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Polydopamine-coated graphene oxide nanosheets embedded in sulfonated poly(ether sulfone) hybrid UF membranes with superior antifouling properties for water treatment

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ABSTRACT

A novel high-performance hybrid ultrafiltration (UF) membrane was fabricated by blending polydopamine-coated graphene oxide (PDGO) nanosheets with sulfonated poly(ether sulfone) (SPES) *via* phase inversion method and tested for the removal of natural organic matter (humic acid; HA) from aqueous solution. The PDGO nanosheets were synthesized *via* self-polymerization of dopamine with GO nanosheets in alkaline tris-buffer solution at room temperature for 24 h and were fully characterized. Hybrid SPES membranes were prepared by incorporating 1–10 wt.% of PDGO, which were further characterized by Raman spectroscopy, surface zeta potential, and field emission scanning electron microscopy to confirm membrane stability without any defects even by adding up to 10 wt.% of PDGO nanosheets. The membranes demonstrated a significant increase in hydrophilicity, water flux, and retention rate for HA (R_{HA}). For instance, water permeability with 5 wt.% PDGO (M5) ($680.7 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) was ca. 1.8-folds that of the pristine SPES membrane ($380.8 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$), while maintaining an HA rejection (R_{HA}) of 91.7% for a 50 ppm HA feed solution. This was accompanied by a distinct increase in surface hydrophilicity of M5, which showed a water contact angle of 27.8° , well below that of pristine SPES membrane (59.1°). The hybrid UF membranes also demonstrated a significant reduction in HA adhesion onto the membrane surface along with a superior antifouling performance for the membrane containing 10 wt.% PDGO, giving irreversible fouling ratio (R_{ir}) of only 6.9% compared to 32.7% for the pristine membrane.

Keywords: Polydopamine; Graphene oxide; Sulfonated poly(ether sulfone); Antifouling membrane; Ultrafiltration

1. Introduction

Rapid population growth, industrialization, and climate change have led to greater demand for freshwater supply, which is expected to increase by 20-30% by around 2050 [1,2]. This situation is further exacerbated by the continuous discharge of organic pollutants into the natural water bodies deteriorating the water quality [3]. Almost 50–60% of natural organic matter (NOM) in the surface water is HA [4,5], which, upon chlorination of water, generates carcinogenic disinfection by-products (DBPs) [6,7]. In order to mitigate water pollution, various environmental protection agencies have adopted strict water quality standards requiring the removal of HA to avoid the formation of DBPs. Among the many purification techniques, ultrafiltration (UF) is of particular interest to remove HA from the contaminated surface water, due to its lower operation costs compared to other treatment processes [8–10]. Despite the growing proliferation of UF technology, its application in water treatment is still limited due to membrane fouling such as adhesion of HA onto the membrane surface that can block membrane's pores and ultimately deteriorate its water permeability [5,11]. Increasing the hydrophilicity of the membrane can help mitigate membrane fouling caused by the relatively hydrophobic NOM compounds [5,9,10]. Surface coating, graft polymerization, and layer-by-layer deposition techniques have all been utilized in membrane surface modification to increase its hydrophilicity [12,13], albeit with technical challenges. Even though surface coating or grafting may not alter the membrane's internal pores, they do reduce its surface pore size significantly. On the other hand, the complex steps involved in graft polymerizations increase the production cost of the membrane [14].

Another pathway to increasing membrane hydrophilicity to boost its antifouling performance is *via* incorporation of hydrophilic nanofillers into the bulk polymer matrix to fabricate hybrid UF membranes. This approach can lead to increased membrane flux due to the creation of new low-resistance pathways for water permeability, which are created through the

interfacial gaps between the imbedded nanofillers and the polymer molecular chains in the hybrid membranes [15]. Several types of hybrid membranes with improved water permeability and antifouling performance have been prepared by imbedding SiO₂, graphene oxide (GO), TiO₂, Zn-MOF, WS₂, and SnO₂-GO nanofillers into different polymeric matrices [16–21]. However, most nanofillers are made of inorganic materials, which are incompatible with the membrane-forming polymers, and hence, a nanofiller loading of higher than 2 wt.% is often reported to cause the agglomeration within the membrane matrix, thereby deteriorating its retention efficiency [15,22]. Consequently, efforts have been made to chemically modify the nanofillers to introduce water holding functional moieties (e.g., –NH₂, –SO₃H, –⁺N(CH₂)₂–SO₃[–]) onto the nanofillers. This approach was tested using several inorganic nanofillers, including SBA-15, GO, titanium dioxide nanotubes (TNTs), CuO, ZnO, and carbon nanofiber, which were utilized in the fabrication of hybrid UF membranes via non-solvent induced phase separation [8,23–27]. Polymer-anchored nanofillers have also been developed via free radical, reversible addition–fragmentation chain-transfer, and surface-initiated atom transfer radical polymerization, which were then blended into the bulk polymer matrices to fabricate hybrid membranes with improved filtration performances [28–30]. However, the synthesis of polymer-grafted nanofillers via these pathways is rather challenging because of the complicated reaction steps involved and the required oxygen-free reaction environment [14]. A simpler alternative involving thin polymer layer coated nanofillers can potentially overcome these challenges.

Polydopamine (PDA) has received increased interest in various fields owing to its versatile adhesion capability, biocompatibility, and hydrophilicity and was used in membrane surface modification without affecting its pore geometry [31,32]. A thin layer of PDA is formed on various substrates *via* the self-polymerization of dopamine (Dopa) in a slightly alkaline tris-buffer solution, introducing the water holding phenolic –OH and –NH moieties onto the

surface [31,32]. Polydopamine-functionalized graphene oxide (rGO-PDA), PDA-GO, PDA and GO have been utilized in the fabrication of different types of membranes for water treatment [33-35]. Alkhouzaam and Qiblawey manufactured PSf hybrid UF membranes using low amounts rGO-PDA via the non-solvent induced separation (NIPS) method [33], but used un-functionalized PSf for membrane fabrication. The flux of the hybrid membrane containing 0.15 wt.% rGO-PDA was $326.5 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$. Alkhouzaam et al., utilized a pressure assisted self-assembly method to fabricate PSf/PDA-GO thin film composite membrane by depositing PDA-GO on PSf substrate [34]. Chen et al. grafted GO covalently onto the surface of UF membranes through a PDA layer mediation. The grafting of GO onto the surface of UF membranes was accomplished in two steps by coating of PDA layer and the subsequent EDS-NHS coupling reaction in an alkaline medium [35].

In this research, we fabricate a new anti-fouling, high-flux, and high HA rejection hybrid UF membrane through the synergic integration of PDA-coated nanofillers within a charged polymer matrix. Since the solute rejection of hybrid membranes has been found to be enhanced by using the charged polymers in membrane fabrication [36–38], the integration of a charged polymer matrix with functional moieties that can induce additional surface charge of the membrane, would lead to hybrid membranes with superior performance at multiple levels including permeability, HA rejection, and fouling resistance. To accomplish this, we synthesized PDA coated GO (PDGO) via self-polymerization of Dopa in alkaline tris-buffer solution at room temperature (23°C). The GO nanosheets were specifically chosen because of their easy synthesis, low cost, hydrophilicity, and the abundant oxygen functional moieties they contain (–OH, –COOH, and epoxide) [22,26,29,38]. The phase-inverted hybrid UF membranes were prepared by combining sulfonated poly(ether sulfone) (SPES), a negatively charged polymer with hydrophilic PDGO nanosheets, to acquire higher hydrophilicity to the barrier layers due to enriched water-holding functional groups such as –SO₃H, –COOH, –OH, and

–NH, while the PDA thin layer promoted a better dispersion and high loading of the PDGO nanosheets into the SPES membrane matrix, resulting in a well dispersed nanosheets, even at PDGO loading up to 10 wt.%. These membranes were thoroughly characterized and their performance was demonstrated in the removal of HA from an aqueous feed mixture.

2. Materials and methods

2.1. Materials

Poly(ether sulfone) (PES; M_w : 58 kDa) was purchased from Goodfellow Co. (UK) and dried in a vacuum oven at 100°C prior to sulfonation. Poly(vinylpyrrolidone) (PVP; M_w : 10 kDa), technical grade HA, dopamine hydrochloride (Dopa. HCl; $\geq 99\%$), tris(hydroxymethyl) aminomethane (Tris; $\geq 99.9\%$), N-methyl pyrrolidone (NMP; 99%), graphite powder (average size < 20 μm), potassium permanganate (KMnO_4 ; 99%), phenolphthalein, sulfuric acid (H_2SO_4 ; 96%), hydrogen peroxide (H_2O_2 ; 30% w/v), sodium hydroxide (NaOH), and hydrochloric acid (HCl; 36.5%) were procured from Sigma-Aldrich (Germany). Absolute ethanol ($\text{C}_2\text{H}_5\text{OH}$; 99.95%) was procured from VWR Co. (UK). Sodium chloride (NaCl), calcium chloride dihydrate ($\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$; 98%), and phosphate buffer saline (PBS) were supplied by Merck Chemicals (Germany). All the chemicals and reagents were used as received without further purification. Ultra-pure water, produced from a Millipore Milli-Q Plus 185 purification system (Millipore Corp., USA) was used. GO nanosheets were synthesized in our lab and 100 ppm HA solution was prepared according to our previously published paper [38].

2.2. Preparation and characterization of sulfonated poly(ether sulfone) (SPES)

SPES was prepared *via* direct sulfonation of PES in 96% H_2SO_4 at 60°C with a slight modification in the reaction conditions [38,39] (**Fig. S1**, Supporting Information). The details for synthesis and characterization of SPES are included in Section S1.

