Original Article

The Use of *Verbascum Thapsus L* as a Biomembrane for Activated Sludge Filtration

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Abstract
Membrane technology is a green technology, but it still faces a pressing problem related to the effect of fabrication materials on the environment. The plant *Verbascum thapsus* L (VTL) was utilized as a biomembrane to reduce chemicals. In this study, VTL was successfully utilized as a membrane for activated sludge separation. The membrane was characterized via scanning electron microscopy (SEM)-EDX, Fourier transform infrared (FTIR), and contact angle measurement. Additionally, the effects of pressures on the fluxes and the rejection ability were studied. The permeability of the bio-based membrane reached 581 L/m^2.h.bar. The VTL membrane was examined for the removal of chemical oxygen demand (COD), protein, and carbohydrate. Accordingly, the maximum COD removal was obtained at a transmembrane pressure of 2.5 bar and reached up to 57%. The protein and carbohydrates rejections raised from 80% and 84% at 0.5 bar to 90% and 98% at 2.5 bar, respectively. The total resistance increased from 87% at a pressure of 0.5 bar to 96% at 2.5 bar. The flux recovery ratio (FRR) for the membrane at working pressures (0.5-2.5 bar) was 96% for 0.5 bar and 70% for 2.5 bar. The physical cleaning showed a flux recovery after three operation cycles. At the end of the filtration experiments, the pressure variation along streamlines over the membrane cross-section was simulated. As a result of this study, the use of a naturally-derived membrane is considered a green technology. The plant-based membrane reduces the use of non-green chemicals. Moreover, VTL has no commercial value and is recognized as an invasive plant species. All of the previous issues made the study worthwhile.
Keywords: *Verbascum thapsus*, Bio-membrane, Green membrane, Activated sludge filtration

1. Introduction

Water is the most valuable substance that is involved in all life activities (1). The water demand increased in the last centuries due to the sharp growth in population, the industrial revolution, and commercial development (2). As a result, the generation of wastewater has also increased. In this context, the need for safe disposal from the generated wastewater has become an urgent issue (3). Wastewater treatment aims to recover water with standard limits before discharging it to the environment (4). In general, the treatment method is classified into physical, chemical, biological, or a combination thereof (5). The physical forces are the predominant forces in the physical treatment units. The typical physical methods are screening, mixing, sedimentation, filtration, and adsorption (6). Membrane technology has become more attractive than conventional separation techniques (7,8) due to high efficiency (9), small area requirements (10), and ease of operation (11).

The membrane technology employs a semipermeable material to separate components (12). It can also be described as a selective barrier between two adjacent phases that regulates the movement of some constituents from one side to another (13). Membrane technology has been utilized in many fields. For example, Dizge et al used a cellulose membrane for drinking water treatment (14). Collivignarelli et al removed non-ionic and anionic surfactants from laundry wastewater (15), and Romero-Dondiz et al employed ultrafiltration and nanofiltration membranes for wastewater recovery in leather industry (16). The membrane is also employed in biodiesel purification (17), fermentation industry (18), gas separation (19), and food industry processes (20).

Even though membrane technology is considered a green technology (14), it still faces a pressing problem related to the effect of fabrication materials on the environment (21, 22). To overcome this issue, many researchers have developed green membranes from natural sources. Saleh et al prepared ceramic membranes from basalt rock (23). Aloulou et al utilized Tunisian natural sand in a ceramic membrane preparation (24). Natural clay (25), fly ash (26), zeolite (27), and phosphate sub-products (28) were used as primary materials for membranes synthesis. Bio-derived solvents were also utilized in the preparation of polymeric membranes (22, 29, 30).

Nature has inspired many researchers to use materials

present in it. Correa and Sens studied the possibility of utilizing wood as a membrane (31). Sens et al successfully employed a helical cross-flow filtration system to achieve an average removal of 70% and apparent color removal of 93% for average turbidity (32). Boutilier et al developed an effective point-of-use device from plant xylem to supply pathogen-free drinking water (33).

