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Filtration of natural organic matter using ultrafiltration membranes for drinking water purposes: circular cross-flow compared with stirred dead end flow

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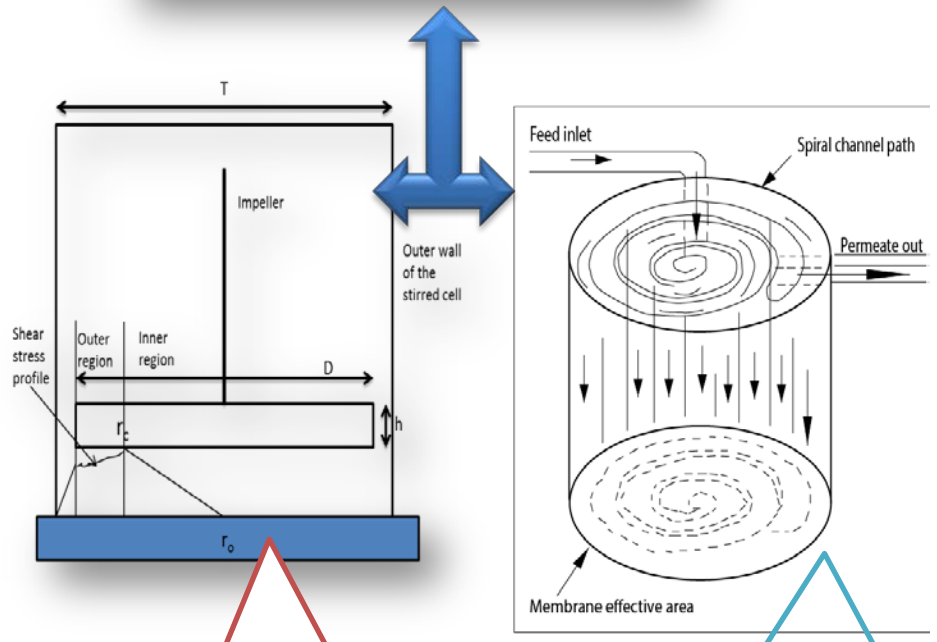
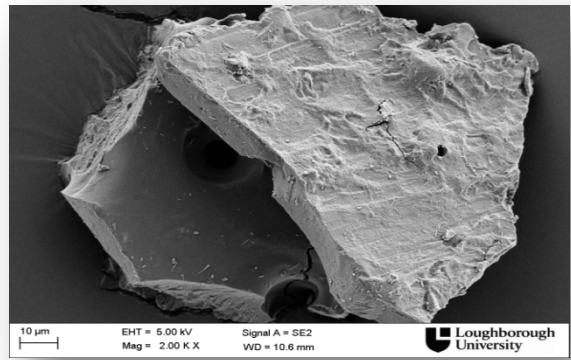
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Graphical abstract

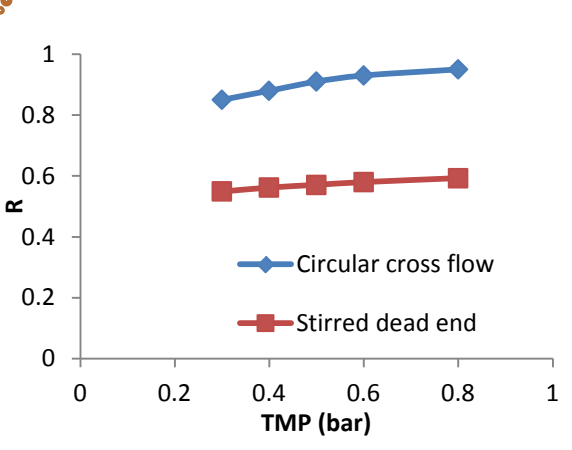
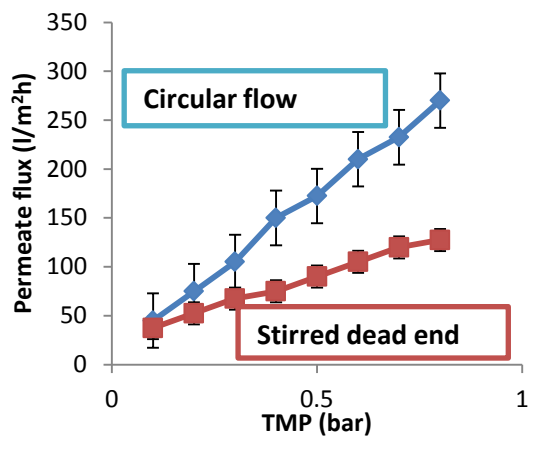
Humic acid particle



Stirred dead end module

Circular cross flow module

Comparison results



Filtration of natural organic matter using ultrafiltration membranes for drinking water purposes: circular cross-flow compared with stirred dead end flow

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Abstract

Application of ultrafiltration membranes for removal of humic acids is investigated below. Membrane filtration processes were compared using two different set-ups: circular flow and stirred dead end flow. The transmembrane pressure, temperature, feed concentration, pH, ionic strength and shear stresses applied on the membrane surfaces were kept constant whilst the permeate flux and solute rejection were measured during the experiments with both set-ups. It was shown that the rejection (both the observed and the true rejection) in the case of circular flow was higher than in the case of dead end flow. The mass transfer coefficients were determined for both set-ups. In the case of stirred dead end, it ranged in from $(2.14-4.72) \times 10^{-6}$ m/s; however, for circular cross flow system, the mass transfer coefficients were found in the range $(2.24-3.22) \times 10^{-5}$ m/s. Comparison of the mass transfer coefficients obtained for both systems showed that it was significantly higher for circular flow systems as compared with stirred dead end system at similar operating conditions. Energy consumed per volume of purified water by circular flow system (0.345 kW) was found to be much lower when by stirred dead end system (0.955 kW). This proved that the performance of circular flow system was more efficient in terms of rejection, mass transfer coefficient and energy consumption.

Key words: Membrane filtration – ultrafiltration - circular cross flow – stirred dead end flow – mass transfer coefficient - humic acid

Nomenclature

C_F humic acid concentration in the feed (kg/m^3)

C_M humic acid concentration at the membrane surface (kg/m^3)

C_P humic acid concentration in the permeate (kg/m^3)

D diffusion coefficient (m^2 / s)

D_t stirred dead end cell diameter (m)

De Dean number (-)

31	d_i	hydraulic diameter (m)
32	d_c	equivalent centreline diameter of curved channel (m)
33	h	height of the impeller blade (m)
34	J_V	volumetric flux (m / s)
35	k	mass transfer coefficient (m / s)
36	L	membrane channel length (μm)
37	Re	Reynolds number ($-$)
38	T	Operating temperature (K)
39	u	Cross flow velocity (m/s)
40	Greek letters	
41	δ	momentum boundary layer ($-$)
42	τ	shear stress (Pa)
43	μ	dynamic viscosity ($Pa \cdot s$)
44	ρ	fluid density (kg / m^3)
45	ω	angular velocity (rad/s)

46 **1. Introduction**

47 Clean and safe drinking water is one of the basic needs for the survival of human beings
48 especially under extreme conditions. Regular sources of drinking water in the events of natural
49 disasters are often polluted by harmful/hazardous components which can cause considerable
50 losses of life. Urgent purification of polluted water under such extraordinary conditions for
51 immediate consumption is a major priority (Chandrappa and Das, 2014). Membrane processes
52 such as microfiltration (MF) and ultrafiltration (UF) have been widely used for water treatment in
53 recent years (Metsämuuronen et al., 2014; Shamsuddin et al., 2014). They have been used as
54 alternative technologies to conventional methods such as coagulation, sedimentation, ozonation,
55 granular activated carbon (Fan et al., 2014), flocculation/chlorination (Bergamasco, 2014) and
56 slow sand filtration (Kaiser et al., 2014) etc. The reason is that membrane processes are not
57 only cost effective but also offer simple operation conditions, high output production with lower
58 energy consumption and chemicals. However, one of the critical issues of membrane
59 technology in water treatment is membrane fouling. Fouling results a considerable decline in

60 productivity over time and is caused by specific interactions between the membrane and the
61 components in feed water (Shi et al., 2014; Xiao et al., 2013). Fouling results in an
62 accumulation of colloidal matter, organic and inorganic compounds, microorganisms on
63 membrane surfaces and within membrane pores (Lee et al., 2014; Xia et al., 2013). This is often
64 referred to as irreversible loss of permeate flux through the membrane (Miller et al., 2014;
65 Yamamura et al., 2014).