2.3. Preparation of polydopamine decorated GO nanosheets

PDGO nanosheets were produced *via* self-polymerization of Dopa-HCl with GO nanosheets in pH = 8.5 tris-buffer solution [31,40]. The details of the synthesis of GO nanosheets have been published previously [38]. Briefly, GO (0.5 g) was initially added into a mixture of 250 ml tris-buffer solution (50 mM) and absolute C₂H₅OH (4:1 v/v), in a neck round bottom flask, and bath-sonicated for 1 h to obtain a well-dispersed suspension. Then, Dopa-HCl (1.0 g) was dissolved in the above suspension by mechanical agitation at 100 rpm, followed by stirring for 24 h on a magnetic stirrer plate. Subsequently, PDGO nanosheets were recovered by vacuum filtration, purified by dispersion and centrifugation in water or absolute ethanol for 30 min at 8000 rpm (this step was repeated 3 times). The well-dispersed suspension of PDGO in C₂H₅OH was added into a glass Petri dish and placed inside an airflow fume hood for 12 h at 23°C (room temperature). The Petri dish, with the black PDGO nanosheets was then placed in a vacuum oven for 8 h at 50°C. The PDGO was then stored in airtight glass vial for further characterizations and application in membrane fabrication.

PDGO was characterized using several techniques as detailed in **Section S2**, Supporting Information. The coating of PDA in PDGO nanosheets was determined from the thermogravimetric analysis (TGA, Section S2) using the following equation:

$$\text{Coating of PD (\%)} = \frac{W_{GO} - W_{PDGO}}{W_{GO}} \times 100 \quad (1)$$

where W_{GO} and W_{PDGO} are the weight residues of GO and PDGO at 700°C (expressed as % of the original sample weight).

2.4. Fabrication and characterization of SPES-PDGO hybrid UF membranes

Hybrid membranes with different loadings of PDGO nanosheets were fabricated using SPES, PVP, and PDGO through the non-solvent induced phase separation (NIPS) process [37,38,41]. Compositions of the dope solutions used in membrane casting are summarized in **Table 1**. A predetermined quantity of PDGO was initially charged into an airtight 50 ml glass

bottle containing NMP and ultrasonicated in a bath sonicator for 30 min at 40 kHz frequency. Then, PVP (2.0 g) and SPES (18 g) were alternatively added and dissolved in the suspension at 200 rpm for 2 h at 60°C. The dope solution was then stirred without heating for an additional 12 h. The trapped air bubbles were removed *via* ultrasonication for 10 min. Using an automatic film applicator (Elcometer 4340, UK) and a metallic casting knife with a gate height of 180 μm , the bubble-free dope solution was then uniformly cast onto a nonwoven polyester support (Novatexx 2471 M, Freudenberg, Germany) mounted atop a glass plate. After 30 s standing time, the glass plate with the cast polymer film was immersed in DI water coagulation bath and left for an additional 1 h. The formed membrane was then transferred to a fresh DI water bath for 48 h to eliminate any residual solvent and water-soluble PVP. Hybrid membranes with 1, 3, 5, and 10 wt.% PDGO were fabricated, which were labeled M1, M3, M5, and M10, respectively. A pristine membrane (M0) was also prepared, for performance benchmarking, from a dope solution without PDGO. Free-standing (without support) membranes were also fabricated for the purpose of conducting cross-sectional SEM and thermal stability analysis. The first indication of a homogeneous PDGO incorporation into the hybrid membranes was obtained from their digital photographs (**Table 2**). The color of the membrane changed from colorless M0 to a uniform light brown upon PDGO loading. The brownish color intensified with higher PDGO loading, although it remained uniform (i.e., no color grading) throughout the membrane surface. An attempt was also made to fabricate the hybrid membranes with PDGO loading higher than 10 wt.%, but the formed membranes showed the defects because the high loading of PDGO caused the casting solution to be too viscous to produce a uniform membrane *via* phase inversion.

The membrane porosity was quantified following a typical gravimetric method [22,23,42]. A membrane coupon ($2 \times 2 \text{ cm}^2$) was soaked in a water bath and excess water was wiped off the membrane surface using a Kimwipe paper. The weight of the wet membrane

sample was measured using a digital analytical balance (Ohaus, UK). The membrane was dried in a vacuum oven at 50°C for 24 h and the weight of the dry membrane was noted. The porosity of the membrane was calculated according to the following equation [22,23,42]:

$$\epsilon (\%) = \frac{W_w - W_d}{A \times l \times \rho_{water}} \times 100 \quad (2)$$

where W_w is the weight of wet membrane sample (g), W_d is the weight of dry membrane (g), A is the area of wet membrane (cm²), l is the thickness of the membrane (cm), and ρ_{water} is the density of water (1.0 g cm⁻³). At least triplicate samples were tested and the average values were reported.

The mean pore radius of the membrane (r_m ; nm) was determined from its pure water flux and porosity using the Guerout-Elford-Ferry equation [42,43]:

$$r_m = \sqrt{\frac{(2.9 - 1.75\epsilon) \times 8\eta l Q}{\epsilon \times A \times \Delta P}} \quad (3)$$

where η is pure water viscosity (8.9×10^{-4} Pa s), Q is permeation volume of pure water per unit time (m³ s⁻¹), A is the effective membrane area (12.5 cm²) and ΔP is the operation pressure (0.1 MPa).

The top surface and cross-sectional morphologies of the membranes were visualized using a high-resolution field emission scanning electron microscope (FESEM, Nova Nano 650, FEI Co.) operated at different acceleration voltages and magnifications. A multi-step solvent exchange procedure was followed for drying the membrane samples before imaging to prevent the collapse of the pores [22]. The dried membrane coupons were mounted on the SEM stubs using a double-sided adhesive carbon tape. Specifically for the cross-sectional SEM images, the free-standing membrane coupons were frozen in liquid N₂ and fractured using a tweezer. All membrane coupons were coated with Au-Pd ultrathin layer (10 nm) in a sputtering machine.

The surface and cross-sectional images were collected at 18 and 30 kV accelerating voltage, respectively.

The surface topography of the membranes was characterized using an atomic force microscope (AFM, CSI, France) connected with a silicon nitride cantilever. The top surface AFM images were recorded in a tapping mode, at a $3\ \mu\text{m} \times 3\ \mu\text{m}$ scanning size. The average surface roughness (r_a ; nm) values were also obtained from the three-dimensional (3D) AFM images using the Gwydion software. The hydrophilicity of the membrane was assessed via contact angle (CA) measurements using a goniometer (Krüss, Germany), following the sessile drop method. A $5.0\ \mu\text{L}$ drop of DI water was dispensed, from an automated micro-syringe with a needle (Hamilton Micro-syringe), on the top surface of a dry membrane coupon fixed atop a glass slide. Then, the CA between the membrane surface and water droplet was determined by the Drop Shape Analysis software from the captured digital images. CA measurements were carried out at five random locations of the membrane sample and the average CA value was reported. The surface free energy (SFE; $-\Delta G_{SL}\ \text{mJ m}^{-2}$) was also calculated from the CA value using the Young-Dupre equation [42,44]:

$$-\Delta G - \Delta G_{SL} = (1 + \cos\theta)\gamma_L^T \quad (4)$$

where θ is the water CA (deg.) and γ_L^T is the surface tension of water ($72.8\ \text{mJ m}^{-2}$).

Nanosheets within the hybrid membrane matrix were identified using a Raman spectrometer (Witec Alpha 300 RAS) fitted with a He-Ne laser (634 nm) and $50\times$ objective lens. The laser beam was focused at the center of the dry membrane sample, fixed on a glass slide, and the acquisition time was set at 30 s.

The surface zeta potential (ζ ; mV) of the membrane was measured using a SurPass Electrokinetic Analyzer (Anton Paar GmbH, Austria). A membrane coupon ($20\ \text{mm} \times 10\ \text{mm}$) was first immersed in $1.0\ \text{mM}$ KCl solution for 24 h, wiped using a Kimwipe paper, fixed on a sample holder using a double-side adhesive silicon tape, then plugged into a cell with a gap

height of 120 μm . The cell was then fixed onto the Electrokinetic analyzer and recirculated with 1.0 mM KCl solution ($\text{pH} = 7$) at 400 mbar for 200 s. Four measurements were made and averaged for each membrane at a $\text{pH} = 7$ and target pressure of 200 mbar. The ζ was then estimated using the Helmholtz-Smoluchowski equation [42,45]. Thermal stability of the membranes was measured using a TGA with a temperature range of 50–750°C at the heating rate of 10°C min^{-1} under a N_2 environment.

The mechanical strength of the membranes was tested using a tensile strength testing machine (5960 series, Instron, USA) to obtain the bursting pressure of each membrane.

2.5. Pure water permeability and filtration performance assessment of the UF membranes

Pure water permeability of the membranes was measured using a stirred dead-end ultrafiltration system (UHP 4370, Sterlitech Co., USA). The wet membrane (4.9 cm in diameter) was first installed in a UF cell connected to a compressed N_2 gas cylinder and 5 L stainless steel reservoir (**Fig. S2c**, Supporting information). The membrane was pre-compacted by filtering DI water at 2.0 bar for 60 min to reach a stable flux. The pressure was then reduced to 1.0 bar and filtration was continued for another 30 min, during which the collected permeate weight was measured using an analytical balance (Ohaus, UK). The pure water permeability (J_w ; $\text{L m}^{-2} \text{h}^{-1} \text{bar}^{-1}$) was calculated using:

$$J_w = \frac{W}{\rho_w \times A \times \Delta t \times \Delta P} \quad (5)$$

where W is the permeate weight (g), ρ_w is the density of water (1.0 g cm^{-3}), A is the membrane area (m^2), Δt is the filtration time (h), and ΔP is the feed pressure (bar).