Verbascum thapsus L (VTL) is a plant that grows in many regions all over the world (34). At the bottom of the plant, there is a large rosette of leaves where the stem grows to reach 2 m or more (35). Its small yellow flowers are densely grouped on a tall stem (36). The presence of oil, glycoside, flavonoids, sesquiterpenes, and verbathasin A made the researchers use the plant in medical practices (37). The plant has limited applications in environmental engineering (38). In Turkey, the plant is unfavorable and classified as an invasive plant species. The farmers remove VTL from their farms by the physical method. Consequently, the accumulated plant wastes are burned near the fields. In this study, VTL was used as a biomembrane for activated sludge filtration. The ability of VTL membrane in removal of COD, protein, and carbohydrate was examined. At the end of the filtration experiments, the pressure variation among streamlines over the membrane cross-section was simulated. From different perspectives, the use of a naturally-derived membrane is considered a green application. The plant-based membrane reduces the use of non-green chemicals. Additionally, VTL has no commercial value and is recognized as an invasive plant species. All of the previous issues made the study worthwhile.

2. Materials and Methods
2.1. Preparation of Plant-Based Biomembrane

VTL was collected from farms within the boundaries of Erdemli district, Mersin, Turkey. The leaf of the plant was separated from the whole plant. VTL leaves were gently washed with distilled water to remove any dust residuals and dried at room temperature for 48 hours. VTL leaves were cut in a circular shape with a diameter of 5.2 cm and used as a bio-membrane in a dead-end filtration system, as shown in Fig. 1.

2.2. Filtration Experiments of Activated Sludge

In this research study, the filtration experiments were accomplished, as discussed in the previous work (39), with some modifications. Briefly, the prepared membrane was supported by a porous steel disk and inserted in a dead-end filtration apparatus (Sterlitech HP 4750, Sterlitech, Kent, WA, USA). The distilled water was passed through the membrane under different pressure values (0.5-2.5 bar) using N2 gas at room temperature. The operating pressure was selected based on the preliminary studies to avoid any damages to the biomembrane. The water passed through the membrane was measured, and the permeate

flux (Jw) was calculated using equation 1 (40, 41).

$$Jw = \frac{V}{A \times t} = \frac{\Delta P}{\mu K_m} \qquad (1)$$

where J_w is the permeate flux (L/m²h), V is the volume of the water passed through the membrane (L), A is the active surface area (m²), t is the time (t), ΔP is the differential pressure across the membrane (bar), μ is the fluid dynamic viscosity (kg/m.s), and K_m is the membrane resistance coefficient (1/m).

The activated sludge was also filtrated, and the flux of permeate was recorded in the same manner as the distilled water flux. The used activated sludge was supplied from a local domestic wastewater treatment plant. The activated sludge characteristics included mixed liquor suspended solids concentration of 4500±150 mg/L, pH of 7.2±0.2, chemical oxygen demand (COD) of 323±5 mg/L, protein concentration of 58±4 mg/L, and carbohydrate concentration of 86±0.5 mg/L. The permeate water from the activated sludge filtration experiments was characterized, and the rejection percentage in the COD, protein, and carbohydrate was calculated using equation 2 (39).

$$\text{Rejection \%} = (1 - \frac{Permeate\ Concentration}{Initial\ Concentration}) \times 100\% \qquad (2)$$

2.3. Characterization of Plant-Based Membrane

The plant-based membrane was characterized using Scanning Electron Microscopy and. Energy Dispersive X-Ray Analysis (SEM-EDX, Zeiss Supra 55 Germany). The membranes were frozen with nitrogen, and a chromium layer was used to coat the membranes. SEM images and SEM-EDX were used to explore the surface morphology and the chemical composition of the membrane. Fourier Transform Infrared Spectroscopy (FTIR, PerkinElmer USA) was used to record the membrane spectrum in the range 400-4000 cm⁻¹. Water contact angle measurement was carried out using a goniometer (One Attension, Biolin scientific instrument).

