

Engineering antifouling UiO-66-NH₂ mixed matrix membranes for geothermal brine pretreatment: Fouling prediction, techno-economic analysis, and environmental assessment

Pra Cipta Buana Wahyu Mustika^{a,b}, Cornelius Satria Yudha^{a,c}, Melisa Christine Prayogo^a, Putu Doddy Sutrisna^b, Sutijan Sutijan^a, Widi Astuti^e, Deliana Dahnum^f, Amir Razmjou^g, Himawan Tri Bayu Murti Petrus^{a,d,e,*}

^a Chemical Engineering Department, Sustainable Mineral Metallurgical Processes, Faculty of Engineering, Universitas Gadjah Mada, Jl. Grafika No.2, Kampus UGM, Yogyakarta, 55281, Indonesia

^b Chemical Engineering Department, Faculty of Engineering, Universitas Surabaya, Jl. Raya Kalirungkut, Kampus UBAYA, Surabaya, 60293, Indonesia

^c Chemical Engineering Department, Vocational School, Universitas Sebelas Maret, Jl. Kolonel Sutarto No 150K, Surakarta, 57126, Indonesia

^d Unconventional Geo-Resources Research Center, Faculty of Engineering, UGM, Jl. Grafika No.2, Kampus UGM, Yogyakarta, 55281, Indonesia

^e Research Center for Mineral Technology, National Research and Innovation Agency (BRIN), Jl. Ir. Sutami Km. 15, Tanjung Bintang, Lampung Selatan, 35361, Indonesia

^f Research Centre for Molecular Chemistry, National Research, and Innovation Agency (BRIN), Building 452, KST BJ Habibie, Tangerang Selatan, Banten, 15314, Indonesia

^g Mineral Recovery Research Centre (MRRC), School of Engineering, Edith Cowan University, Joondalup, Perth, WA, 6027, Australia

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ABSTRACT

This study develops polysulfone/UiO-66-NH₂ mixed-matrix membranes (MMMs) for silica removal from geothermal brine and evaluates their antifouling behavior, fouling mechanisms, techno-economic feasibility, and environmental implications. Incorporation of UiO-66-NH₂ improved membrane hydrophilicity, decreasing the water contact angle from 80.8° to 47.6°, and increased permeability from 158.94 to 376.03 L m⁻² h⁻¹ bar⁻¹. FTIR and XRD confirmed successful UiO-66-NH₂ incorporation, while SEM revealed a well-developed porous structure. The optimized MMM achieved a maximum FRR of 82.94%, showing that backwashing restored flux by reopening pores narrowed by reversible intrapore silica deposition. Hermia-derived modeling confirmed that silica fouling was dominated by internal pore narrowing, rather than surface cake formation. Techno-economic analysis showed that mixed matrix membrane ultrafiltration reduced pretreatment costs to 0.91 and 0.47 USD/m³ at 100 and 1000 m³/day, respectively, compared with 1.29 and 0.68 USD/m³ for conventional coagulation–flocculation–clarification, but remained less cost-efficient than commercial ultrafiltration at 0.80 and 0.43 USD/m³. Both membrane-based routes reduced energy demand and greenhouse gas emissions by approximately 53–54% relative to conventional treatment, although the mixed-matrix membrane system showed slightly greater impacts than commercial ultrafiltration and increased water consumption by approximately 47–48%. These results reveal a clear trade-off between improved antifouling performance and modest economic, energy, emission, and water-use penalties relative to commercial ultrafiltration.

1. Introduction

To address the escalating crisis of carbon emissions, sustainable development must prioritize a synergy between economic growth, environmental stewardship, and social responsibility. Achieving net-

zero carbon emissions by 2050 requires a multi-faceted approach involving energy transition, green finance, and digitalization [1]. Within this framework, geothermal energy has emerged as a critical substitute for fossil fuels [2]. Unlike solar or wind energy, which are subject to intermittency caused by external weather factors, geothermal energy

* Corresponding author at: Chemical Engineering Department, Sustainable Mineral Metallurgical Processes, Faculty of Engineering, Universitas Gadjah Mada, Jl. Grafika No.2, Kampus UGM, Yogyakarta, 55281, Indonesia.

E-mail address: bayupetrus@ugm.ac.id (H.T.B.M. Petrus).

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offers baseload capability due to its consistent availability. While the sector faces barriers to investment, technology, and awareness of adaptation, recent studies confirm that enhancing the efficiency of existing geothermal systems remains a viable strategy for sustainable energy generation [3,4].

The efficiency of geothermal operations is significantly hindered by scaling (inorganic fouling), defined as the deposition of mineral precipitates pipelines during brine extraction. Depending on the well's geological characteristics, the primary constituents of this scaling include silicates, carbonates, sulfides, and sulfates [5–7]. The deposition of these minerals reduces system efficiency by impeding fluid flow during reinjection. Economically, this necessitates frequent maintenance and pipe replacement [8]. While chemical inhibitors are currently employed to mitigate scaling, they do not offer a comprehensive resolution to the precipitation issue [9].

Beyond power generation, geothermal brine presents an opportunity for resource valorization through the extraction of critical minerals, specifically lithium. The integration of Direct Lithium Extraction (DLE) represents a mature, high-efficiency technology for this purpose [10]. Compared to conventional methods, DLE offers significant technical advantages: (1) Temporal Efficiency: Reduction of production timelines from months to hours, (2) Spatial Efficiency: A smaller physical footprint via compact system design, (3) Resource Efficiency: Elimination of evaporative ponds, resulting in lower water and chemical consumption, (4) Quality Assurance: Potential for improved product yield and purity [11].

While DLE presents significant advantages, its operational success is contingent upon rigorous feed pre-treatment. Specifically, the complete removal of dissolved minerals is required to prevent downstream system fouling. Ultrafiltration technology is widely recognized for its reliability in this domain, offering high separation efficiency for suspended solids, including nanoparticles (NPs) and colloidal particles [12]. Despite its separation capabilities, the long-term performance of UF membranes in geothermal brine applications is compromised by severe fouling. The primary inorganic fouling agent is amorphous silica. The inorganic fouling trajectory is governed by a sequential three-stage mechanism. Initially, saturation occurs when solute concentration exceeds the solubility limit at the membrane interface. This thermodynamic instability triggers the polymerization of monomeric silicic acid into oligomers. Finally, these species undergo colloidal fusion, coalescing into a dense, impermeable gel layer on the membrane surface [13,14].

Standard operational adjustments, such as modulating flow rate, transmembrane pressure, and temperature, have proven insufficient to completely resolve inorganic fouling. The persistence of this issue is attributed to concentration polarization (CP). CP is an unavoidable hydrodynamic phenomenon where retained solutes accumulate at the membrane surface, creating a boundary layer of high concentration [15,16]. This localized supersaturation drives the silica polymerization process described above.

Given that mechanical controls alone are insufficient to suppress concentration polarization, tuning membrane physicochemical properties has become essential. While surface modification can adjust hydrophilicity or surface charge, its effects are often limited to the outer layer and may be compromised by coating instability or pore blockage. In contrast, matrix blending provides a more integrated route by embedding functional additives within the membrane structure, enabling simultaneous control of hydrophilicity, pore architecture, transport pathways, and antifouling behavior.

Accordingly, strategies to improve membrane performance and antifouling behavior have long relied on bulk modification through membrane matrix blending. Amphiphilic polymer blends, such as Pluronic F127 incorporated into PES by Wang et al. [17], Zhao et al. [18], and Pandey et al. [19], have been shown to improve membrane structure, separation performance, and fouling resistance. Similar improvements have been reported for functional polymer blends, including SPSF/PES [20] and PSFNA/PSF [21] systems, as well as PEG-modified

PSf membranes, where hydrophilicity and pore structure strongly depend on additive composition and molecular weight [22,23]. Nanoparticle incorporation, including TiO₂ and GO, has further demonstrated the ability to enhance membrane morphology, selectivity, and antifouling properties [24–26]. These studies establish blending as an effective route for membrane optimization; however, conventional additives may still face limitations related to leaching, aggregation, or restricted functionality. This motivates MOF-based mixed matrix membranes as an integrated bulk-modification strategy to address these challenges.

Table 1 shows that MOF-reinforced UF-MMMs consistently improve membrane performance through increased hydrophilicity, higher porosity and water uptake, and more favorable surface charge. Across different MOFs and polymer matrices, optimal filler loading is generally found within 0.1–1 wt%, beyond which aggregation and increased dope viscosity may hinder solvent/non-solvent exchange and limit permeability enhancement. Reported improvements remain substantial but variable, with permeability increases of 37–419% and FRR values ranging from 52.9% to 98.7%, mainly attributed to electrostatic repulsion and hydration-layer protection.

However, the antifouling evaluations summarized in Table 1 are still dominated by organic or biological foulants, including HA, NOM, BSA, milk powder, sodium alginate, oil-in-water emulsions, and dyes. The behavior of MOF-based UF-MMMs under inorganic fouling, particularly silica scaling in geothermal brine, remains insufficiently understood. This gap is critical because dissolved silica is mainly present as weakly charged or neutral monosilicic acid at near-neutral pH, making electrostatic repulsion less effective and shifting inorganic fouling control toward hydration effects and siloxane-gel formation.

In the present work, UF-grade MMMs were developed for geothermal brine pretreatment through the incorporation of UiO-66-NH₂ nanoparticles into a polysulfone (PSf) matrix. PSf was selected as the base polymer due to its favorable physicochemical properties, including high thermal stability, mechanical strength, chemical resistance over a broad pH range, and excellent film-forming capability [27,28]. However, its intrinsic hydrophobicity remains a key limitation in filtration applications. To address this challenge, UiO-66-NH₂ was introduced to enhance membrane hydrophilicity [27,29]. Moreover, the organic functional groups associated with the nanoparticle ligands are expected to promote favorable interfacial compatibility with the polymer matrix, contributing to improved membrane structure and performance. Therefore, this study advances the field by positioning UiO-66-NH₂/PSf MMMs not only as a modified UF material, but as a pretreatment platform evaluated through inorganic fouling analysis, performance prediction, techno-economic perspective, and environmental impact assessment.

2. Materials and methods

2.1. Materials

All chemicals employed in this study were of high purity and used without further purification. Polysulfone (PSf) pellets (Mw $\approx$ 35,000; Mn $\approx$ 16,000), zirconium (IV) chloride (ZrCl₄, 99.9%), 2-aminoterephthalic acid (ATA, 99%), *N,N*-dimethylformamide (DMF, 99.8%), methanol (MeOH, $\geq$ 99.8%) were also purchased from Sigma-Aldrich. Ultrapure water was supplied by WaterOne®, while laboratory-produced deionized water with a conductivity of $\pm 15 \mu\text{S}\cdot\text{cm}^{-1}$ was used in certain experiments.

2.2. Fabrication of UiO-66-NH₂

UiO-66-NH₂ was synthesized via a solvothermal method using zirconium precursors and ATA ligands, with HCl serving as a modulator in a DMF solvent system. Briefly, ZrCl₄ (2.5 g, 10 mmol) was dissolved in 100 mL of DMF, to which 20 mL of concentrated HCl was added. In a separate vessel, ATA (1.82 g, 10 mmol) was dissolved in 200 mL of DMF

Table 1

Reported ultrafiltration mixed matrix membranes (MMMs) and their evaluated separation and antifouling performance.