66 Various approaches for minimising membrane fouling and concentration polarization had been
67 proposed. These include chemical methods such as modification of membrane surface to
68 minimise interactions between the membrane and the deposits (Kochkodan et al., 2014),
69 physical method such as mechanical scouring (Zhou et al., 2014), and hydrodynamic methods
70 such as improved module design and fluid flow arrangements in order to reduce solute
71 deposition on the membrane (Saxena et al., 2009). A useful method in overcoming
72 concentration polarization is a creation of flow instabilities (Kaur and Agarwal, 2002). The use of
73 eddies, Taylor vortices during pulsation rotating membrane filter (Charcosset, 2006) and
74 secondary flow (Dean vortex flow) (Jaffrin, 2012) are among these options. However, the
75 drawback of such rotating module systems is that more energy is required for scaling up and,
76 thus they have limited large-scale development for commercial purposes. Chung et al. (1993a)
77 and Chung et al. (1993b) studied an alternative method to create centrifugal vortices which
78 result from the onset of unstable flow in spiral wound membrane ducts. At sufficiently low flow
79 rates (that is at Reynolds numbers below some critical value) the velocity in the curved channel
80 flow is approximately stream wise parabolic. However, at higher Reynolds number (or Dean
81 number) above a critical value, centrifugal instabilities cause secondary flow containing stream
82 wise oriented Dean vortices similar to Taylor vortices. The presence of these vortices enhance
83 back migration through convective flow away from the membrane surface, depolarising the
84 solute build up near the membrane surface, thus resulting in an increase of membrane
85 permeation rate (Çulfaz et al., 2011; Moll et al., 2007; Moulin et al., 1999). Al-Bastaki and Abbas
86 (2001) reviewed methods of improving membrane performances and reducing fouling by the
87 presence of fluid instabilities. These techniques had proven to be successful in other
88 applications such as gas-liquid contactors for blood oxygenation (Tanishita et al., 1975). The
89 presence of vortices results in an improved oxygen transfer by a factor from 2 to 4 (Moulin et al.,
90 1996). Ghogomu et al. (2001) studied the performance of several curved membrane channels
91 designs and found that the mass transfer was improved compared to classical models. At the
92 same time the curved channels were showed to be more energy efficient. This was caused by
93 the formation of Dean vortices, which proved to be effective in reducing both the concentration
94 polarization and fouling.

95 It is well known that membrane module configurations can have a noticeable impact on filtration
96 processes (Nassehi et al., 2010). However, the extent to which one system performs

97 better/worse under similar conditions cannot be easily quantified. Therefore, it is essential to
98 develop conditions for comparison of the performance of different membrane systems with
99 respect to hydrodynamics (Reynolds number, membrane surface shear, etc.) and operating
100 conditions such as feed concentration, pH, transmembrane pressure (TMP) etc, while
101 comparison of these different membrane systems is still possible. In order to compare two
102 different membrane systems informative comparison has to be made with adequate
103 experimental details provided, which were missing in some of the earlier papers, e.g., see
104 discussions by Becht et al. (2008).

105 Below ultrafiltration of humic acid was studied using a stirred dead end cell (model XFUF07601;
106 Merck Millipore, Darmstadt, Germany) and a circular flow device (Amicon, Massachusetts, USA).
107 The transmembrane pressure, temperature, feed concentration, pH, ionic strength and shear
108 stresses on membrane surfaces were kept constant whilst the permeate flux and percentages
109 of solute rejection were measured during the experiments with both systems. Humic acid
110 concentration was fixed at 30 mg/l at pH between 7 and 8 and salt concentration at 0.01M NaCl.
111 The ultrafiltration data are compared in terms of permeate fluxes and solution rejections as well
112 as the effects of convective mass transfer in the stirred dead end and circular flow devices. The
113 obtained results showed noticeable differences under controlled experimental conditions. The
114 objective is to demonstrate a significant improvement in the mass transfer coefficient and
115 energy consumption in the case of circular flow as a result of the presence of secondary flows
116 (Dean vortices). The TMP was kept reasonably small (less than 2 bar) in our experiments to
117 imitate emergency situations (e.g., natural disasters) when the high pressure filtration
118 equipment is not available and portable water filtration kits are used for drinking water purposes
119 (Shamsuddin et al., 2014).

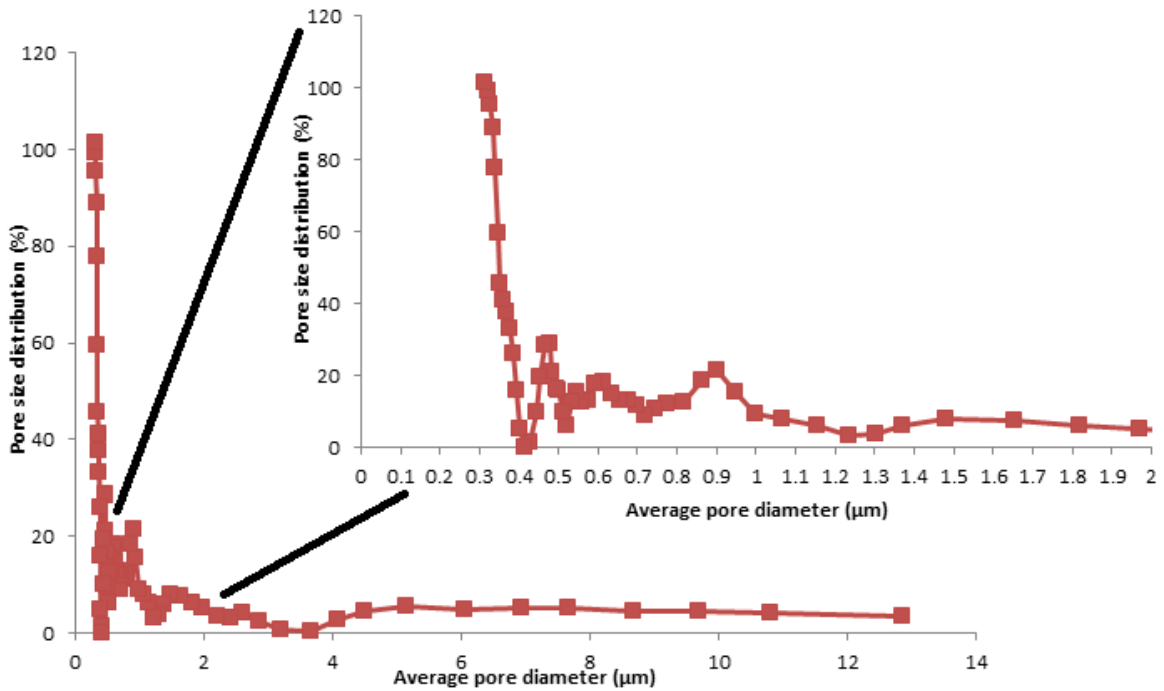
120 **2. Experimental**

121 **2.1 Materials**

122 Experiments were performed with Microdyn-Nadir (Wiesbaden, Germany) regenerated cellulose
123 membrane (UC100: RC100) with molecular cut off (MWCO) of 100kDa and porosity of 54%.
124 The porosity was determined using pycnometric method (Palacio et al., 1999). The membrane
125 samples were first soaked with deionized water for one hour, water was changed every 20
126 minutes interval to remove any wetting agents. Membrane pore size distribution shown in Figure
127 1 was measured using bubble point test (Nassehi et al., 2010).

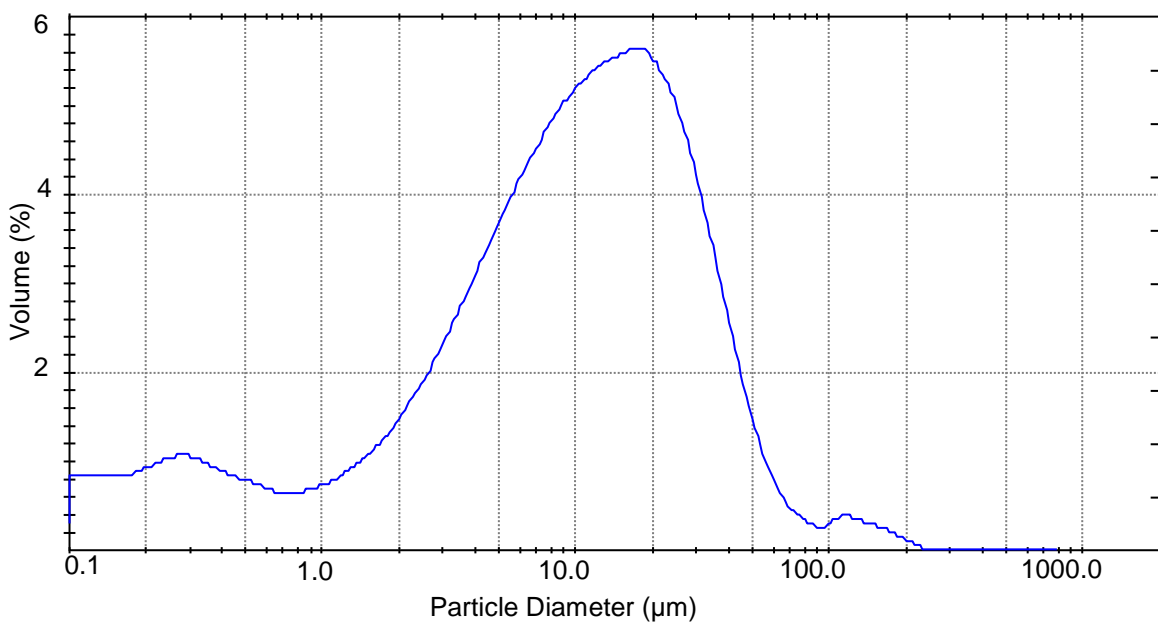
128 Humic acid was purchased from Sigma-Aldrich (Dorset, UK). The concentration of humic acid in
129 the aqueous feed solution was fixed and the concentration in the permeate was evaluated. The
130 effect of fouling was investigated at concentration of 30 mg/l of humic acid. The artificial
131 contaminated water was prepared i.e. 30 mg of humic acid and in 1000 ml of deionized water.