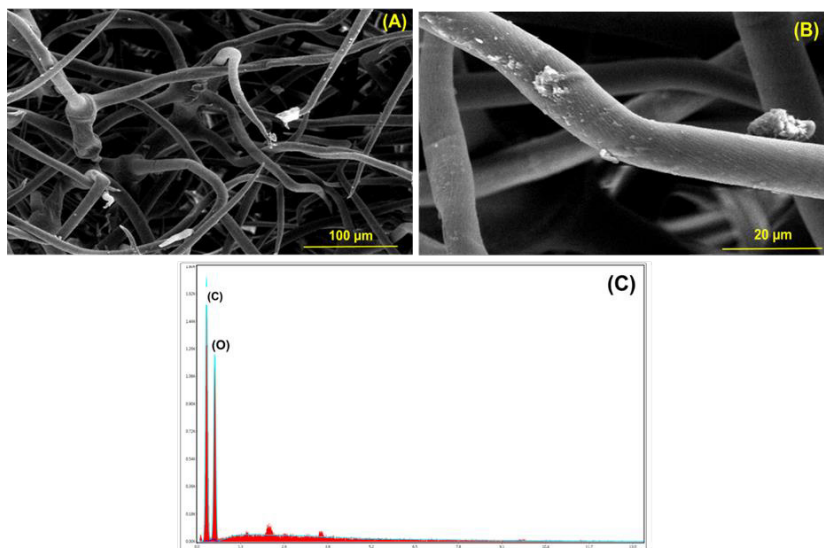**Fig. 1.** A Schematic Diagram of the Bio-membrane System Applied in This Study.

Fig. 2. SEM Images for (A) VTL (100 μm), (B) VTL (20 μm), (C) SEM-EDX Results.

2.4. Model Development

The hydrodynamic part of the filtration process was modeled using the Darcy equations for the fluid flow in the porous material. COMSOL Multiphysics V 5.5 was used as a simulator software. Equation 3 shows the Darcy equation used in the model (42).

$$u = -\frac{K}{\mu(c)}\nabla P \qquad (3)$$

where u is the velocity of the fluid flow through the porous material (m/s), K, μ(c), and P are the permeability (m^2), the dynamic viscosity (Pa/s), and the gradient in the pressure (Pa), respectively.

For the boundary conditions, the liquid was chosen to be distilled water with a density of 1000 kg/m^3 and a dynamic viscosity of 1.002×10^{-3}. SEM image was used as a basis to represent the porous material. Firstly, it was converted to binary format using Image J software and then exported to COMSOL Multiphysics. The input pressure was selected to be the same as the operating pressure (0.5-2.5 bar). The pressure at the output side was selected to be zero. The rest of the boundaries were selected to be a wall with no-slip conditions. To have significant results, the model was made of small triangle meshes. The change in pressure over the cross-section of the plant and the water streamline were determined.

3. Results and Discussion

3.1. Characterization of Plant-Based Membrane

The morphology of the bio-based membrane was characterized. The membranes have a neural network shape. The network consists of a number of nodes that comprised start points for cylindrical rods with different lengths and sizes (Fig. 2A and B). SEM-EDX analysis was also done. The plant-based membrane is formed from 48.15% carbon and 51.85% oxygen by weight. The

obtained results for SEM-EDX are shown in Fig. 2C.

