than the second stage membrane, but smaller than the first stage membrane are trapped in the recycling loop. Secondly, this configuration needs an additional colour removal process between the retentate of the second stage and the recycling to the feed, which is implemented through the added Hydrogen Peroxide.

Table 3.19: Results of the two-stage process with recycling

	Sugar Mass Flow [kg h^{-1}]	**Water Mass Flow** [kg h^{-1}]	**ICUMSA-Colour**
Pre Feed	26.5	73.5	278.99
Feed 1	29.3	80.5	287.62
Permeate 1	23.19	64.65	191.44
Retentate 1	6.11	15.85	650.24
Feed 2	23.19	78.96	194.42
Permeate 2	20.39	74.41	147.11
Retentate 2	2.8	4.55	509.85
Water	0	14.31	0
H_2O_2	0	2.45	0
Retentate $+H_2O_2$	2.8	7	368.08

In table 3.19, the sugar concentration and colour are shown. The colour of the retentate steam of the first stage is with 650.24 a lot darker than the process without recycling, which was measured at 274.6. This is, however, logical, because there is less retentate and, therefore, the concentration of coloured molecules increases.

Table 3.20: Comparison of the two-stage process with recycling

	Single-Stage	**Two-Stage without Recycling**	**Two-Stage with Recycling**
Feed Sugar [kg h^{-1}]	27.20	26.50	26.50
Permeate Sugar[kg h^{-1}]	19.52	16.00	20.39
total Sugar Loss [kg h^{-1}]	**7.68**	**10.50**	**6.11**
relative Sugar Loss [%]	**28.24**	**39.62**	**23.06**

As shown in table 3.20, the sugar loss is remarkably lower when adding recycling. However, this recycling will produce an extensive consumption of hydrogen peroxide. Overall, the two-stage process is only limited suitable, because there is an enourmous sugar loss or large quantities of chemicals are necessary. Therefore, it is essential to study alternative secondary processes, like nanofiltration, to achieve better colour removal, while not losing much sugar.

Chapter 4

Conclusion and Outlook

While investigating the properties of different membranes regarding their ability to separate coloured molecules from sugar, several essential factors were considered. These factors included the membrane material, the molecular weight cut off, the charge, and the wettability of the membrane as well as the pH and the temperature of the sugar syrup. It is notable that even though the sucrose's molecular weight is only 342 $g\,mol^{-1}$, there is a sugar rejection evident when using membranes of 120 kDa. Therefore, the structure of the molecules also has a crucial impact on the separating capacity. Consequently, when using membranes with a smaller MWCO, the sugar rejection tends to increase. However, since the coloured molecules are formed during the Maillard reaction or caramelisation, the composition of these components is not specific, and a gain in sugar rejection does not automatically result in an increased colour rejection. The HPLC analysis of the solution showed that even if there is a colour rejection happening, there is no decrease in hydroxymethylfurfural. Therefore, there are several different components responsible for the colour. Furthermore, there was a test resulting in a sugar rejection larger than the colour rejection, which suggests smaller coloured molecules. Therefore, nanofiltration membranes, which keep the sugar in the retentate and let the smaller coloured molecules pass, should be further investigated.
The trials conducted, show that it is possible to reduce the sugar solution's colour by up to 60%, however at a rather high sugar rejection. As discussed in previous chapters, there has to be a compromise between colour and sugar rejection for industrial applications, because a large sugar rejection leads to an uneconomical amount of sugar loss. Furthermore, there is a shift in sugar composition to smaller sugars happening at a lower MWCO, which produces a higher sugar rejection. The temperature plays a significant role in the results, where an increase in temperature leads to a lowered viscosity. Due to the enhanced fluidity, there is a reduction in colour and sugar rejection. Additionally, there is a rather obvious correlation of the pH value and the colour rejection, where tests at higher pH values produce a more extensive colour rejection. These tests are only practical if the pH is lowered again after the ultrafiltration because ofthe pH dependency of the colour.

Furthermore, the charge of the membrane is an essential factor to consider. The positively and neutrally charged PVDF membranes with a MWCO of 100 kDa produced somewhat different results, where the positive charge led to a higher rejection in sugar and colour.
When investigating multi-stage processes, there was, despite having several positive effects, a large loss in sugar, due to the accumulation of two individual retentate streams. In order to compensate this enormous loss, a recycling configuration of one retentate has to be implemented. However, if using chemicals to remove the colour further, the process becomes rather chemical-intensive.