132 The solution was mixed in an ultrasonic bath for 20 minutes to achieve a uniform distribution of
133 humic acid in the feed. The pH was adjusted between 7 and 8. Concentrations of humic acid
134 were evaluated spectrophotometrically using a UV spectrophotometer (model: Mini 1240 UV-
135 VIS, Shimadzu, Milton Keynes, UK) with the absorbance measured at 254 nm. The overall
136 humic acid concentration was determined by comparison of the absorbance data with an
137 appropriate calibration curve. The particle size distribution of particles of humic acid was
138 evaluated using a laser light scattering instrument for particle size determination (Malvern
139 Instruments, Malvern, UK) and the result is presented in Figure 2 at pH between 7 and 8.



140

141 Figure 1 Pore size distribution of regenerated cellulose (RC) membrane used in this study.



142

143 Figure 2 Size distributions of particles of humic acid in solution.

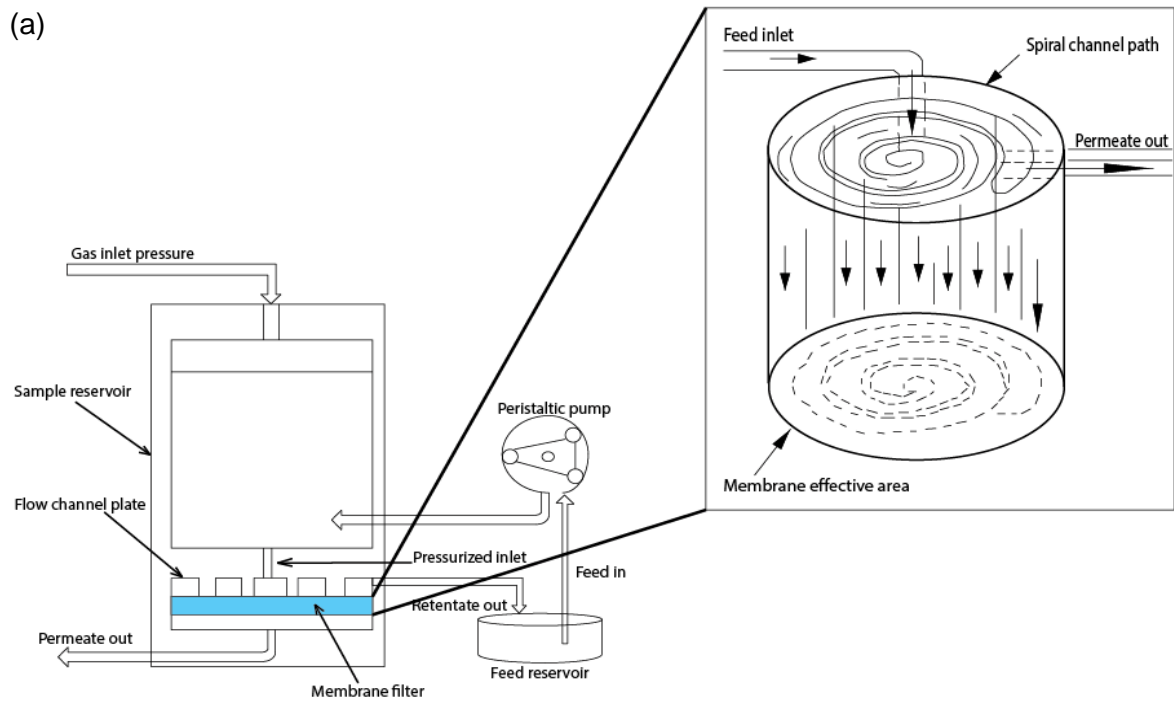
144 **2.2 Membrane filtration set-ups**

145 Two membrane configurations used are shown in Figure 3: circular cross flow systems (Figure
146 3 (a)) and the stirred dead end (Figure 3 (b)).

147 A 600 ml circular cross flow filtration cell (Amicon, Massachusetts, USA) (Figure 3 (a)) was
148 used which provides 40 cm² filtration surface area. The feed suspension circulated using a
149 peristaltic pump (Watson-Marlow Ltd, Cornwall, UK) in the range 83-250 ml/min. The retentate
150 was recovered and recycled back into the feed reservoir at room temperature. The suspension
151 was pumped in a circular channel over the membrane. The channel consists of three spirals of
152 varying radii from 1 cm to 4.1 cm (Figure 3 (a)) with the channel spacing approximately 1 cm.
153 The spiral channel has a width of 9.5 mm, height of 0.38 mm and length of 760 mm. The
154 pressure inside the vessel was adjusted to a predetermined inlet pressure.

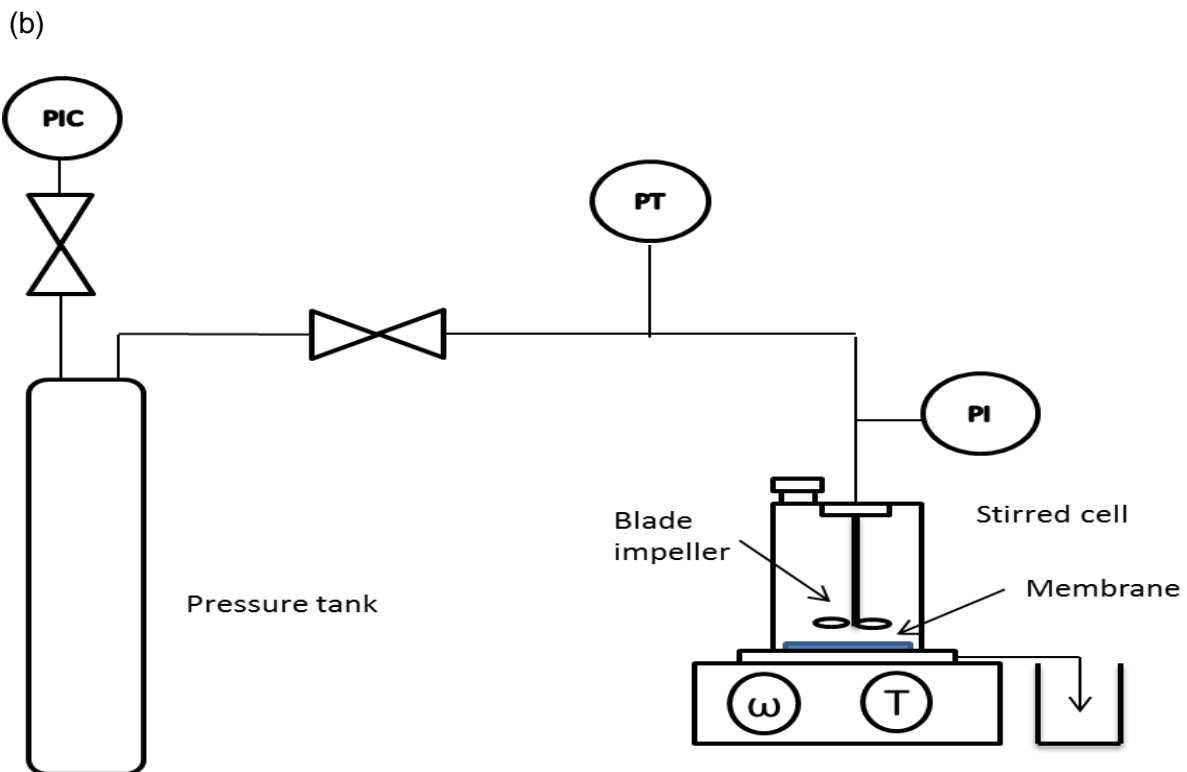
155 A stirred dead end filtration Figure 3 (b) cell (Merck Milipore, Darmstadt, Germany) with a
156 diameter of 76 mm and an effective membrane area of 40 cm² was used. The flat blade paddle
157 impeller has a height of 9 mm and a diameter of 65 mm. The feed suspension was added into
158 the feed reservoir prior to the filtration process with maximum feed volume of 300 ml. The stirrer
159 speed was measured using a digital tachometer (Shenzhen Ever Good Electronic Co Ltd,
160 Shenzhen, China). Air from a compressor was supplied to a glass apparatus containing the UF
161 membrane at the bottom of the cell. The pressure in the filtration cell was monitored by a
162 pressure gauge and controlled using a pressure regulator (model 8286; Porter Instrument Co.,
163 Hatfield, USA). The pressure gauges for the stirred dead end and circular flow modules were
164 calibrated. Calibration was made by cross-checking the gauges with an accurate gauge of
165 varying pressures.