The functional groups present on the VTL surface were recorded using Fourier Transform Infrared Spectroscopy (FT/IR-6700, Jasco) and are shown in Fig. 3. The peaks extended over five bands reflect that the sample may be a complex molecule. The presence of a peak at a band 3333.36 cm^{-1}, which is located in the range 3200-3600 cm^{-1} refers to hydrogen bond O-H stretching (43). The recorded peaks under 3000 cm^{-1} indicate the presence of aliphatic compounds. The functional group, C-H stretch for CH$_2$ and CH$_3$ groups, was noticed at 2850.27 and 2917.77 cm^{-1} bands. The mono distributed alkyne with C= C stretch was recorded at 2115.53 cm^{-1} (44,45). The carbonyl compounds (aldehydes) were noticed at the functional group, C=C stretch, at the band of 1728.87 cm^{-1}. The conjugated alkene class, which was attributed to C=C stretching, was noticed near 1633.41 cm^{-1}. Johar et al recorded strong peak at 1623.77cm^{-1} and related it to the H-O-H bending (46). The peaks at the bands 1426.10
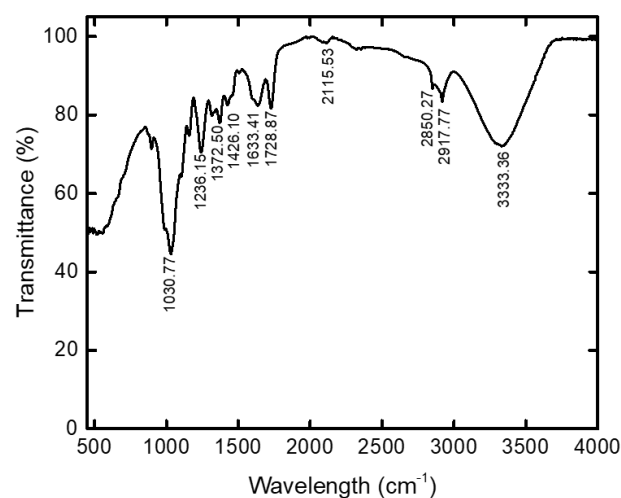

Fig. 3. FTIR Spectrum for VTL Membrane.

and 1372.50 cm^{-1} may correspond to O-H functional group. The strong peak at 1030.77 cm^{-1} may be related to sulfoxide class with functional group, S=O stretch. Yalvaç et al recorded similar results for VTL (38).

3.2. Plant-Based Membrane Permeability for Distilled Water

The permeability of the plant-based membrane was determined by measuring the distilled water passed through the membrane under different transmembrane pressure. Water flux increased at higher transmembrane pressure from 339 L/m^{2}h at 0.5 bar to 1387 L/m^{2}h at 2.5 bar. The permeability of the plant-based membrane reached 580 L/m^{2}hbar, and the membrane resistance coefficient was 5.8×10^{11} m^{-1}. Similar results were obtained in previous studies (47,48). The linearity between the pressure and the water flux is shown in equation 1, and Fig. 4 infers that the flux can reach the maximum values at the highest potential transmembrane pressure. The water contact angle for the VTL plant was found to be 112±2°. According to Cheryan, the above-mentioned result is valid for the distilled water only where the risk of fouling is negligible. In the case of natural water/wastewater, the operation at higher pressure may exacerbate the membrane fouling (49).

3.3. Effect of the Operating Pressure on the Rejection and the Permeate Flux

The fluxes of the plant-based membrane at different transmembrane pressures (0.5-2.5 bar) were recorded and shown in Fig. 5A. The fluxes at the steady state were convergent, but in general, the fluxes increased with the increases in the pressure. The steady-state flux was 45 L/m^{2}h at 0.5 bar, while the flux at 2.5 bar reached 50 L/m^{2}h. The membrane capabilities in the COD, protein, and carbohydrate reduction at different operational pressures were also explored (Fig 5B). Accordingly, the maximum COD removal was obtained at a transmembrane pressure of 2.5 bar and reached 57%. The minimum rejection was 13% at a pressure of 0.5 bar. For the protein and carbohydrate rejection, the membrane showed high rejection performance at all operating pressures. In both, the rejection increased with the increases in the applied pressure. The protein rejection raised from 80% at 0.5 bar to 90% at 2.5 bar. The rejection of protein showed a low increment at pressures 1.0, 1.5, and 2.0 bar. The rejection of carbohydrates increased slowly with the increases in the pressure (from 84% at 0.5 bar to 89% at 2 bar). Then, the removal increased to reach 98% at 2.5 bar.

