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## Re-use of waste cotton textile as an ultrafiltration membrane

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### ABSTRACT

Textile industry produces millions of tons of waste annually, which is predominantly incinerated or landfilled. Cotton textile comprises a quarter of total textile production, and although being renewable, its production is highly chemical- and water-intensive, rising the need for effective waste cotton textile recycling. This study presents an investigation whether it is possible to utilize waste cotton textile as a cellulose source for the fabrication of cellulose membranes. The effect of casting thickness and cellulose concentration on the prepared membranes' performance was studied. Membranes cast from 2 wt% casting solutions exhibited the highest permeabilities of 1.11 and 3.09 m<sup>3</sup>/(m<sup>2</sup>·s·Pa) for 300 and 150 μm casting thickness, respectively, but poor adhesion stability and low retention. Membranes cast from solutions of higher concentrations (5, 6, and 7 wt%) resulted in membranes with more stable performance. The permeability values for 300 μm membranes were in the range of 0.27 – 0.39 m<sup>3</sup>/(m<sup>2</sup>·s·Pa) and for 150 μm 0.51 – 0.67 m<sup>3</sup>/(m<sup>2</sup>·s·Pa). The retention values of these six membranes were relatively close to each other, showing 80 – 92% retention of polyethylene glycol (PEG) 35 kDa. Three most promising membranes (5, 6, and 7 wt% cast at 150 μm) were additionally characterized, showing negative zeta potential within –23 – –35 mV range at pH 7 and contact angles of very hydrophilic material (14 – 16°). Overall, the results showed that very hydrophilic ultrafiltration membranes having attractive permeability and retention properties can be made from textile waste. 1 m<sup>2</sup> of cotton bed linen is enough to produce approximately 20 m<sup>2</sup> of cellulose membrane.

### 1. Introduction

The continuous and steady growth of the textile market can be attributed to several reasons. The main reason behind is the growth of population itself [1]. Another reason is shortening of the fashion cycle. The textiles are also becoming cheaper than before compared to the other consumer goods. According to [2], the clothing purchase in EU-28 has increased by 40% in less than twenty years. Even in Finland, where the clothing consumption is smaller compared to other European countries, the number of owned goods has doubled since the 1990s [2]. Among with the growth of the textile industry, the challenges attributed to handling the environmental, energy, and resource related issues arise.

Textile market is currently dominated (63%) by synthetic fibers, which are mainly produced from petrochemicals, generating significant carbon dioxide (CO<sub>2</sub>) emissions. The next widespread is cotton, of which production is associated with water depletion and toxic pollution, due to extensive use of pesticides [2–4]. According to the estimations, the global consumption of cellulosic fibers is going to reach 5.4 kg per capita level by 2030, whereas the forecasted availability is going to comprise only 3.1 kg of cotton per capita [1].

Textile industry is known to be chemical-, water-, and energy-intensive. The global impact of the textile industry was reported by the Ellen MacArthur Foundation in 2017 [5]. According to the forecasts, textile industry will continue to grow, raising the urgent need for

**Abbreviations:** [Emim][OAc], 1 – ethyl – 3 – methylimidazole acetate; ATR, Attenuated total reflection; CA, Contact angle; DI, Deionized (water); DMSO, Dimethylsulfoxide; FTIR, Fourier-Transform Infrared (spectroscopy); IL, Ionic Liquid; MWCO, Molecular Weight Cut-Off; NOM, Natural organic matter; PEG, Polyethylene glycol; TCI, Total Crystalline Index; TOC, Total Organic Carbon.

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efficient recycling as one of the key directions towards sustainable development [1,6]. Currently, 95% of the wasted post-consumer textile is not recycled but either incinerated or landfilled, which causes additional environmental problems. The recycled part is mainly going through down-cycling routines, resulting in product with lower quality, which limits its further utilization [3,4,7].

Due to its' properties, cotton textile is the most prospective material for efficient recycling, as it consists solely of cellulose which can be utilized variously, from reinforced composites to completely different products [8]. Because of cellulose's attractive properties, recycling of waste cotton, which can no longer serve any purpose, into various value-added products with adjustable features and sustainable utilization potential can be considered upcycling [9,10]. One of the possible yet not researched options is production of cellulose membranes which is possible once appropriate solvent medium is found. Dissolution of cellulose in ionic liquids (IL) and IL-cosolvent systems is widely discussed nowadays [11–13]. A favorable choice is the use of 1-ethyl-3-methylimidazole acetate ([Emim][OAc]), the low-corrosive and -toxic IL, mixed with dimethylsulfoxide (DMSO), giving the middle-viscous and -cheap system for preparation of membrane casting solutions [12,14,15]. As resulted membranes consist of cellulose, they can be useful in many applications due to cellulose's hydrophilicity, biocompatibility and relatively good stability. The hydrophilicity makes the cellulose membrane to be a feasible option for instance for treatment of different water streams in the pulp and paper industry and biorefinery streams [16–19].