166 All experiments were conducted at a temperature of 22°C (±2°C). Each experiment was carried
167 out with new, clean membrane samples, which were pre-treated as described above. Filtration
168 experiments were conducted for 1 hour and 20 min for each module. Samples of permeate
169 were collected at regular intervals of 1 minute.



170

171



172

173 Figure 3 (a) experimental set up for circular cross flow system with a sketch of circular channel
 174 path, and (b) experimental set up for stirred dead end system.

175 **2.3 Membrane thickness measurement**

176 The purpose of measuring the thickness of the membranes is to evaluate if there is any effect of
 177 membrane compaction throughout the filtration processes. The measurements were carried out

178 by using a surface/height profiling technique. A non-contact scanning surface topography
179 instrument, Talysurf CLI 2000 (Taylor Hobson, Leicester, UK), was used to create a profile of
180 the membranes thickness. The membrane samples were mounted on a glass surface as a
181 reference point. Data were collected one point at a time with each point having a discrete X, Y
182 and Z location. A non-contact chromatic length aberration (CLA) gauge with range of 3 mm and
183 resolution of 100 nm, and speed of 30 mm/sec was selected. For this purpose light was directed
184 by a beam splitter through a spectral aberration lens onto the surface (that included the
185 membrane sample). The lens then splits the light into different wavelengths and at any point on
186 the surface only a certain wavelength was in focus. Light was reflected from the surface to a pin
187 hole which allowed only the wavelength in focus to pass through. A spectrometer deflected the
188 light onto a sensor to interpolate spatial position of the data point. The thickness of membrane
189 was evaluated before and after filtration experiment for compaction.

190 **2.4 Operating conditions of circular cross flow system**

191 The formation of Dean vortices, which generate secondary flow perpendicular, appear in the
192 curved channel because of the centrifugal force. If Reynolds number of the system is higher
193 than the critical Reynolds number, when Dean vortices start to form according to Brewster et al.
194 (1959). The critical Reynolds number can be found experimentally. Reynolds number in the
195 circular flow system (Re_{TCF}) can be defined similar to the fluid flow in a pipe as follows:

$$196 \quad Re_{TCF} = \frac{\rho u d_i}{\mu}, \quad (1)$$

197 where ρ is the density of the solution, u is the cross flow velocity of the fluid, d_i is the equivalent
198 hydraulic diameter and μ is the dynamic viscosity of the fluid.

199 Dimensions of the spiral channel provided by the manufacturer are: 9.5 mm (width), 0.38 mm
200 (height) and 760 mm (length). The peristaltic pump has a flow rate of 1.67 ml per revolution.
201 Therefore, the rate of flow in the spiral channel can be calculated by dividing the pump flow rate
202 with the volume of the spiral channel. Then, multiply it with the length of the spiral channel gives
203 the cross flow velocity. The pump rotation speed of the experiments in circular cross-flow
204 system ranged from 83-250 ml/min. Hence, the cross flow velocity was calculated in the range
205 between 0.383 m/s to 1.156 m/s. The hydraulic diameter of the spiral channel can be calculated
206 according to Kaur and Agarwal (2002):

$$207 \quad d_i = 4 \frac{Area}{Perimeter}. \quad (2)$$

208 The hydraulic diameter equals to 0.0745 cm so the Reynolds number of the system fall in the
209 range between 287 and 861 which lies in the laminar flow region. Dean number (De) is
210 determined as a product of Reynolds number and the square root of the diameter ratio and

211 gives the ratio of the viscous force to the centrifugal force which shows the fluid motion in a
212 curved pipe or channel:

$$213 \quad De = Re_{TCF} \sqrt{\frac{d_i}{d_c}} \quad , \quad (3)$$

214 where d_c is the diameter of curvature of the path of the channel (equivalent centerline diameter).

215 The ratio of radii of the channel does not remain constant and varies in the range between 0.50
216 and 0.70. The equivalent centerline diameter is determined as the average radius of the
217 channel. Therefore d_c of the circular flow system is 4.51 cm. The critical Reynolds number of our
218 system ranges from 33 to 45 according to the data obtained in Brewster et al. (1959). The
219 Reynolds number of our circular cross flow system is much higher than the critical Reynolds
220 number. Therefore, Dean vortices occurred under the operating conditions implemented.

221 **2.5 Operating conditions of the stirred dead end system**

222 The Reynolds number of stirred dead end system is calculated as:

$$223 \quad Re_{SDE} = \frac{\rho \omega r_t^2}{\mu} \quad , \quad (4)$$

224 where ω is the angular velocity, and r_t is the radius of the stirred dead end system.

225 Reynolds number of the stirred dead end system is in the turbulent flow region that is $Re \sim$
226 77,427. In order to compare the circular cross-flow system with the stirred dead end system, all
227 the operating conditions has to be similar. All other parameters solution concentration, pH,
228 pressure, ionic strength and temperature of both systems were kept identical. However, the
229 liquid mixing and the flow profile differed because there is a difference between the circular
230 cross flow Reynolds number (laminar flow) and the rotational Reynolds number (turbulent flow).
231 So, it has been decide to maintain similar shear stresses on the surface of the membrane in
232 both circular cross flow system and stirred dead end system instead of keeping the identical
233 Reynolds numbers. This allowed a direct comparison between the two systems.

234 The determination of the shear stress for the circular cross flow system (τ_{TCF}) is relatively
235 easier by solving the force balance across the membrane in the circular flow system according
236 to Becht et al. (2008):

$$237 \quad \tau_{TCF} = \frac{\Delta P d_i}{4L} \quad , \quad (5)$$

238 where ΔP is the pressure drop across the membrane (transmembrane pressure) and L is the
239 membrane channel length:

$$240 \quad \Delta P = \frac{P_i + P_o}{2} - P_p \quad , \quad (6)$$

241 where P_i is the inlet pressure, P_o is the outlet pressure and P_p is the permeate pressure. The
 242 shear stress across the circular cross flow membrane under operating pressure of 1 bar is
 243 approximately 12 Pa.

244 In order to obtain the shear stress across the membrane for the stirred dead end system, the
 245 cell was divided into two regions which were the inner and outer ones, respectively. The shear
 246 stress reaches the peak at the critical radius of the impeller and begins to decrease in the outer
 247 region. The shear stress across the membrane is the average value of that in the inner and the
 248 outer region. The critical radius of the impeller is calculated by the correlation found by
 249 Kosvintev et al. (2005) as follows:

$$250 \quad r_c = \frac{D_i}{2} 1.23 \left(0.57 + 0.35 \frac{D_i}{D_t} \right) \times \left(\frac{h}{D_t} \right)^{0.036} n_b^{0.116} \frac{Re_d}{1000 + 1.43 Re_d} \quad , \quad (7)$$

251 where D_t is the diameter of the stirred cell, D_i is the impeller diameter n_b is the number of stirred
 252 blades.

253 According to the definition given by Kosvintev et al. (2005) the shear stresses in the inner and
 254 outer region are determined by Eq. (8) and (9), respectively:

$$255 \quad \tau_i = 0.825 \mu \omega r \frac{1}{\delta} \quad r < r_c \quad , \quad (8)$$

$$256 \quad \tau_o = 0.825 \mu \omega r_c \left(\frac{r_c}{r} \right)^{0.6} \frac{1}{\delta} \quad r > r_c \quad , \quad (9)$$

257 where δ is the momentum boundary layer ($\delta = \sqrt{\frac{\mu}{\rho \omega}}$).

258 The critical radius of the stirred dead end system in our case equals to 2.37 cm. Hence, the
 259 stirrer speed is determined as 750 rpm which corresponds to the shear stress across the
 260 membrane in stirred dead end system of approximately 12 Pa. The obtained value is
 261 comparable to the value of shear stress of circular cross-flow system. Therefore, all the
 262 operating conditions are similar for both systems which give a possibility of direct and
 263 reasonable comparison between two systems.

264 **2.6 Mass transfer coefficients for the two systems**

265 Mass transfer coefficient quantifies the resistance to the diffusion transfer in a boundary layer at
 266 the liquid/membrane interface (Cussler, 2009), and it is an important parameter for comparing
 267 circular cross-flow system and stirred dead end system. A high value of mass transfer
 268 coefficient value means high mass transfer which is the desirable outcome. The diffusion
 269 coefficient (D) humic acid particles in the deionized water are the ratio of molar flux and the
 270 driving force for diffusion and it was determined by the Stokes-Einstein (Einstein, 1905):

271 $D = \frac{k_B T}{6\pi\mu r_s}$, (10)

272 where r_s is the average radius of humic acid particles, T is the operating temperature and k_B is
 273 Boltzmann constant ($1.38 \times 10^{-23} \text{ m}^2 \text{ kg s}^{-2} \text{ K}^{-1}$). Based on the Stokes-Einstein equation, the
 274 diffusion coefficient is determined to be $1.255 \times 10^{-12} \text{ m}^2/\text{s}$.

