Membrane-based conceptual design of reuse water production from candy factory wastewater

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ABSTRACT

Intense pressure on water resources has led to efforts to reuse reclaimed processing wastewater in the food industry. There are tight rules for water quality, but efficient separation technologies such as reverse osmosis possess good possibilities for water reuse. This study developed a membrane-based reuse water concept for wastewater from the candy industry emphasizing the pre-treatment stage in the concept to reduce fouling. The wastewater contained suspended solids, sugar compounds and the ingredients for candy gelation, which had a tendency to foul membranes, making pre-treatment essential for a successful concept. Cross-rotational ultrafiltration, which featured enhanced fouling prevention for membranes, functioned well for the removal of challenging substances. Conventional filtration technologies were impractical due to a low flux, even when the viscosity of the wastewater was reduced using surfactants. The wastewater had a high chemical oxygen demand, meaning that there was a strong fouling potential for reverse osmosis membranes, but also high osmotic pressure. A spiral wound reverse osmosis functioned well when the wastewater was pre-treated, and it produced good quality water with respect to all the other studied parameters except the chemical oxygen demand. However, chemical oxygen demand rejection was 99% since the concentration in the wastewater was originally very high.

Key words: cross-rotational, food industry, fouling, membrane filtration, wastewater, water reuse

HIGHLIGHTS

• A membrane-based concept was successfully applied for water reuse from wastewater.
• Cross-rotational ultrafiltration technology enabled chemical-free pre-treatment for wastewater.
• Spiral wound reverse osmosis achieved good filtration performance with developed pre-treatment.
• Techno-economic evaluation is available for the functional concept.
**INTRODUCTION**

Wastewater reuse has raised interest in industry since it is a dual-benefit strategy involving increased safety and sustainability by removing impurities from discharged water and enabling new sources of water for industrial operations. This is important especially in water-scarce areas, alleviating pressures on groundwater and other freshwater bodies. The food sector has one of the highest consumptions of water, mainly due to continuous cooling and heating systems and cleaning processes, and it is one of the largest effluent producers per unit of production (Vourch et al. 2008; Simate et al. 2011; Suarez et al. 2014). Hence, intense pressure on water resources has led to efforts to reuse reclaimed processing wastewater in the food industry. Permeates and concentrates can both be suitably reused, which may reduce the intake of raw water and provide savings on raw water processing costs (Chen et al. 2019). The agro-food industry requires water widely for irrigation with lower water quality requirement than that of drinking water, which is required for food processing itself (Quist-Jensen et al. 2015). However, water quality also matters in irrigation. For example, when using low quality irrigation water it is necessary to apply extra water to prevent salt accumulation in the soil, which can have a negative effect on the crops (Letey et al. 2011; Quist-Jensen et al. 2015).

Wastewaters from the food industry are characterised by high organic matter (OM), suspended solids (SS), salts, oils, fats, and cleaning agents if they are not previously segregated, along with seasonal changes in flow and composition (Suarez et al. 2014). A range of biological and physico-chemical methods have been investigated for the treatment of wastewater from food industry wastewater before discharge into the aquatic environment. A combination of biological methods, such as digestion and composting, is a popular choice for wastewater treatment (Rais & Sheoran 2015). Membrane separation has been applied in the food and chemical industry for several decades, but its application in some areas such as the sugar industry is still under research (Rafik et al. 2015).