In conclusion, it is possible to remove up to 60% of the colour; however, there is a significant amount of sugar loss, when producing a higher colour rejection. Therefore, it is more economical to reduce the colour by 30% to 40% at a reasonable sugar rejection and investigate alternative colour removal methods. Treatments after the primary UF stage can be adsorption using activated carbon, oxidisation using hydrogen peroxide, a second UF stage, or even a nanofiltration stage to remove smaller coloured molecules and keep the sugar in the retentate. Therefore, further work has to be conducted investigating secondary colour removing technologies, especially nanofiltration membranes, which separate smaller coloured molecules.

List of Tables

List of Figures

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Appendix A

Experimental Data

In the following chapters the measured and calculated data of the conducted experiments is provided. The data is divided into multiple sections.In chapter A.1 the data for the calculation of the flux is presented, in chapter A.2 the measured sugar concentration, conductivity and pH as well as the calculated sugar rejection is shown, and in chapter A.3 the measurements of the colour is provided.

A.1 Experimental Data and Calculations of Flux

Table A.1: Experimental data and calculations of flux

Membrane	Feed pH	Temperature [°C]	Time [min]	Mass [g]	Flux [kg m^{-2} h^{-1}]
			0	38.8	70.5
			10	112.7	47.625
			20	193.5	43.125
			30	231.2	42
			40	288	41.25
			50	365.7	40.125
			60	393.7	40.125
			70	447	37.5
			80	530.6	36
			90	554.4	35.25
			100	589.2	34.5
			110	646.6	34.125
			120	676	33.75
M-183	4.29	30	130	717.8	33
			140	784.2	32.625
			150	806.7	32.25
			160	844.6	32.25
			170	905.4	31.5
			180	931.3	31.125
			190	958.5	30.75
			200	1032	30
			210	1056	30
			220	1085.8	29.25
			230	1139.7	28.875
			240	1161.5	28.125
			250	1196.6	28.5
			260	1225.4	27.75

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux** $[\mathrm{kg\,m^{-2}\,h^{-1}}]$
M-183	4.42	50	0	11	-
			10	240.7	172.28
			20	409.5	126.6
			30	550.9	106.05
			40	667.3	87.3
			50	771.8	78.38
			60	869.4	73.2
			70	955.3	64.43
			80	1031.5	57.15
			90	1105.5	55.5
			100	1178	54.38
M-183	9.50	50	0	24.6	-
			10	158	90.95
			20	269	83.25
			30	371.6	76.95
			40	472	75.3
			50	561	66.75
			60	648.2	65.4
			70	732	62.85
			80	814	61.5
			90	892.1	58.58
			100	972.3	60.15
			110	1046	55.28
			120	1118.8	54.6
			130	1188.3	52.13
			140	1257.4	51.83

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Mass [g]	Flux [kg m^{-2} h^{-1}]
M-183	3.31	50	0	15	-
			10	208.2	131.727
			20	323.9	86.775
			30	442.6	89.025
			40	526.3	62.775
			50	600.4	55.575
			60	673.6	54.9
			70	745.2	53.7
			80	815.7	52.875
			90	882	49.725
			100	949.5	50.625
			110	1015.6	49.575
			120	1087.6	54
			130	1157.4	52.35
			140	1230	54.45
M-180	4.37	50	0	16	-
			10	467.5	338.625
			20	837.8	277.725
			30	1155.8	238.5
			32	1214.7	220.875
M-180	4.48	30	0	4	-
			10	226	166.5
			20	412.7	140.025
			30	577.4	123.525
			40	728.3	113.175
			50	869.2	105.675
			60	1004.2	101.25
			70	1113.2	81.75
			80	1258.4	108.9

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux [kg m^{-2} h^{-1}]**
M-180	9.34	50	0	15	-
			10	281.7	200.025
			20	485.7	153
			30	661	131.475
			40	820	119.25
			50	976.2	117.15
			60	1123.2	110.25
M-180	3.15	50	0	10	-
			10	296.1	214.575
			20	502.6	154.875
			30	674.6	129
			40	835	120.3
M-183	4.57	30	0	3	-
			10	291.8	216.6
			20	500.6	156.6
			30	161.2	120.9
			40	308.8	110.7
			50	135.2	101.4
			60	263.7	96.375
			70	121.5	91.125