HA solutions of various concentrations (5 to 100 ppm) were initially filtered through the hybrid membrane M5 at 1.0 bar feed pressure to find a suitable concentration of the foulant

solution for assessing HA rejection (R_{HA} ; %) and antifouling capability of the hybrid membranes during long-term filtration operation. The obtained results are depicted in **Fig. S3d**, Supporting Information. The membrane M5 maintained an R_{HA} value of 96.4 – 91.7% for all HA feed solutions in the concentration range from 5 to 50 ppm. However, R_{HA} was dropped to 79.3% when 100 ppm HA solution was used. Hence, a 50 ppm HA solution was henceforth utilized. The solution contained NaCl (7 mM) and $CaCl_2$ (1.0 mM), a composition that is well supported by previously reported membrane fouling studies in the literature [46-48]. The antifouling capability of the membrane was studied using the 50 ppm HA solution as follows: First, DI water was filtered through the membrane for 30 min at 1 bar and pure water permeability (J_w ; $L\ m^{-2}\ h^{-1}\ bar^{-1}$) was measured as detailed above. Second, an HA solution was filtered for 3 h at 1.0 bar pressure and, 300 rpm mechanical stirring and its permeability was recorded (J_{HA} ; $L\ m^{-2}\ h^{-1}\ bar^{-1}$). Finally, the used membrane was thoroughly washed inside the filtration cell with DI water-PBS solution for 30 min at 300 rpm stirring speed, after which the post-cleaning water permeability ($J_{w,pc}$; $L\ m^{-2}\ h^{-1}\ bar^{-1}$) was measured by filtering DI water for another 30 min at 1.0 bar pressure. From the three permeability values, three key parameters; the flux recovery ratio (FRR; %), the reversible fouling ratio (R_r ; %) and the irreversible fouling ratio (R_{ir} ; %) were calculated as follows [20,38,49]:

$$FRR\ (\%) = \frac{J_{w,pc}}{J_w} \times 100 \quad (6)$$

$$R_r\ (\%) = \frac{(J_{w,pc} - J_{HA})}{J_w} \times 100 \quad (7)$$

$$R_{ir}\ (\%) = \frac{(J_w - J_{w,pc})}{J_w} \times 100 \quad (8)$$

The membrane's HA rejection rate (R_{HA} ; %) was also determined as follows [20,42]:

$$R_{HA} (\%) = \frac{(C_{feed}^0 - C_{permeate}^t)}{C_{feed}^0} \times 100 \quad (9)$$

where (C_{feed}^0) and ($C_{permeate}^t$) are the concentrations of HA in the feed and permeate solutions in ppm, respectively, and are determined from the absorbance of the solution at 254 nm using a UV-Vis spectrophotometer (UV-1800, Shimadzu, Japan).

3. Results and discussion

3.1. Synthesis and characteristics of PDGO nanosheets

PDGO nanosheets were produced *via* self-polymerization of Dopa on GO nanosheets in alkaline tris-buffer solution in the presence of atmospheric oxygen (**Scheme S1**, Supporting information). The self-polymerization of Dopa and subsequent coating of a thin layer of PDA on GO were performed in three steps: (i) the deposition of Dopa molecules onto the GO through generated $NH_3^+COO^-$ ion-pairs; (ii) the construction of PDA precursors *via* oxidative polymerization of Dopa in the presence of O_2 and (iii) the strong adhesion of PDA thin layer onto the GO with further growth of PDA precursors during self-polymerization of Dopa for 24 h at 23°C. At the end, PDGO nanosheets with phenolic –OH and –NH moieties were produced [32,40,50]. The decoration of PDA thin-layer on GO was identified through the ATR-FTIR spectra of GO and PDGO nanosheets, shown in **Fig. 1a**. The broad peak at 3395 cm^{-1} in the FTIR spectrum of GO is associated with the stretching vibration of –OH moieties, whereas the peak at 1730 cm^{-1} is the stretching vibration of C=O moieties in GO. The characteristic peak at 1620 cm^{-1} demonstrates the aromatic stretching vibration of C=C moieties. The absorption bands at 1210, 1047 and 985 cm^{-1} are attributed to the stretching vibration of C–O (epoxy), C–O (alkoxy) and C–O–C, respectively [22,39,42]. Upon the decoration of PDA thin-layer on GO nanosheets, the characteristic peaks for the stretching vibration of O–H, C=O, aromatic C=C and C–O (alkoxy) moieties are slightly shifted and located at 3340, 1710, 1592 and 1028

cm⁻¹, respectively.

New peaks have emerged in the ATR–FTIR spectrum of PDGO while a narrow peak at 1710 cm⁻¹ for C=O stretching vibration is identified (**Fig. 1a**). The absorption peaks from 3130 to 2927 cm⁻¹ belong to stretching vibration of N–H moieties. The peaks at 1430 and 1270 cm⁻¹ are attributed to stretching vibration of C–N moieties in indoles and deformation vibration of N–H moieties. The peak at 1345 cm⁻¹ is attributed to the bending vibration of phenolic C–OH moieties of PDA thin-layer. These characteristic peaks suggest the successful decoration of PDA thin-layer on GO and the formation of PDGO nanosheets as depicted in **Scheme S1**. The bands for C=C and N–H moieties at 1590 and 1509 cm⁻¹ couldn't be distinguished due to the overlap of aromatic stretching vibration peaks of C=C in GO [40,50,51].

TGA was conducted to determine the PDA content in PDGO. As depicted in **Fig. 1b**, the decomposition of GO and PDGO occurred in three stages between 50 –700 °C, as deduced from the three distinct stages of weight loss observed. The first stage of weight loss, between 50 – 150 °C, was due to the evaporation of bounded water [51,52], whereas the decomposition of oxygen-containing moieties caused the second stage of weight loss, between 150 – 300 °C. The third stage (600 – 700 °C) exhibited a sharp weight loss for PDGO compared to GO and is attributed to the PDA decomposition. [38,51]. The weight percent of GO and PDGO residues at 700 °C were 38 wt.% and 28.4 wt.%, respectively. The coating of PDA in PDGO nanosheets was calculated to be 25.3% (as per eqn. (1)).

The ζ of PD, GO, and PDGO was determined from their aqueous suspensions and found to be –19.7, –25.4, and –34.2 mV, respectively (**Fig. S2d**, Supporting information). The charge in terms of ζ on PD is attributed to the deprotonation of phenolic O–H and indole N–H of PD, whereas the charge on the GO can be attributed to the deprotonation of oxygen-containing functional moieties (–COOH, phenolic –OH) [53]. The synergistic effect of PD and GO is

evident in the increased ζ , thanks to the enriched phenolic $-\text{OH}$ and $-\text{NH}$ moieties on GO upon PDA coating.

The surface morphologies and microstructures of GO and PDGO were studied using FE-SEM and TEM imaging (**Fig. 2(a-d)**). The FE-SEM images display the transformation of GO microstructure from an exfoliated sheet-like structure with some wrinkles to a rougher surface upon the PDA thin-layer coating. TEM image of PDGO indicates a slightly thick sheet with a smaller size. This can be attributed to the size reduction of GO during ultrasonication in tris-buffer solution, the decoration of PDA thin-layer, and an adequate interfacial interaction among GO and PDA thin-layer [40,51–53]. The characterization results thus far collectively corroborate the synthesis of PDGO nanosheets.

3.2. The presence of PDGO nanosheets in the hybrid membranes

As depicted in **Fig. 3**, two types of interactions can be observed between PDGO nanosheets and the SPES polymer matrix, suggesting a good dispersion of the former in the latter: i) the hydrogen bonding interactions between $-\text{SO}_3\text{H}$ groups of SPES and phenolic ($-\text{OH}$) moieties of the PDGO, and ii) the electrostatic interactions between $-\text{SO}_3^-$ groups of SPES and the NH_2^+ moieties of PDGO [43,54].

The loading of PDGO nanosheets in the hybrid membranes was also confirmed via Raman spectra shown in **Fig. S3a**, Supporting information. For M0, peaks at 1077 and 1110 cm^{-1} can be assigned to symmetric and asymmetric stretching vibrations of $\text{O}=\text{S}=\text{O}$ groups [41,55], while those at 1150 and 1205 cm^{-1} are attributed to the stretching vibrations of $\text{C}-\text{O}-\text{C}$ moieties. The doublet peak at 1582 and 1603 cm^{-1} is due to the vibration of benzene rings [43,55]. Notably, the Raman spectrum of M5 displayed two distinct peaks at 1332 and 1604 cm^{-1} , which are characteristic D and G bands of PDGO nanosheets [42,43].

Further evidence of PDGO presence in the membranes can be obtained from the TGA data as shown in **Fig.S3b**, Supporting information. All the membranes (pristine and hybrid)

demonstrated three distinct weight-loss stages in the range of 50–750°C. These stages are attributed to i) the evaporation of adsorbed moisture traces (50–150°C), ii) the pyrolysis of functional moieties such as $-\text{SO}_3\text{H}$, $-\text{COOH}$, $-\text{NH}$, and $\text{C}=\text{O}$ in the membrane's matrix (300 – 490 °C), and iii) the degradation of PES and PDGO (>560 °C) [39,48,51]. However, the weight-loss of hybrid membranes in the first and second stages was slightly higher than the pristine membrane due to the integration of hydrophilic moieties ($-\text{COOH}$, $-\text{OH}$ and $-\text{NH}$) into the membrane matrix after PDGO loading. Thermal decomposition temperature (T_d , °C) was also offset upon PDGO loading. The T_d of M0 was 620 °C, while the T_d of M1, M3, M5 and M10 was 630, 636, 640, and 652 °C, respectively. The higher T_d of the hybrid membranes is attributed to the presence of the thermally-stable carbon skeleton of PDGO.

3.3. Influence of PDGO nanosheets loading on the porosity, mean pore radius and morphology features of the membranes

The ε (%) and r_m (nm) of the membranes were tuned upon the loading of PDGO (Table 2). The obtained values reveal higher ε and r_m of the hybrid membranes than the pristine one. Both ε and r_m values increased systematically with PDGO loading up to 5 wt.%, but then decreased marginally with the next-step loading of 10 wt.%. The trends of ε and r_m can be correlated with the thermodynamic instability and kinetic hindrance caused by the viscosity of casting solution at increased PDGO loading [8,27,31,38]. Up to 5 wt.% loading, the presence of PDGO with rich hydrophilic moieties ($-\text{OH}$, $-\text{COOH}$, and $-\text{NH}$) resulted in an increased hydrophilicity of dope solution. This created a thermodynamic instability and induced rapid phase separation during membrane formation, resulting in membranes with high ε and r_m values. However, this thermodynamic enhancement was curtailed at 10 wt.% loading because

of the dominant effect of kinetic hindrance, slowing down the phase separation and resulting in a slightly denser M10 membrane with lower ε and r_m values than M5 [22,26,30].