Nanofiltration (NF) and reverse osmosis (RO) are efficient technologies for the separation of dissolved components present in food processing streams, such as impurities in wastewaters. RO is a process that ensures the highest water quality, especially regarding desalination. With NF as high as 98.7%, the reduction of chemical oxygen demand (COD) has been achieved for wastewater from a confectionery plant in the Silesian province (Puszczało & Marszalek 2019). A major inefficiency of membrane processes is fouling, which causes a reduction in the RO and NF membrane permeability as a result of the accumulation of colloidal and particulate matter, organic macromolecules (organic fouling), sparingly soluble inorganic compounds (inorganic fouling, scaling), and microorganisms (biofouling) on the membranes (Tang et al. 2011; Yu et al. 2020). Membranes are sensitive; for example, to oil, chlorine (Cl₂), aluminium (Al), iron (Fe) and manganese (Mn). Membrane scaling is also a critical challenge in membrane filtration, particularly for applications involving scaling precursors at high concentrations, such as calcium (Ca) and sulphate (SO₄²⁻) (Wang et al. 2016; Kyllönen et al. 2017). Membrane fouling and scaling can lead to reduced productivity, deteriorated permeate quality, increased energy consumption and treatment costs, as well as shorter membrane life spans (Tang et al. 2011).
In the water reuse concept, pre-treatment is carried out to help the final stage of reuse water production by removing substances that are detrimental to RO or NF membranes. Control of the dissolved components may require different technologies, depending on how they impact RO membranes (de Oliveira & Schneider 2019). The first step in reuse water production is usually SS separation from dissolved components and water. Traditional coarse filtration equipment, such as belt filtration, sand filters or dissolved air flotation (Zouboulis et al. 2007; Šereš et al. 2015) are used for the removal of particular impurities. These technologies can be aided by coagulation and flocculation enabling particles to form larger-size clusters, facilitating faster and better SS removal (Fritzmann et al. 2007; Aderibigbe et al. 2017). Less is known about the removal of suspended solids as well as assistance of flocculation for the wastewater of the confectionery industry.

The water quality after coarse filtration is not as good as the quality after tighter filters, such as microfiltration (MF) or ultrafiltration (UF) (Prihasto et al. 2009). Ninety-nine percent turbidity removal has been achieved for wastewater from a sugar refinery by means of a tubular alumina ceramic MF membrane with a pore diameter of 0.2 μm (Šereš et al. 2015). In the fruit juice processing industry, MF is used to remove SS from solutions, such as fibrous concentrated pulp, but also fat and high molecular weight proteins, and spoilage micro-organisms (Bhattacharjee et al. 2017). Apart from SS, these components should be removed prior to NF or RO in the production of reused water. In the dairy industry, MF is used to clarify cheese whey, as well as to de-fat and reduce the microbial load of milk. UF instead can be utilized to fractionate milk for cheese generation; the concentrate contains proteins, fat and certain insoluble and bound salts, while the permeate contains lactose and solvent salts (Bhattacharjee et al. 2017).

Although MF and UF have been found to be suitable for removing impurities in the food industry, the technologies are characterized by significant membrane fouling, which limits their applicability (Rais & Sheoran 2015). Apart from the feed water quality, membrane characteristics and filtration circumstances, such as the crossflow velocity, pressure, temperature, and the pH, affect fouling. A potential method to reduce fouling is to increase the shear on the membrane by means of rotation (Lee & Lueptow 2003; Kyllönen et al. 2012; Zhu et al. 2016), or ultrasonic cavitation (Kyllönen et al. 2005; Heikkinen et al. 2017). Kyllönen et al. (2012) observed that shear-induced rotational UF-filtration produced high fluxes and appropriate quality water for reuse in paper making processes. Only a little membrane fouling was observed. Huuhilo et al. (2001) used the similar type of cross rotational UF filter and found the higher the rotational frequency the less fouling occurred. In order to limit the formation of a fouling layer, a rotating disk module (RDM) was employed for the dead-end UF of juice from electroporated sugar beet (Zhu et al. 2015). The permeate flux increased when increasing the rotation speed. RDM-assisted crossflow UF was studied in full recycling and concentration modes with sugar beet juice, obtained by cold pulsed electric field-assisted pressing. A flux decline in crossflow filtration with no assistance was observed indicating the need for RDM for effective anti-fouling technology (Zhu et al. 2016). Less is known about rotation-assisted UF as a pre-treatment for challenging wastewater that contains high concentrations of suspended solids and sugar compounds as well as ingredients for gelation.

In this research, the pre-treatment of wastewater from a candy factory for RO was investigated targeting the reuse water concept applicable to the confectionery industry.

**METHODS**

**Wastewater and analysis**

A candy factory sent two batches of original wastewater for the studies. The batches were treated individually so that the first batch was used to build up the concept and the second batch was used to execute the best concept in series. The same batch was always used when technologies for the separation steps were compared. The wastewater contained all the ingredients possible in candies such as sugars, jellies, starch, essences and dyes. pH of the wastewater was not changed in the study since it was expected to be a large expense in industrial scale.