3.4. Plant-Based Membrane Fouling Resistance and Flux Recovery

The hydraulic resistance for the membrane due to the fouling aspect manifested as a decline in the permeate flux in the case of fixed operation pressure. Or as an increment in the transmembrane pressure in case of constant permeate flux(50). The total fouling resistance (R_t) may be divided into reversible (R_r) and irreversible resistance (R_{ir}) (51). Fig. 6A shows the total, reversible, and irreversible fouling resistance at different transmembrane pressures. The total resistance increased from 87% at a pressure value of 0.5 bar to 96% at 2.5 bar. The irreversible resistance (R_{ir}) at a pressure of 0.5 bar was at the minimum
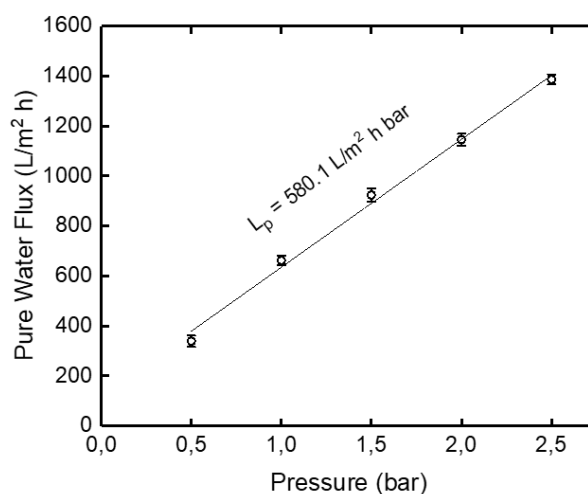

Fig. 4. Distilled Water Flux at Distinct Pressure Values.
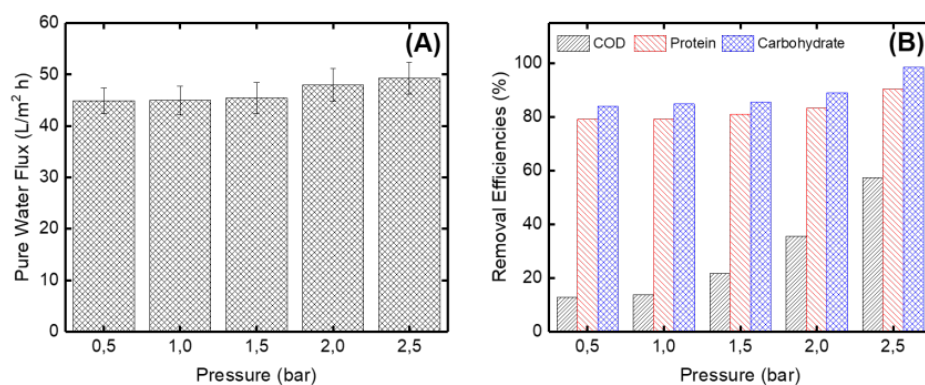