MMMs (opt. loading)	Pure Water Permeability [$\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$]	Flux recovery ratio [%]	Water Contact Angle [$^{\circ}$]	Detail Testing Condition	Ref
MIL-101@PVDF (0.1%wt)	170 $\rightarrow$ 360 (+111%)	47 $\rightarrow$ 77.7	88 $\rightarrow$ 68 (–20%)	• Dead-end @ $P = 0.10$ MPa • 1000 ppm BSA	[30]
MOF-5@PVDF (0.5%wt)	13.2 $\rightarrow$ 68.6 (+419%)	• BSA = 70.6 $\rightarrow$ 89.7 • HA = 68.7 $\rightarrow$ 92.4	98.6 $\rightarrow$ 52.3 (–47%)	• Dead-end @ $P = 0.10$ MPa • BSA or HA	[31]
TMU-5@PES (0.1%wt)	44.4 $\rightarrow$ 60.7 (+37%)	25.5 $\rightarrow$ 98.7	67.2 $\rightarrow$ 57.5 (–14%)	• Dead-end @ $P = 0.30$ MPa • O/W emulsion	[32]
UiO-66-NH ₂ @PES (0.1%wt)	11.7 $\rightarrow$ 16.7 (+43%)	59.2 $\rightarrow$ 95.2	65 $\rightarrow$ 50 (–23%)	• Dead-end @ $P = 0.30$ MPa • 1000 ppm Milk Powder	[33]
MOF-303@PES (0.75%wt)	37.4 $\rightarrow$ 81.5 (+117.8%)	73.4 $\rightarrow$ 85	69.2 $\rightarrow$ 52.9 (–23.5%)	• Dead-end @ $P = 0.20$ MPa • 1000 ppm BSA	[34]
MIL-125-Ti-NH ₂ @PES (0.5%wt)	270.4 $\rightarrow$ 560.7 (+107%)	32.9 $\rightarrow$ 52.9	90.6 $\rightarrow$ 81.6 (–9.9%)	• Dead-end @ $P = 0.15$ MPa • 1000 ppm CR	[35]
UiO-66@PS (0.6%wt)	110 $\rightarrow$ 211 (+92%)	34.2 $\rightarrow$ 87.7	86.6 $\rightarrow$ 53.5 (–38.2%)	• Dead-end @ $P = 0.10$ MPa • 20 ppm HA	[36]
Ti-MOF @PS (1%)	65 $\rightarrow$ 150 (+131%)	82 $\rightarrow$ 100	82 $\rightarrow$ 62 (–24.4%)	• Dead-end @ $P = 0.10$ MPa • 100 ppm HA	[37]
Co-MOF @PS (4%)	60 $\rightarrow$ 276 (+360%)	45 $\rightarrow$ 82.5	72 $\rightarrow$ 52 (–28%)	• Dead-end @ $P = 0.10$ MPa • 20 ppm NOM	[38]
MIL-125-Ti @PS (0.5%wt)	55.2 $\rightarrow$ 104.5 (+89.4%)	81.41 $\rightarrow$ 94.79	89.5 $\rightarrow$ 67.5 (–24.6%)	• Dead-end @ $P = 0.20$ MPa • 1000 ppm BSA	[39]

under continuous stirring until complete dissolution was achieved. The precursor solutions were combined and homogenized via ultrasonication for 20 min. The mixture was transferred to a Teflon-lined autoclave and placed in a furnace. The temperature was increased to 120 °C at a heating rate of 2 °C/min and maintained for 8 h to promote crystallization, followed by a 12 h cooling period. After reaction completion, the UiO-66-NH₂ particles were recovered by centrifugation at 6800 rpm for 20 min, followed by sequential washing with DMF to remove unreacted ATA and residual HCl, and subsequently with MeOH to ensure purity. Finally, the obtained product was dried at 90 °C for 24 h, yielding UiO-66-NH₂ powders for further use. The fabrication process is schematically illustrated in Fig. 1.

2.3. Fabrication of MMMs

The MMMs were fabricated using a combined evaporation-induced

phase separation (EIPS) and non-solvent-induced phase separation (NIPS) approach. A predetermined amount of UiO-66-NH₂ (0, 0.04, 0.12, 0.20, 0.28 g) was first dispersed in 20 mL of DMF and sonicated for 10 min to achieve uniform nanoparticle dispersion. Subsequently, 4 g of PSf pellets were added, and the mixture was stirred at 60 °C for 6 h until complete polymer dissolution. The resulting dope solution was sonicated for an additional 15 min to remove entrapped air and then cooled to room temperature.

The dope was cast onto a nylon-coated glass plate at a controlled thickness of ± 200 μm using calibrated insulating layers. Solvent removal proceeded in two stages: (i) partial evaporation in an oven at 60 °C for 10 min to initiate EIPS, followed by (ii) immersion in deionized water for 24 h to induce full solvent–non-solvent exchange via NIPS. The membranes were subsequently rinsed thoroughly with ultrapure water and dried at ambient conditions prior to characterization and use. A detailed schematic of the MMM fabrication process is presented in Fig. 2.

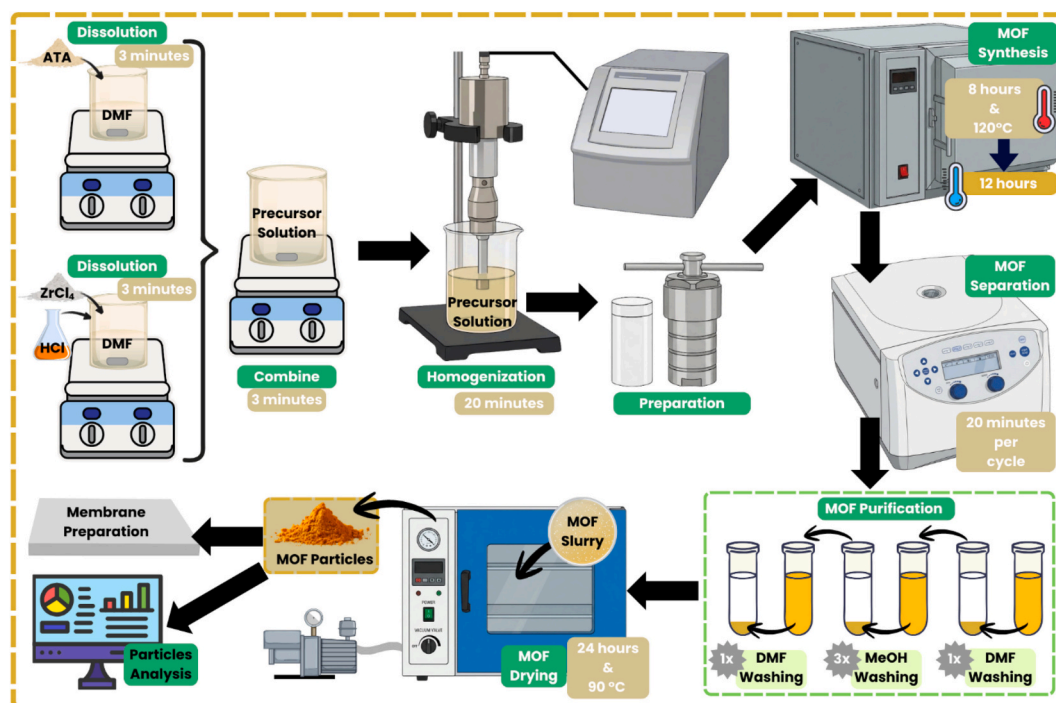


Fig. 1. Schematic illustration of UiO-66-NH₂ synthesis via the solvothermal method.

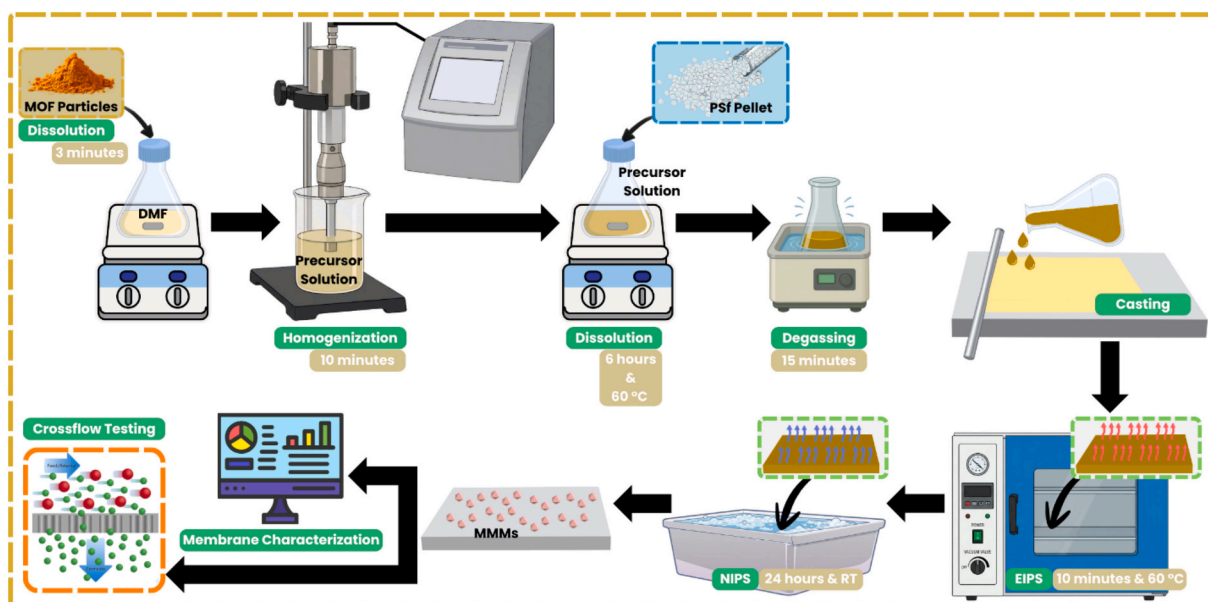


Fig. 2. Schematic illustration of PSF-based MMM fabrication incorporating UiO-66-NH₂ as the filler, produced via phase inversion initiated by EIPS followed by NIPS.

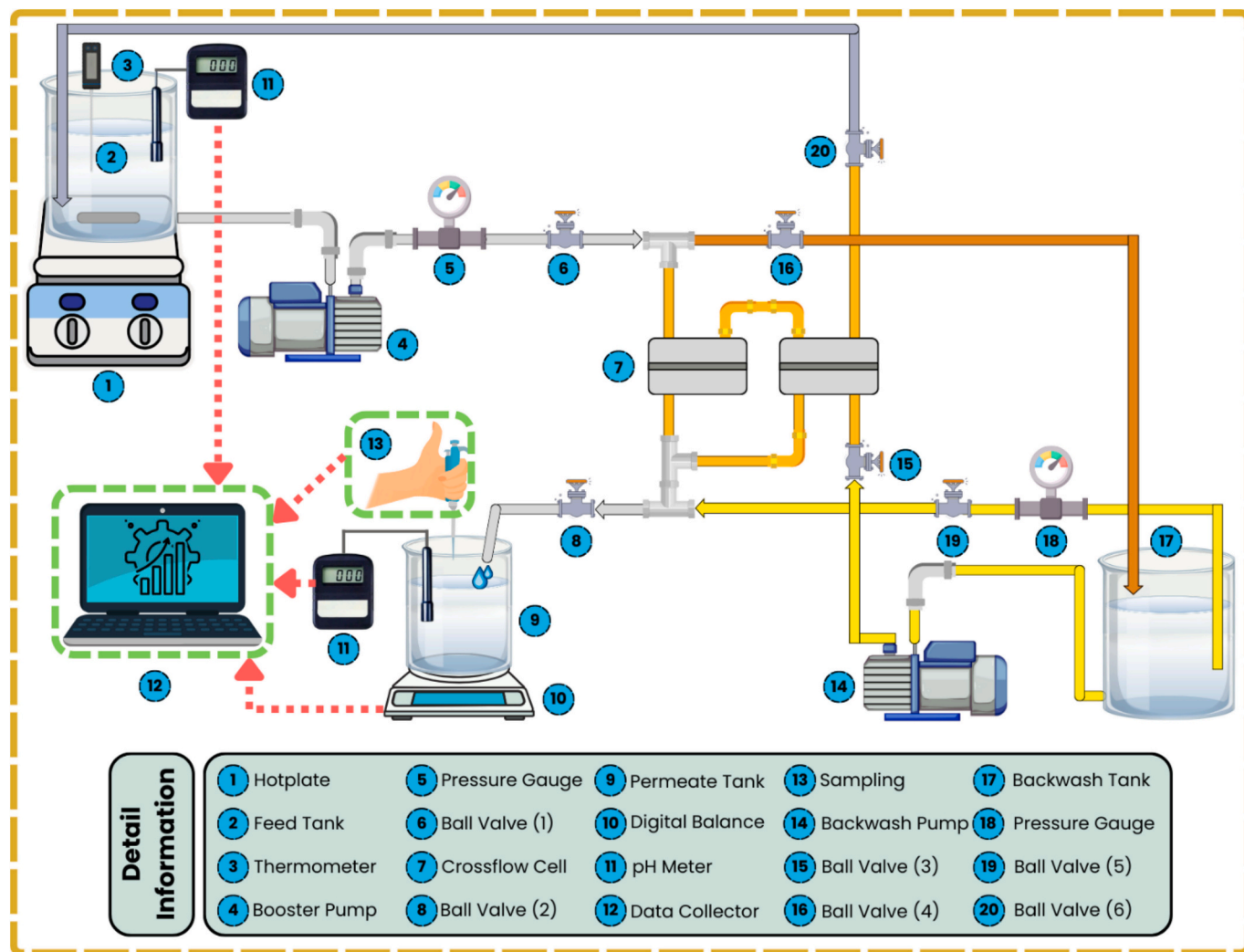


Fig. 3. Schematic illustration of MMM testing in a crossflow filtration system using geothermal brine as the feed.