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux [kg m^{-2} h^{-1}]**
K-328	4.45	30	0	1.5	-
			10	34.6	24.825
			20	61.8	20.4
			30	86.2	18.3
			40	109	17.1
			50	130.6	16.2
			60	151.5	15.675
			70	171.9	15.3
			80	191.4	14.625
			90	210.7	14.475
			100	229.7	14.25
			110	248.1	13.8
			120	266.3	13.65
			130	284.2	13.425

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Mass [g]	Flux [kg m^{-2} h^{-1}]
K-328	4.45	50	0	1.7	-
			10	45	32.475
			20	84.9	29.925
			30	122.4	28.125
			40	157.7	26.475
			50	190.6	24.675
			60	222.6	24
			70	253.5	23.175
			80	283.8	22.725
			90	313.9	22.575
			100	343	21.825
			110	372	21.75
			120	400.5	21.375
			130	429	21.375
			140	456.1	20.325
			150	483.5	20.55
			160	510.2	20.025
			170	536.6	19.8
			180	562.7	19.575
			190	587.8	18.825
			200	612.9	18.825
			210	637.9	18.75
			220	662.3	18.3
			230	686.7	18.3

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux [kg m^{-2} h^{-1}]**
K-328	2.95	50	0	0	-
			10	32.7	24.525
			20	63.2	22.875
			30	90.6	20.55
			40	116.5	19.425
			50	141.9	19.05
			60	165.9	18
			70	188.7	17.1
			80	211.7	17.25
			90	234.3	16.95
			100	256.7	16.8
			110	278.8	16.575
			120	300.7	16.425
			130	322.6	16.425
			140	343.8	15.9
			150	365.2	16.05
			160	385.8	15.45
			170	406.1	15.225
			180	425.7	14.7
			190	445.5	14.85
			200	465.2	14.775
			210	484.5	14.475

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Mass [g]	Flux [kg m^{-2} h^{-1}]
K-328	9.64	50	0	3	-
			10	94.2	68.4
			20	179.2	63.75
			30	259	59.85
			40	335.3	57.225
			50	408.7	55.05
			60	479.2	52.875
			70	547	50.85
			80	611.3	48.225
			90	672.9	46.2
			100	734	45.825
			110	790	42
			120	845.5	41.625
			130	898.2	39.525
			140	949.5	38.475
			150	998	36.375
			160	1044.6	34.95
P-707	4.49	30	0	8	-
			10	190.4	136.8
			20	324	100.2
			30	437.4	85.05
			40	537.8	75.3
			50	626.8	66.75
			60	709.7	62.175
			70	787.1	58.05
			80	861.5	55.8
			90	932.8	53.475
			100	1002.1	51.975
			110	1070	50.925
			120	1135.7	49.275
			130	1199.7	48

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Mass [g]	Flux [kg m^{-2} h^{-1}]
P-707	4.49	50	0	9	-
			10	217.3	156.225
			20	402.8	139.125
			30	573.2	127.8
			40	732.6	119.55
			50	882.6	112.5
			60	1026.5	107.925
			70	1165.2	104.025
P-707	3.16	50	0	8.8	-
			10	228	164.4
			20	412	138
			30	577.8	124.35
			40	730	114.15
			50	872.4	106.8
			60	1007.9	101.625
			70	1134.3	94.8
			80	1256.4	91.575
P-707	10.15	50	0	8	-
			10	203.3	146.475
			20	368.2	123.675
			30	519.4	113.4
			40	665.5	109.575
			50	804.1	103.95
			60	937	99.675
			70	1063.8	95.1

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux** $[\mathrm{kg\,m^{-2}\,h^{-1}}]$
			0	3	-
			10	97.7	71.025
			20	175.9	58.65
			30	247.7	53.85
			40	314.7	50.25
			50	377.2	46.875
			60	437.1	44.925
			70	494.1	42.75
			80	549.7	41.7
			90	603.5	40.35
UH050	4.43	30	100	657	40.125
			110	709.4	39.3
			120	760.8	38.55
			130	810.6	37.35
			140	860	37.05
			150	907.7	35.775
			160	953.8	34.575
			170	998.8	33.75
			180	1043.8	33.75
			190	1087.4	32.7
			200	1131.2	32.85