275 According to the stagnant film model for concentration polarization in a membrane system
 276 (Jonsson and Boesen, 1977; Van der Berg et al., 1989; Zydney 1997) the volumetric water flux,
 277 J_V , can be written as:

278 $J_V = k \ln \left(\frac{C_M - C_P}{C_B - C_P} \right)$, (11)

279 where $k = D/\delta$ is the ratio of the diffusion coefficient D and the thickness of the concentration
 280 polarization layer, δ ; C_M , C_B and C_P are the solute concentration at the membrane surface, the
 281 solute concentration in the bulk phase and the solute concentration in the permeate,
 282 correspondingly. Concentration C_M is always higher than C_B due to the concentration
 283 polarization. Eq. (11) can be rearranged as follows:

284 $\ln \left(\frac{1 - R_{obs}}{R_{obs}} \right) = \ln \left(\frac{1 - R}{R} \right) + b \left(\frac{J_V}{u^a} \right)$, (12)

285 where $R = 1 - \frac{C_P}{C_M}$ is the true rejection and $R_{obs} = 1 - \frac{C_P}{C_B}$ is the observed rejection. The velocity
 286 variation method (Jonsson and Boesen, 1977; Van der Berg et al., 1989; Zydney 1997) is used
 287 to calculate the mass transfer coefficient for circular cross flow module. By plotting a linear
 288 relationship between $\ln(1 - R_{obs})/R_{obs}$ vs. J_V/u^a , the true rejection R can be calculated by
 289 extrapolation to an ordinate axis, and also the value of b can be determined graphically i.e. the
 290 slope of each flux (Jonsson and Boesen, 1977; Van der Berg et al., 1989; Zydney 1997). Once
 291 R and b were found, the mass transfer coefficient, both k_n and k_m can be determined from Eqs.
 292 (11-12). The value $a = 0.33$ (Van der Berg et al., 1989) is used as the flow condition in the
 293 circular channel path was laminar; and $a = 0.567$ (Mehta and Zydney, 2006) is used for
 294 turbulent conditions in the stirred dead end flow system.

295 To convert the angular velocity (ω) for stirred dead end flow, to a linear velocity (u) in order to
 296 suit the velocity variation method, the following equation is used:

297 $u = r \times \omega$, (13)

298 where r is the critical radius of the impeller i.e. 0.0237m, u is the linear velocity in m/s, and ω is
 299 the angular velocity in rad/s. The linear velocities were calculated to range between 0.390 m/s
 300 to 1.581 m/s.

301 3. Results and discussion

302 3.1 Filtration results

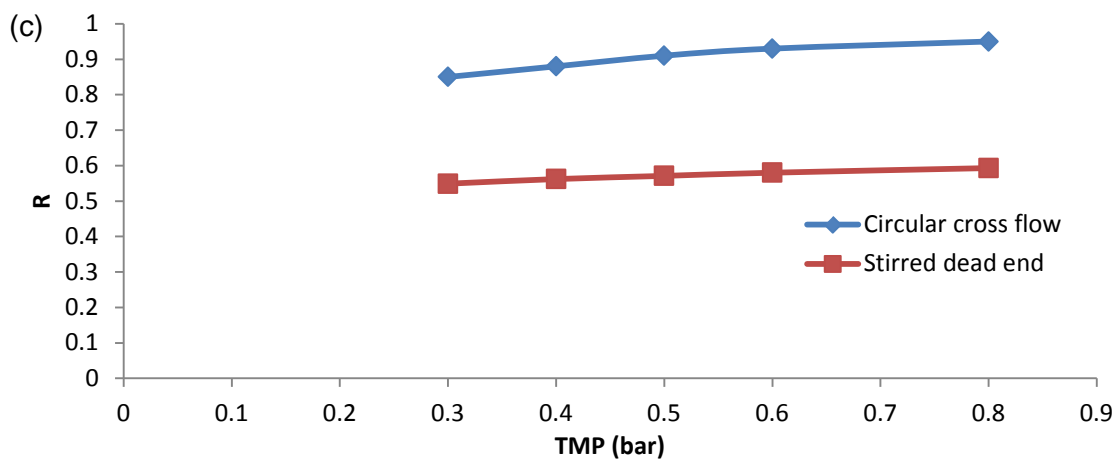
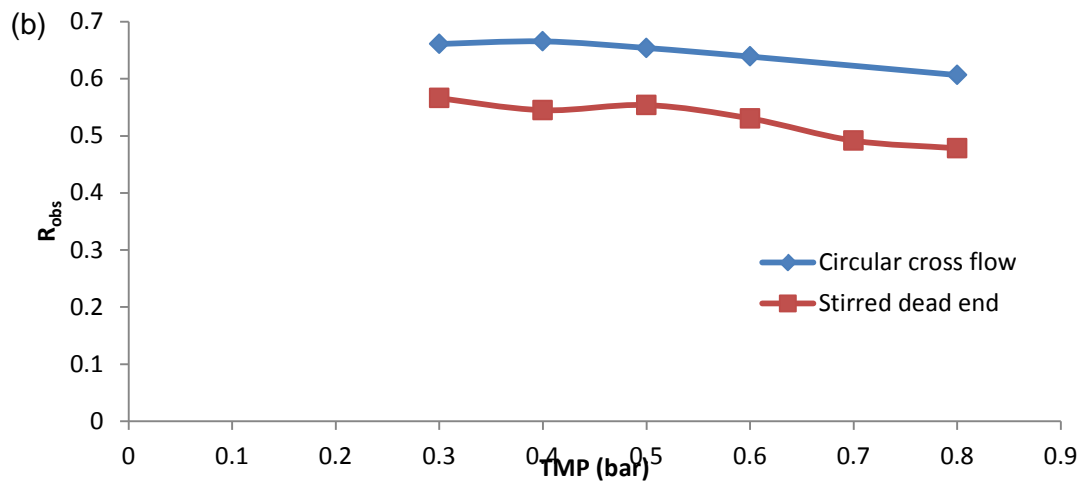
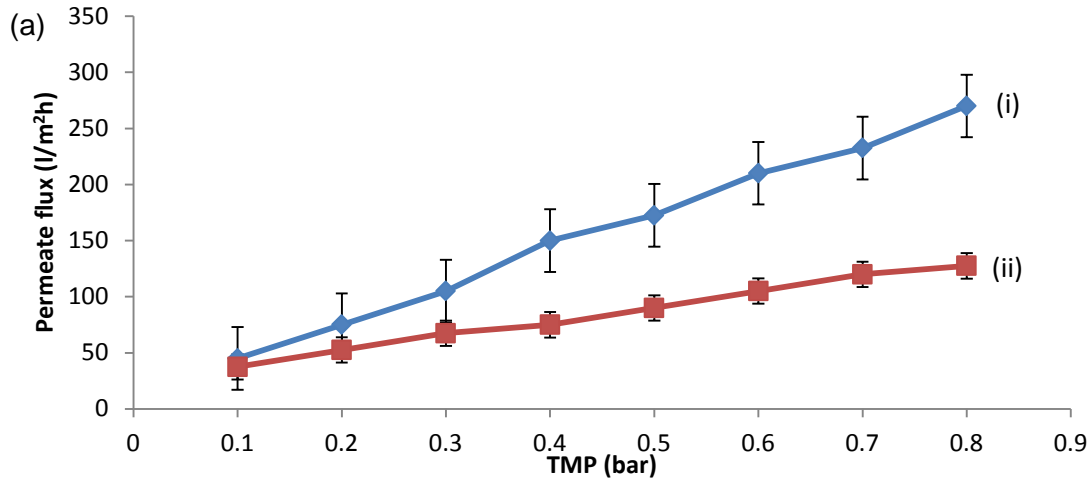
303 As stated earlier, all experiments were conducted with a humic acid at concentration 30 mg/l.
304 The rotational Reynolds number in the stirred dead end system calculated using Eq. (4) at a
305 stirrer speed of 750 rpm was 77,427 that the flow was turbulent. In the circular flow system the
306 cross flow stream through the channel was laminar and the cross flow velocities were varied
307 from 0.385 to 1.156 m/s. The Reynolds numbers (determined using Eq. (1)) for the circular flow
308 system were in the range of 287 to 861. The shear stress at the membrane surface in both
309 systems was approximately equal to 12 Pa according to Eq. (5-9). The maximum filtration time
310 was 1 hour and 20 min for both systems. Since the duration of the filtration experiment was
311 relatively short, the variation of concentration in these circumstances would not be considerable
312 (less than 5%) over that period of time. Hence, it was possible in the first approximation to
313 neglect this variation to simplify the calculations.