2.4. Structural and functional characterization

Surface morphology and elemental distribution of the membranes were examined using scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM–EDX) [Thermo Scientific, Axia ChemiSEM System and JEOL, JCM 7000]. Transmission electron microscopy (TEM) was employed to examine the internal structure [FEI Company, Tecnai G2 20]. Nitrogen adsorption–desorption analysis was conducted to obtain comprehensive information on the material's textural properties, including specific surface area, pore volume, and pore size distribution [Micromeritics, TriStar II Plus]. Prior to analysis, approximately 80 mg of the powdered sample was loaded into a glass analysis tube and degassed under dynamic vacuum at 150 °C for 3 h. Surface chemical functionalities were identified by Fourier-transform infrared (FTIR) spectroscopy [Shimadzu, IRTracer-100] over the wave-number range of 500–4500 cm^{−1}. The incorporation of UiO-66-NH₂ into the membrane matrix was verified by X-ray diffraction (XRD) analysis [Malvern Panalytical, Aeris]. Membrane wettability was assessed by measuring the static water contact angle using a contact angle meter [Dataphysics OCA20]. Hydrodynamic size and zeta potential of UiO-66-NH₂ were measured using a HORIBA nanoPartica SZ-100-S.

2.5. Evaluation of membrane performance

Filtration experiments were performed using two crossflow membrane modules connected in series, each providing an effective membrane area of 7.06 cm². The feed solution was circulated through the system using a pressure pump, and all tests were conducted at ambient temperature. Membrane operation was maintained at a transmembrane pressure of 4 bar for 1 h.

Permeability tests were conducted alternately using deionized water and geothermal brine. Following each geothermal brine run, the membrane was subjected to physical cleaning via backwashing at 4 bar for 15 min to remove accumulated foulants and restore permeability before subsequent testing. A schematic of the MMM membrane testing setup is presented in Fig. 3.

The membrane flux (J) was determined using Eq. (1).

$$J = \frac{V}{A \times t} \quad (1)$$

where V is the permeate volume (L), A is the effective membrane area (m²), t is the filtration time (h). The effectiveness of membrane cleaning was assessed by calculating the flux recovery rate (FRR) using Eq. (2), where J_{w1} is the initial pure water permeability and J_{w2} is the pure water flux measured after cleaning. In addition, J_{w2} also denotes the flux of the contaminated solution (L.m^{−2}.h^{−1}.bar^{−1}) used to evaluate fouling behavior. The reversible fouling rate (RFR), irreversible fouling rate (IFR), and total fouling rate (TFR) were determined using Eqs. (3), (4), and (5), respectively.

$$\text{FRR} = \left(\frac{J_{w2}}{J_{w1}} \right) \times 100\% \quad (2)$$

$$\text{RFR} = \left(\frac{J_{w2} - J_p}{J_{w1}} \right) \times 100\% \quad (3)$$

$$\text{IFR} = \left(1 - \frac{J_{w2}}{J_{w1}} \right) \times 100\% \quad (4)$$

$$\text{TFR} = \left(1 - \frac{J_p}{J_{w1}} \right) \times 100\% \quad (5)$$

Table 2

Symbols and definitions used in the four fouling prediction models.

Symbol	Information	Dimension
J_0	Initial permeate flux	s.m ^{−1}
J_{pss}	Steady-state permeate flux	s.m ^{−1}
J_p	Permeate flux	s.m ^{−1}
K_c	Complete blocking model constant for crossflow filtration	m ^{−1}
K_{gl}	Gel layer model constant for crossflow filtration	m ^{−1}
K_i	Intermediate blocking model constant for crossflow filtration	m ^{−1}
K_s	Standard blocking model constant for crossflow filtration	m ^{−1}
t	Time	s

2.6. Membrane fouling prediction

In this study, inorganic fouling behavior is interpreted using fouling and adsorption models derived from the Hermia framework [40], as further developed by Field et al. [41]. This modeling approach enables the classification of fouling mechanisms into complete blocking, intermediate blocking, standard blocking, and gel layer formation, each representing a distinct mode of flux decline. The applicability of the Field model to the present work is supported by shared experimental conditions, including convective-dominated transport, cross-flow filtration configuration, and flat-sheet membrane operation [41].

Silica scaling is governed by a multistep mechanism in which dissolved silicic acid first reaches supersaturation, adsorbs onto the membrane surface, and subsequently undergoes polymerization to form a gel-like or film layer. These sequential processes correspond well with the assumptions underlying the Hermia-based models, allowing the selected equations to capture the transition from pore blocking to surface layer formation. Accordingly, the integration of these models provides a systematic and physically consistent framework for linking flux-decline behavior to the dominant inorganic fouling mechanisms observed in this research.

Silica polymerization can be described as an equilibrium process, as expressed in Eq. (6).



2.6.1. Complete blocking

The complete blocking mechanism assumes that silica species reaching the membrane surface irreversibly seal open pores through direct deposition and polymerization at the pore entrances. As fouling progresses, the number of available pores decreases, while the flow velocity through the remaining unblocked pores remains unchanged. To account for this behavior, a term describing the rate of silica deposition and pore sealing at the pore mouths is incorporated into the model. The resulting flux-decline behavior associated with complete blocking is mathematically described by Eq. (7) (Table 2).

$$J_p = J_{\text{pss}} + (J_0 - J_{\text{pss}})V \quad (7)$$

$$V = e^{-K_c J_0 t} \quad (8)$$

2.6.2. Gel layer formation

Gel layer formation represents a fouling mechanism distinct from pore-blocking processes. In this mechanism, polymerized silica accumulates on the membrane surface to form a continuous gel or cake layer, while the intrinsic membrane resistance remains essentially unchanged. The overall filtration resistance therefore comprises the constant membrane resistance and an additional resistance arising from the silica gel

layer. The flux-decline behavior associated with this mechanism is described by Eq. (9) (Table 2).

$$J_p = \frac{1}{2(J_0(U-1))} \left(J_0 J_{pss}(U-1) - J_{pss}^2 + \sqrt{4J_0^2 J_{pss}^2 (U-1) + (J_0^2 J_{pss}^2 (1-U) + J_{pss}^2)^2} \right) \quad (9)$$

$$U = e^{J_{pss}^2 K_{gt} t} \quad (10)$$

2.6.3. Intermediate blocking

The intermediate blocking mechanism assumes that incoming silica species do not uniformly seal membrane pores. Instead, pore blockage occurs probabilistically, with silica particles or polymerized clusters depositing either at pore entrances or on previously deposited material. This stochastic deposition results in partial pore obstruction and gradual flux decline. The corresponding flux–time relationship for intermediate blocking is described by Eq. (11) (Table 2).

$$J_p = \frac{J_0 J_{pss} Y}{J_{pss} + J_0(Y-1)} \quad (11)$$

$$Y = e^{K_i J_{pss} t} \quad (12)$$

2.6.4. Standard blocking

The standard blocking mechanism assumes that dissolved or polymerized silica penetrates into the membrane pores and deposits along the pore walls, leading to a progressive reduction in effective pore volume without complete pore sealing. This internal pore constriction results in a characteristic flux decline, which in classical filtration theory yields a linear relationship between filtration time and the ratio of time to permeate volume. The flux behavior associated with standard blocking is described by Eq. (13) (Table 2).

$$J_p = \frac{J_0}{(1 + Z J_0^{0.5})^2} \quad (13)$$

$$Z = K_s t \quad (14)$$

3. Results and discussion

3.1. Structural and functional characterization

The morphology of the synthesized UiO-66-NH₂ nanoparticles is shown in Fig. 4.A. TEM observations indicate particle sizes in the range of 1–2 μm with predominantly irregular morphologies. Localized particle overlap and agglomeration are evident, which can be attributed to the rapid synthesis process. Nevertheless, in several regions the nanoparticles retain well-defined cubic features characteristic of the UiO-66 framework, indicating that the underlying crystalline structure is preserved despite partial aggregation. The particle size was further evaluated by DLS, revealing a mean hydrodynamic diameter of 288.7 ± 14.4 nm for UiO-66-NH₂, while the particles exhibited a negative zeta potential of −11.4 mV, as shown in Fig. S1.

The textural properties of UiO-66-NH₂ were evaluated by N₂ adsorption–desorption analysis (Fig. 4B), which exhibited a Type I isotherm with a steep uptake at (p/p⁰ < 0.1) confirming its predominantly microporous structure. Applying the conventional BET fitting range (p/p⁰ = 0.05–0.30) yielded an apparent surface area of 1312 m² g^{−1} and a total pore volume of 0.793 cm³ g^{−1}. The t-plot analysis further gave a micropore volume of 0.559 cm³ g^{−1}, accounting for

approximately 70% of the total pore volume, with micropore and external surface areas of 1120 and 192 m² g^{−1}, respectively, while the mean pore diameter calculated from (4V/S) was 2.42 nm. The high

accessible surface area is consistent with HCl-modulated UiO-66-NH₂ reported by Katz et al., where HCl increased the BET area from 830 to approximately 1200 m² g^{−1} through missing-linker defect formation [42]. In contrast, Sun et al. [43] and Wang et al. [44] reported lower values of 778 and 765 m² g^{−1} using water and acetic acid modulators, respectively, while Mohammadi et al. obtained 375 m² g^{−1} despite HCl modulation [45].

The FTIR spectrum of UiO-66-NH₂ particles is shown in Fig. 4.C. The analysis provides detailed information on the functional groups present, confirming the chemical structure of the material. The characteristic low-frequency vibrations associated with the Zr–O bonds within the inorganic Zr₆ cluster appear at 531 and 645 cm^{−1}, corresponding to μ₃-O and Zr–(OC) stretching modes. The C–H out-of-plane bending vibration of the benzene ring is observed at 793 cm^{−1}. The presence of amino-functionalized linkers is indicated by the C–N stretching band at 1203 cm^{−1}. Symmetric and asymmetric carboxylate stretching vibrations are identified at 1327 and 1567 cm^{−1}, respectively, while the aromatic C=C skeletal vibration is seen at 1523 cm^{−1}. Additionally, the primary amine (–NH₂) group exhibits weak to moderate symmetric and asymmetric stretching bands at 3318 and 3459 cm^{−1}, confirming successful amine functionalization of the UiO-66-NH₂ framework. The FTIR findings align with those reported in previous studies and further verify the successful synthesis of UiO-66-NH₂ particles [46–48].

Besides that, Fig. 4.C presents the XRD pattern of the synthesized UiO-66-NH₂. The material exhibits high crystallinity, as indicated by the characteristic reflections at 2θ = 7.4° and 8.5°, corresponding to the (111) and (200) planes. Additional well-resolved peaks at 12.0°, 14.1°, and 17.1° assigned to the (220), (311), and (400) planes further confirm the ordered framework structure. Higher-angle reflections at 22.3° and 25.7° ((511) and (600) planes) along with the higher-order peaks at 30.7° and 43.5° ((711) and (755) planes) demonstrate the long-range structural regularity and indicate the formation of well-defined crystallites with minimal amorphous contributions. These observations are consistent with the findings reported by Zhang et al. and Liu et al., who similarly demonstrated the high crystallinity of UiO-66-NH₂ in their studies [48,49].