The top surface SEM images displayed in **Fig. S4**, Supporting information, show minor differences in surface morphology between the pristine and hybrid membranes. Tiny irregular pores were observed on the top surface of M0 and surface pore size as well as density values were increased with PDGO loading up to 5 wt.%. In contrast, the surface pore size slightly decreased in M10, due to the delayed phase inversion of the highly viscous M10 dope solution and subsequent hindrance in the extension of the pore structures, as discussed earlier. The cross-sectional SEM images presented in **Fig. 4** show that all the membranes have typical asymmetric porous structures with a compact skin layer and porous sublayer with finger-like structure. These finger-like pores elongated towards the bottom upon PGDO loading, which was triggered by the thermodynamic instability of the blended dope solutions and subsequent rapid phase separation, resulting in the hybrid membranes having increased porosity [22,25,31,42]. The observed variations in the morphological structures were due to the synergic effect of GO and the coated hydrophilic PDA layer of PDGO.

The 3D topology images of M0, M1, M5 and M10 are presented in **Fig. 5** and r_a values are provided in **Table 2**. The roughness of the hybrid membranes significantly increased (from 6.1 nm for M0 to 14.6 nm for M10) due to the rapid phase separation of the highly viscous dope solutions containing hydrophilic PDGO. The SPES molecular chains were most likely compacted upon contact with DI water, inducing the formation of the polymer nodules with increased roughness [20,41,42]. Furthermore, some of the hydrophilic PDGO nanosheets migrated towards the skin layers of the nascent membrane during phase inversion, as indicated by the dark brown color of the hybrid membranes at higher PDGO loading (**Table 2**), increasing the membrane surface roughness.

3.4. Influence of PDGO nanosheets loading on surface charge, hydrophilicity and mechanical stability of the membranes

The surface charge of the membranes was analyzed using their ζ at neutral pH. All the membranes had a negative ζ (**Table 2**), which increased with PDGO loading. For example, the ζ of 10 wt.% PDGO loaded M10 was -27.8 mV, which is almost two-fold higher than the M0. This is due to $-\text{SO}_3\text{H}$ moieties of the SPES polymer in the barrier layers, which is combined with the additional functional moieties ($-\text{COOH}$, $-\text{OH}$, and $-\text{NH}$) in the hybrid membranes (**Fig. 3**) [31, 37, 38, 43, 56, 57]. These functional moieties in the barrier layer are deprotonated in an aqueous medium at neutral pH, imparting an increasingly negative surface charge for the hybrid membranes [53]. The surface charge of the hybrid membranes is, therefore tunable with the extent of PDGO loading.

The hydrophilicity of the membranes was determined *via* their static water CA, provided in **Table 2**. The CA of M0 was 59.1° , demonstrating hydrophilic nature owing to the water holding $-\text{SO}_3\text{H}$ moieties on its barrier layer [8, 38, 42, 42, 56]. The PDGO loading enriched the hybrid membrane surfaces with water-holding functional moieties ($-\text{COOH}$, $\text{O}-\text{H}$, and $\text{N}-\text{H}$) [41, 57, 58], and consequently, the CA systematically decreased with PDGO loading, reaching 27.8° for M10. The presence of water-holding moieties in the surface enabled the capture and penetration of water droplets into the hybrid membrane matrix, leading to an enhanced wettability [57-60]. The mechanical stability of both membranes (pristine M0 and hybrid M10) was assessed by conducting pure water filtration at different feed pressure in the range of 1 to 4 bar. The water flux data for both membranes is depicted in **Fig. S5**, Supporting Information. The water flux was consistently increased with feed pressure up to 4 bar. The maximum PDGO (10 wt.%) loaded membrane M10 had the water flux $1138.7 \text{ L m}^{-2} \text{ h}^{-1}$, which confirmed the production of robust and mechanically stable hybrid membranes from SPES and PDGO nanosheets. The mechanical strength results of the membranes are depicted in **Fig. S6**,

Supporting Information. The results show that the mechanical strength was tunable with the loading of PDGO nanosheets into the matrix of hybrid membranes. The bursting pressure of pristine and hybrid membranes was 1.29 (M0), 1.25 (M1), 1.62 (M3), 0.56 (M5) and 0.65 MPa (M10). Both pure water flux data at feed pressure of 1 to 4 bar and the bursting pressure values confirmed that the fabricated membranes were mechanically stable for use in filtration of HA solution at 1 bar feed pressure.

3.5. Pure water and HA permeability, HA rejection and antifouling capability

The filtration performance of PDGO embedded membranes was studied by first measuring their J_w at 300 rpm and 1.0 bar (gauge) pressure. **Fig. 6(a)** shows J_w of M0 and the hybrid membranes. The J_w of M0 was $\sim 381 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$, which is relatively high owing to the high ε (65.2%) and the promotion of water permeation by the hydrophilic $-\text{SO}_3\text{H}$ moieties within the membrane matrix [38,43,56,61]. The J_w has significantly increased with PDGO loadings into the SPES membrane matrix. For example, M5 exhibited a J_w of $680.7 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$, almost 1.8-fold that of M0 (**Fig. 6a**). This increase in J_w of the hybrid membranes is primarily due to their higher ε and hydrophilicity in comparison to the pristine membrane. The water molecules captured by the hydrophilic functional moieties ($-\text{SO}_3\text{H}$, $-\text{COOH}$, $-\text{NH}$, and $-\text{OH}$) of the hybrid membranes would permeate faster through the hydrophilic channels [8,31,60,62]. However, J_w slightly dropped for M10 ($515 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$), albeit being still higher than M0. This drop is due to the slightly denser structure, reduced r_m , and reduced ε of M10, as discussed earlier (**Table 2, Fig. S4**). Another membrane with 5 wt.% loading of unmodified GO (MG) was also fabricated, and its J_w was measured for comparing to those of M0 and M5 under similar conditions (**Fig. S3c**, Supporting information). The J_w of the MG membrane was $473.3 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$, which is much lower than M5 ($680.7 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$). PDA coated GO (PDGO) nanosheets at 5 wt.% loading was liable for inducing additional

thermodynamic instability of blended casting solution and fast phase inversion during a precipitation process. As a result, highly porous membrane M5 including more tiny pores was obtained (**Fig. S4e**). DI water was filtered at a faster rate under constant feed pressure of 1 bar because the membrane M5 was more hydrophilic and large number of surface pores. On the other hand, a less porous membrane MG was produced from blended casting solution containing 5 wt.% of GO nanosheets and SPES (**Fig. S7**, Supporting Information). The density of small pores on the surface of membrane MG was declined and the size of the resulting pores slightly reduced. This could be due to the decline in thermodynamic instability of GO blended casting solution and subsequently, the phase inversion became slow. Thus, less porous membrane MG was obtained and DI water was slowly passed through this membrane MG at constant 1 bar feed pressure. Finally, significantly lower J_w was attained for membrane MG. It is worth mentioning here that the fabrication of a 10 wt.% GO (corresponding to M10) was unsuccessful because of the very high viscosity of the casting solution and the creation of pinholes in the produced membrane.

J_{HA} ($\text{L m}^{-2} \text{h}^{-1} \text{bar}^{-1}$) of the membranes using 50 ppm HA solution with NaCl and CaCl_2 was then evaluated. The J_{HA} values were well below their corresponding J_w values (**Fig. 6b**), which can be attributed to either concentration-polarization or cake-layer formation *via* the adhesion of HA molecules on the membrane surface [21,63], although the contribution of concentration-polarization can be ignored as the filtration experiments were carried out at 300 rpm stirring. Similar to J_w , J_{HA} was lowest for M0 ($167.7 \text{ L m}^{-2} \text{h}^{-1} \text{bar}^{-1}$) and systematically increased with PDGO loadings (**Fig. 6b**) due to the thick hydration layer formed on the hybrid membranes surface as a result of the presence of water-holding functional moieties ($-\text{SO}_3\text{H}$, $-\text{COOH}$, $-\text{OH}$ and $-\text{NH}$) of PDGO. This hydration layer acted to weaken the interactions between HA molecules and the hydrophilic membrane surface, resulting in the formation of a loose cake-layer and the subsequent increase in water permeability [38,43,64]. The R_{HA} of the

membranes, averaged over a 3 h filtration test, is presented in **Fig. 7**. In spite of their higher J_{HA} , the hybrid membranes showed an increased R_{HA} with PDGO loading, thanks to the electrostatic repulsion of the negatively charged HA by the similarly charged hybrid membranes (**Table 2**) [8,38,42,43,64]. The maximum rejection (92.4 %) was observed for M10 with the highest negative ζ (−27.8 mV; **Table 2**).