Literature research showed that so far there is only one study existing, which reports the preparation of membranes from waste textile resources. In that study the IL-extracted keratin from wool was used in the fabrication of blended nanofiltration membranes [20]. However, to the best of our knowledge there seems to be no papers reporting a preparation of membranes directly from the cotton textile. Thus, in this study the aim was to investigate the possibility to make a filtration membrane from wasted cotton textile. The flat sheet membranes were prepared via wet phase inversion method from 100% cotton bed linen dissolved in the mixture of [Emim][OAc]-DMSO using the procedure developed and presented in our previously reported study [15]. The usability of the produced membranes was tested by measuring the water permeability and retention of polyethylene glycols (PEGs). In addition, hydrophilicity, zeta potential, membrane porosity, and chemical structure of the fabricated membranes were characterized.

## 2. Materials and methods

### 2.1. Materials

The non-dyed bed sheet labeled as 100% cotton was purchased from a local supermarket and used for preparation of casting solution and membrane fabrication. It was preliminary cut in approximately to  $1.5 \times 1.5$  cm pieces and used directly without any pretreatment. Membrane casting solutions were prepared using the mixture of ionic liquid – 1-ethyl-3-methylimidazolium acetate, 95% ([Emim][OAc], CAS # 143314–17–4, Iolitec Ionic Liquids Technologies GmbH) and DMSO (CAS # 67–68–5, Merck KGaA, Darmstadt, Germany). Flat-sheet membranes were cast on nonwoven polypropylene/polyethylene carrier material Viledon® Novatexx 2484 ( $60 \text{ L}/(\text{s}\cdot\text{m}^2)$  air permeability,  $85 \text{ g}/\text{m}^2$  weight per unit area,  $300/200 \text{ N}/5 \text{ cm}$  maximum tensile force along/across, 25/30% elongation at maximum tensile force along/across, and 0.12 mm thickness, Freudenberg, Germany).

Ultra-pure deionized (DI,  $15 \text{ M}\Omega$ ,  $0.5 - 1 \mu\text{S}/\text{cm}$ ) water was produced by CENTRA-R 60/120 system (Elga purification system, Veolia Water, UK). DI water was used in washing of the membranes, in measuring the membrane permeability, and as a non-solvent in membrane manufacturing as well as in preparation of all solutions.

In the measurement of the retention of the manufactured membranes, polyethylene glycol (PEG) of two different molecular weights were used as a model compound. The PEGs (PEG 20, approximate  $M_w$

20,000 g/mol and PEG 35, approximate  $M_w$  35,000 g/mol) were purchased from Merck KGaA (Darmstadt, Germany; CAS # 25322–68–3).

### 2.2. Methods

#### 2.2.1. Membrane preparation

Homogeneous solutions of cellulose in [Emim][OAc]-DMSO were prepared with different concentrations of 2, 5, 6, and 7 wt% by stirring cotton textile shreds overnight at constant heating in oil bath at  $70^\circ\text{C}$ . As a result of dissolution, visually transparent homogeneous solutions were formed as shown in Fig. S1.

The prepared solutions were cast at room temperature on the carrier material placed on a glass plate, by spreading an appropriate amount with Automatic Film Applicator L (BYK-Gardner USA) with casting knife (with casting thickness 150 or 300  $\mu\text{m}$ ) at 50 mm/s speed. Eight different types of membranes were cast, as two parameters, casting solution concentration and casting thickness, varied. For future convenience, Table 1 summarizes the casting parameters variations and gives codes for the membrane types produced.

Without a preceding dry phase inversion in the air, the films were immediately immersed in the coagulation bath of DI water at  $0^\circ\text{C}$  temperature. In order to guarantee a complete phase separation, the membranes were kept in the coagulation bath overnight. After that, they were washed with water and used for analyses without drying if not stated otherwise.

#### 2.2.2. Membrane permeability and retention measurements

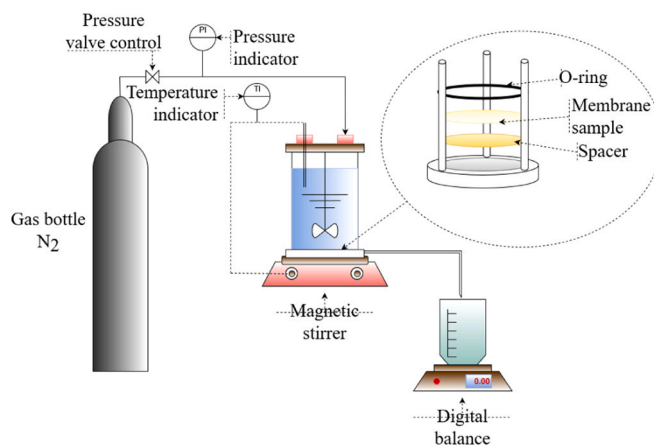
Permeability and retention of the prepared membranes were measured with the Amicon dead-end stirring cell equipment (Millipore, USA, Cat No.: XFUF07611, diameter of the stirring device 60 mm). The circular membrane sample with effective filtration area of  $0.0040 \text{ m}^2$  was cut and placed in the Amicon filtration cell (Fig. 1).