To further verify the incorporation of UiO-66-NH₂ into the polymer matrix, FTIR and XRD analyses were performed on the synthesized MMMs, as shown in Fig. 4.D and E, respectively. The FTIR spectra of the MMMs (Fig. 4.D) exhibit weak but discernible symmetric and asymmetric stretching vibrations of primary amine (–NH₂) groups in the range of 3300–3660 cm^{−1} for membranes M1–M4, indicating the presence of amine-functionalized MOF fillers within the membrane matrix. Prominent absorption bands at approximately 3048 cm^{−1} and 1249 cm^{−1} correspond to aromatic C–H stretching and aromatic C–N stretching vibrations, respectively, which are consistently observed across all MMM samples.

Additional evidence of UiO-66-NH₂ incorporation is provided by characteristic peaks at approximately 534 and 738 cm^{−1}, attributed to Zr–O stretching and Zr–O framework (node) vibrations, respectively. The aromatic ring skeletal vibration (C=C) is identified at 1466 cm^{−1}, while coordinated carboxylate linkers are confirmed by the presence of symmetric and asymmetric stretching bands in the range of approximately 1277–1488 cm^{−1}. Concurrently, Shirazian et al. identified

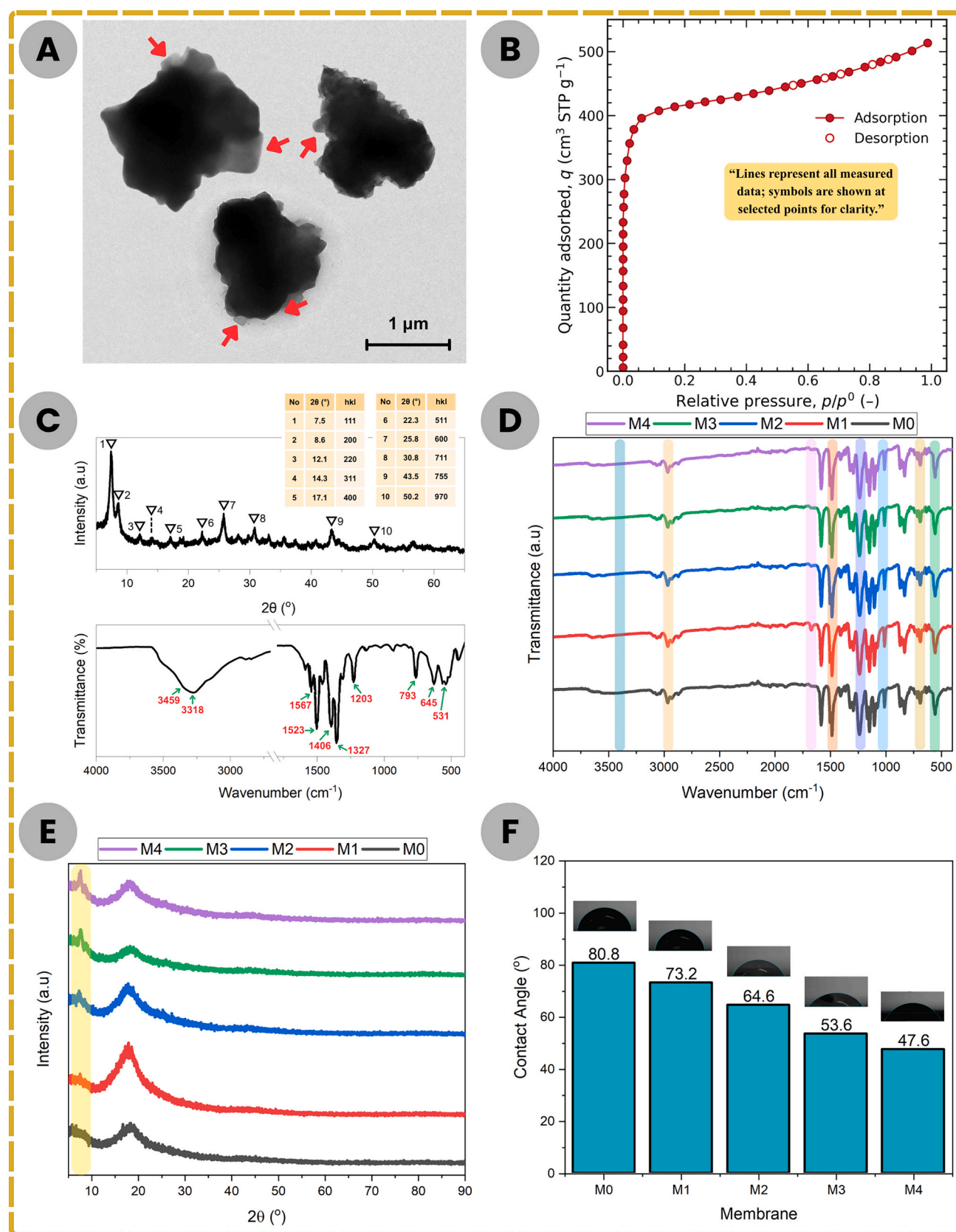


Fig. 4. (a) TEM image of UiO-66-NH₂ nanoparticles; (b) N₂ adsorption-desorption isotherms measured at 77 K and pressures up to 1 bar; (c) XRD diffractogram and FTIR spectrum of UiO-66-NH₂ nanoparticles; (d) FTIR spectra of pristine membranes and MMMs; (e) XRD diffractograms of pristine membranes and MMMs; (f) water contact angle measurements of pristine membranes and MMMs.

coordinated carboxylate peak pairs; however, these features were observed within the $1400\text{--}1600\text{ cm}^{-1}$. Additionally, characteristic shifts were noted for --NH_2 stretching vibrations at $3200\text{--}3400\text{ cm}^{-1}$ and C—H stretching modes at $2800\text{--}2900\text{ cm}^{-1}$ [50].

These FTIR observations are further supported by XRD analysis (Fig. 4.E), which confirms the presence and structural integrity of UiO-66-NH₂ within the mixed matrix membrane (MMM). A distinct diffraction peak at $2\theta \approx 7.4^\circ$, corresponding to the (111) crystallographic plane of UiO-66-NH₂, is clearly observed, indicating the retention of its characteristic crystalline framework after incorporation into the polymer matrix. In addition, the (200) crystallographic plane appears at $2\theta = 8.61^\circ$; however, its intensity is significantly reduced due to overlap with the broad amorphous halo of the polymer. As a result, this peak is only discernible in membranes with higher UiO-66-NH₂ loading (M3 and M4), where the contribution of the crystalline filler becomes more pronounced.

Comparable diffraction features have been reported by Xu et al., who observed the (111) and (200) planes of UiO-66-NH₂ at $2\theta = 7.34^\circ$ and 8.48° , respectively [51]. The close agreement between the diffraction peak positions in this study and those reported in the literature indicates that the crystalline structure of UiO-66-NH₂ is well preserved during membrane fabrication. The reduced peak intensity and partial masking of MOF reflections in the MMMs can be attributed to the low filler content and the dominant amorphous nature of the polymeric matrix. Overall, the XRD results confirm the successful incorporation of UiO-66-NH₂ nanoparticles into the membrane without significant loss of crystallinity, supporting their structural stability and functional contribution within the MMM.

The effect of UiO-66-NH₂ incorporation on membrane surface wettability was assessed by static water contact angle measurements (Fig. 4.F). The M0 exhibited a relatively hydrophobic surface with a contact angle of 80.8° . With increasing UiO-66-NH₂ loading, the contact angle progressively decreased to 73.2° , 64.6° , 53.6° , and 47.6° for membranes containing 1, 3, 5, and 7 wt% filler, respectively, demonstrating a systematic enhancement in membrane hydrophilicity. This improvement can be attributed to the combined effects of filler chemistry and surface morphology. The abundance of hydrophilic functional groups on UiO-66-NH₂ promotes their migration toward the membrane surface during phase inversion, thereby modifying surface chemistry [52]. In parallel, the incorporation of nanoparticles induces a rougher, wrinkled surface topology, which enhances wettability. Moreover, the hydrophilic groups within the UiO-66-NH₂ framework readily form hydrogen bonds with water molecules, facilitating water penetration and diffusion into the mixed matrix membrane [53].

The surface morphology and elemental composition of the MMMs were characterized via SEM-EDX (Fig. 5). The M0 exhibited a smooth surface (Fig. 5.A); however, the incorporation of UiO-66-NH₂ nanoparticles induced higher surface roughness (Fig. 5.B–E). This morphological shift is attributed to the inherent challenges of achieving ideal filler dispersion within the PSf matrix, despite pre-dispersion in DMF. To further confirm particle agglomeration and localized Zr accumulation within the membrane structure, cross-sectional SEM-EDX analysis was performed by comparing M0 and M3, as presented in Fig. S2. The M3 cross-section reveals distinct Zr-rich agglomerates within the membrane pores, highlighted by the green circle, indicating localized accumulation of UiO-66-NH₂ particles. Similar increases in roughness were reported by Miao et al. for UiO-66-NH₂@PSf hollow fiber membranes. Furthermore, nanoparticle addition appeared to promote pore enlargement, likely due to localized filler agglomeration [54]. This contrasts with the findings of Nayeri et al., who observed reduced porosity as nanoparticles occupied nanovoids within the polymer structure [55]. Finally, EDX analysis showed an increasing Zr mass fraction with increasing nominal UiO-66-NH₂ loading, providing compositional evidence for successful filler incorporation into the membrane. To further evaluate the membrane surface after prolonged operation, the surface morphology and elemental distribution after four cycles of geothermal brine filtration

were examined, as shown in Fig. 6.A–E. The membrane appearance after filtration suggests that part of the silica was retained within the membrane structure, particularly in the pore region. This observation is supported by the quantitative EDX comparison before and after filtration: Si was not detected in any membrane before testing, whereas after filtration the residual Si content remained low, only 0.4–0.8 wt%. Normalization to the PSf backbone showed that the Si/S ratio increased from 0 to 0.063, 0.066, 0.136, 0.098, and 0.067 for M0–M4, respectively, while normalization to the MOF tracer gave Si/Zr ratios of 0.80, 1.33, 0.75, and 0.36 for M1–M4. These low Si/S and Si/Zr ratios confirm that silica deposition occurred only as a minor residual deposit after filtration, indicating that backwashing effectively removed most surface-associated silica but could not completely eliminate silica trapped within narrowed pores. This finding was evident in previous research by Cheng et al., who tested the use of commercial polyethersulfone (PES)-based KOCH membranes in groundwater pretreatment [56].

3.2. Membrane performance

Membrane performance was evaluated using geothermal brine samples provided by the research collaborator PT Geo Dipa Energi (GDE) Dieng. The samples were collected from a reinjection fluid storage pond after thermal energy extraction for turbine operation. During cooling in the conveyance canal and within the storage pond, a substantial fraction of dissolved silica precipitated, resulting in a remaining SiO₂ concentration of approximately 400 ppm [57]. This behavior is consistent with previous observations reported by Pambudi in studies on geothermal energy systems in the Dieng field [58]. A schematic of the sampling location is shown in Fig. 7.A, and photographic views of the facility and silica settling pond are presented in Fig. 7.B–D.

The silica separation performance of the membranes over four filtration–backwash cycles is summarized in Fig. 8.A, while the corresponding average permeability values are shown in Fig. 8.B. Overall, the incorporation of UiO-66-NH₂ nanoparticles enhanced the membranes' resistance to inorganic fouling; however, a gradual decline in permeability was still observed for all membranes over successive filtration cycles (Fig. 8.A). However, the presence of UiO-66-NH₂ clearly enhanced membrane permeability, consistent with the increased hydrophilicity indicated by the reduced contact angle discussed in the previous section.