Fig. 5. (A) Inclination-dependent Flux Profile for the Activated Sludge Under Different Pressure Values, (B) COD, Protein, and Carbohydrate Removal Efficiencies Under Different Pressure Values.
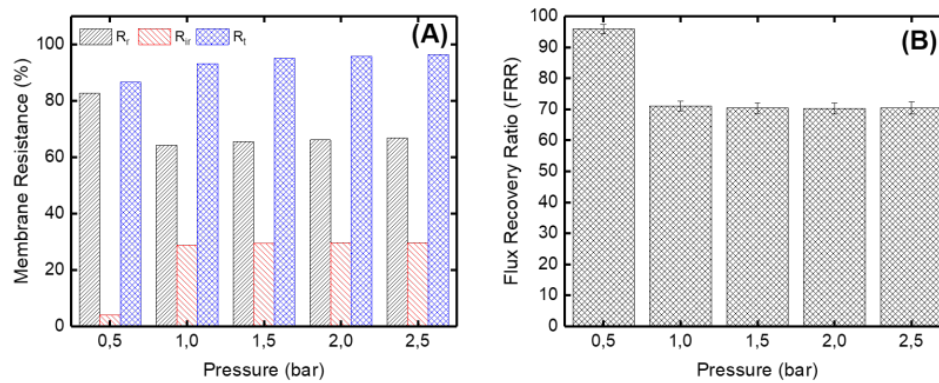
Fig. 6. (A) Fouling Resistance for the Activated Sludge Under Different Pressure Values (B) Flux Recovery Rate for the Activated Sludge Under Different Pressure Values.

level (4%), while the reversible resistance (R_r) was at the maximum level (83%). In contrast, R_{ir} and R_r almost were 30% and 66% at an operational pressure of 2.5 bar.

The flux recovery ratios (FRRs) for the membrane at working pressures (0.5-2.5) were calculated by obtaining the flux at the end of the filtration experiments. Fig. 6B shows the recovery of the flux at the pressure of 0.5-2.5 bar. Accordingly, FRR had an inverse relationship with the transmembrane pressure. At lower pressure values (0.5 bar), FRR was approximated to be 96%, while the values decreased to reach 70% at the pressure of 2.5 bar.

3.5. Physical and Chemical Cleaning of Biomembrane

The fluxes declined in three stages. For the first 10 minutes, fluxes decreased rapidly, followed by gradual flux decreases to reach the steady state. The membrane pore blocking is the mechanism that causes rapid initial drop, which incremented with the increases in the applied differential pressure (52). After that stage, the cake layer formation at the membrane surface demonstrated additional permeate flux resistance, followed by gradual inclination over the filtration time (53).

Membrane cleaning is a way to recover the flux and eliminate fouling of the membranes. The cleaning was carried out using physical and chemical cleaning methods. According to Katsoufidou et al, physical cleaning is the simplest way to recover the flux (54). However, the flux may not be recovered by the physical cleaning alone (55). Chemical cleaning should be applied to remove the organic and inorganic materials, which cannot be removed via physical cleaning (56). In this study, physical cleaning was firstly applied by distilled water filtration for 10 minutes. Then, the plant-based membrane was cleaned chemically using 0.1 M sodium hydroxide followed by distilled water. Subsequently, activated sludge was filtered again for 120 minutes. The physical cleaning showed a flux recovery after three operation cycles. The initial and steady-state fluxes were 695 and 20 L/m²h, respectively. The flux recovery after chemical cleaning was the same after physical cleaning. The initial flux after the chemical cleaning was 585 L/m²h and gradually dropped to reach

14.8 L/m²h at the end of the filtration experiments (Fig. 7).

3.6. Model Results

The streamlines of the water flow through the plant-based membrane are shown in Fig. 8. The streamlines flow from the inlet to the outlet. According to the SEM images of the plant-based membrane sample, the water is collected at specific points where the flow rate is greater and the membrane is present in two locations. The pressure gradient through the membrane exposed to the pressure of 2.5 bar is larger than the pressure gradient for the same membrane under the pressure of 0.5 bar (Fig. 8). explained the increase in flux with the pressure increase.

3.7. Comparison with other Biomembranes

The prepared VTL membrane was compared with other biomembranes. Different types of membranes were used in separation of different types of materials. Table 1 shows the biomembrane application.