Prior to the filtration experiments, each membrane was compacted for 60 s at 100 kPa, 120 s at 200 kPa, 180 s at 300 kPa, 240 s at 400 kPa, and 1200 s at 500 kPa. This also ensured that the solvents used in the membrane manufacturing were completely rinsed from the membrane pores. Solvents' removal was checked through the measurement of total organic carbon (TOC) content in a permeate sample collected during the

**Table 1**

Casting parameters combinations and assigned membrane codes.

Casting thickness, $\mu\text{m}$	Cellulose concentration in casting solution, wt.-%			
	2	5	6	7
150	2/150	5/150	6/150	7/150
300	2/300	5/300	6/300	7/300



**Fig. 1.** Schematic configuration of Amicon dead-end filtration system.

membrane compaction at maximum pressure of 500 kPa.

Pure water permeability of the membranes was measured at  $25 \pm 0.5$  °C and determined as a slope of plotted four flux values, measured at 100, 200, 300, and 400 kPa pressure and calculated using the Eq. (1):

$$J = \frac{Q_p/1000}{A \cdot \tau}$$

where  $J$  – tested membrane's flux ( $\text{m}^3/\text{m}^2 \cdot \text{s}$ ),  $Q_p$  – the gravimetric flow of water permeate through the membrane (g/s),  $A$  – the area of membrane sample ( $\text{m}^2$ ),  $\tau$  – the time of collection of the permeate (s).

To demonstrate the differences between the different membrane samples also membrane hydraulic resistance values ( $R_m$ ) were calculated at different transmembrane pressures from the slope of pure water flux versus transmembrane pressure difference Eq. (2):

$$R_m = \frac{\Delta P_T}{J \cdot \mu}$$

where  $R_m$  – the hydraulic resistance of the membrane sample (1/m),  $\Delta P_T$  – the transmembrane pressure (Pa),  $J$  – the pure water flux ( $\text{m}^3/\text{m}^2 \cdot \text{s}$ ), and  $\mu$  is the dynamic viscosity of water at 25 °C ( $8.90 \cdot 10^{-4}$  Pa·s) [21].

For the retention study two model solutions of PEG 20 and PEG 35 were prepared with concentration of 300 ppm and filtered through the membrane at a pressure that was set for each membrane individually in order to have approximately same model solutions' flux around  $0.83 \text{ m}^3/(\text{m}^2 \cdot \text{s})$ . Throughout all the measurements, the stirring speed was maintained at 300 rpm using the magnetic stirrer with a rpm indicator and the temperature was kept at  $25 \pm 0.5$  °C. The samples of feed, retentate, and permeate were collected and analyzed for TOC content with a Shimadzu TOC analyzer (TOC-L series, Japan). Membrane retentions were calculated out of measured TOC content in the samples with the Eq. (3):

$$R = \left(1 - \frac{2 \cdot C_p}{C_f + C_r}\right) \cdot 100$$

where  $C_p$ ,  $C_f$  and  $C_r$  are the total organic carbon concentrations in the permeate, feed and retentate (mg/L) respectively.

### 2.2.3. Examination of hydrophilicity of the membranes

For the assessment of membranes hydrophilicity static contact angle (CA) of selected membranes was measured based on the captive bubble method. Nearly 3–4  $\mu\text{L}$  of air bubble volume was placed by the means of U-shaped needle on the surface of the tested membrane attached to a piece of glass with double sided tape and submerged into DI water at the room temperature. For each membrane sample six independent measurements of CA were made at different points with the average value of recorded data taken as final CA. The CA was measured with KSV CAM 101 equipment (KSV Instruments Ltd., Finland) connected to a CCD camera (DMK 21F04, The Imaging Source Europe GmbH, Bremen, Germany). To determine the CA, the obtained images were treated by curve fitting analysis with CAM 2008 software.