For the M0, a pronounced permeability decline was observed during each filtration cycle. After the first cycle, permeability decreased from $158.94\text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ to $49.82\text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ (0.31 of the initial value). Backwashing partially restored permeability to approximately 70% of the initial value, but subsequent cycles exhibited progressively lower recovery. By the end of the fourth cycle, permeability was reduced to $21.30\text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$, corresponding to only 13% of the initial membrane performance, despite partial recovery after each backwash.

Membranes incorporating UiO-66-NH₂ exhibited higher initial and average permeabilities, with the most pronounced improvement observed for M3, followed by M4, M2, and M1 (Fig. 8.B). For M3, the initial permeability was $392.62\text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$, which decreased to $109.93\text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ after the first cycle (0.28 of the initial value). Backwashing restored permeability to approximately 83% of the initial value, and although permeability declined in subsequent cycles, recovery after each backwash remained higher than that of M0. At the end of the fourth cycle, M3 retained approximately 17% of its initial permeability, indicating improved flux performance relative to the pristine membrane.

The performance trends of M0 and M1 were broadly similar, although M1 exhibited more stable permeability recovery. Despite having a lower initial permeability ($142.99\text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$), M1 maintained higher relative permeability after each cycle compared with M0. After the first cycle, M1 retained 47% of its initial permeability, and backwashing restored permeability to 93%. Consistent recovery was

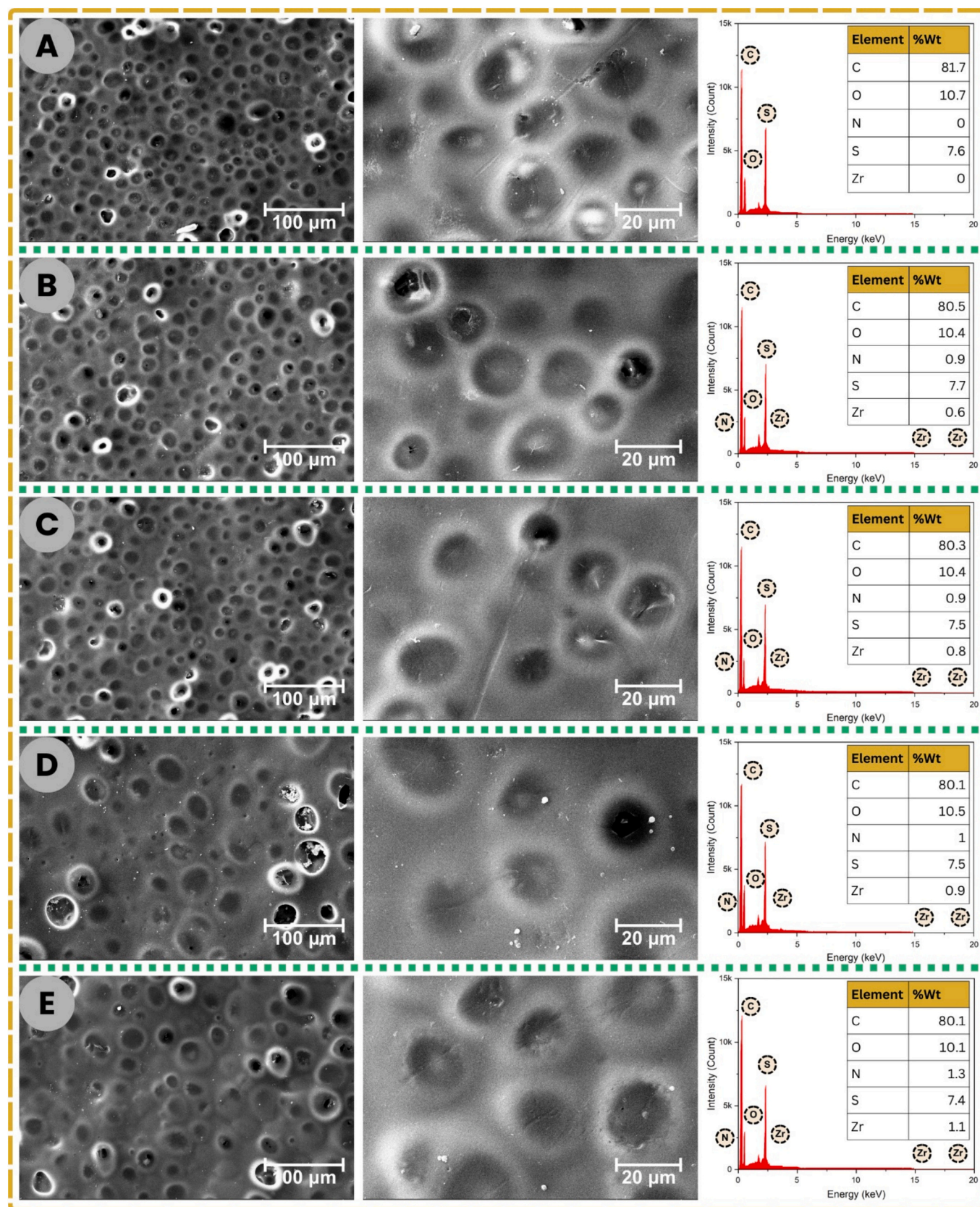


Fig. 5. Surface morphology (500 $\times$ and 1000 $\times$ magnification) and elemental composition of membranes M0–M4 before geothermal brine filtration.

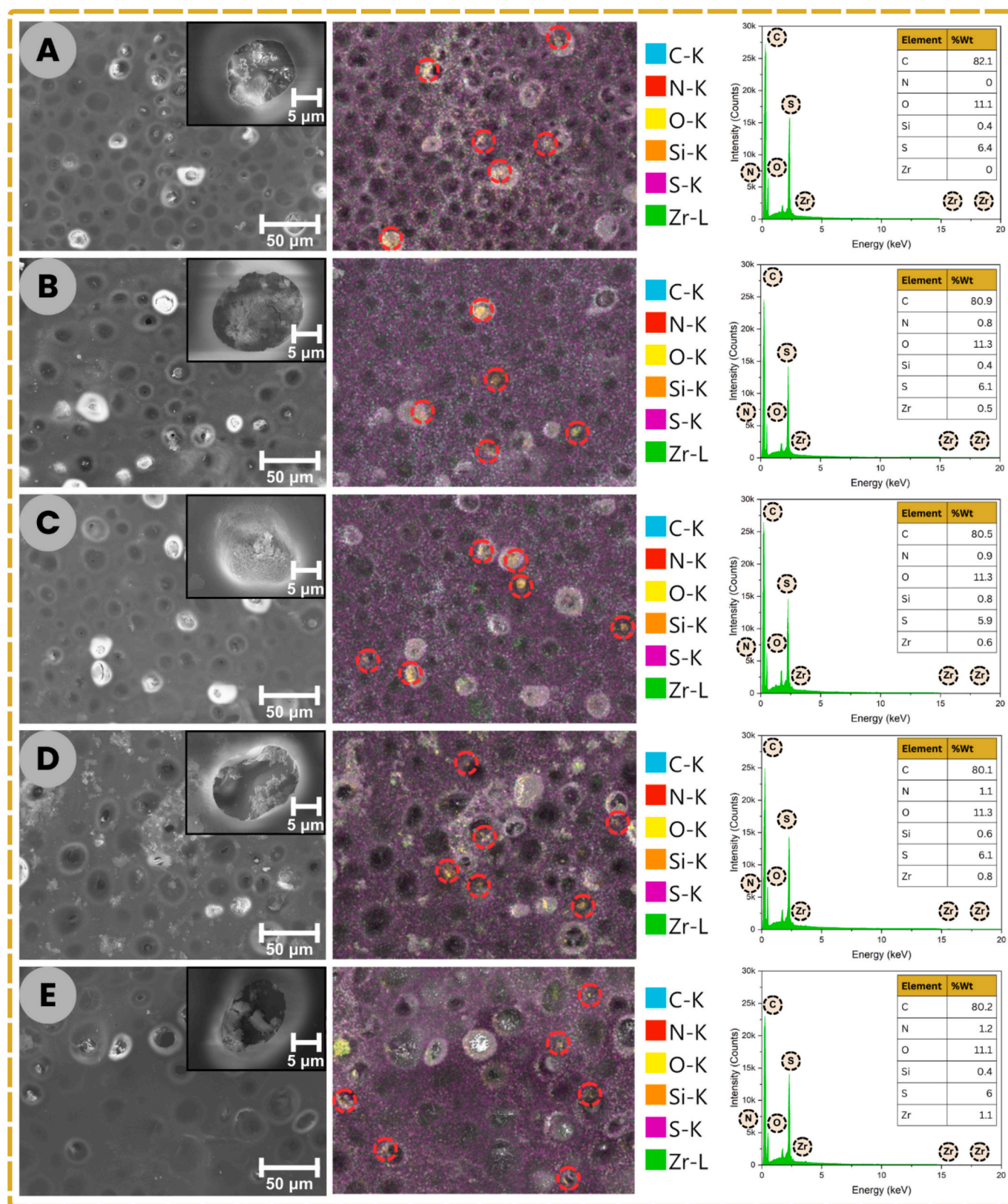


Fig. 6. Surface morphology (500 × and 5000 × magnification) and elemental composition of membranes M0–M4 after geothermal brine filtration.

also observed in cycles 3 and 4, with permeability recoveries of 78% and 75%, respectively.

Overall, these results indicate that while UiO-66-NH₂ incorporation does not significantly suppress inorganic fouling, it enhances membrane

permeability and improves flux recovery after backwashing, thereby contributing to more stable membrane performance during repeated geothermal brine filtration cycles.

The antifouling performance analysis was conducted to elucidate the

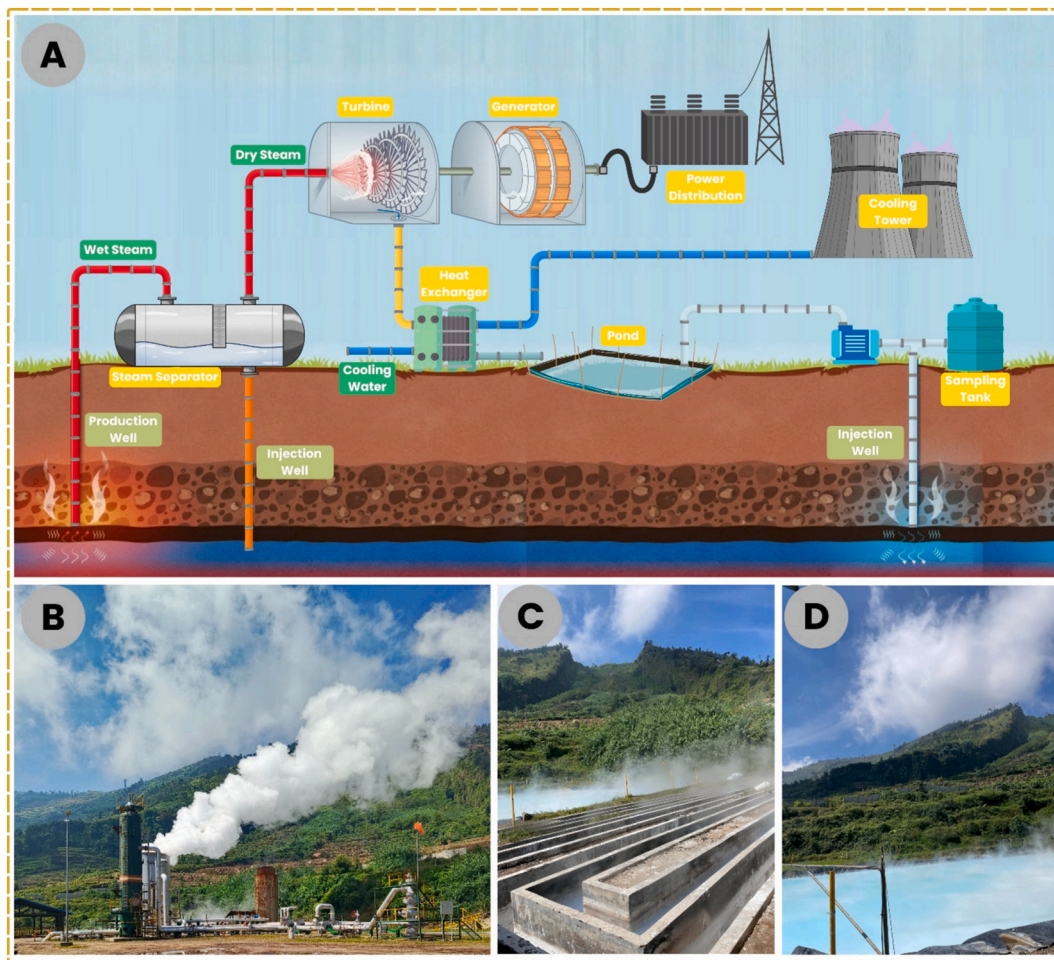


Fig. 7. Schematic representation of the PT Geo Dipa Energi (GDE) Dieng geothermal power plant (a), view of the reinjection site (b), and views of the conveyance canal and reinjection fluid storage pond prior to reinjection (c–d).