The antifouling properties of the membranes were assessed by examining their FRR (%). This was accomplished by filtering a 50 ppm HA solution for 3 h and subsequently washing the membrane using DI water–PBS solution, then measuring $J_{w,pc}$ of the membranes, (**Fig. 6b**). The $J_{w,pc}$ was higher for the hybrid membranes than for the pristine M0 indicating that cleaning with DI water-PBS solution efficiently detached the weakly adhered HA molecules from the hybrid membranes' surfaces. The FRR values of the pristine and hybrid membranes were then calculated (per Eqn. 6) and are presented in **Fig. 8a**. The FRR of the membranes progressively increased with the PDGO loading, reaching 93.4% for the M10 membrane. This indicates an improvement in the antifouling nature of the hybrid membranes, which is attributed to their improved hydrophilicity and the electrostatic repulsion between the negatively charged HA molecules and the membrane surface [8,31,60,62]. The membranes' antifouling capacity was then evaluated in more detail by calculating R_r and R_{ir} values using J_w , J_{HA} , and $J_{w,pc}$. The resultant R_r and R_{ir} values of all the membranes are plotted in **Fig. 8b**. The pristine membrane had the highest R_{ir} value (32.7%), indicating the strong, irreversible adhesion of a portion of the HA molecules onto the surface, owing to the latter's low hydrophilicity. The R_{ir} of the hybrid membranes was considerably and consistently reduced upon incorporating more loadings of PDGO. For instance, M10 exhibited the lowest value of R_{ir} (6.9 %). On the other hand, the R_r values of the hybrid membranes were significantly increased with increased PDGO loading (**Fig. 8b**). The adhesion of HA molecules and the subsequent development of a robust cake layer on the membrane surface are believed to be

minimized in the hybrid membranes with high PDGO loading due to the formation of a thick hydration layer. The loosely attached HA molecules and cake layer were effectively washed off the surface of the hybrid membrane, resulting in a substantial improvement in the R_f values for all these membranes. The surface SEM images of membrane M10 before and after UF of 50 ppm HA solution followed by PBS solution cleaning were recorded to confirm the minimum adhesion of HA molecules and effective washing-off the surface of the hybrid membrane (**Fig. S8**, Supporting Information). A slight variation in surface texture of membrane M10 was observed after UF of 50 ppm HA solution and PBS solution cleaning. On the other hand, the small pores were still visible on the surface of the membrane without significant alteration after use in UF of 50 ppm HA solution and washing (**Fig. S8b**)

The pristine M0 and the maximum 10 wt.% PDGO loaded membrane M10 were tested in the UF of 50 ppm BSA solution containing 1 mM CaCl_2 and 3 mM NaCl solution for 3 h at 1 bar. The variation in flux versus time during filtration of 50 ppm BSA solution at 1 bar and 300 rpm is depicted in **Fig. S9**, Supporting Information. The J_w , BSA solution permeability (J_{BSA} ; $\text{L m}^{-2} \text{h}^{-1} \text{bar}^{-1}$) and $J_{w, \text{pc}}$ values for both membranes are presented in **Fig. S10**, Supporting Information. Both membranes rejected more than 85% of BSA from its 50 ppm solution at 1 bar (**Fig. S10a**). The hybrid membrane M10 was more efficient in BSA rejection than pristine M0 due to its higher surface charge ($\zeta = -34.2 \text{ mV}$). Furthermore, the antifouling ability of pristine M0 and M10 was assessed in term of FRR (%) after BSA solution filtration and subsequently washing with DI water–PBS solution. FRR values for M0 and M10 membranes are depicted in **Fig. S10b**. FRR of M10 was 89.1% while FRR of M0 was 80.7%. These results again indicate a better antifouling ability of hybrid membrane M10 during BSA solution filtration. This is ascribed to the improved hydrophilicity and the electrostatic repulsion between the negatively charged BSA molecules and the membrane surface. In addition, the

presumed hydration layer on the surface of hydrophilic membrane M10 could not be allowed strong adhesion of BSA molecules. So, the loosely adhered BSA molecules were detached by simple washing with DI water–PBS solution.

3.6. Stability of the membranes in cyclic UF of humic acid solution

A comparative assessment of the filtration performance stability of the pristine and PDGO-loaded membranes was conducted over three UF cycles of a 50 ppm HA solution using the M0 and M10 membranes at 1 bar feed pressure and 300 rpm mechanical stirring. M10 was selected for its considerable J_w ($\sim 515 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) and high R_{HA} (92.4%). The variation in flux with time during these filtration cycles is presented in **Fig. S11**, Supporting Information. Both membranes were tested under three consecutive filtrations cycles and the obtained values of J_w , J_{HA} , and $J_{w,pc}$ for each cycle are shown in **Fig. 10 (a and b)**. The FRR, R_{HA} and R_{ir} for both membranes in each cycle are also listed in **Table 3**. In the third filtration cycle, a 59% reduction in J_w (381 to $146 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) was observed for M0 while the drop was only 18.5% (515 to $414.4 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) for M10. Similar trends were observed for J_{HA} and $J_{w,pc}$ of both membranes. Likewise, FRR of M0 was as low as 38.2% in the third cycle while that for M10 was more than double (77.5%). Moreover, M10 showed a significant reduction in its irreversible fouling, R_{ir} , in the 3rd cycle when compared to M0 (61.6% and 22.5%, respectively). These findings show a sustained reduction in the surface adhesion of HA on the barrier layer of the hybrid membrane. The HA cake layer that formed on M10 was obviously more easily removed during the cleaning with DI water–PBS solution than in the case of M0, and this remained the case over three filtration cycles. Similarly, R_{HA} ability of M10 was also maintained over 3 cycles. In the third cycle, R_{HA} of M10 was noticeably higher ($> 85\%$) than that of M0 (71%). This indicates that the negative surface charge of M10 remains largely unaltered after three consecutive filtration cycles. These results demonstrate a stable performance (J_w , R_{HA} , and fouling resistance) of the PDGO-loaded membrane in UF of HA

solution.

To identify the leaching of PDGO nanosheets from the hybrid membrane, pure DI water was filtered through the membrane M10 (with the maximum, 10 wt.%, loading of PDGO nanosheets) for 3 h at 1 bar. The absorbance of both 0.2 mg mL⁻¹ PDGO suspension and permeated pure water sample were separately recorded at different wavelength in the range of 200 to 300 nm. The related data is shown in **Fig. S12**, Supporting Information. The absorbance of the permeated water at 220 nm was almost negligible in comparison to the absorbance of 0.2 mg mL⁻¹ PDGO suspension. Thus, the leaching of PDGO nanosheets from the hybrid membrane was ruled out during the long-term filtration operation.

Finally, in order to benchmark the performance of M5 membrane (claimed as the best PDGO-SPES membrane candidate) against that of the other recently-reported hybrid membranes containing other nanofillers, a plot (**Fig. 10**) was generated showing J_w and R_{HA} and a comparison table showing all the relevant data/parameters related to the filtration experiments and the membrane composition is included in Supporting Information (**Table S2**). The nanofillers in the other hybrid membranes include TNTs-SO₃H-PSf [8], GOSiO₂-NH₂-SPES [38], sulfonated hyperbranched polyethersulfone-modified halloysite nanotubes (SPHNTs)-PES [64], oxidized multi-walled carbon nanotubes (OCNTs)-PVDF [65], layered double hydroxides (LDH)-PVDF [66], Exfoliated-MoS₂ (eMoS₂)-PEI [67], titanium dioxide (TiO₂)-PES [68], and flower-like MoS₂ (FMoS₂)-PES [69]. **Fig. 10** reveals the often-reported permeability-selectivity trade-off for most of the membranes in the literature. For instance, low J_w values were reported for high R_{HA} membranes, such as OCNTs-PVDF, eMoS₂-PEI, TNTs-SO₃H-PSf, and GOSiO₂-NH₂-SPES. The M5 membrane, on the other hand, exhibited a good balance of relatively high selectivity ($R_{HA} = 91.7\%$), compared to the other membranes, while yielding a higher J_w than all the other membranes. This demonstrates that the trade-off between high J_w and R_{HA} was effectively abated through the synergic effect of SPES and PDGO.

Coupled with its stable performance and antifouling properties, these findings show the great promise for PDGO-SPES membrane.

4. Conclusions

Mussel-inspired PDGO nanosheets were successfully synthesized via self-polymerization of Dopa in an alkaline solution of tris-buffer with GO nanosheets. The 25.3% loading of PDA onto the GO nanosheets resulted in an improvement in the surface charge of PDGO nanosheets ($\zeta = -34.2$ mV). The hybrid SPES UF membranes were produced with the loading of 1–10 wt.% PDGO into the SPES matrix. The hybrid membranes showed a typical asymmetric structure with dense skin layer and finger-like pores in the sublayer. The surface pore density on the skin layer increased and the finger-like pores elongated towards the bottom of the membranes upon PDGO loading. The hybrid membrane M10, with the maximum (10 wt.%) PDGO loading, displayed the highest surface charge ($\zeta = -27.8$ mV) and hydrophilicity ($CA = 27.8^\circ$) of all hybrid membranes. The hybrid membrane M5 had the maximum water permeability ($680.7 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$) because of its highly porous structure (ε : 84.7%, and r_m : 20.6 nm). The hybrid membranes were efficient in removing more than 88% of HA from a 50 ppm solution and a remarkable antifouling performance was observed during the three cycles of HA solution filtrations coupled with washing using a DI water-PBS solution. Both reversible and irreversible fouling were significantly reduced with 5 wt.% or more PDGO loading into the SPES membrane. Thus, the synergic actions of PDGO and SPES were accountable for the high water permeability and selectivity, as well as the stable antifouling performance of the hybrid UF membranes. Tailor-made hybrid UF membranes can be easily produced *via* phase-inversion using this approach for effective NOM removal from the contaminated surface water. Such hybrid membranes may serve as the support layers while fabricating high-performance thin-film nanofiltration membranes for saline water desalination.

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.

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Table 1. Composition of the casting solution along with the thickness for each of the fabricated UF membranes.

Component	Membrane				
	M0	M1	M3	M5	M10
SPES (wt.%)	18	18	18	18	18
PVP10 (wt.%)	2	2	2	2	2
PDGO (wt.%)*	0	1	3	5	10
NMP (wt.%)	80	79.82	79.46	79.1	78.2
Thickness (μm)	132 ± 3.2	133 ± 3.5	135 ± 4.0	136 ± 3.7	138 ± 2.8

*Control (pristine) membrane

** Weight percentage of PDGO is calculated relative to the weight of SPES.

Table 2. Surface properties and structural characteristics of hybrid membranes with standard deviation (SD).