### 2.2.4. Examination of the zeta potential of the membranes

The streaming potential which characterizes the charge of the membrane surface was measured with the SurPASS Electrokinetic Analyzer (Anton Paar GmbH, Graz, Austria) with an adjustable gap cell method and using 0.001 M KCl solution as a background electrolyte. The membranes were preliminary stored at approximately 5 °C for 24 h in a fridge. Before the start of the experiment the solution pH was shifted to 7.5 by addition of 0.1 M KOH solution and then automatically titrated to 2.7 with use of 0.05 M HCl solution as the analysis was carried on. The final value of the zeta potential was calculated automatically by SurPASS software based on the Helmholtz-Smoluchowski equation.

### 2.2.5. Examination of chemical structure of the membranes

To identify the functional groups presented and assess changes in crystallinity, which occurred through solution preparation and

membrane formation, the FTIR analysis was performed with the Frontier MIR/FIR Spectrometer (PerkinElmer Inc.) with the universal diamond crystal ATR module in the range  $400 - 4000 \text{ cm}^{-1}$  of wave number with the spectra resolution of  $4 \text{ cm}^{-1}$ . Air-dried samples of selected membranes were recorded in pentaplicate and averaged. For the graphical representation all the spectra were processed with ATR correction, baseline correction and normalization. The ratio of the not-normalized absorption bands  $A_{1428}/A_{897}$  was used to calculate Lateral Order Index (LOI), as was proposed by Nelson and O'Connor [22,23].

### 2.2.6. Estimation of membrane porosity

Membrane porosity ( $\varepsilon$ ) was determined via gravimetric method with following Eq. (4):

$$\varepsilon = \frac{\frac{(w_w - w_d)}{\rho_H}}{\frac{(w_w - w_d)}{\rho_H} + \frac{w_d}{\rho_c}} \cdot 100$$

where  $\varepsilon$  - the membrane porosity (%),  $w_w$  and  $w_d$  – the weight of wet and dry membrane, respectively (g),  $\rho_H$  – the density of water at 25 °C ( $0.997 \text{ g/cm}^3$ ),  $\rho_c$  – the density of cellulose ( $1.5 \text{ g/cm}^3$ ) [24].

## 3. Results and discussion

### 3.1. Membranes performance

The casting result differed depending on the chosen parameters. Casting solutions with cellulose concentrations of 5, 6, and 7 wt% resulted in formation of uniform film, showing even adhesion to carrier material, whereas 2 wt% membranes showed poor adhesion probably related to the use of too low concentration of casting solutions. However, it was still possible to perform filtration experiments with the membranes cast from 2 wt% solutions. For filtration experiments three samples of each membrane were used, and the results presented in Fig. 2 are the averaged values of pure water permeabilities from individual experiments.

It can be seen that all the membranes cast at 150 and 300  $\mu\text{m}$  show the same trend of permeabilities. With increase of concentration of cellulose in the membrane matrix, the permeability decreases, and the highest values are obtained with the 2/300 and 2/150 membranes. The hydraulic resistances of the membranes cast with a lower cellulose concentration were also clearly lower compared to the membrane cast from the solutions containing higher amounts of cellulose (Fig. 3.). As the cellulose concentration increases in the solvent, the polymer concentration at the nonsolvent interface is higher during precipitation. Consequently, a membrane with lower porosity is obtained [25]. From the values in Table 3 it can be seen that experimental results are in good

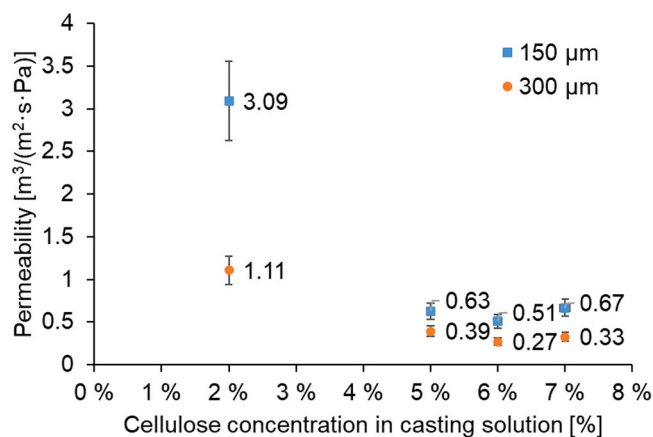
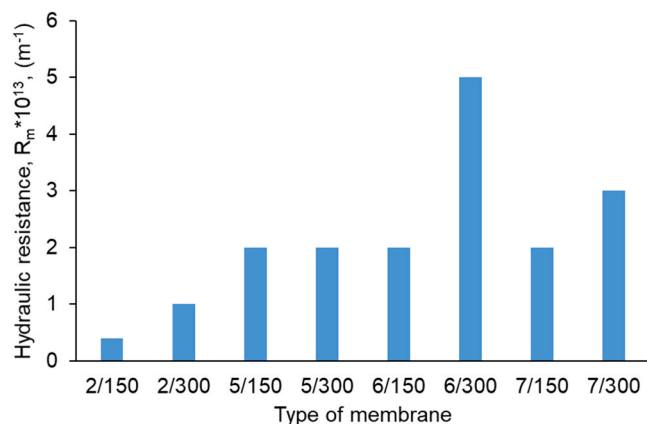


Fig. 2. Pure water permeabilities of the tested membranes all measured in the Amicon ultrafiltration cell at 25 °C and mixing rate approximately 300 rpm.