long-term fouling behavior and cleaning efficiency of the membranes. Fig. 8.C summarizes the FRR, RFR, IFR, TFR, which collectively describe the extent, reversibility, and severity of fouling during silica filtration. Overall, the membranes exhibited markedly different antifouling behaviors, with FRR values ranging from 56.63% to 82.94%, indicating varying degrees of fouling reversibility. Among the tested membranes, M2 showed a noticeably lower permeability recovery after backwashing, as reflected by its substantially reduced RFR compared with M1, M3, and M4. This behavior suggests the presence of particle agglomeration within the membrane structure, leading to pore blockage and pore narrowing. Such conditions hinder the removal of polymerized silica from the pores during backwashing and are consistent with the relatively high IFR observed for M2 (43.37%), indicating dominant irreversible fouling.

In contrast, membranes exhibiting higher RFR values demonstrated predominantly reversible fouling, which can be effectively mitigated by physical cleaning. Notably, the low IFR and favorable RFR observed for M3 indicate that fouling was largely weakly bound [59]. This behavior can be attributed to the homogeneous dispersion of nanoparticles within the membrane matrix, which minimizes pore clogging and facilitates the removal of silica deposits during backwashing. In addition, the presence of amine functional groups associated with the nanoparticles likely reduces silica adhesion by increasing membrane hydrophilicity and electrostatic repulsion, both on the membrane surface and within the pore structure [60].

The combined effects of nanoparticle dispersion and surface functionalization on fouling mechanisms are schematically illustrated in

Fig. 8.D. Collectively, these results demonstrate that appropriate nanoparticle incorporation can effectively shift fouling behavior from irreversible to predominantly reversible, thereby enhancing membrane cleanability and long-term operational stability.

3.3. Membrane fouling prediction

To complement the experimental antifouling analysis, membrane fouling behavior was quantitatively predicted using the modeling framework described in Section 2.5, with the considered inorganic fouling mechanisms schematically illustrated in Fig. 9.B–E. Permeability–time data were fitted to the selected fouling models following the computational workflow shown in Fig. 9A. Nonlinear regression was performed using MATLAB R2025b, employing the *lsqnonlin* function with the Trust-Region-Reflective (TRR) algorithm. Model performance was assessed using the coefficient of determination (R^2), root mean squared error (RMSE), and normalized root mean squared error (NRMSE).

Fitting results and statistical metrics for the four Hermia models are presented in Tables S1–S4 and summarized in Fig. S3. The cake/gel layer model consistently showed the poorest agreement across all membranes and cycles (mean $R^2 = 0.49$ – 0.72 ; NRMSE = 17.0–22.9%), suggesting that flux decline was not governed by the formation of a coherent surface cake or gel layer, but rather by fouling within the membrane pore structure. Although complete blocking produced the highest mean R^2 values (0.77–0.91), it was excluded on mechanistic grounds because the model assumes that each incoming particle independently seals a pore

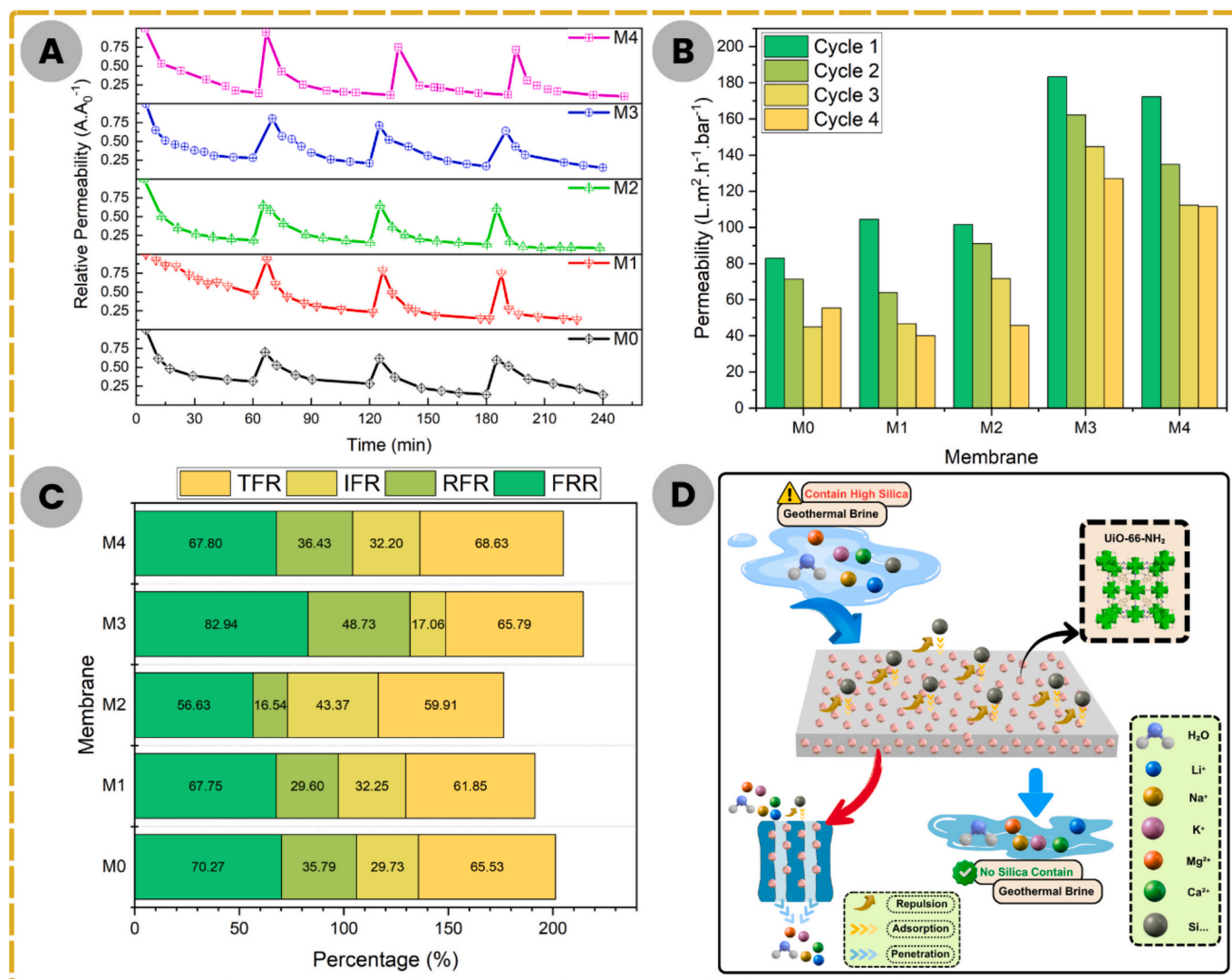


Fig. 8. (a) Time-dependent filtration performance of membranes M0–M4 over four test cycles; (b) average permeability of membranes M0–M4 during four test cycles; (c) evaluation of average fouling parameters (FRR, RFR, IFR, and TFR) for membranes M0–M4 after four test cycles; and (d) schematic illustration of silica removal from geothermal brine using MMMs.

entrance without particle superposition, which requires discrete species comparable to or larger than the pore size. Since the feed was dominated by dissolved and oligomeric silicic acid, this assumption is unlikely to be valid. Accordingly, mechanism identification was focused on intermediate and standard blocking, both of which represent internal deposition and differed from complete blocking by only $\Delta R^2 \leq 0.03$ across the dataset.

The two membranes exhibited distinct stability in their dominant fouling mechanisms (Fig. 10). For M0, intermediate blocking dominated the first cycle ($R^2 = 0.85$, NRMSE = 13.3%), followed by a persistent shift to standard blocking from cycle 2 onward ($R^2 = 0.87, 0.92$, and 0.92 ; NRMSE = 12.7, 10.0, and 10.1%). Once the initial fouling layer had formed, subsequent flux decline was therefore governed mainly by progressive constriction of the internal pore walls rather than by surface coverage. In contrast, M3 did not exhibit the same mechanistic lock-in. Intermediate blocking dominated cycles 1 and 4 ($R^2 = 0.89$ and 0.98), whereas standard blocking prevailed in cycles 2 and 3 ($R^2 = 0.84$ and 0.95). Overall fit quality improved across the filtration sequence, with NRMSE values of 9.7, 13.5, 7.7, and 4.3%. The reappearance of intermediate blocking in the final cycle suggests that hydraulic cleaning effectively restored the pore structure between cycles and limited the cumulative internal constriction observed for M0.

This alternating behavior reflects the progressive evolution of silica speciation during each filtration cycle. Monomeric silicic acid is considerably smaller than the membrane pores and can initially penetrate the pore network with minimal restriction, consistent with the exclusion of complete blocking as the dominant mechanism. As filtration continues, concentration polarization and localized supersaturation promote polymerization into oligomeric and eventually colloidal silica, both inside the pore channels and at the membrane surface. Consequently, the dominant resistance gradually shifts from transient surface deposition toward internal pore obstruction. Silica formed during the early stage of each cycle remains relatively loose and can be partially removed by backwashing, so the prevailing fouling mechanism depends on the extent to which deposits from the previous cycle are displaced.

Pore accessibility strongly influences the location of silica deposition. Wang et al. reported that pore clogging progressively intensified during silica filtration and ultimately became a greater contributor to flux decline than cake-layer formation [61]. Similarly, Springer et al. found that a relatively open 100 kDa membrane was primarily affected by internal fouling, whereas a tighter 10 kDa membrane was dominated by surface cake formation [62]. Both findings are consistent with the poor fit of the cake/gel layer model across the present dataset (Fig. S3), indicating that fouling resistance developed predominantly within the

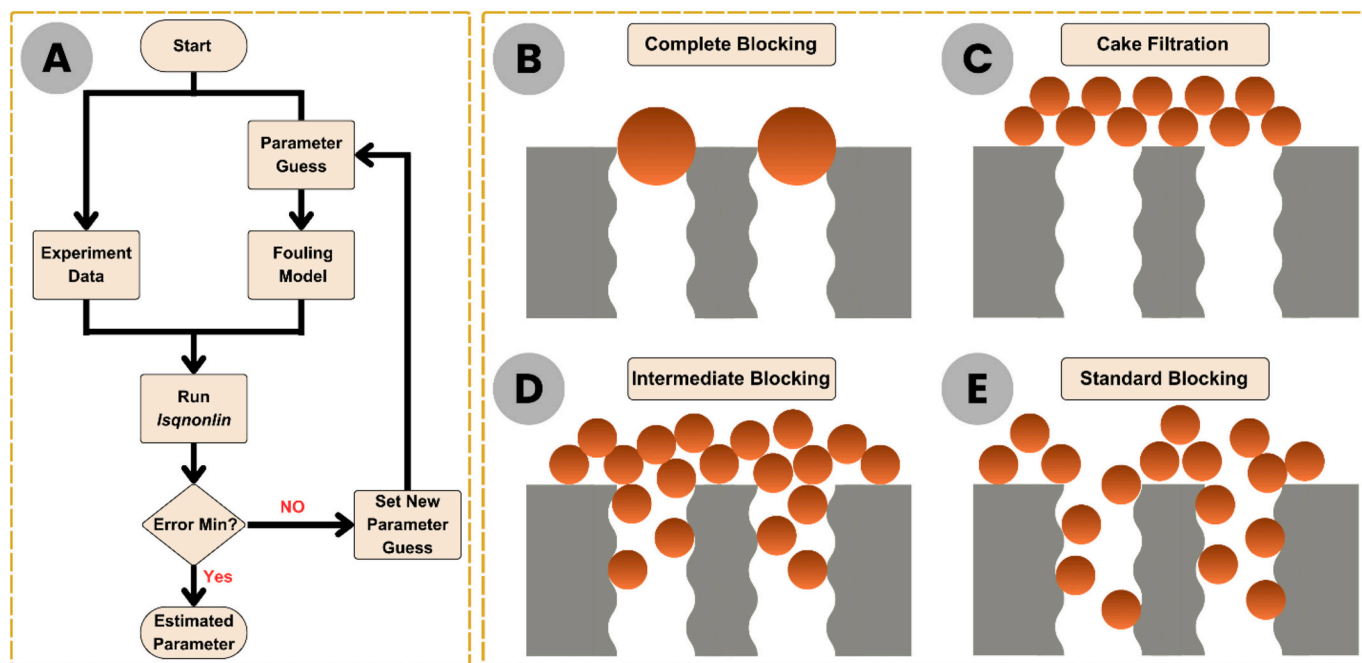


Fig. 9. (a) Workflow for parameter estimation using a numerical optimization approach; (b-e) Representative inorganic fouling mechanisms occurring in MMMs.