The wastewater was characterized by means of pH, conductivity, particle size distribution, dry solids content (DS), suspended solids (SS), turbidity, chemical oxygen demand (COD), total nitrogen (N-total), total phosphorous (P-total), Brix degree, polyphenols, proteins, viscosity, osmotic pressure, and elementary analysis. An elementary analysis included calcium (Ca), sodium (Na), potassium (K), sulphur (S), aluminium (Al), manganese (Mn), ferrous (Fe) phosphorous (P), strontium (Sr), barium (Ba), chromium (Cr), copper (Cu), nickel (Ni), zinc (Zn), lead (Pb), arsenic (As), cadmium (Cd), antimony (Sb), mercury (Hg), and boron (B). The same analyses were carried out for the feeds of each filtration stage, as well as for permeates and concentrates of the filtration stages when applicable.
The pH and conductivity were measured using standard handheld meters, VWR pH 100 for pH and VWR EC 300 for conductivity. The DS was analysed by drying an unfiltered sample in a glass beaker at 105 °C using standard procedure SFS 3008, and the SS was analysed using standard procedure SFS-EN 872 with Whatman ME25 (0.45 μm) filter papers. The COD was analysed using a DR3900 spectrophotometer with LCK314 (COD 15–150 mg/l), LCK114 (150–1000 mg/l) or LCK014 (COD 1,000–10,000 mg/l). The N-total was analysed using a DR3900 spectrophotometer with LCK158 (LATON total N 1–16 mg/l) and the P-total was analysed using a DR3900 spectrophotometer with LCK350 (PO4-P 2–20 mg/l). Phenols were determined using a DR3900 spectrophotometer with LCK345 (phenols 0.05–5 mg/l) or LCK346 (phenols 5–200 mg/l). The Brix degree was analysed using an ATAGO Master-10 α refractometer (0.0–10%), and proteins were analysed with a Bio-Rad DC Protein Assay (similar to Lowry assay, max. 1.42 g/l). The osmotic pressure was measured using a Vapro 5600XR Vapor Pressure Osmometer (2.4–24 bar), and the viscosity was measured using a Brookfield LVDV-II + Pro Extra Rotator Viscosimeter (1.5–30,000 cP).

An elementary analysis was conducted for the feed from a nitrohydrochloric acid extraction carried out using SFS-EN 13650, SFS-EN 13657 and SFS-EN ISO 15587-1 methods and analysed as permeates. The permeates were analysed using inductively coupled plasma optical emission spectrometry (ICP-OES), wherein the procedure was carried out using standard SFS-EN ISO 11885, or inductively coupled plasma mass spectrometry (ICP-MS) using standard SFS-EN ISO 17294-2. The particle size distribution of the feed samples was determined using a Malvern Mastersizer 2000 analyser, with a measurement range from 0.02 to 2000 μm. The turbidity was measured using an HACH 2100 AN IS Turbidimeter.

**Suspended solids removal**

Microfiltration (MF) for SS removal included various filters with various pore sizes; 0.2 μm MV020 (Microdyn Nadir, Germany), 5 μm Millipore LS, 12–25 μm Schelicher & Schuell 589 Black ribbon, and metallic 400 mesh with 37 μm openings (Karanor Oy, Finland). MF was carried out in a Millipore filtration cell with a maximum batch volume of 300 ml. The results from these experiments were verified at larger scale using 5 and 10 μm filter bags in a HiFlux TWP-4250 housing.

Coagulation and flocculation chemicals were used to form larger-size clusters of particles in wastewater, facilitating faster and better removal of SS during settling and filtration. Coagulation and flocculation experiments either alone or in combination were carried out in one litre jars using Kemira Flocculator 2000 equipment with programmable sequencing for fast mixing, slow mixing, flocc formation, and settling. Femnolocs F105 and K5060 as well as Superflocs C492 and C1592RS were tested using the dosages of 56.5 ppm Fe³⁺, i.e. a 500 ppm product for F105, and a 10 ppm active compound added as 0.1% solution for the others. Aluminium coagulants were not studied since the low feed pH was out of the optimum range for aluminium based coagulants. Instead, the feed pH favoured the usage of iron coagulants. Settling was also carried out in a measuring glass to check the applicability of clarification for subsequent MF and UF.