**Fig. 3.** Membrane hydraulic resistance calculated for each type of membrane tested in the Amicon ultrafiltration cell at 25 °C and mixing rate approximately 300 rpm.

agreement with theoretical assumptions. However, when these values are considered, it has to be taken into account that the method used here to estimate the membrane porosity does not show the porosity given by the pores, which really lead through the membrane. The results also reveal that the membranes cast at 150  $\mu\text{m}$  show noticeably higher permeabilities than the membranes of the same concentrations but cast at 300  $\mu\text{m}$ . This difference can be seen also from the hydraulic resistances of the membranes. The reasons for this might be in the physical structure of the membranes originating both from the different thicknesses of the layers and the possible differences in the macrovoids structures [26].

The retention values measured with PEG 20 and PEG 50 model solutions for all the manufactured membranes are presented in Fig. 4, a and b respectively. The lowest retentions were obtained with the 2/150 and 2/300 membranes. Membranes with the 5, 6, and 7 wt% cellulose concentration showed small difference between values measured for 150- and 300- $\mu\text{m}$  casting thicknesses.

In order to evaluate whether the membranes prepared in this study are competitive compared to the commercial membranes and also

regenerated cellulose membranes prepared by other research groups, their performance was compared to results published by other research groups and to the RC70pp (Alfa Laval) membrane (Fig. 5.). The membranes prepared by other researchers were picked for comparison based on the existence of common parameters with current study, i.e. the same cellulose concentration in casting solution or similar composition of solvent mixture. It is vital to mention that presented comparison serves only as nominal, since the presented combination of cellulose source, solvent system, casting parameters, and performance measurement parameters differs from one study to another, almost never coincides, and can be found in details in the original articles [12,27]. Information about performance of RC70pp membrane was taken from the works that reported it earlier [28,29].

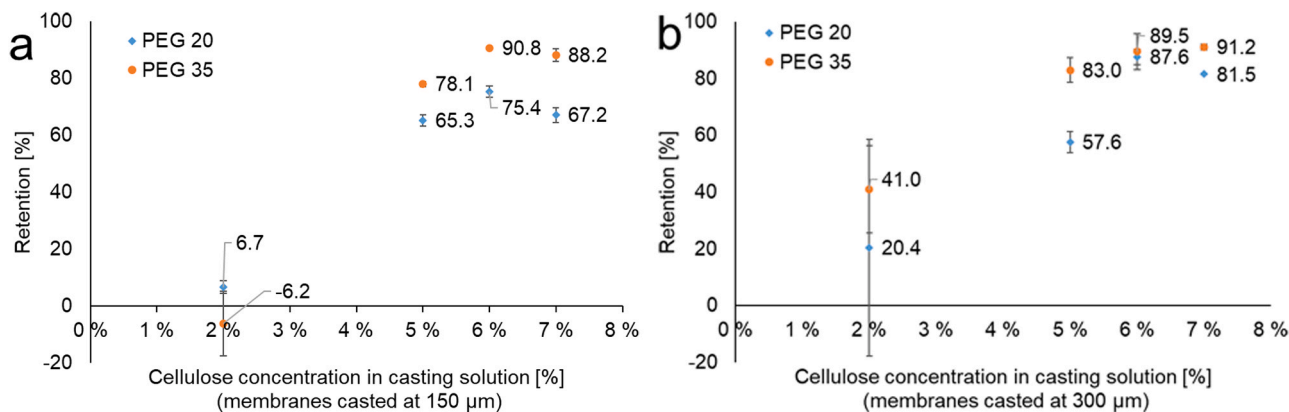
It can be seen from Fig. 5 that the membranes prepared within this study from the waste cotton show performance that is close to both lab-made membranes. Membrane performance depends mostly on the membrane morphology, which is in turn dependent on the polymer concentration, casting thickness, etc. [13]. The “Durmaz” membrane in Fig. 5 was formed from casting solution with 8 wt% cellulose in the mixture of [Emim][OAc]-DMSO and it had significantly lower pure water permeability and higher retention compared to the membranes prepared within current study [27]. The “Durmaz” membrane retention was evaluated with Blue Dextran (20 kDa). The “Livazovic” membrane was prepared from 5 wt% cellulose (Avicel pH101 microcrystalline cellulose) solution in pure [Emim][OAc] which had the same pure water permeability as the membranes prepared in this study (6/300 and 7/300 in Fig. 5) but the retention was lower based on the evaluation with PEG molecules [12]. The commercial RC70pp membrane demonstrates more or less the same permeation rate as the membranes prepared within this research, however, the 90% retention was achieved with significantly smaller molecules [28,29].