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux [kg m^{-2} h^{-1}]**
UH050	4.42	50	0	4	-
			10	116.6	84.45
			20	224.9	81.225
			30	328	77.325
			40	427.2	74.4
			50	522.7	71.625
			60	615.6	69.675
			70	704.1	66.375
			80	790.7	64.95
			90	874.4	62.775
			100	956.6	61.65
			110	1036	59.55
			120	1113.9	58.425
UH050	9.73	50	0	4.6	-
			10	152.9	111.225
			20	284.2	98.475
			30	406.4	91.65
			40	520.5	85.575
			50	626.8	79.725
			60	728.2	76.05
			70	825.7	73.125
			80	918.9	69.9
			90	1008.4	67.125
			100	1096.4	66

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux [kg m^{-2} h^{-1}]**
			0	3	-
			10	86.1	62.325
			20	165.2	59.325
			30	239.9	56.025
			40	311.6	53.775
			50	381	52.05
			60	448.3	50.475
			70	513.5	48.9
			80	577.4	47.925
UH050	3.08	50	90	639.4	46.5
			100	700.4	45.75
			110	760	44.7
			120	819.2	44.4
			130	877	43.35
			140	934	42.75
			150	989	41.25
			160	1043.8	41.1
			170	1098.4	40.95
			180	1148.9	37.875

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Mass [g]	Flux [kg m^{-2} h^{-1}]
			0	1.5	-
			10	52.4	38.175
			20	95	31.95
			30	134	29.25
			40	171	27.75
			50	206.2	26.4
			60	240	25.35
			70	272.8	24.6
			80	304.7	23.925
			90	335.9	23.4
			100	367.2	23.475
			110	397.4	22.65
UH020	4.54	30	120	426.8	22.05
			130	455.8	21.75
			140	484.6	21.6
			150	512.8	21.15
			160	540	20.4
			170	566.7	20.025
			180	593.2	19.875
			190	619	19.35
			200	644.8	19.35
			210	669.9	18.825
			220	695	18.825
			230	719.4	18.3
			240	743.6	18.15

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux** $[\mathrm{kg\,m^{-2}\,h^{-1}}]$
UH020	4.52	50	0	2	-
			10	83.6	61.2
			20	160.8	57.9
			30	233.5	54.525
			40	303.2	52.275
			50	370.5	50.475
			60	435.6	48.825
			70	498.4	47.1
			80	560.2	46.35
			90	620.2	45
			100	679	44.1
			110	736	42.75
			120	792	42
UH020	3.01	50	0	2.1	-
			10	67.8	49.275
			20	126.4	43.95
			30	181.2	41.1
			40	233.4	39.15
			50	284	37.95
			60	334.3	37.725
			70	384.5	37.65
			80	431.5	35.25
			90	478.4	35.175
			100	524.5	34.575
			110	569.3	33.6
			120	613.4	33.075
			130	656.2	32.1
			140	698.3	31.575

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Mass [g]	Flux [kg m^{-2} h^{-1}]
UH020	10.67	50	0	7	-
			10	178	128.25
			20	319.8	106.35
			30	447.5	95.775
			40	564.5	87.75
RC10PE	4.74	50	0	6	-
			10	176	127.5
			20	337.5	121.125
			30	489.3	113.85
			40	632.6	107.475
			50	767.4	101.1
			60	895.3	95.925
RC10PE	5.59	30	0	1.5	-
			10	68.9	50.55
			20	135.8	50.175
			30	201.8	49.5
			40	266.9	48.825
			50	330.4	47.625
			60	393.2	47.1
			70	454.6	46.05
			80	515.8	45.9
			90	576.2	45.3
			100	636.1	44.925
			110	694	43.425
			120	752.6	43.95
			130	811.5	44.175
			140	868.9	43.05
			150	925.4	42.375
			160	981.6	42.15
			170	1036.4	41.1
			180	1091.9	41.625

Table A.1: Experimental data and calculations of flux (Continued)

Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**Mass [g]**	**Flux** $[\mathrm{kg\,m^{-2}\,h^{-1}}]$
RC10PE	5.44	50	0	5	-
			10	176.6	128.7
			20	332.7	117.075
			30	480.9	111.15
			40	625.6	108.525
			50	765.8	105.15
			60	902.2	102.3
			70	1036	100.35
			80	1162.4	94.8
RC10PE	3.24	50	0	4	-
			10	157.9	115.425
			20	299.7	106.35
			30	432.4	99.525
			40	559.7	95.475
			50	681.8	91.575
			60	799.8	88.5
			70	917.2	88.05
			80	1030.7	85.125
			90	1139.2	81.375
			100	1244.2	78.75
RC10PE	9.50	50	0	4	-
			10	150.1	109.575
			20	292.7	106.95
			30	431.6	104.175
			40	567.7	102.075
			50	699.4	98.775
			60	827.9	96.375
			70	952.8	93.675
			80	1075.5	92.025
			90	1194.7	89.4
			100	1311.9	87.9

A.2 Experimental Data of Concentration, Conductivity, and pH

Table A.2: Experimental data and calculations of concentration, conductivity, and pH

Membrane	Feed pH	Temperature °C	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
M-183	4.29	30	20	25	779	4.24	27.2	737	4.25	0.0809
			50	24	788	4.19	27.3	731	4.24	0.1176
			80	24.4	798	4.19	27.7	726	4.23	0.1062
			110	24.5	799	4.22	27.8	718	4.25	0.1155
			140	24.3	799	4.27	28.2	714	4.24	0.1259
			170	24.3	801	4.26	28.5	704	4.28	0.1383
			200	24.6	799	4.29	28.8	696	4.28	0.1368
			230	24.4	800	4.25	29.1	686	4.26	0.1528
			260	24.4	800	4.28	29.5	680	4.24	0.1615
M-183	4.42	50	20	25.8	767	4.26	26.6	774	4.33	0.0264
			40	25.7	760	4.29	26.8	766	4.32	0.0338
			60	25.6	757	4.26	27.1	762	4.31	0.0448
			80	25.7	763	4.25	27.4	758	4.32	0.0517
			100	25.6	758	4.25	27.6	751	4.32	0.0657

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature °C	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
M-183	9.50	50	20	25.6	920	9.89	27	1005	9.7	0.0554
			40	25.5	912	10	27.3	1007	9.33	0.0556
			60	25.4	909	9.61	27.5	1012	9.22	0.0696
			80	25.6	913	9.61	27.7	1015	9.15	0.0691
			100	25.5	914	9.62	27.9	1016	9.2	0.0794
			120	25.5	919	9.63	28.2	1022	9.09	0.0860
			140	25.5	920	9.71	28.7	1025	9.23	0.0957
M-183	3.31	50	20	25.9	1014	3.25	27	1003	3.25	0.0443
			40	25.6	1012	3.29	27.1	978	3.26	0.0519
			60	25.5	1013	3.26	27.3	974	3.32	0.0590
			80	25.5	1018	3.27	27.6	963	3.27	0.0659
			100	25.2	1015	3.25	27.8	955	3.27	0.0870
			120	25.4	1018	3.26	28.1	950	3.27	0.0863
			140	25.7	1015	3.25	28.7	947	3.26	0.0854
M-180	4.37	50	20	26.3	747	4.39	26.6	779	4.4	0.0075
			32	26.4	741	4.44	27.2	785	4.42	0.0075
M-180	4.48	30	20	26.6	717	4.47	26.6	742	4.44	0.0000
			40	26.3	718	4.46	26.5	745	4.44	0.0113
			60	26.2	716	4.46	26.6	747	4.45	0.0113
			80	26.2	717	4.47	26.7	751	4.44	0.0150

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature °C	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
M-180	9.34	50	20	26.2	939	9.48	27	1042	9.29	0.0224
			40	26.3	933	9.5	27.2	1053	9.26	0.0259
			60	26.8	932	9.5	27.6	1080	9.21	0.0147
M-180	3.15	50	20	20.7	1176	3.15	21.4	1147	3.15	0.0096
			40	20.7	117.3	3.18	21.8	1150	3.21	0.0327
M-183	4.57	30	20	22.1	709	4.52	22.5	712	4.48	0.0264
			40	21.7	714	4.5	22.6	709	4.46	0.0356
			60	21.5	720	4.47	22.9	706	4.47	0.0487
			70	21.5	720	4.51	23	706	4.48	0.0611
K-328	4.45	30	20	20.2	836	4.5	26.6	733	4.42	0.2406
			40	19.8	867	4.48	26.8	729	4.39	0.2556
			70	19.4	883	4.43	27.2	724	4.39	0.2761
			100	19.4	899	4.45	27.2	722	4.39	0.2868
			130	19	902	4.41	29.1	714	4.39	0.3015