Parameter	Membrane				
	M0	M1	M3	M5	M10
ε (%)	65.2±1.6	70.8±1.2	76.1±1.3	84.7±1.8	78.6±2.2
r_m (nm)	15.7±0.6	17.3±0.4	19.7±0.2	20.6±0.3	18.4±0.2
r_a (nm)	6.1±0.2	7.6±0.3	10.9±0.4	11.5±0.4	14.6±0.5
CA (deg.)	59.1±2.1	52.3±2.3	43.5±1.9	36.9±1.5	27.8±1.4
ζ (mV)	-14.5±1.1	-18.3±0.9	-23.1±0.7	-25.7±1.2	-27.8±1.6
Membrane digital image					

Table 3. Cyclic efficiency of membranes M0 and M10 during UF of 50 ppm HA solution with 1.0 mM CaCl_2 and 7.0 mM NaCl.

M0				M10		
Cycle	FRR (%)	R_{ir} (%)	R_{HA} (%)	FRR (%)	R_{ir} (%)	R_{HA} (%)
1	67.3	32.7	87.9	93.4	6.9	92.4
2	41.4	58.4	75.2	81.3	19.7	89
3	38.2	61.6	70.9	77.5	22.5	85.8

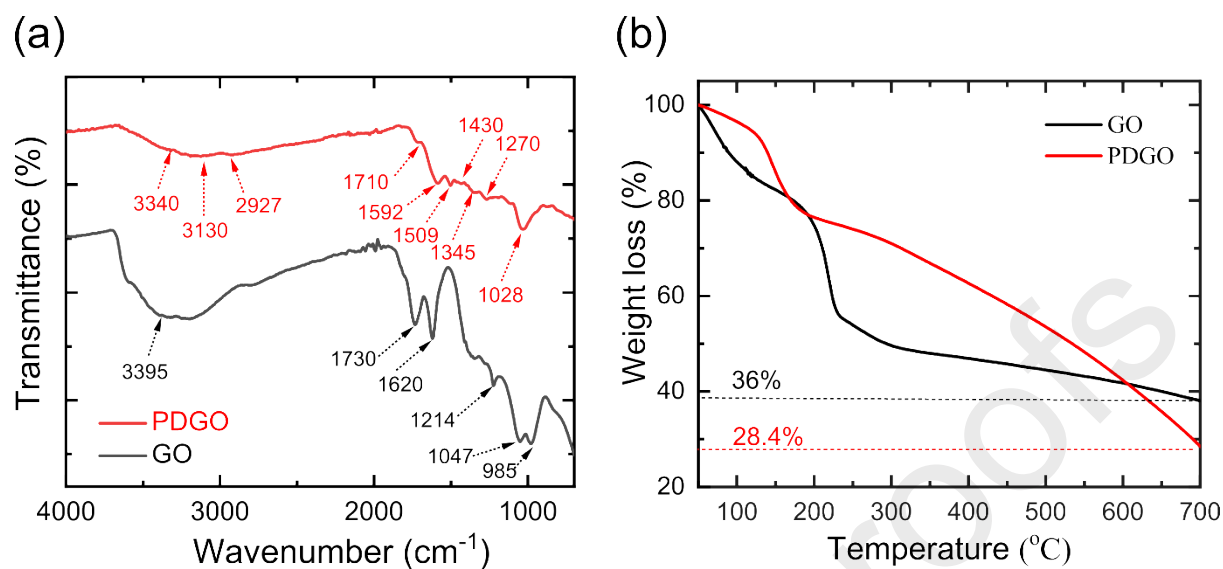


Fig. 1. (a) ATR-FTIR spectra and (b) TGA data (in the temperature range of 50 – 700 $^{\circ}\text{C}$) of GO and PDGO nanosheets.

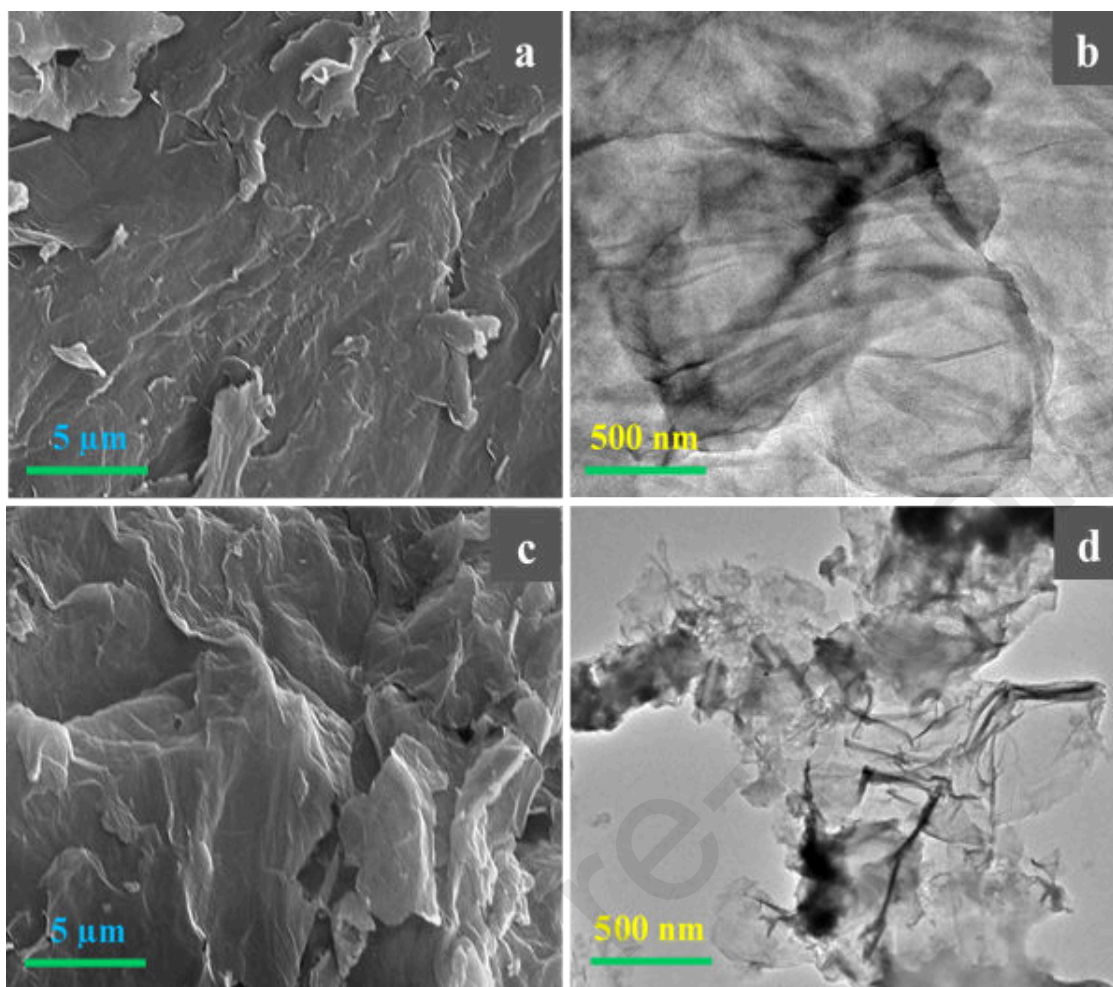


Fig. 2. Surface FE-SEM images of GO (a) and PDGO (c) nanosheets and TEM images of GO (b) and PDGO (d) nanosheets.

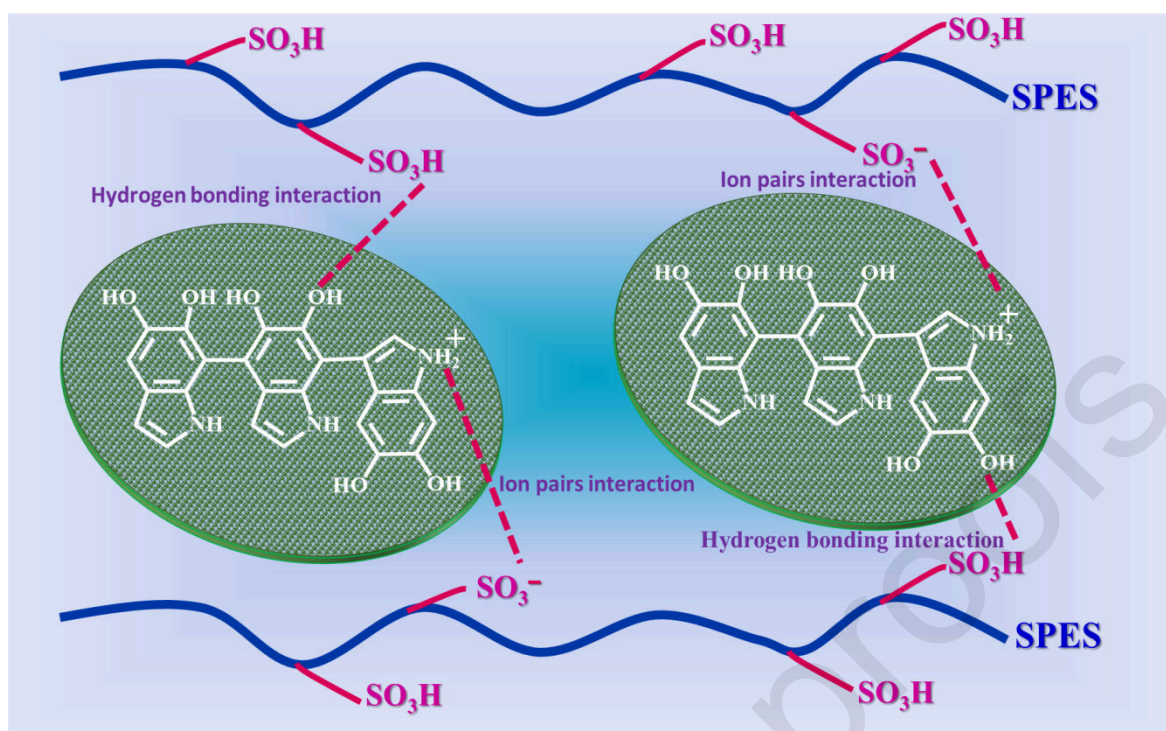


Fig. 3. Schematic presentation of the PDGO nanosheets imbedded in the matrix of SPES membrane.