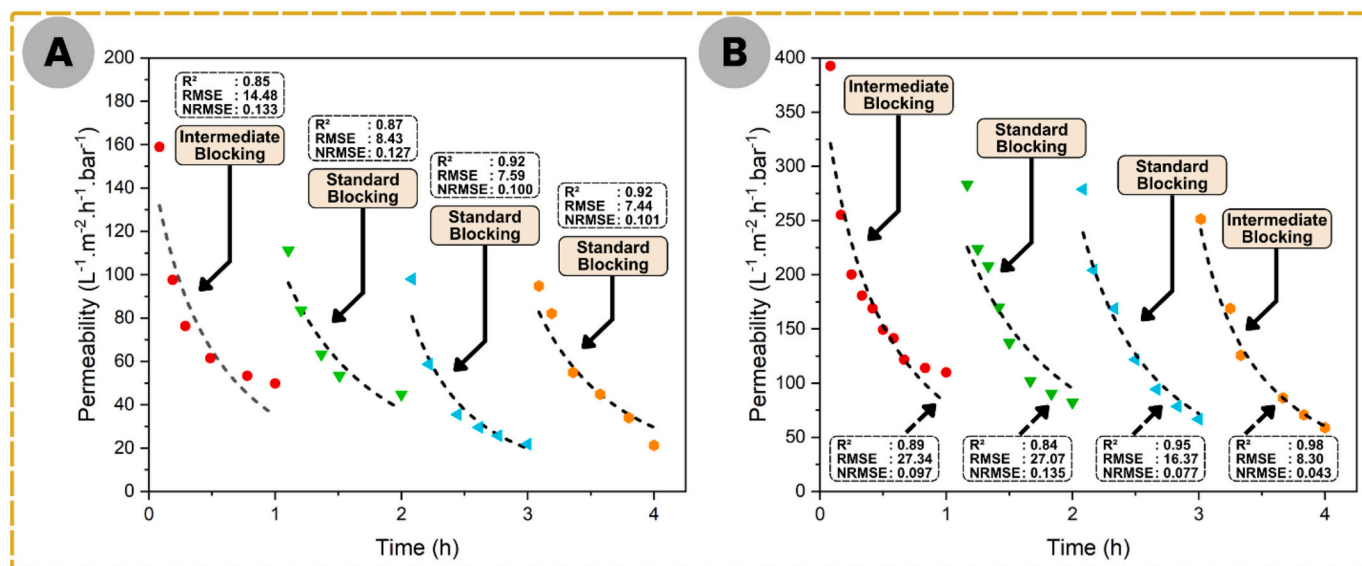


Fig. 10. Comparison of permeability profiles and fouling model fitting for (A) M0 and (B) M3.

pore structure rather than as a distinct surface layer.

In M3, UiO-66-NH₂ further modifies this behavior through additional interactions at the membrane–solution interface. Its amine groups facilitate acid–base interactions and hydrogen bonding with silicic acid, while its high specific surface area provides additional nucleation sites that influence where silica growth begins and how strongly the resulting deposits adhere. Electrostatic effects are expected to be secondary because, given the high pK_a of silicic acid (~9.8), silica species in near-neutral geothermal brine are predominantly uncharged and are therefore only weakly repelled by the negatively charged membrane surface. Farahbakhsh et al. similarly reported only modest changes in zeta potential after incorporating NH₂-UiO-66 into a polysulfone UF matrix, from –11 to –14 mV for the unmodified membrane to –13 to –15 mV for the modified membrane above the isoelectric point [63]. Collectively, these effects explain the different fouling behaviors of the two

membranes. In M0, silica polymerization within the unmodified pore network generated deposits that could not be fully removed by backwashing, causing the mechanism to remain dominated by standard blocking from the second cycle onward. In M3, the amine-functionalized framework produced deposits that remained more accessible to hydraulic cleaning, allowing partial recovery of the pore structure between cycles and enabling the dominant mechanism to return to intermediate blocking in the final cycle.

The model constants (Fig. S4) and their evolution from the first to the fourth cycle (Fig. S5) further quantify this distinction. Considering the two mechanistically valid models, M0, M3, and M4 remained close to their initial values ($K_4/K_1 = 0.93$ – 1.43 for K_i and K_s), indicating that backwashing removed most of the accumulated deposits. In contrast, M1 and M2 showed 7.7- and 2.7-fold increases in K_i and 5.9- and 2.1-fold increases in K_s , respectively, indicating progressive silica accumula-

tion within the pore network that could not be fully removed by hydraulic cleaning. For M0, when the kinetic analysis was limited to the standard blocking regime dominating cycles 2–4, K_s increased by 82% from cycle 2 to cycle 3 (6.78×10^{-2} to $12.34 \times 10^{-2} \text{ m}^{-1}$), followed by a 34% decrease from cycle 3 to cycle 4 (to $8.11 \times 10^{-2} \text{ m}^{-1}$). This decline is attributed to the backwashing step after cycle 3, which likely removed a substantial fraction of the retained foulants and reduced the available sites for further interaction between bulk silicic acid and residual deposits. Across the full filtration sequence, however, the overall change remained minimal ($K_4/K_1 = 0.93$). This value should not be interpreted as improved fouling resistance because, by cycle 4, M0 permeability had already reached a low quasi-steady plateau, where the fitted rate constant becomes inherently suppressed. Thus, the apparent reduction reflects a saturation effect rather than genuine fouling mitigation.

3.4. Techno-economic analysis

A screening-level techno-economic analysis was conducted to evaluate the scale-up relevance of UiO-66-NH₂ MMMs as a pretreatment unit for geothermal brine. The analysis was limited to the pretreatment stage, where the main function is to reduce silica-rich colloids, suspended solids, and fouling precursors before downstream lithium recovery. Three scenarios were compared under an equivalent process basis: Scenario 1, conventional coagulation–flocculation–clarification pretreatment (S1); Scenario 2, commercial UF membrane pretreatment with periodic cleaning (S2); and Scenario 3, UiO-66-NH₂ MMM-based UF pretreatment with periodic cleaning (S3). The schematic representation of each scenario is shown in Fig. 11A–C.

The process assumptions were developed from the experimental results obtained in this study and supporting information from relevant literature, technical references, cost databases, and vendor-based data where applicable. To provide a scale-up perspective, the TEA was evaluated at representative feed capacities of 100 (C1) and 1000 m³/day (C2), corresponding to demonstration and industrial-scale implementation. All scenarios were assessed using a common design basis, including continuous operation for 24 h/day with scheduled service downtime, equivalent to 330 operating days/year. The recovery was

assumed to be 90–98% for S1 and 85–95% for S2 and S3, representing the treated brine volume generated by each pretreatment route. All costs were normalized to a 2026 reference cost year, assuming an electricity price of 0.06–0.09 USD/kWh and a discount rate of 10%. The project lifetime was set at 15 years, comprising 2 years of construction and 13 years of operation. These general assumptions were kept consistent across all scenarios, while technology-specific parameters were applied according to each process configuration. The major equipment items included in the TEA for scenarios S1–S3 are summarized in Tables S5–S7, while the detailed CAPEX and OPEX breakdowns for C1 and C2 under each scenario are provided in Tables S8–S10.

As shown in Fig. 11, Scenario 1 employs four stirred units for conventional coagulation–flocculation–clarification pretreatment. This configuration is designed to remove up to 95% of colloidal silica while recovering 93.3% of the geothermal brine as treated brine. The geothermal brine is first conditioned to the target alkaline condition, typically pH ~ 8.0 to 8.5, NaOH to promote effective silica management and subsequent coagulation–flocculation. At this pH range, silica particles can remain colloidally stable due to electrostatic repulsion; however, beyond approximately pH 9, the diffuse double layer may become compressed and silica particles may begin to partially dissolve, indicating the onset of silica solubilization [64]. The coagulation step is then conducted under rapid mixing by adding PAC at 17 mg/L for 30–60 s, aiming to neutralize the surface charge of silica colloids and initiate micro-floc formation. These micro-flocs are subsequently enlarged during flocculation using PAM at 14 mg/L under slow mixing for 30–60 min [65,66]. The chemical dosage must be carefully controlled to avoid suspension re-stabilization [67,68]. Finally, the formed large and dense flocs are separated in the clarifier under very low mixing or quiescent conditions, allowing solids to settle and clarified brine to be recovered.

In contrast, Scenarios 2 and 3 use more compact UF membrane-based pretreatment configurations. S2 employs a commercial 30 kDa PVDF UF membrane, while S3 uses the developed MMM UF membrane. Based on the design assumptions, S2 achieves a removal efficiency of approximately 85% with 89% geothermal brine recovery, whereas S3 provides a slightly higher removal efficiency of 88% with 92% recovery. Both membrane-based scenarios incorporate periodic water flushing/

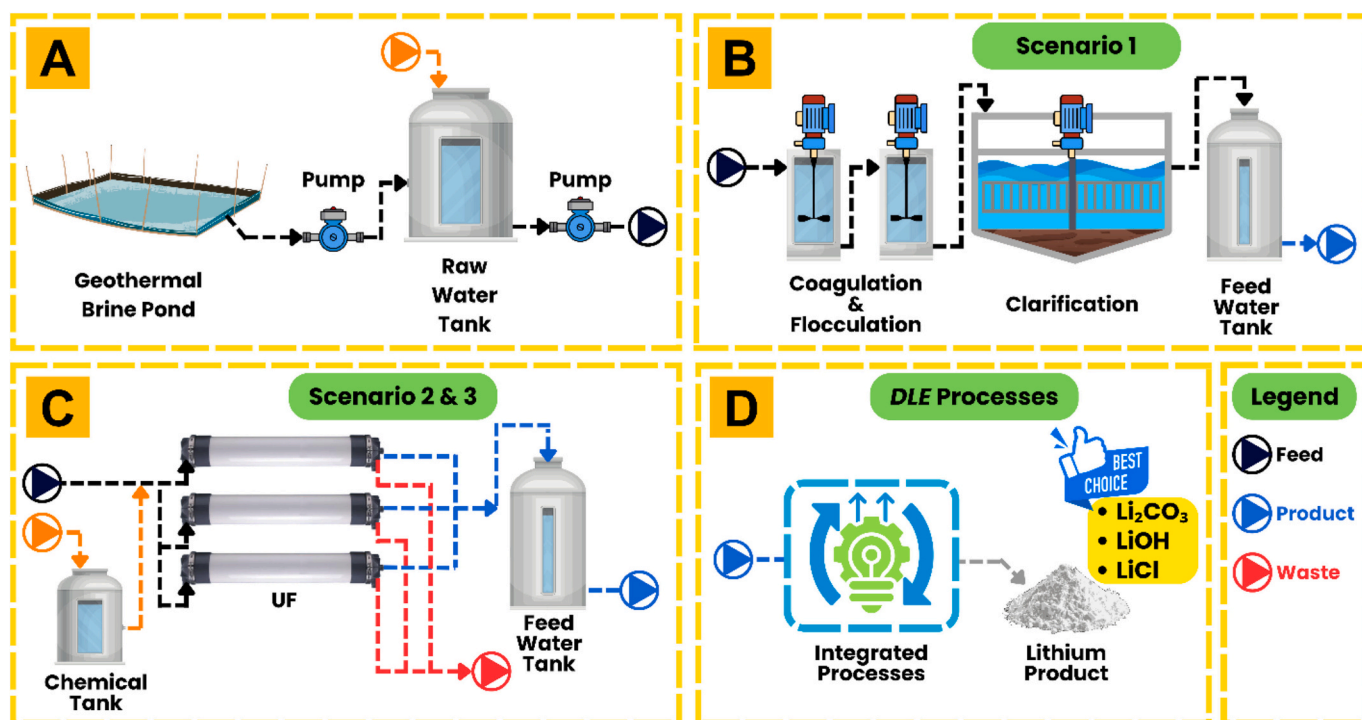


Fig. 11. (a) illustrates geothermal brine preparation, (b) Scenario 1, (c) Scenario 2 & 3, and (d) Direct Lithium Extraction.