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature °C	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
K-328	4.45	50	20	21.6	862	4.4	26.8	770	4.44	0.1910
			40	21.2	864	4.44	27	760	4.47	0.2090
			60	21.1	862	4.43	27.3	755	4.47	0.2185
			90	21.1	872	4.43	27.6	749	4.47	0.2271
			120	21.1	868	4.41	27.8	745	4.49	0.2355
			150	21.1	871	4.43	28.3	734	4.43	0.2410
			180	21.2	866	4.39	28.6	758	4.41	0.2509
			210	21.3	871	4.44	28.9	717	4.44	0.2552
			230	21.2	868	4.37	29.1	714	4.42	0.2664
K-328	2.95	50	20	19.5	1306	3.04	26.3	1021	3.04	0.2500
			40	19.3	1343	3.03	26.6	1013	3.04	0.2662
			60	19.2	1339	2.94	26.6	1007	2.98	0.2782
			90	19.2	1363	2.96	26.7	995	2.98	0.2782
			120	19.1	1342	2.94	26.9	986	2.99	0.2846
			150	19.2	1370	3.01	27.3	974	3.02	0.2862
			180	19.1	1360	2.95	28.5	963	3.04	0.3004
			210	19.2	1370	2.97	28.5	953	3.04	0.3263

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature °C	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
K-328	9.64	50	20	23.6	2170	9.82	26.2	3070	9.44	0.0888
			40	23.5	2180	9.85	26.5	3130	9.35	0.1031
			60	23.5	2170	9.75	26.8	3200	9.26	0.1132
			80	23.7	2190	9.77	27	3240	9.13	0.1157
			100	23.5	2200	9.73	27.4	3300	9.03	0.1296
			120	23.5	2220	9.63	27.7	3350	8.97	0.1423
			140	23.5	2240	9.64	28	3420	8.86	0.1516
			150	23.7	2240	9.59	28.1	3430	8.8	0.1536
			160	23.5	2260	9.62	28.3	3490	8.73	0.1637
P-707	4.49	30	20	26.5	704	4.46	26.8	749	4.44	0.0185
			40	26.4	703	4.47	26.9	750	4.45	0.0149
			60	26	705	4.44	27	751	4.43	0.0335
			80	26	706	4.45	27.1	755	4.43	0.0370
			100	26	710	4.45	27.2	755	4.43	0.0406
			120	26	713	4.45	27.6	756	4.43	0.0441
			130	25.9	712	4.44	27.9	760	4.43	0.0616
P-707	4.49	50	20	26.4	712	4.45	26.9	788	4.48	0.0186
			40	26.4	715	4.45	27	795	4.48	0.0186
			60	26.5	715	4.45	27.1	809	4.49	0.0185
			70	26.4	718	4.45	27.2	817	4.48	0.0258

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature °C	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
P-707	3.16	50	20	26.2	900	3.16	26.4	915	3.16	0.0187
			40	25.8	906	3.16	26.6	906	3.2	0.0227
			60	25.8	906	3.17	26.8	903	3.21	0.0301
			80	25.8	910	3.17	26.9	901	3.21	0.0373
P-707	10.15	50	20	26.2	2160	10.3	26.6	2510	10.11	0.0150
			40	26.1	2130	10.3	26.8	2570	10.02	0.0188
			60	26.1	2130	10.27	27.1	2620	9.98	0.0261
			70	26.1	2150	10.26	27.1	2700	9.96	0.0369
UH050	4.43	30	20	25.6	753	4.47	27.3	743	4.43	0.0623
			40	25.1	760	4.46	27.5	739	4.43	0.0806
			60	25	762	4.44	27.6	735	4.43	0.0909
			80	24.9	767	4.44	27.8	731	4.42	0.0978
			100	24.8	769	4.42	28	728	4.41	0.1079
			120	24.8	772	4.42	28.2	719	4.4	0.1143
			140	24.8	772	4.41	28.5	719	4.4	0.1206
			160	24.8	774	4.41	28.7	717	4.4	0.1298
			180	24.8	774	4.4	28.9	709	4.38	0.1359
			200	24.8	774	4.41	29.2	702	4.4	0.1419