The comparison presented above proves the suitability of waste cotton textile as a material for membrane preparation as it shows competitive performance to referenced laboratory-made membranes prepared from commercially available cellulose feedstock, like cotton linter from fibers and microcrystalline cellulose [12,27]. Based on the square masses of the used substrate and the final membrane product (7/150), 7 g of cellulose is needed to produce 1  $\text{m}^2$  of membrane. Thus,

**Table 3**  
Membranes and cotton textile characterization.

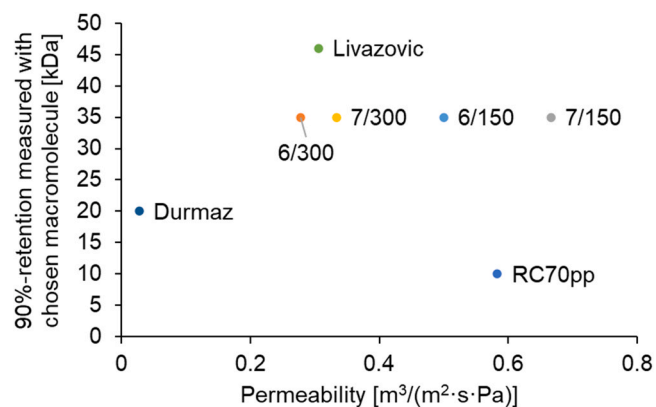
Sample	PWP, $\text{m}^3/(\text{m}^2 \cdot \text{s} \cdot \text{Pa})$	$R_m \cdot 10^{13}$ , $1/\text{m}$	PEG 35 kDa retention, %	Porosity, %	Contact angle, °	Zeta potential at pH 7, mV	LOI $A_{1428}/A_{897}$
Textile	nd	nd	nd	nd	nd	nd	0.86
5/150	0.63	2	78.1	59.2	$15.6 \pm 2.8$	-25	0.60
6/150	0.51	2	90.8	56.3	$16.3 \pm 5.7$	-23	0.47
7/150	0.67	2	88.2	49.6	$14.0 \pm 5.6$	-35	0.50

nd – no data



**Fig. 4.** Retention measurements of membranes cast at 150  $\mu\text{m}$  (a) and 300  $\mu\text{m}$  (b) all measured in the Amicon ultrafiltration cell at 25 °C and mixing rate approximately 300 rpm.





**Fig. 5.** Permeability and the retention values of the membranes prepared within current research and membranes which were chosen for comparison; membranes chosen for comparison are named after first authors Durmaz [27] and Livazovic [12], information about RC70pp was taken from [28,29]; membranes prepared in this research are signed as cellulose wt% concentration in casting solution/casting thickness in  $\mu\text{m}$ .

it can be evaluated that  $1 \text{ m}^2$  of bed linen is enough for production of approximately  $20 \text{ m}^2$  of membrane.

### 3.2. Membrane characterization results

Aiming to assess the changes occurred in the chemical composition and crystallinity of cellulose, as cotton textile was dissolved and the membrane was prepared, FTIR spectra of non-woven carrier material, untreated cotton textile, and two selected membranes prepared in this study are presented in Fig. 6. The spectra were recorded from all the membrane samples, and all of them can be divided into two groups: showing similar peaks as either 5/150 membrane or 2/150 membrane (Fig. 6). The difference seen between the spectra of the membranes made from 2 wt% and 5 wt% cellulose solutions was that in the spectrum of the 2/150 membrane small peaks at  $1710$ ,  $1570$ ,  $870$ , and  $720 \text{ cm}^{-1}$  were present. These peaks do not belong to cellulose and are distinctively present in the spectrum of the carrier material. Same peaks were found in all the  $300 \mu\text{m}$  membranes, which shrank during the drying and from which the cellulose skin layer was partly separated from the carrier material. In the spectra of 5/150, 6/150, and 7/150 membranes (represented as 5/150 membrane spectrum in Fig. 6) the peak at  $1734 \text{ cm}^{-1}$  can be seen. This might be interpreted as a sign of

**Table 2**

General interpretation of FTIR spectra, [35–37].