**Membrane filtration**

The best surfactant and dosage for viscosity reduction was determined by preliminary tests. Based on the results, the surfactant applied for membrane filtration was sodium dodecylbenzenesulfonate (DBS) at 140 mg/l dose. The MF permeate was treated with surfactant for UF, while the UF permeate was treated with surfactant for RO.

UF membranes UP150, UP020, UP005 and NP010 (Microdyn-Nadir, Germany) with pore sizes of 150, 20, 5 and 1 kDa, respectively were selected for Millipore filtrations, where the suitable pore size of the membrane was investigated for a pre-treatment of NF or RO. pH range for the used UF membranes are 0–14. Based on these tests, a 150 kDa membrane UP150 was selected for pre-treatment. UP150 was applied both in a 2.5′′ pressure vessel with a spiral wound 2540 element and a maximum pressure of 40 bar and a Valmet Ultrafilter CR (Figure 1) 250 filter (Valmet, Finland) with a maximum pressure of 10 bar. The cross-rotational filter created a high crossflow and turbulence on the membrane surface with special rotors leading to fouling reduction. In the Valmet Ultrafilter CR 250 test unit there were two membranes with a total filtration area of 0.09 m² and between the membranes a rotor operated at 1000 rpm. The used rotation speed is down-scaled from the industrial scale equipment. It was earlier tested and now required by the manufacturer.

The NF and RO experiments were conducted in a SEPA CF plate-and-frame crossflow laboratory filtration unit with a membrane area of 140 cm² and a maximum operating pressure of 1,000 psi (69 bar) allowing the high pressure needed for RO. The membranes used were an NF90 (Dow Filmtec, USA) with an MWCO of ~400 Da and a BW30LE (Dow Filmtec, USA), with an MWCO of ~100 Da. pH range for the NF/RO membranes is 2–11. The membrane flat sheets were rinsed before filtration with deionized water and stored overnight at 4 °C in a glass bottle filled with deionized water. The salt
rejection at a 2000 ppm concentration was determined based on conductivity measurements of permeate at pH 8 and with a 15% recovery using sodium chloride (NaCl). Filtrations were carried out at 25 ± 1 °C and a feed flow of 6.5 ± 0.1 L/min, which generated a cross flow velocity of 0.6 m/s. RO filtrations of the pre-treated wastewater were verified using 2,540 spiral wound element with a maximum pressure of 40 bar. All research items regarding all process steps are shown in the scheme of the study (Figure 2).

Techno-economic evaluation (TEE)
The techno-economic evaluation (TEE) of the investment for an industrial process studied here included the evaluation of capital costs (CAPEX), annual operating expenses (OPEX), revenues and profits. CAPEX can be roughly divided into the sum of the fixed capital investment (FCI) and working capital investment (WCI) (Sinnott 1999). Typical values for the WCI of, for example, chemical plants are between 10 and 20% of the FCI. CAPEX costs were estimated based on the experimental data and with percentages and shares from literature (Peters & Timmerhaus 1991).

OPEX was grouped into direct manufacturing costs (DMC), fixed manufacturing costs (FMC) and general expenses (GE) and was calculated as a sum of those. Table 1 shows the types of cost items as grouped into these categories (Turton et al. 2012). Unit capacity in this project was 3,500 L/h (84 m³/day). No costs were estimated for buildings, land, site extensions or wastes produced. Estimation for energy consumption was calculated based on the study.

Figure 1 | Schematic figure of the Valmet Ultrafilter CR by courtesy of the manufacturer (Rosenberg et al. 2019).

Figure 2 | Scheme of the study.
In TEE, parallel to the mass balance of inputs and outputs, market prices of energy, materials, labour costs are additionally required. Different sources were used to retrieve information concerning the market prices (Table 2).

In order to carry out a CAPEX estimation in the TEE, price information on the system components used (machines, equipment, etc.) was needed (Table 3).

CAPEX is the sum of the fixed capital investment (FCI) and working capital investment (WCI). In the calculations, the value of Total FCI (TFCI), which is the sum of the total direct costs and total indirect costs, have been used. In WCI calculations, value of 10% has been used. Thus, WCI is of 10% of FCI.

**RESULTS AND DISCUSSION**

**Suspended solids removal**

The candy factory wastewater contained a lot of SS and DS. The SS concentration of the wastewater was 4.4 g/L, while the DS concentration was 69 g/L. The average particle size of the wastewater was 41 μm but it also contained small particles starting from 1 μm (Figure 3).