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature °C	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
UH050	4.42	50	20	26	765	4.4	27.7	780	4.43	0.0511
			40	26.1	770	4.4	27.9	780	4.44	0.0578
			60	26.1	764	4.37	28.2	779	4.42	0.0645
			80	26.1	766	4.4	28.4	774	4.44	0.0745
			100	26	764	4.38	28.7	768	4.44	0.0845
			120	26.2	764	4.39	28.9	768	4.42	0.0871
UH050	9.73	50	20	27.3	1050	9.88	28.3	1156	9.58	0.0250
			40	27.2	1039	9.87	28.5	1162	9.55	0.0389
			60	27.2	1040	9.86	28.7	1169	9.54	0.0456
			80	27.2	1046	9.83	29	1175	9.5	0.0523
			100	27.3	1044	9.82	29.3	1169	9.43	0.0586
UH050	3.08	50	20	26.1	1323	3.04	28	1269	3.07	0.0645
			40	25.5	1346	3.09	28.3	1252	3.12	0.0893
			60	25.5	1329	3.08	28.5	1249	3.12	0.0989
			80	25.5	1340	3.12	28.7	1237	3.12	0.1053
			100	25.5	1335	3.08	28.9	1227	3.13	0.1115
			120	25.5	1344	3.12	29.1	1215	3.12	0.1176
			140	25.5	1336	3.07	29.4	1201	3.12	0.1237
			160	25.4	1333	3.07	29.5	1193	3.13	0.1361
			180	25.3	1335	3.08	29.6	1181	3.13	0.1424

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
UH020	4.54	30	20	22.6	839	4.68	26.4	735	4.53	0.1439
			40	22	839	4.64	26.6	731	4.53	0.1667
			60	21.8	845	4.61	26.9	724	4.52	0.1805
			80	21.7	852	4.59	27.1	719	4.51	0.1933
			100	21.6	853	4.54	27.3	713	4.5	0.2030
			120	21.6	857	4.55	27.6	709	4.51	0.2088
			140	21.5	858	4.55	27.7	702	4.5	0.2210
			160	21.5	860	4.55	27.9	697	4.5	0.2238
			180	21.5	859	4.54	28.2	690	4.5	0.2294
			200	21.5	861	4.55	28.5	683	4.5	0.2376
			220	21.6	860	4.54	28.8	677	4.5	0.2421
			240	21.5	861	4.53	29	667	4.5	0.2535
UH020	4.52	50	20	23.1	808	4.49	26.7	767	4.48	0.1250
			40	23	822	4.55	27.1	759	4.53	0.1386
			60	23	812	4.56	27.4	751	4.53	0.1513
			80	23	815	4.59	27.9	740	4.58	0.1606
			100	23.1	809	4.62	28.3	731	4.59	0.1720
			120	23.2	812	4.64	28.8	717	4.6	0.1802

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
UH020	3.01	50	20	22.3	1309	2.93	26.6	1093	2.98	0.1489
			40	22.2	1331	2.96	26.9	1078	3	0.1654
			60	22.1	1322	2.95	27.5	1065	3	0.1784
			80	22.2	1323	2.93	27.7	1049	3.01	0.1927
			100	22.2	1321	2.93	28	1032	2.99	0.1986
			120	22.2	1317	2.92	28.5	1012	2.98	0.2071
			140	22.3	1319	2.93	28.9	999	2.98	0.2175
UH020	10.67	50	10	25.9	3790	10.96	27	4570	10.7	0.0336
			20	25.8	3990	10.96	27.1	4620	10.68	0.0444
			30	25.7	4000	10.96	27.2	4640	10.64	0.0517
			40	25.7	4020	10.97	27.5	4610	10.64	0.0551
RC10PE	4.74	50	20	19.5	2670	4.63	21.1	2690	4.72	0.0625
			40	19.8	2660	4.69	21.4	2680	4.82	0.0616
			60	19.9	2670	4.68	21.8	2670	4.75	0.0701