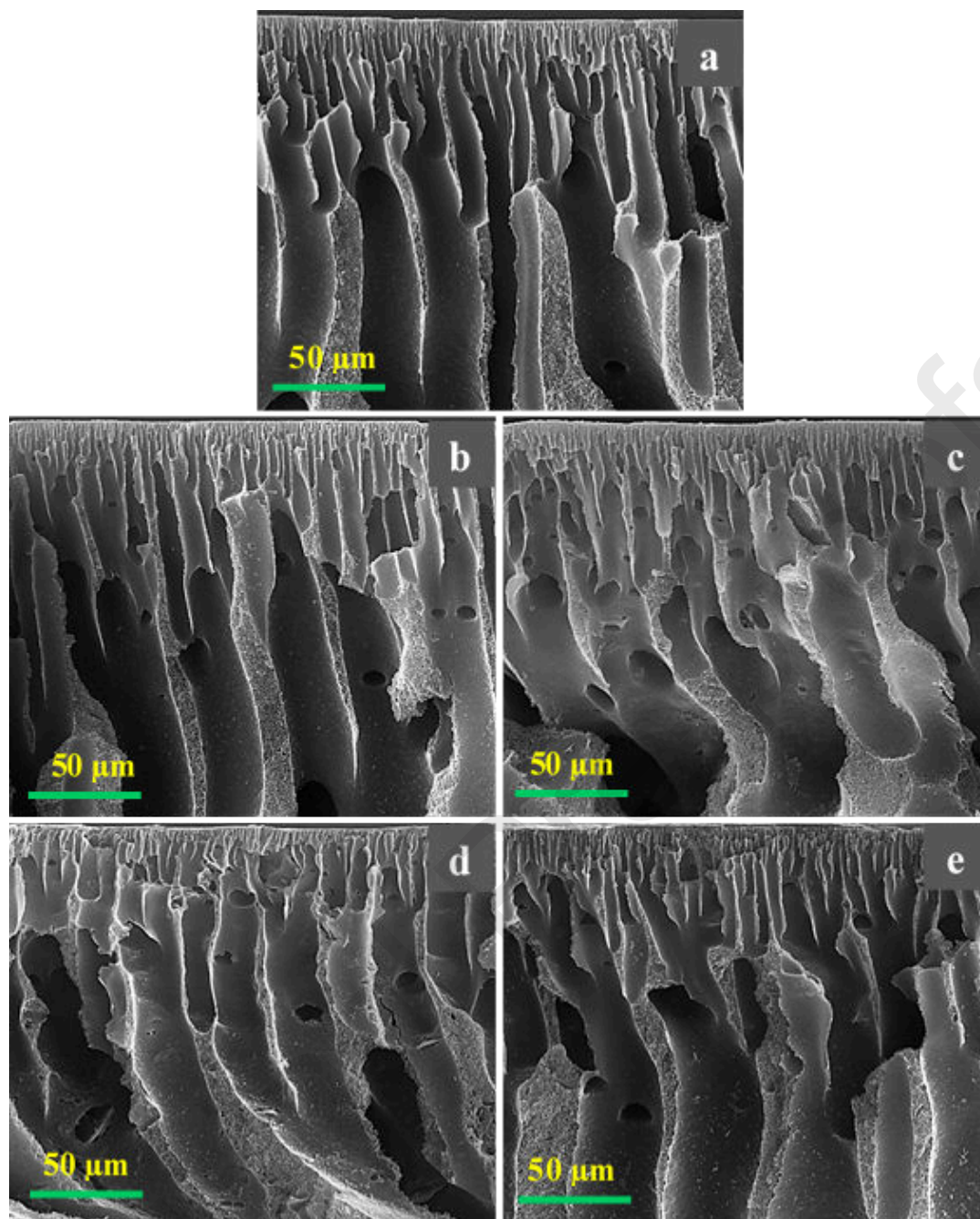


Fig. 4. Cross-sectional SEM images of M0 (a) and hybrid membranes M1(b); M3(c); M5 (d); and M10 (e).

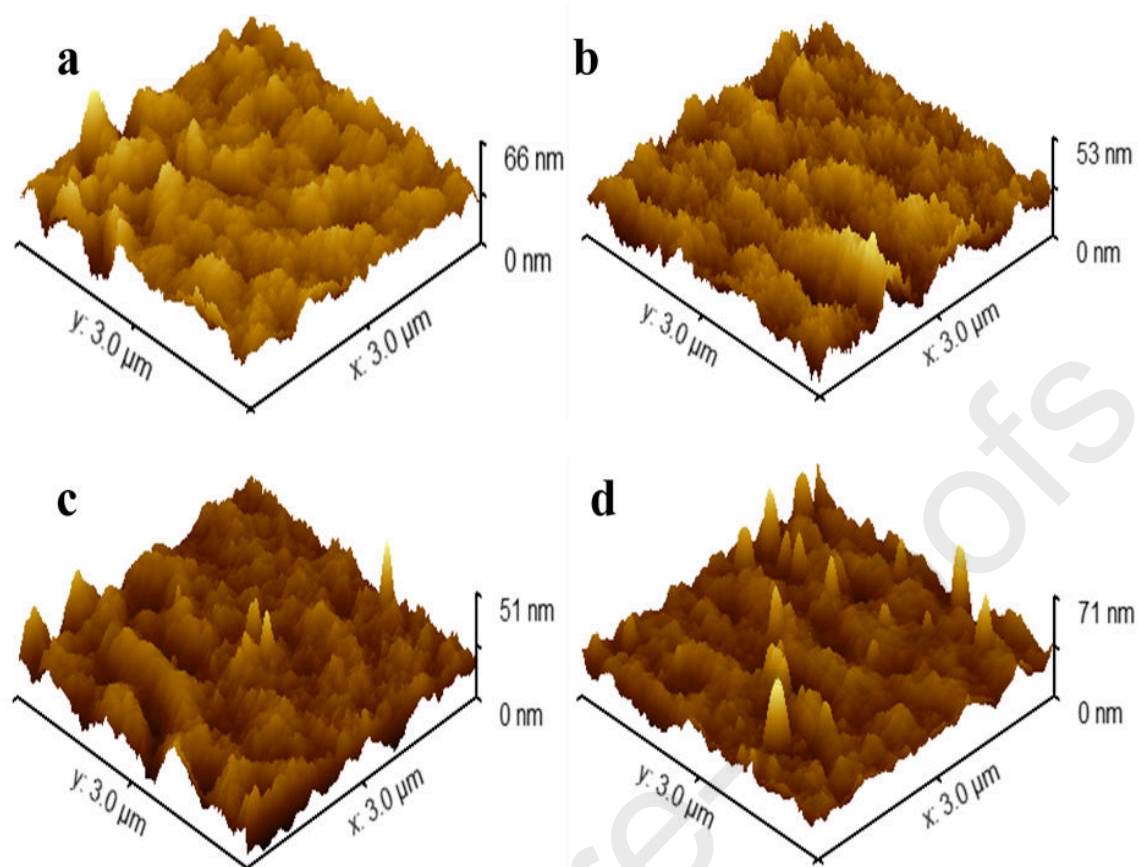


Fig. 5. 3D AFM images of (a) M0 and hybrid membranes M1 (b); M5 (c); and M10 (d).

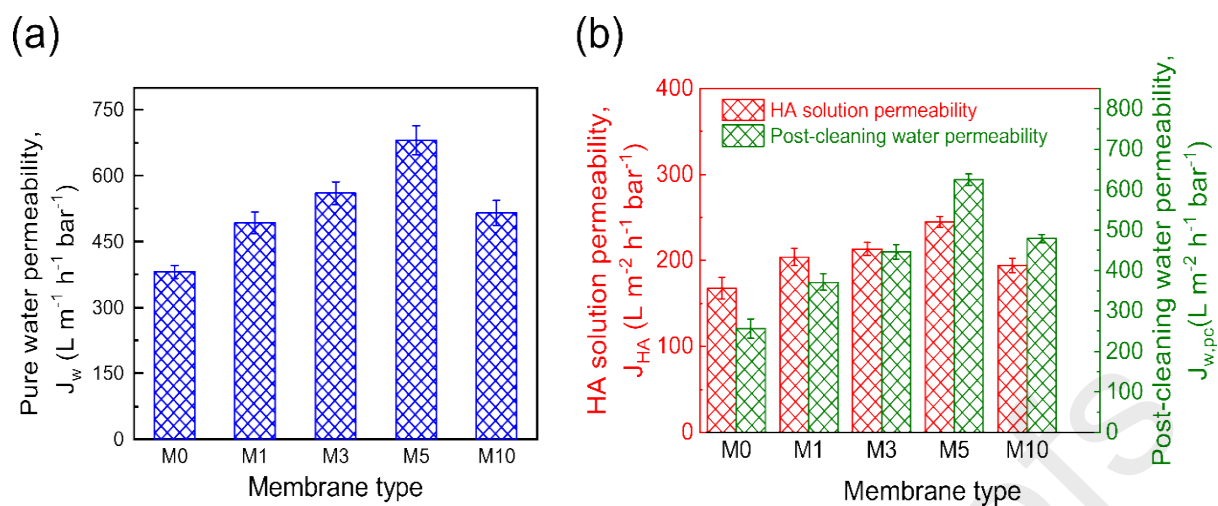


Fig. 6. (a) J_w ; (b) J_{HA} $J_{w,pc}$ of M0 and hybrid M1, M3, M5, and M10 membranes.