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**Table 1 | Cost items as grouped into OPEX categories**

| Direct manufacturing costs (DMC) | Price or calculation |
|----------------------------------|----------------------|
| CWF – Wastewater as feedstock    | No price             |
| CRM – Other raw materials        | No price             |
| CUL – Utilities                  | Actual prices        |
| COL – Operating labour           | 960 h/year (320 days; 3 hours/day) |
| CDS – Direct supervisory and clerical labour | 0.18C_{OL} |
| CML – Maintenance and repairs    | 0.02FCI              |
| COS – Operating supplies         | 0.003FCI             |
| CLE – Laboratory charges         | 0.15C_{OL}           |
| CPH – Patents and royalties      | 0.03OPEX             |

Fixed manufacturing costs (FMC)

| C_{D} – Depreciation           | 0.067FCI |
| C_{LT} – Local taxes and insurances | 0.032FCI |
| C_{PO} – Plant overhead costs  | 0.708C_{OL} + 0.036FCI |

General expenses (GE)

| C_{AD} – Administration costs | 0.177C_{OL} + 0.003FCI |
| C_{DI} – Distribution and selling costs | 0.11OPEX |
| C_{RD} – Research and development | 0.05OPEX |

*OPEX can be determined when the bold costs together with FCI are known or can be estimated (Turton et al. 2012).

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**Table 2 | Prices and costs used in the study**

| Input                  | Price/costs (in €) | Source          |
|------------------------|--------------------|-----------------|
| Electricity            | 0.13               | EUROSTAT 2021   |
| Operating labour, average hour wage in EU | 28.5 | EUROSTAT 2020 |
| Waste water            | 0                  |                 |
| Membrane cleaning chemical | 9,200         |                 |
| Membranes              | 5,600              |                 |
| Output                 | Water              | 0               |
| Sugar and sugar components | 0               |                 |

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Settling with the first batch wastewater was done to check the possibility of clarification as a very first pre-treatment step for membrane filtration. The turbidity of the solution in settling could be clearly reduced, from 7500 NTU to 150 NTU in supernatant, but only about 20% of clear solution was obtained after 1 hour settling in measuring glass tests, after which settling become even slower (Figure 4). Therefore, settling cannot be used for total SS recovery on its own, but can be considered a part of the solid/liquid separation process when circumstances allow a long enough settling time. In this research, settling was not used as an additional step in pre-treatment. Centrifugation, which is also a possible technology, was not studied here.

Since the wastewater contained small size particles, flocculation was studied to improve the settling or filtration of SS. However, with the Fennoflocs F105 and K5060 as well as the Superflocs C-492 and C1592RS, only minor flocculation was seen, and the decrease of turbidity and the improvement of settling in the flocculator remained negligible. Hence, subsequent suspended solids removal was carried out with no assistance from flocculation chemicals.

Filtration was considered as a main pre-treatment technology for pure water production with either NF or RO. The turbidity results of lab scale filtrations indicated that MF with a 5 μm filter still produced too much turbidity at 240 NTU for RO filtration where the turbidity is recommended to be less than 1 NTU. An absolute 0.2 μm filter removed turbidity ending in 3.4 NTU, which was close to the value needed for NF and RO. Larger-scale experiments with a second batch of wastewater verified that filter bags of 5 and 10 μm produced permeates with too much turbidity. The turbidity results on a larger scale were even worse than on the lab scale, producing filtrates with a turbidity of 1900 NTU and 1930 NTU, respectively.

**Table 3** | CAPEX estimation of the filtration system

| System component                                           | Price/costs (in €) | Process step |
|------------------------------------------------------------|-------------------|--------------|
| Valmet Ultrafilter CR CR1010/30                             | 400,000           | UF           |
| RO system with SW30XLE-400i membrane elements               | 100,000           | RO           |
| Four pieces of membrane elements (37 m²)                   | 24,000            | RO           |