314 Substantial differences between the two systems were found (see Figure 5). According to Eq.
315 (5), shear stress in the circular cross flow increases with increase in transmembrane pressures
316 and cross flow velocities. Figure 5 (a) shows that within the range of Re number, where the flow
317 conditions remain laminar (<1500), the observed permeation fluxes were much greater as
318 compared to the stirred dead end system. This is due to the effects of Dean vortices, which
319 destroy (at least partially) the concentration polarization on the membrane surface to increase,
320 and reduces the thickness in the membrane boundary layer, improves solute mass transfer
321 away from the membrane surface into the bulk phase.

322 In this work, the observed rejection (R_{obs}) was experimentally measured. However due to the
323 concentration polarization this value is not accurate. The true concentration of solute at the
324 membrane surface is higher than in the bulk phase, thus the real rejection was calculated using
325 Eq. (12) using the velocity variation method (Jonsson and Boesen, 1977; Van der Berg et al.,
326 1989; Zydney, 1997). According to the data presented in Figure 5 (b) the observed rejections
327 (R_{obs}) for stirred dead end and the real rejections (R) were found to be low (Figure 5 (c)).
328 Accumulation of solute on the membrane surface as a result of concentration polarization effect
329 is likely to take place as the mass transfer coefficients calculated for the stirred dead end were
330 significantly lower than the circular cross flow.

331 In the case of circular cross flow, higher R_{obs} means more solute was rejected by the membrane
332 caused by a concentration increase in the concentration polarization layer due to higher solvent
333 permeation and solute retention. This can be seen from Figure 5 (c) where the true rejection
334 values in circular cross flow are higher than stirred dead end. The presence of circular channel
335 path induces the formation of Dean vortices to disrupt rapid solute build-up on the membrane

336 surface by creating flow instabilities. Consequently, mass transfer coefficients of solutes away
337 from the membrane into the bulk phase were found to be higher.



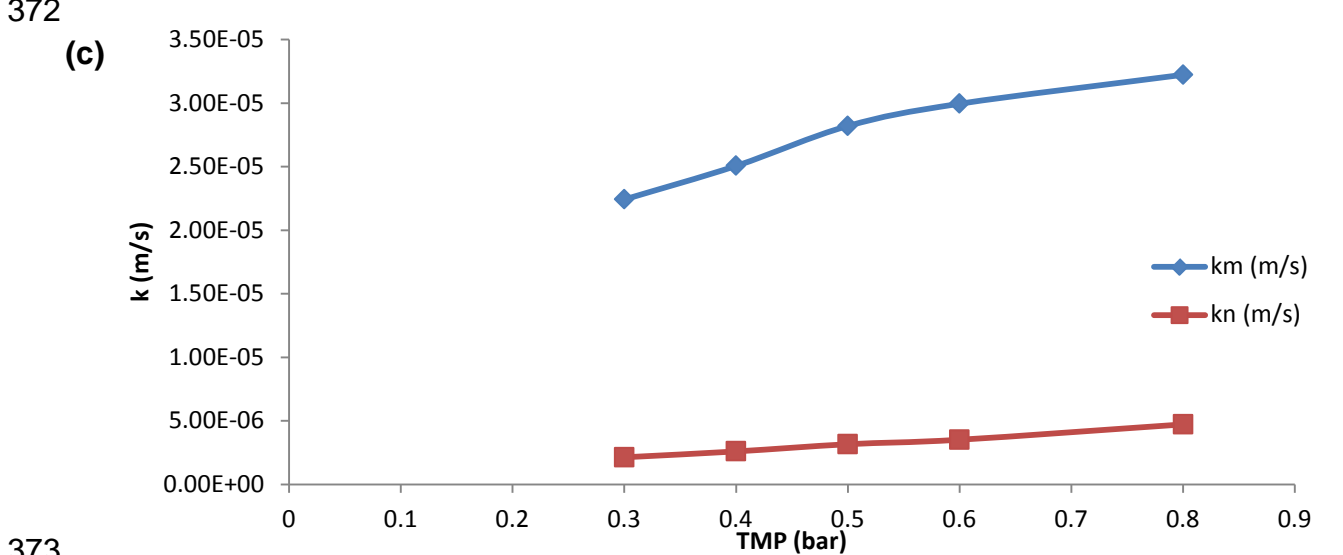
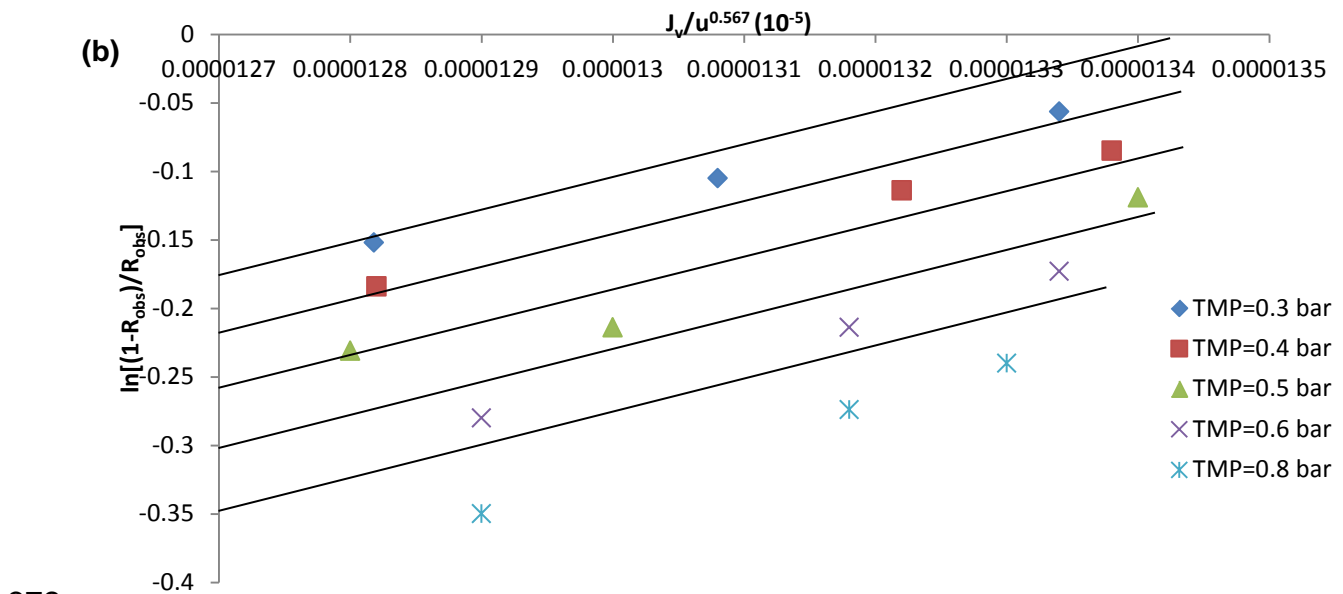
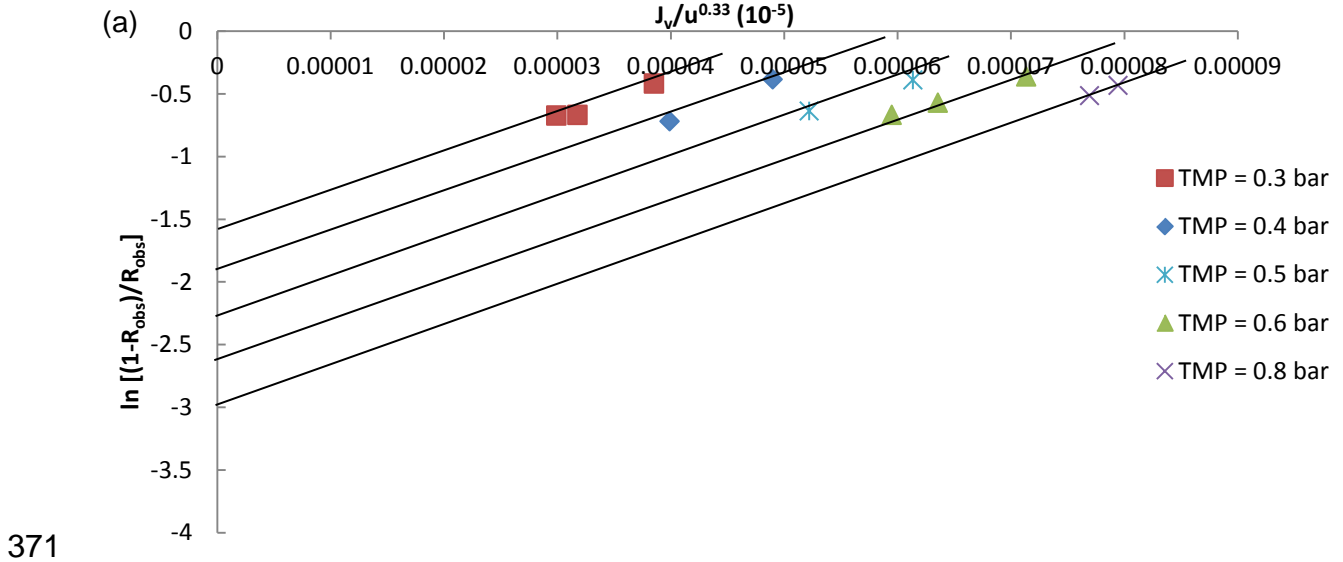
341 Figure 5 (a) Flux v/s TMP for both systems (i) circular cross flow, and (ii) stirred dead end, (b)
342 effect of TMP on humic acid observed rejections (R_{obs}), (c) effect of TMP on humic acid true
343 rejections (R) for circular cross flow system and for stirred dead end flow system.