4. Conclusion

In this study, VTL was successfully utilized as a membrane for activated sludge separation. Based on the SEM images, the membranes have a neural network
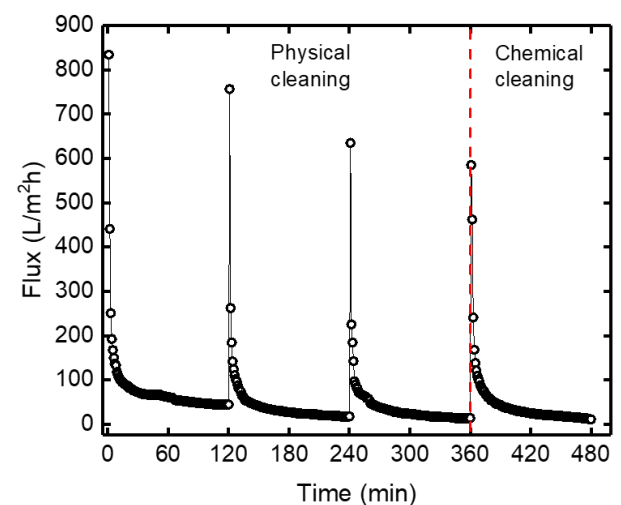

Fig. 7. Effects of Physical and Chemical Cleaning on the Flux Recovery.

Table 1. Comparison Between Different Types of Biomembranes

Membrane	Application	Efficiency	Reference
VTL based- membrane	Activated sludge (removal of COD, protein, and carbohydrate)	COD: 57% Protein: 90% Carbohydrate:98%	This study
Aspergillus carbonarius M333	Textile wastewater	Color: 91%	(48)
Bio-based polymers fabricated from isosorbide-incorporated poly (arylene ether)s	Water treatment	BSA >99%	(57)
Bio-derived ultrathin membrane	Solar-driven water purification	Organic pollutant >99.8%	(58)
Bio-ceramic hollow fiber membranes prepared from cow bone waste	Future perspective: seawater desalination	-	(59)
Bio-inspired membrane with adaptable wettability	Oil/water separation	Oil purity >99%	(60)
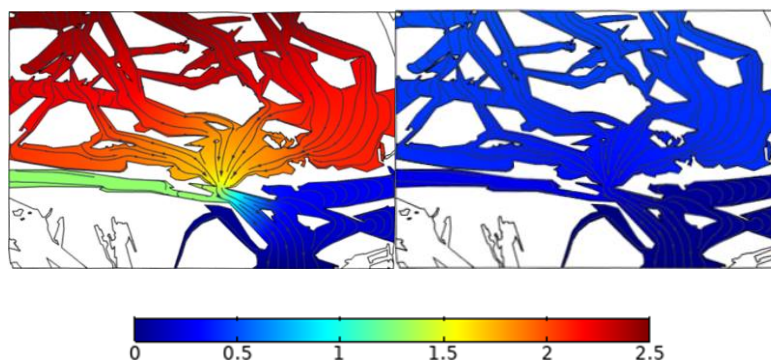
Fig. 8. Pressure gradient and the stream line at 2.5 bar (Left side) and 0.5 bar (right side) over the plant-based membrane.

shape. The network consisted of a number of nodes, which comprised start points for cylindrical rods with different lengths and sizes. The VTL-based membrane was examined for the removal of COD, protein, and carbohydrate. The plant-based membrane showed a high performance in activated sludge separation, especially in protein and carbohydrate rejection. The total resistance increased from 87% at 0.5 bar to 96% at 2.5 bar. The FRR for the membrane was 70% at 2.5 bar. The physical cleaning showed a flux recovery after three operation cycles without needing any chemicals. Additionally, the pressure gradient over the plant-based membrane cross-section was modeled. The utilization of the plant as a membrane can be considered as an eco-friendly application from different perspectives. The first one is to reduce the chemicals needed in the membrane fabrication process, which leads to a reduction in the cost. Moreover, the use of plants with no commercial value may reduce the air pollution that originated from similar plant burning. However, the plant-based membranes still need further researches before entering the application stage from sustainability and durability aspects. In addition, the synthesis of nanomaterials should be studied to increase efficiency.

Conflict of Interest Disclosures
The authors declare that there are no known conflicts of interest associated with this publication.

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