**Figure 3** | Particle size distribution of the candy factory wastewater.
Ultrafiltration
Since wastewater contained impurities with a large molecular weight such as proteins and gelation-making substances, laboratory-scale UF filtration tests with Millipore equipment were conducted to determine the pore size needed to supplement impurity removal for NF and RO. Apart from fouling reduction in NF and RO, sugars could be made purer in the concentrate of NF and RO. The rejection of proteins, and the N-total and P-total, increased when decreasing the pore size starting from 150 kDa and ending at 1 kDa. Proteins were rejected by the membrane with pore sizes of 150 kDa 66%, 20 kDa 85%, 5 kDa 92% and 1 kDa 92%. Additionally, no effect was seen on conductivity of the UF permeates and minor effects were noted in COD and Brix concentrations, meaning that salt and sugars were not rejected by UF, which was an expected result. Based on the quality results, no clear decision on the pore size of the pre-treatment could be made. Instead, the flux up to WR 80% with a UP150 membrane was clearly higher than with the other studied UF membranes. The permeability was double with the UP150 than with the UP020. Hence, based on the flux results supported by quality results, UP150 with a pore size of 150 kDa was selected for the pre-treatment method in the subsequent conceptual design.

UF with 150 kDa membrane was applied on a larger scale to study the fluxes in two different element constructions, and to verify the quality results obtained at the laboratory scale. A spiral wound element with a 150 kDa membrane produced unacceptable fluxes as a workable pre-treatment method. Instead, a cross-rotational UF system using the same 150 kDa membrane performed very well (Figure 5, left). The flux of the cross-rotational UF was further improved by pre-filtering the wastewater with a 5 μm cartridge filter. It is noteworthy that the UF produced a concentrate that was jelly-like in both cases, suggesting the rejection of gelation-making compounds into the concentrate (Figure 5, right). The cross-rotational filter created high crossflow and turbulence on the membrane surface with special rotors causing fouling reduction, and the jelly created less problems with cross-rotation during concentration than with crossflow only.

Viscosity decrease
The viscosity of the wastewater itself was 2.1 cP, which was double the value of the viscosity of water measured by a Brookfield LVDV-II + pro Extra rotator Viscometer. The removal of SS using MF produced a 19% and UF 27% decrease in viscosity. When an SDBS surfactant was added to the MF permeate or to the UF permeate in order to help the subsequent UF or RO, the viscosity decreased further by 16 and 2%, respectively. The UF concentrate was jelly-like, and it was not possible to measure the high viscosity reliably using similar measurement methods as used for wastewater. The measurement gave only a value of 8.4 cP. Although the surfactant reduced the viscosity of the feeds of UF and RO, the result was not as expected when studying the effect of surfactant in filtration. The flux was not improved when concentrating but the surfactant seemed to act as a foulant. The SDBS reduced 40% of UF flux when using a 150 kDa membrane (Figure 6, left) but did not affect rejections (N, P, protein, COD, polyphenols, Brix). A similar flux decrease using SDBS in the UF permeate; that is, the
feed to RO, was obtained when using RO with a BW30LE membrane (Figure 6, right). The significance of surfactant on fouling was not further investigated since the use was discarded from the water purification concept.

**Nanofiltration and reverse osmosis**

After cross-rotational UF using a 150 kDa membrane, NF and RO were examined for sugar concentration using an SEPA CF unit. NF90 produced performance results as good as BW30LE (Figure 7, left). In both cases, the flux decreased linearly during concentration mainly due to the osmotic pressure increase in the concentrate. The high sugars content gave the wastewater high COD and Brix values but also a high osmotic pressure, varying from 5.7 to 9.2 bar. Thus, concentration to the target recovery WR > 70% required increasing the hydraulic pressure first at WR 30% from 20 bar to 30 bar and then at WR 66% to 40 bar. This was close to the maximum operational pressure for the NF90 and BW30 membranes. Fluxes at WR 70% were still good, 10 LMH. Apart from the fluxes, the rejections were pretty similar using NF and RO technologies. However RO led to slightly lower permeate conductivity than NF, 210 μS/cm and 310 μS/cm, respectively. Thus, RO was selected for reuse water production with the spiral wound element (Figure 7, right).

**Water qualities**

Cross-rotational UF produced good rejections of turbidity, proteins, polyphenolic compounds, and N-total, which were similar results to the laboratory-scale UF filtrations (Table 4). Additionally, a UF membrane with a pore size of 150 kDa allowed...
**Figure 7** | NF and RO for wastewater purification with Sepa CF module (left) and RO with spiral wound element (right).