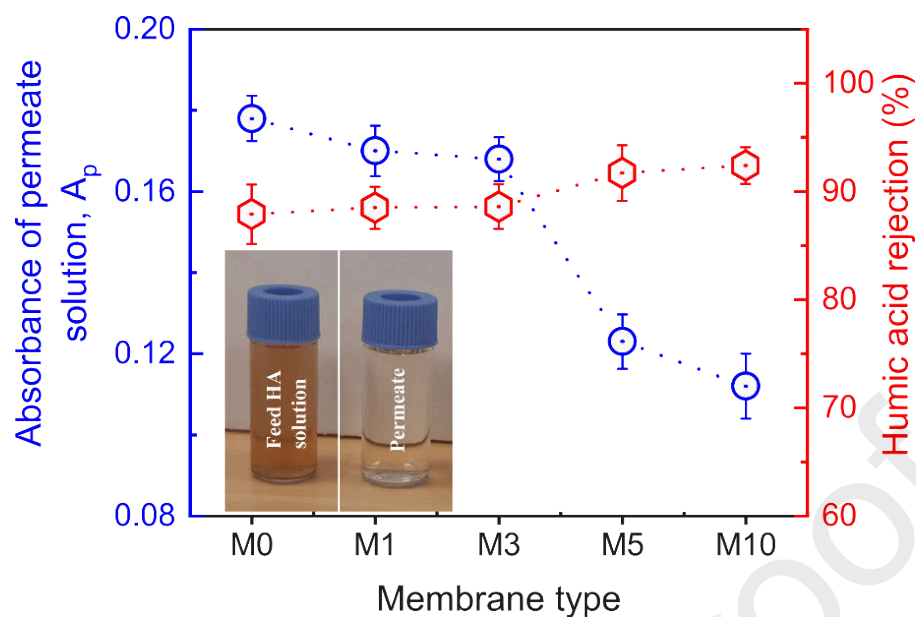


Fig. 7. Absorbance of permeate solutions (A_p) and R_{HA} (%) by pristine M0 and hybrid membranes M1, M3, M5 and M10. Inset is a digital photograph of 50 ppm HA feed and permeate solution after filtration using the M5 membrane for 3 h.

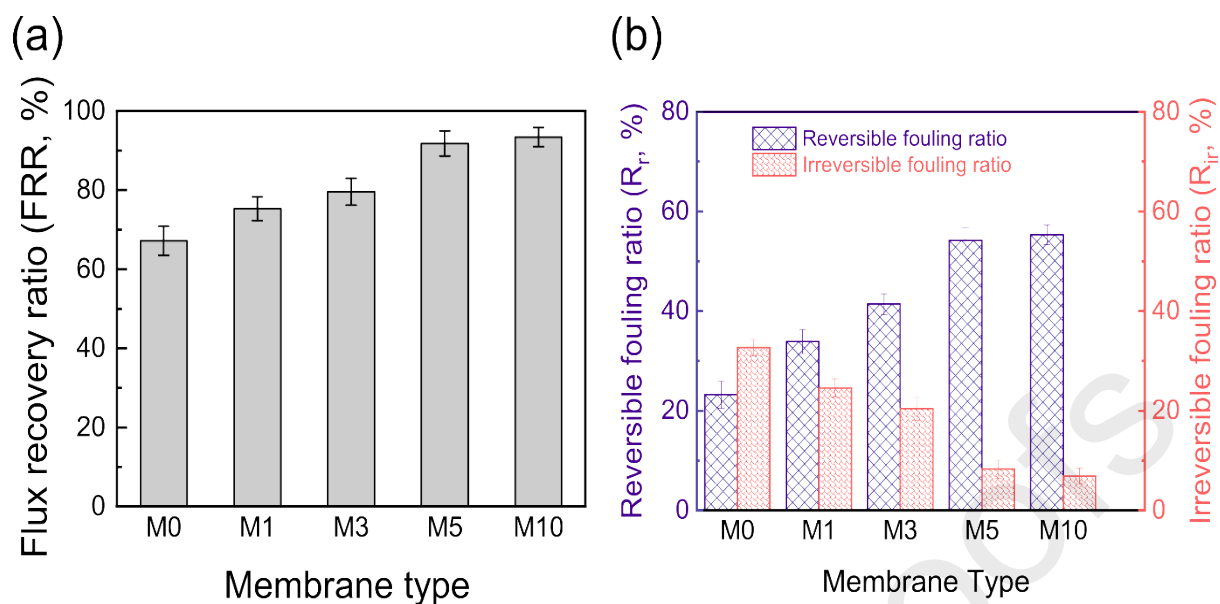


Fig. 8. (a) FRR, (b) R_r and R_{ir} values obtained from pure water permeability, 50 ppm HA solution J_{HA} and $J_{w, pc}$ of pristine and hybrid membranes.

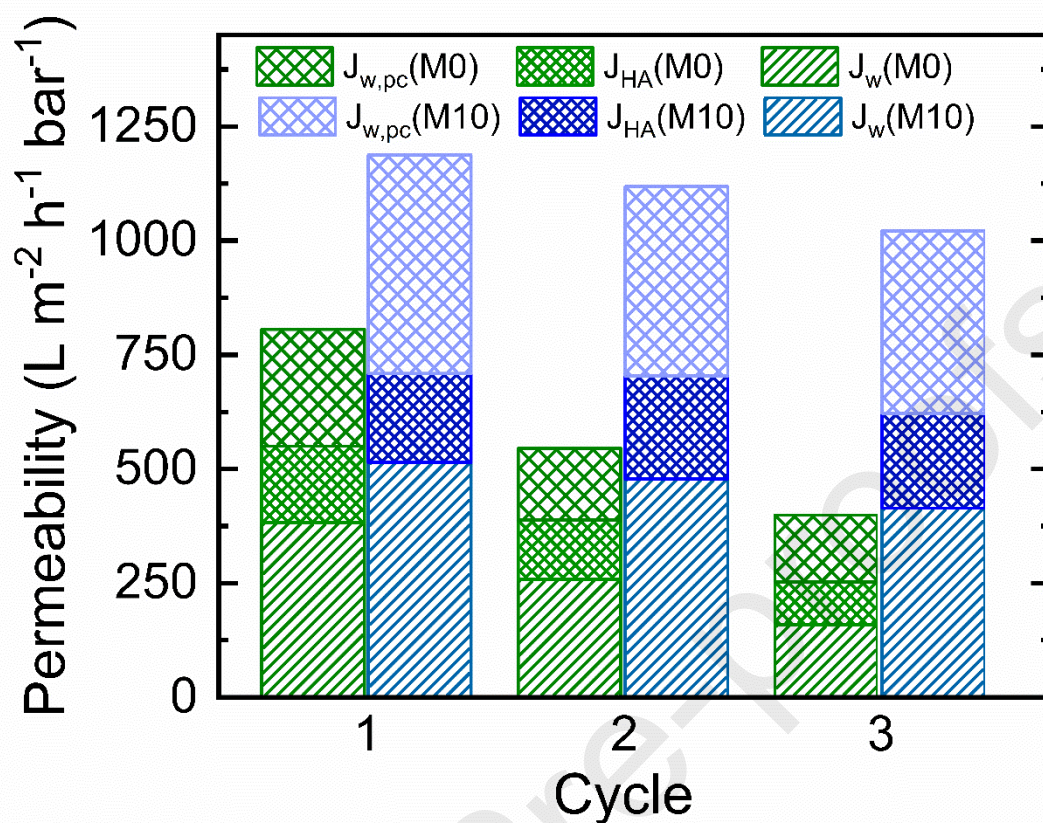


Fig. 9. J_w , J_{HA} and $J_{w,pc}$ of (a) M0 and (b) M10 for 3 filtration cycles of 50 ppm HA solution (3 h in each cycle) at 1 bar feed pressure, and 300 rpm mechanical stirring followed by cleaning with DI water–PBS solution.

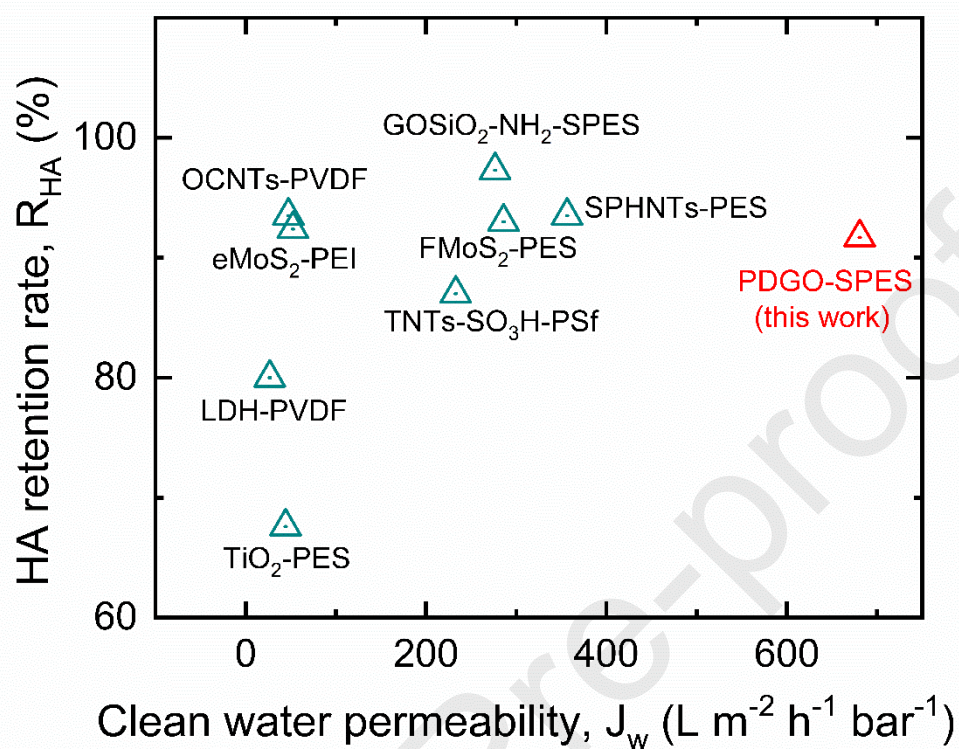


Fig. 10. Performance comparison of M5 hybrid PDGO-SPES membrane with other recently-reported hybrid membranes in the literature, in terms of J_w and R_{HA} (%) [8,38, 64–69].



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Polydopamine-coated graphene oxide nanosheets embedded in sulfonated poly (ether sulfone) hybrid UF membranes with superior antifouling properties for water treatment

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