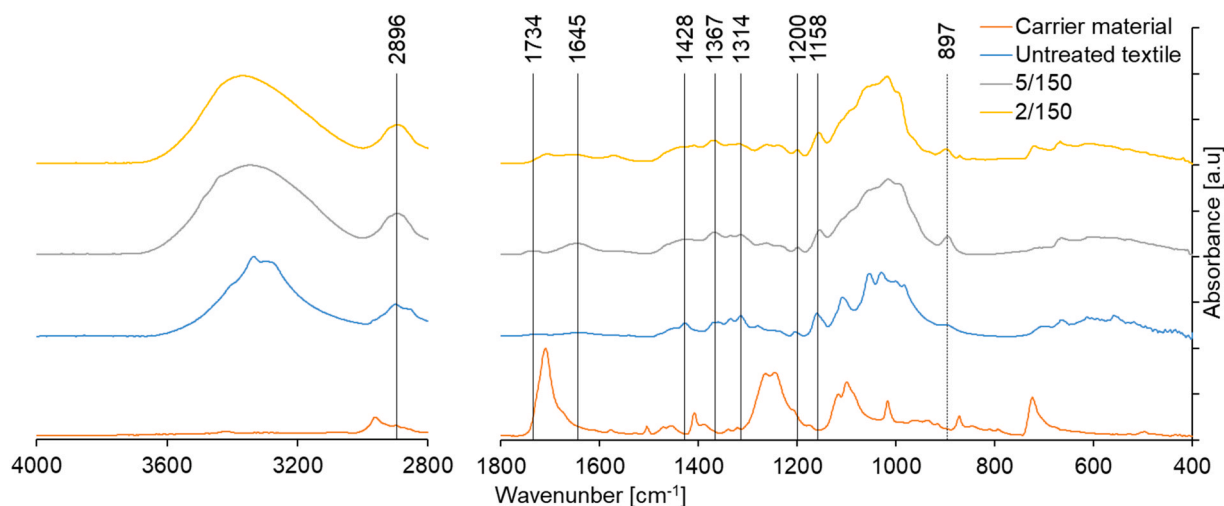
Wavenumber ( $\text{cm}^{-1}$ )	Assignments
3600–3000	OH stretching broad peak
2896	CH symmetric stretching
1734	CO stretching
1645	OH bending/adsorbed $\text{H}_2\text{O}$
1428, 1367	CH bendings
1314	$\text{CH}_2$ wagging
1278 <sup>a</sup>	CH deformation
1158, 1107 <sup>a</sup> , 1054, 1029 <sup>a</sup> ,	asymmetric CO stretchings in C-O-C and C-O-H
983	fragments
660–500	OH out-of-plane bending

<sup>a</sup> peaks are distinctive only in untreated cotton textile.

derivatization side-reaction happening during cellulose dissolution, which is already discussed in a number of works [30–32]. Increased intensity of peak at  $897 \text{ cm}^{-1}$ , which is assigned to the amorphous region in cellulose, is in the agreement with the earlier reported changes in cellulose structure due to dissolution and regeneration processes [33, 34]. Assignments of the cellulose-belonging peaks from the untreated textile and membranes with spectrum as of the 5/150 membrane are described in Table 2.

Considering the membrane performance described earlier, it seems reasonable to pay more attention to membranes cast at  $150 \mu\text{m}$ , as they are offering higher permeability and almost the same retention, than the membranes of the same concentration but cast at  $300 \mu\text{m}$ . Membranes cast from the 2 wt% casting solutions showed high permeability but poor retention, so it was decided to characterize the hydrophilicity and surface zeta potential of the membranes formed from solutions with higher concentrations. Thus, the characterization was done for the membranes cast from the solutions with 5, 6, and 7 wt% concentrations of cellulose at  $150 \mu\text{m}$ . Contact angle and surface zeta potential measurements were performed on undried membranes, whereas for FTIR measurements the membranes were air-dried overnight. LOI was calculated from ATR- and baseline-corrected spectra without normalization.

As has been mentioned earlier, the commercial regenerated cellulose membrane RC70pp (Alfa Laval) was chosen for comparison with the membranes prepared in this study, and its' characteristics were found in other reported works [28,29]. The following values are combined for the RC70pp membrane from these two works: membrane showed  $-30 \text{ mV}$  zeta potential at pH 7,  $0.58 \text{ m}^3/(\text{m}^2 \cdot \text{s} \cdot \text{Pa})$  permeability at 1 bar, molecular weight cut-off (MWCO) of 10 kDa, as announced by Alfa Laval,



**Fig. 6.** FTIR spectra of nonwoven support material, untreated cotton textile, and representative membranes recorded via using the Perkin Elmer Frontier spectrometer with universal ATR module of diamond crystal at a resolution of  $4 \text{ cm}^{-1}$  in the absorbance mode.

membrane hydraulic resistance  $0.25 \cdot 10^{13} \text{ m}^{-1}$ , and contact angle  $11^\circ$ . From the values presented in Table 3, it can be seen, that membranes, prepared within the scope of this study, show lower retention and higher hydraulic resistance than the commercial membrane.