**Table 4** | Quality of the feed and the produced permeates with rejections in CR 150 and RO

| Parameter                  | Feed | CR 150 permeate | Rejection (%) | RO permeate | Rejection (%) | Chemical quality/EU |
|----------------------------|------|-----------------|--------------|-------------|--------------|---------------------|
| pH                         | 3.9  | 4.0             | 3.3          | 92.5        | 2.5          |                     |
| Turbidity, NTU             | 3600 | 1.6             | 100          | 95.7        | 0.1          |                     |
| Conductivity, mS/cm        | 2.2  | 2.1             | 4.5          | 0.17        | 91.9         |                     |
| COD, mg/L                  | 77500| 62500           | 19.4         | 180         | 99.7         |                     |
| P-total, mg/L              | 2.6  | 1.1             | 57.7         | 0.1         | 90.9         |                     |
| N-total, mg/L              | 870  | 39              | 95.5         | 0.5         | 98.7         | b                   |
| Proteins, g/L              | 4.0  | 0.8             | 80.0         | 0.0         | 100          |                     |
| Polyphenols, mg/L          | 24   | 2.3             | 90.4         | 0.1         | 95.7         |                     |
| Brix, wt-%                 | 6.6  | 5.7             | 13.6         | 0           | 100          |                     |
| Osmotic pressure, bar      | 5.7  | 5.4             | 5.3          | 0           | 100          |                     |
| Ca, mg/L                   | 82   | 83              | 0            | 2.3         | 97.2         |                     |
| Na, mg/L                   | 590  | 570             | 3.4          | 0           | 96.5         | 200                 |
| K, mg/L                    | 13   | 11              | 15.4         | < 1         | > 91         |                     |
| P, mg/L                    | 4.8  | 2.9             | 39.6         | 0.25        | 91.4         |                     |
| S, mg/L                    | 120  | 110             | 8.3          | 1.9         | 98.3         | c                   |
| Fe, mg/L                   | 2.4  | 2.0             | 16.7         | 0.01        | 99.5         | 0.2                 |
| Al, mg/L                   | 0.8  | 0.91            | 0            | < 0.05      | > 94         | 0.2                 |
| Mn, mg/L                   | 0.05 | 0.05            | < 0.05       | < 0.05      | > 94         | 0.2                 |
| Zn, mg/L                   | 0.29 | 0.35            | 0            | < 0.05      | > 85         |                     |
| Ba, mg/L                   | 0.42 | < 0.05          | 88.1         | < 0.05      | > 96         |                     |
| Sr, mg/L                   | 1.5  | 1.5             | 0            | < 0.05      | > 96         |                     |
| B, mg/L                    | 1.0  | < 0.1           | 90.0         | < 0.1       | < 1          | 1.0                 |
| As, μg/L                   | 2.3  | 2.5             | 0            | < 1         | > 60         | 10                  |
| Cd, μg/L                   | < 1  | < 1             | < 1          | < 1         | < 1          | 5.0                 |
| Cr, μg/L                   | 96   | 31              | 67.7         | < 1         | > 96         | 50                  |
| Cu, μg/L                   | 540  | 420             | 22.2         | < 1         | > 99         | 2.0                 |
| Hg, μg/L                   | < 1  | < 1             | < 1          | < 1         | < 1          | 1.0                 |
| Ni, μg/L                   | 61   | 33              | 45.9         | < 1         | > 97         | 20                  |
| Pb, μg/L                   | 22   | 14              | 36.4         | < 1         | > 93         | 10                  |
| Sb, μg/L                   | 1.5  | < 1             | < 1          | < 1         | < 1          | 5                   |

Limits for the chemical quality of water intended for human consumption in European Union (EUR-Lex 1998).

*Below measurement range.

1NO₃ 50 mg/l, NO₂ 0.5 mg/l.

SO₄ 250 mg/l means S ~83 mg/l.
salts and sugars to pass, which was an expected result. The P-total rejection was 58%. Fe and Al concentrations in the UF permeate were a bit high for the subsequent spiral-wound RO. However, no fouling was seen in short RO filtrations. The Ca concentration was low enough at up to a WR of 70%, thus scaling was not seen. RO produced colourless water having low concentrations of compounds with respect to all the other studied chemical quality parameters (EUR-Lex 1998) except COD. This was only moderate regardless of 99% rejection, since the concentration of the wastewater was originally very high (Table 1, Figure 8). There are no limits for organic compounds in drinking water but the discharge limit for COD in the EU is 125 mg/L (EC 2019). However, purified RO water containing some concentration of sugar compounds could find reuse application in industry.