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

Membrane	Feed pH	Temperature [°C]	Time [min]	Permeate C_S [°Brix]	Permeate Conductivity [µS cm^{-1}]	Permeate pH	Retentate C_S [°Brix]	Retentate Conductivity [µS cm^{-1}]	Retentate pH	Sugar Rejection
RC10PE	5.59	30	20	25	759	5.58	27.3	716	5.55	0.0876
			40	25.1	757	5.6	27.5	711	5.55	0.0806
			60	25.1	757	5.58	27.6	708	5.56	0.0873
			80	25.2	755	5.59	27.8	704	5.55	0.0870
			100	25.2	754	5.58	28	700	5.56	0.0935
			120	25.5	753	5.6	28.1	696	5.55	0.0893
			140	25.3	752	5.6	28.3	691	5.56	0.0996
			160	25.4	751	5.58	28.4	687	5.56	0.1025
			180	25.5	751	5.57	28.6	682	5.57	0.1021
RC10PE	5.44	50	20	26.2	736	5.54	27	755	5.48	0.0260
			40	26.1	736	5.55	27.2	751	5.51	0.0333
			60	26	733	5.55	27.4	746	5.5	0.0441
			80	26.1	738	5.56	27.7	739	5.53	0.0474
RC10PE	3.24	50	20	26.6	936	3.18	27.8	920	3.22	0.0327
			40	26.5	928	3.19	28	913	3.23	0.0468
			60	26.6	932	3.19	28.1	912	3.23	0.0500
			80	26.6	926	3.19	28.4	902	3.22	0.0534
			100	26.6	930	3.19	28.7	888	3.21	0.0634

Table A.2: Experimental data and calculations of concentration, conductivity, and pH (continued)

				Permeate			Retentate			
Membrane	**Feed pH**	**Temperature [°C]**	**Time [min]**	**C_S [°Brix]**	**Conductivity [µS cm^{-1}]**	**pH**	**C_S [°Brix]**	**Conductivity [µS cm^{-1}]**	**pH**	**Sugar Rejection**
RC10PE	9.50	50	20	26	932	9.74	27.4	952	9.36	0.04412
			40	26	928	9.73	27.5	947	9.32	0.05109
			60	26.1	929	9.68	27.6	942	9.27	0.05091
			80	26.1	927	9.65	27.9	940	9.23	0.05435
			100	26.2	926	9.66	28.1	925	9.15	0.06093

A.3 Experimental Data of Colour

Table A.3: Experimental data of colour

Membrane	pH	Temperature [°C]	Feed Colour [IU]	Permeate Colour [IU]	Retentate Colour [IU]
M-183	4.29	30	273.3	139.24	289.98
M-183	4.42	50	246	177	232
M-183	9.50	50	449	339	641
M-183	3.31	50	244	180	226.3
M-180	4.37	50	270.5	190.4	274.6
M-180	4.48	30	288.3	194.1	317.4
M-180	9.34	50	522.8	418.1	615.1
M-180	3.15	50	293.7	187.1	286.4
M-183	4.57	30	267.6	143.5	188.3
K-328	4.45	30	281.5	118.3	255.9
K-328	4.45	50	250	152.8	233.9
K-328	2.95	50	220	159.5	205.5
K-328	9.64	50	455.2	220.4	599.4
P-707	4.49	30	275.6	248.9	318.9
P-707	4.49	50	254.7	177.4	276
P-707	3.16	50	204.2	157.5	211.2
P-707	10.15	50	473.7	490.7	760.4
UH050	4.43	30	269.6	168.9	300.6
UH050	4.42	50	254.1	176.2	267.1
UH050	9.73	50	546.5	421.5	696.8
UH050	3.08	50	261.8	199.6	242.6
UH020	4.54	30	276.4	149.7	283.9
UH020	4.52	50	253.2	149.5	249
UH020	3.01	50	223	148.8	241.2
UH020	10.67	50	610.8	564.6	1072.2
RC10PE	4.74	50	258.9	189.7	275.5
RC10PE	5.59	30	448.56	334.76	512.46
RC10PE	5.44	50	450.5	380.16	491.66
RC10PE	3.24	50	416.74	336.36	422.8
RC10PE	9.50	50	809.4	754.34	1005.89