As it was briefly discussed in 3.1 subsection, the increase in cellulose concentration in theory should lead to the formation of membrane with lower porosity. From the values in Table 3 it can be seen that experimental results are in good agreement with theoretical assumptions.

Membrane hydrophilicity affects permeability, retention, and fouling behavior of membranes [38]. The contact angles of membranes cast from 5, 6, and 7 wt% casting solutions are presented in Table 3 and are quite close to each other, demonstrating that all the membranes can be classified as very hydrophilic. Therefore, the hydrophilicity of membranes might not account for any differences in water permeability or retention observed here, and differences exist more likely due to structural changes.

From Table 3 it can be seen that all the tested membranes are negatively charged at pH 7. Based on the FTIR spectra, the reason for this is at least partly the acetylation of hydroxyl groups during the membrane manufacturing process. The C=O stretch can be seen at  $1734 \text{ cm}^{-1}$  (Fig. 6). As TOC results showed no traces of solvents in permeate collected during the membrane compaction step, we might assume that C=O stretch comes from acetylated fragments of cellulose, as was reported previously, and not from the traces of solvents used in the process [31,32].

Permeability of ultrafiltration membranes is dependent on their microcrystalline structure, and highly crystalline membranes usually show lower permeabilities than more amorphous ones [39]. Thus, it is important to understand membrane's crystallinity, and the easiest way of assessment is to calculate the crystallinity index. First suggested by Nelson and O'Connor [22,23], the LOI is still widely used for the assessment of crystalline fraction of cellulose. The LOI is revealed from the ratio at  $1428 \text{ cm}^{-1}$  in-plane symmetric bending, characteristic for cellulose  $I_\beta$  crystal, and C-H deformation in  $\beta$ -glycosidic linkages at  $897 \text{ cm}^{-1}$ , specific for amorphous cellulose regions ( $A_{1428}/A_{897}$ ) [33,40]. From the values presented in Table 3, it can be seen that LOI values for prepared membranes are relatively close to each other. There is no correlation found between LOI and permeability values, however, it seems that LOI values are inversely proportional to retention values showed by these membranes with PEG 35 kDa measurements (i.e., the lower the LOI value, the higher the retention was). As LOI is correlated to the overall degree of order in cellulose, it can be concluded that there is less crystalline fraction in the membrane matrix than in original cotton textile. This is in the agreement with previous studies of cellulose crystallinity changes after dissolution in ILs and subsequent regeneration [34,41].

#### 4. Conclusions

This study aimed to test the usability of waste cotton textile for the fabrication of cellulose membranes. Waste cotton textile was dissolved in the mixture of [Emim][OAc] – DMSO. Two parameters, casting thickness and cellulose concentration in casting solution, were varied during the membrane fabrication process. Characterization of the prepared membranes was done by measuring water permeability, PEG retentions, and carrying out ATR-FTIR, hydrophilicity, and zeta-potential measurements. Additionally, membrane hydraulic resistance and porosity were calculated. The results showed that it is possible to prepare an ultrafiltration membrane from waste cotton textile without any pretreatment. However, use of too low concentration of cellulose seems unreasonable, as membranes prepared from the solution with 2 wt% cellulose concentration showed poor adhesion stability and uneven performance. More stable membranes were achieved from the solutions with 5, 6, and 7 wt% cellulose concentration. Membranes cast at  $150 \mu\text{m}$  thickness showed higher permeabilities and approximately same retentions as membranes cast at  $300 \mu\text{m}$ . Membranes with 5%, 6%, and

7% concentration of cellulose cast at  $150 \mu\text{m}$  showed water permeabilities in the range of  $0.51 - 0.67 \text{ m}^3/(\text{m}^2 \cdot \text{s} \cdot \text{Pa})$  and their retention of PEG 35 kDa is close to 90%. All the prepared membranes prepared were very hydrophilic and had negative surface charge based on the streaming potential measurements. Based on our results and preliminary calculations,  $1 \text{ m}^2$  of cotton bed linen should be enough for production of approximately  $20 \text{ m}^2$  of cellulose membrane. This study demonstrates well that membrane manufacturing from waste textile is a potential process for upcycling of waste materials.

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#### CRediT authorship contribution statement

**Anastasiia Lopatina:** Conceptualization, Methodology, Investigation, Writing - original draft, Writing - review & editing. **Ikenna Anugwom:** Conceptualization, Investigation, Writing - review & editing. **Hervé Blot:** Investigation, Methodology. **Ángela Sánchez Conde:** Investigation. **Mika Mänttari:** Conceptualization, Methodology, Resources, Writing - review & editing, Supervision, Project administration. **Mari Kallioinen:** Conceptualization, Methodology, Resources, Writing - review & editing, Supervision, Project administration, Funding acquisition.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2021.105705.

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