TEE calculations
DMC, FMC and GE were calculated as 280,000 €, 280,000 € and 120,000 € respectively for the filtration system. The annual OPEX obtained in TEE for the filtration system was 680,000 € in total. Figure 9 shows a detailed breakdown of OPEX.

Total FCI was calculated at € 1.9 million and WCI € 0.19 million, which was 10% of FCI. CAPEX costs were thus estimated as € 2.09 million. The overall picture of CAPEX for the process studied is shown in Figure 10.

CONCLUSIONS
The food sector has one of the highest consumptions of water, mainly due to the continuous cooling, heating, and cleaning processes, and it is one of the largest effluent producers per unit of production. There are tight rules for water quality in the food industry, but high efficiency separation technologies, such as RO, have good chances of producing good quality water for reuse. However, wastewater has a great tendency to foul membranes, making pre-treatment essential for successful water reuse. This study developed a membrane-based reuse water concept for wastewater from the confectionery industry; that is, candy factory wastewater, emphasizing the pre-treatment stage in the concept to reduce fouling. Solutions were studied on the pre-treatment of this challenging industrial wastewater, which contains high concentrations of SS and sugar compounds, measured as the Brix and chemical oxygen demand in this study, as well as ingredients for candy gelation.

The wastewater contained SS, which was difficult to separate from the solution. Flocculation did not aid settling, or filtration. Thus, although settling produced clear supernatant, the time needed for sufficient separation was long. Hence, it was not used as an additional technology in pre-treatment. Typical 5 and 10 μm MF filters were not sufficient for SS removal of the wastewater, but 0.2 μm was needed to produce a clear permeate for subsequent filtration. Since substances for gelation also needed to be removed from the wastewater before NF or RO, UF was studied as a potential pre-treatment technology. A 150 kDa membrane produced the best flux of the studied UF membranes. Using the cross-rotational technology with enhanced fouling prevention for membranes, the jelly-like concentrate was not an issue. Conventional spiral wound UF
produced fluxes that were too low for use in pre-treatment. Additionally, there was no positive effect either for the UF or RO performance if the viscosity of the wastewater was reduced using surfactants.
NF90 produced performance results as good as BW30LE regarding fluxes and permeate qualities when the wastewater was pre-treated as designed. In both cases, the flux decreased linearly during concentration mainly due to the osmotic pressure increase in the concentrate. Concentration to the target recovery WR at >70% required a high hydraulic pressure of 40 bar, which was close to the maximum operational pressure for NF90 and BW30 membranes. Thus, membranes with higher pressure limits, such as sea water membranes, could be considered for the concentration of sugar-containing solutions. RO was selected for spiral wound studies, and it produced good quality water with respect to the other studied parameters except the chemical oxygen demand. This was only moderate regardless of 99% rejection, since the concentration of the wastewater was originally very high.

The accuracy of TEE methodology depends very much on the accuracy of the available inventory data. Incorrect estimates or model assumptions, outdated data and data gaps are sources of uncertainty. Optimization of the filtration process in the future may lead to significant changes in the inventory data and also in the TEE calculation. As already established, no turnover was expected from water or sugar produced nor savings coming from wastewater fees. This led to annual OPEX of €680,000/year. Energy consumption (electricity) was the largest cost factor. CAPEX was estimated at €2.09 million for a system treating 84 m³ water per day. As the process was expected to run in the same industrial park as the wastewater supplier, several CAPEX cost factors could be excluded. Otherwise, the CAPEX would be even higher.

Wastewater from the food industry can be considered a new source of water for industrial operations. If wastewater is required to be treated for reuse, the studied system was considered costly but workable. Permeates and concentrates can be both suitably reused with the fit-for-purpose principle, which may reduce the intake of raw water and provide savings on the processing costs. When the candy factory wastewater is pre-treated using cross-rotational UF, the final stage spiral wound RO can perform well and good quality reuse water can be produced while 99% of sugars are recovered as a concentrate.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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