344 **3.2 Mass transfer coefficient**

345 Both stirred dead end flow and circular cross flow systems, the mass transfer coefficients were
346 determined using Eqs. (11-12). The mass transfer coefficients calculated for stirred dead end
347 (k_n) are between $(2.14-4.72) \times 10^{-6}$ m/s. For circular cross flow system, the mass transfer
348 coefficients (k_m) for humic acid solution calculated from our experiments fall in the range $(2.24-$
349 $3.22) \times 10^{-5}$ m/s. These values are determined from data presented in Figure 6 (a-b) in which
350 velocity variation method was used. A linear relationship was observed and all straight lines
351 which fitted the data points showed almost identical gradient. An extrapolation to an ordinate
352 axis will determine the value of the true rejection (R) at various pressures. According to our data
353 the calculated values of R increased with the transmembrane pressures increase as seen in
354 Figure 5(c).

355 Comparison of the mass transfer coefficients for both the systems proves that the mass transfer
356 coefficients for circular flow systems were significantly higher as compared with stirred dead end
357 system at similar operating conditions.

358 In the case of vortex flow ultrafiltration with Taylor vortices, the mass transfer coefficients were
359 in the range of $(0.4-4.0) \times 10^{-5}$ m/s according to Agarwal (1997), whereas for cross flow filtration
360 the mass transfer coefficients were around $(1.0-5.0) \times 10^{-6}$ m/s (Muller et al., 2003; Kaur and
361 Agarwal, 2002). The method used by Kaur and Agarwal (2002) to assess the mass transfer
362 coefficient is debatable as the true rejections (R) could not be extrapolated from just one
363 crossflow velocity. This was the difference in our approach used: the velocity variation method
364 as shown in Figure 6 (a-b) at different crossflow velocities was used, the values of mass
365 transfer coefficients (k_m) and (k_n) and the true rejections (R) were graphically calculated.
366 Meanwhile Figure 6 (c) shows the relationship between mass transfer coefficients (k) and
367 TMPs for both systems. A linear relationship was expected as mass transfer would increase as
368 TMPs increases. The values of k_m are always higher when compared with k_n at similar
369 operating conditions, hence we conclude that circular cross flow system is better than stirred
370 dead end.



374 Figure 6 Velocity variation plot for (a) circular cross flow system, and (b) stirred dead end flow
 375 system, $\ln [(1-R_{obs})/ R_{obs}]$ as a function of $J_v/u^{0.33}$ and $J_v/u^{0.567}$ respectively, at various
 376 transmembrane pressures, (c) mass transfer coefficient, k_m and k_n against TMPs.

377 **3.3 Membrane thickness measurements**

378 Table 1 shows the membrane thicknesses and permeate qualities for both systems before and
 379 after filtration experiments. An increase in the membrane thickness less than 0.2% could be due
 380 the deposition of humic acid particles on the membrane surface but it is not significant enough
 381 to cause noticeable membrane fouling. No significant fouling was observed on the surface of
 382 the membranes in both systems due to low feed concentration (~30mg/l), but traces of humic
 383 acid can be seen in permeate when experiments were conducted with the stirred dead end
 384 module.

385 Table 1 Comparison of membrane thickness and permeate qualities between circular flow and
 386 stirred dead end systems.

Membrane system	Membrane thickness (before)	Membrane thickness (after)	% Thickness increase/decrease for imposed conditions	Permeate turbidity (NTU)	Average humic acid concentration in permeate (mg/l)
circular flow	273.18 ±0.05 um	273.70 ±0.05 um	±0.19% Increase	0.09±0.01	1.354±0.05
Stirred dead end flow	274.35 ±0.05 um	274.91 ±0.05 um	±0.20% Increase	0.17±0.05	13.780±0.05

387

388 Under similar operating conditions with constant TMPs of 0.5 bars for both modules, the overall
 389 permeate qualities were determined. The results shows that permeate turbidity and
 390 concentration of HA were much lower when circular flow was used. Both turbidity and humic
 391 acid concentration in permeate were less than 0.10 NTU and 2 mg/l respectively which meets
 392 the drinking water standard from WHO. In stirred dead end system however, solute penetration
 393 through the membrane causes higher turbidity and humic acid concentration in the resulting
 394 permeates. It was found that 19% calculated from transmission data represents the amount of
 395 humic acid observed in the permeate as seen from Table 1. Figures 2 and 3 give indications
 396 that smaller solute size might contribute to the transmission of HA through the pores and into
 397 the permeate.

398 The energy consumption for both systems were calculated and compared at similar operating
 399 conditions used. For stirred dead end system, in order to produce 12 Pa of shear stress the
 400 system required stirrer speed of 750 rpm which results a Reynolds number of 77,427. Since

401 Reynolds number is higher than 10^4 i.e. turbulent regimes, the power number N_T is independent
402 of the Reynolds number, and viscosity is no longer a factor to be taken into account; the
403 following equation was used to calculate the amount of power consumption (McCabe et al.,
404 2001):

$$405 \quad P = N_T \omega^3 D_i^5 \rho \quad , \quad (14)$$

406 where ω is angular velocity, and D_i is the diameter of the impeller.

407 The total energy consumption calculated was 0.955 kW per volume of purified water. Meanwhile
408 the total energy consumption for the circular cross flow system calculated was found 0.345 kW
409 per volume of purified water. It was assumed for calculation of energy consumption that it is
410 proportional to pressure drop across the membrane multiplied by the flow rate of feed for 'a thin
411 channel flow' designs according to Bird et al. (1960). This calculation proved that energy
412 consumed by circular cross flow system was much less than by stirred dead end system. This
413 conclusion on energy consumption by modules with Dean vortices is in line with other authors
414 Moulin et al. (1999) and Manno et al. (1998). These authors concluded that the presence of
415 Dean secondary flow, even at fixed amount energy dissipated would result in more permeation
416 fluxes compared to other conventional modules.

417 **4. Conclusions**

418 Ultrafiltration of humic acid solutions using two different configurations (circular flow and stirred
419 dead end set-ups) were studied and compared. Flow arrangement in both systems is
420 considerably different and the method of comparison has been suggested, which allows
421 comparison under similar conditions. Both systems were compared in terms of convective mass
422 transfer in the filtration experiments, permeate fluxes, rejections and energy consumed per
423 volume of purified water. The presence of Dean vortices in circular flow system due to the
424 curved geometry of the channel creates flow patterns which resulted in reducing of the effects of
425 concentration polarization and, hence, fouling in the filtration processes and higher rejection.
426 The mass transfer coefficients for stirred dead end system, (k_n) were determined to range from
427 $(2.14-4.72) \times 10^{-6}$ m/s; however, for circular cross flow system, the mass transfer coefficients
428 (k_m) were in the range $(2.24-3.22) \times 10^{-5}$ m/s. Energy consumed per volume of purified water by
429 circular flow system (0.345 kW) was calculated to be much lower as compared to the stirred
430 dead end system (0.955 kW). This proved that not only the performance of circular flow system
431 was better in terms of mass transfer coefficient, and rejections but also more energy efficient.

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435 **6. References**

- 436 Agarwal, G. P. "Analysis of proteins transmission in vortex flow ultrafilter for mass transfer
437 coefficient." *Journal of Membrane Science* 136(1), 1997: 141-151.
- 438 Al-Bastaki, N, and A Abbas. "Use of fluid instabilities to enhance membrane performance: a
439 review." *Desalination* 136, 2001: 255-262.
- 440 Becht, N.O, D.J Malik, and E.S Tarleton. "Evaluation and comparison of protein ultrafiltration
441 test results: Dead-end stirred cell compared with a cross-flow system." *Separation and*
442 *Purification Technology*, 62, 2008: 228-239.
- 443 Bergamasco, R. "Reduction of trihalomethanes using combined process
444 coagulation/flocculation/membranes in water treatment." *36th Symposium on*
445 *Biotechnology for Fuels and Chemicals* . 2014.
- 446 Bird, R.B, W.E Steward, and E.N Lightfoot. *Transport Phenomena*. New York: Wiley, 1960.
- 447 Blatt, W.F, A Dravid, A.S Michaels, and L Nelsen. "Solute polarization and cake formation in
448 membrane ultrafiltration: causes, consequences, and control techniques." *Membrane*
449 *Science and Technology*, 1970: 47-97.
- 450 Brewster, D.B, P Grosberg, and A.H Nissan. "The stability of viscous flow between horizontal
451 concentric cylinders." *Proceedings of the Royal Society of London. Series A.*
452 *Mathematical and Physical Sciences* 251(1264), 1959: 76-91.
- 453 Chandrappa, R, and D.B Das. *Sustainable Water Engineering: Theory and Practice*. John Wiley
454 & Sons, 2014.
- 455 Charcosset, C. "Membrane processes in biotechnology: an overview." *Biotechnology advances*,
456 24(5), 2006: 482-492.
- 457 Chung, K-Y, R Bates, and G Belfort. "Dean vortices with wall flux in a curved channel
458 membrane system. 4. Effect of vortices om permeation fluxes of suspensions in
459 microporous membrane." *Journal of Membrane Science* 81 (1), 1993a: 139-150.
- 460 Chung, K-Y, W.A Edelstein, and G Belfort. "Dean vortices with wall flux in a curved channel
461 membrane system. 6. Two dimensional magnetic resonance imaging of the velocity field
462 in a curved impermeable slit." *Journal of Membrane Science* 81 (1-2), 1993b: 151-162.
- 463 Çulfaz, P.Z, M Haddad, M Wessling, and R.G.H Lammertink. "Fouling behavior of
464 microstructured hollow fibers in cross-flow filtrations: Critical flux determination and
465 direct visual observation of particle deposition." *Journal of Membrane Science*, 372(1),
466 2011: 210-218.

- 467 Cussler, E.L. *Diffusion: Mass Transfer in Fluid Systems*. Cambridge: Cambridge University
468 Press., 2009.
- 469 Einstein, A. "Über die von der molekularkinetischen Theorie der Wärme geforderte Bewegung
470 von in ruhenden Flüssigkeiten suspendierten Teilchen." *Annalen der Physik (in German)*
471 322 (8): , 1905: 549-560.
- 472 Fan, X, et al. "Performance of an integrated process combining ozonation with ceramic
473 membrane ultra-filtration for advanced treatment of drinking water." *Desalination* 335(1),
474 2014: 47-54.
- 475 Ghogomu, J.N, C Guigui, J.C Rouch, M.J Clifton, and P Aptel. "Hollow-fiber membrane module
476 design: comparison of different curved geometries with Dean vortices." *Journal of*
477 *Membrane Science* (181), 2001: 71.
- 478 Jaffrin, M.Y. "Hydrodynamic techniques to enhance membrane filtration." *Annual Review of*
479 *Fluid Mechanics*, 44, , 2012: 77-96.
- 480 Jonsson, G, and C.E Boesen. "Concentration polarization in a reverse osmosis test cell."
481 *Desalination*, 21, 1977: 1-10.
- 482 Kaiser, E, C Prasse, M Wagner, K Bröder, and T.A Ternes. "Transformation of Oxcarbazepine
483 and Human Metabolites of Carbamazepine and Oxcarbazepine in Wastewater
484 Treatment and Sand Filters." *Environmental Science & Technology*, 48(17), 2014:
485 10208-10216.
- 486 Kaur, J, and G.P Agarwal. "Studies of protein transmission in thin channel flow module: the role
487 of dean vortices for improving mass transfer." *Journal of Membrane Science* 196, 2002:
488 1-11.
- 489 Kochkodan, V, D.J Johnson, and N Hilal. "Polymeric membranes: Surface modification for
490 minimizing (bio) colloidal fouling." *Advances in colloid and interface science*, 206, 2014:
491 116-140.
- 492 Lee, W, H.W Lee, J.S Choi, and H.J Oh. "Effects of transmembrane pressure and ozonation on
493 the reduction of ceramic membrane fouling during water reclamation." *Desalination and*
494 *Water Treatment*, 52(4-6), 2014: 612-617.
- 495 Manno, P, P Moulin, J.C Rouch, M Clifton, and P Aptel. "Mass transfer improvement in helically
496 wound hollow fibre ultrafiltration modules Yeast suspensions." *Separation and*
497 *Purification Technology*, 14, 1998: 175-182.

- 498 McCabe, W.L, J.C Smith, and P Harriott. *Unit Operations of Chemical Engineering*. New York:
499 McGraw-Hill Companies, 2001.
- 500 Mehta, A, and A.L Zydney. "Effect of membrane charge on flow and protein transport during
501 ultrafiltration." *Biotechnology Progress* 22(2), 2006: 484-492.
- 502 Metsämuuronen, S, M Sillanpää, A Bhatnagar, and M. Mänttari. "Natural organic matter removal
503 from drinking water by membrane technology." *Separation & Purification Reviews* 43, no.
504 1 (2014): 1-61.
- 505 Miller, D.J, S Kasemset, D.R Paul, and B.D Freeman. "Comparison of membrane fouling at
506 constant flux and constant transmembrane pressure conditions." *Journal of Membrane*
507 *Science*, 454, 2014: 505-515.
- 508 Moll, R, D Veyret, F Charbit, and P Moulin. "Dean vortices applied to membrane process: Part I.
509 Experimental approach." *Journal of Membrane Science*, 288(1), 2007: 307-320.
- 510 Moulin, P, J.C Rouch, C Serra, M.J Clifton, and P Aptel. "Mass transfer improvement by
511 secondary flows: Dean vortices in coiled tubular membranes." *Journal of Membrane*
512 *Science* 114 (2) , 1996: 235-244.
- 513 Moulin, P, P Manno, J.C Rouch, C Serra, M.J Clifton, and P Aptel. "Flux improvement by Dean
514 vortices: ultrafiltration of colloidal suspensions and macromolecular solutions." *Journal of*
515 *Membrane Science*, 156(1), 1999: 109-130.
- 516 Muller, C.H, G.P Agarwal, Th Melin, and Th Wintgens. "Study of ultrafiltration of a single and
517 binary protein solution in a thin spiral channel module." *Journal of Membrane Science*
518 227, 2003: 51-69.
- 519 Nassehi, V, D.B Das, I.M.T.A Shigidi, and R.J Wakeman. "Numerical analyses of bubble point
520 tests used for membrane characterisation: model development and experimental
521 validation." *Asia-Pacific Journal of Chemical Engineering*, 2010: 1-13.
- 522 Palacio, L, P Pradanos, J.I Calvo, and A Hernandez. "Porosity measurements by a gas
523 penetration method and other techniques applied to membrane characterization." *Thin*
524 *Solid Films* 348 , 1999: 22-29.
- 525 Saxena, A, B.P Tripathi, M Kumar, and V.K Shahi. "Membrane-based techniques for the
526 separation and purification of proteins: an overview." *Advances in colloid and interface*
527 *science*, 145(1), 2009: 1-22.

- 528 Shamsuddin, N, D.B Das, and V.M Starov. "Membrane-based point-of-use water treatment
529 (PoUWT) system in emergency situations." *Separation & Purification Reviews (just*
530 *accepted)*. DOI:10.1080/15422119.2014.973967, 2014.
- 531 Shi, X, G Tal, N.P Hankins, and V Gitis. "Fouling and cleaning of ultrafiltration membranes: A
532 review." *Journal of Water Process Engineering, (1)*, 2014: 121-138.
- 533 Tanishita, K, P.D Richardson, and P.M Galletti. "Tightly wound coils of microporous tubing:
534 progress with secondary-flow blood oxygenator design." *ASAIO Journal 21 (1)*, 1975:
535 216-223.
- 536 Van der Berg, G.B, I.G Racz, and C.A Smolders. "Mass transfer coefficients in cross-flow
537 ultrafiltration." *Journal of Membrane Science, 47*, 1989: 25-51.
- 538 Xia, S, Y Zhou, R Ma, Y Xie, and J Chen. "Ultrafiltration of humic acid and surface water with
539 tubular ceramic membrane." *Desalination and Water Treatment* , 2013: 1-8.
- 540 Xiao, F, P Xiao, W.J Zhang, and D.S Wang. "Identification of key factors affecting the organic
541 fouling on low-pressure ultrafiltration membranes." *Journal of Membrane Science 447*,
542 2013: 144-152.
- 543 Yamamura, H, K Okimoto, K Kimura, and Y. Watanabe. "Hydrophilic fraction of natural organic
544 matter causing irreversible fouling of microfiltration and ultrafiltration membranes." *Water*
545 *Research (54)*, 2014: 123-136.
- 546 Zhou, Z, F Meng, H Lu, Y Li, X Jia, and X He. "Simultaneous alkali supplementation and fouling
547 mitigation in membrane bioreactors by on-line NaOH backwashing." *Journal of*
548 *Membrane Science, 457*, 2014: 120-127.
- 549 Zydney, A.L. "Stagnant film model for concentration polarization in membrane systems." *Journal*
550 *of Membrane Science, 130*, 1997: 275-281.