backwashing and chemical cleaning to remove accumulated foulants and restore membrane productivity. [69,70]. For M3, the cleaning protocol is expected to recover permeability to an FRR of approximately 83–94%, which theoretically approaches the practical recovery limit of the membrane after repeated fouling–cleaning cycles. Controlled NaOH cleaning can enhance silica removal; however, excessive exposure should be avoided because it may compromise the polymer matrix and UiO-66-NH₂ stability [71]. The complete cleaning cycle, including chemical treatment and water flushing/backwashing, requires approximately 4 h. To maintain continuous pretreatment operation during this downtime, multiple membrane units are operated alternately.

Based on the available technical design data, CAPEX and OPEX were estimated for S1–S3 at C1 and C2. CAPEX represents the procurement and installation costs required before plant operation, while OPEX represents recurring operating costs, including variable and fixed components [72]. The total capital investment (TCI) was estimated using a screening-level factorial approach, where the purchased equipment cost (PEC) was multiplied by a Lang factor of approximately 3.6. At C1, S1 showed the highest PEC of 53,900 USD, corresponding to a TCI of 194,040 USD. In comparison, S2 and S3 had lower PEC values of 30,100 USD and 35,300 USD, resulting in TCIs of 108,360 USD and 127,080 USD, respectively. At C2, the PEC increased to 270,139.92 USD, 150,857.36 USD, and 176,919.09 USD for S1, S2, and S3, corresponding to TCIs of 972,503.71 USD, 543,086.49 USD, and 636,908.74 USD, respectively. Thus, compared with S1, S2 and S3 reduced TCI by approximately 44% and 35%, respectively, at both capacities, confirming the lower initial investment required for membrane-based pretreatment.

The annual cost structure shows that annualized CAPEX remained the dominant contributor in most cases, although its relative contribution decreased with scale. At C1, annualized CAPEX accounted for 60.0%, 53.7%, and 55.9% of the total annual cost for S1, S2, and S3, respectively, corresponding to 25,511.17 USD/year, 14,246.50 USD/year, and 16,707.69 USD/year; the remaining 40.0%, 46.3%, and 44.1% originated from OPEX. At C2, the CAPEX contributions decreased to

56.8%, 50.4%, and 53.9%, despite their absolute values increasing to 127,855.06 USD/year, 71,399.58 USD/year, and 83,734.39 USD/year, respectively. Consequently, the total annual cost increased from 42,511.78 → 225,038.06 USD for S1, 26,515.47 → 141,641.92 USD for S2, and 29,876.74 → 155,240.52 USD for S3 when scaling from C1 to C2.

The OPEX profile further differentiates the three scenarios. For S1, total OPEX increased from 17,000.61 USD at C1 to 97,183.00 USD/year at C2, with electricity remaining the dominant component (7880.01 → 52,533.37 USD; 46.4–54.1%), followed by labor (5105.00 → 13,427.52 USD) and waste handling (1501.01 → 15,010.05 USD). S2 exhibited lower OPEX of 12,268.97 → 70,242.34 USD/year. At C1, labor was dominant (5105.00 USD; 41.6%), whereas at C2 waste handling became the largest contributor (29,695.38 USD; 42.3%), followed by labor (13,427.52 USD; 19.1%), chemicals (9950.00 USD; 14.2%), and electricity (9396.21 USD; 13.4%). A similar transition occurred for S3, where OPEX increased from 13,169.05 USD to 71,506.13 USD/year. Labor dominated at C1 (5105.00 USD; 38.8%), followed by waste handling (2969.62 USD; 22.5%), insurance (1450.00 USD; 11.0%), electricity (1409.43 USD; 10.7%), and maintenance (1240.00 USD; 9.4%). At C2, waste handling became dominant (29,696.21 USD; 41.5%), followed by labor (13,427.52 USD; 18.8%), chemicals (9950.00 USD; 13.9%), electricity (9396.21 USD; 13.1%), and insurance (7267.01 USD; 10.2%). These results indicate that membrane-based systems reduce capital intensity, but their OPEX remains sensitive to waste handling, chemicals, and maintenance.

The scale-up from C1 to C2 was evaluated using a nonlinear power-law cost relationship to capture economies of scale, such that a 10-fold increase in treatment capacity does not produce a proportional increase in CAPEX. Although scaling exponents (n) are equipment-specific, values of approximately 0.6–0.7 are commonly adopted for preliminary cost estimation. Reported values for water-treatment equipment vary considerably, including 0.539 for coagulation units, 0.713 for UF systems, 0.676 for clarifiers, 0.088 for pumps, 0.655 for storage tanks, and 0.696 for stirred tanks (Fig. S6A–F) [73–75]. Considering

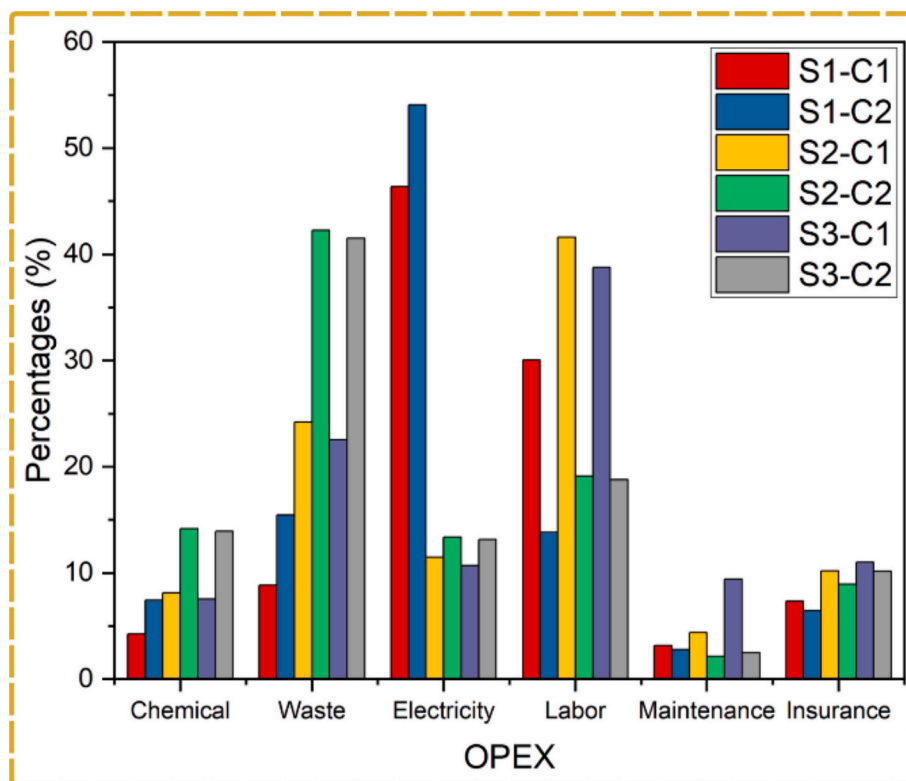


Fig. 12. Relative contribution of annual operating cost components to total OPEX for S1–S3 at C1 and C2.

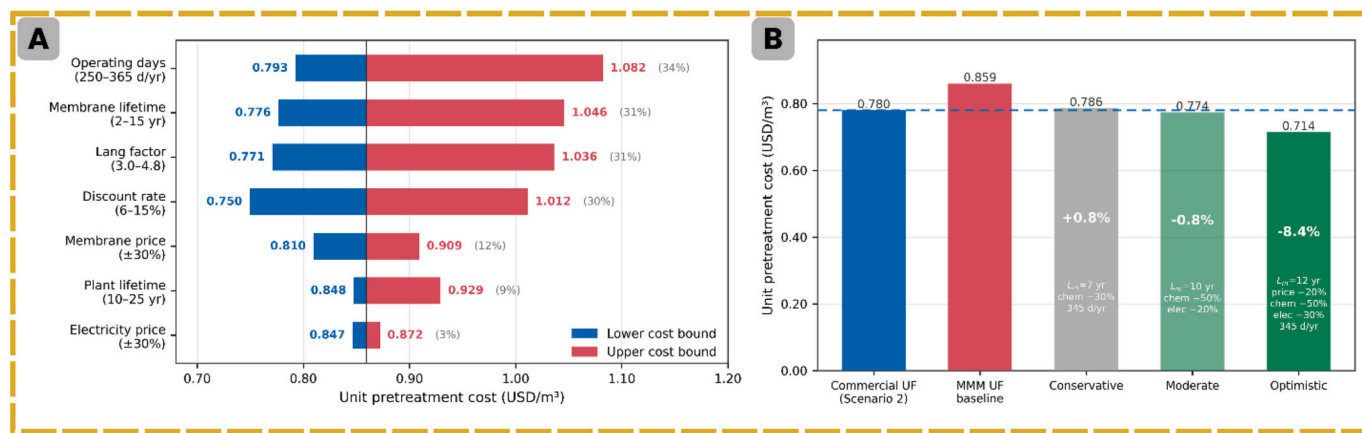


Fig. 13. Techno-economic sensitivity and pathways to cost competitiveness of membrane-based geothermal brine pre-treatment: (A) one-factor-at-a-time sensitivity analysis of unit pre-treatment cost and (B) pathway scenarios for achieving cost competitiveness of MMM-based UF relative to commercial UF.

this variability and the screening-level nature of the present TEA, a uniform and conservative scaling exponent of $n = 0.7$ was selected as a practical basis for scale-up. Consequently, increasing capacity from C1 to C2 by a factor of 10 increases the corresponding equipment cost by only approximately 5.0-fold, rather than tenfold. In contrast, flow-dependent OPEX components, including chemicals and waste handling, were estimated proportionally to the treated brine volume, while supporting equipment and installation costs were adjusted using the scaling exponent. As shown in Fig. 12 in percentage terms and fully detailed in Tables S7 to S9, pretreatment costs per unit decrease from 1.29 to 0.68 USD/m³ for S1, 0.80 to 0.43 USD/m³ for S2, and 0.91 to 0.47 USD/m³ for S3 when scaling from C1 to C2. This corresponds to cost reductions of approximately 47.3%, 46.3%, and 48.4%, respectively. Overall, scaling up improves the economic performance of all scenarios, with S2 showing the lowest unit pretreatment cost at both capacities, while the feasibility of S3 remains strongly governed by controlling waste handling, cleaning demand, membrane lifetime, and replacement-related costs.

To evaluate the robustness of the TEA for C1, a one-factor-at-a-time sensitivity analysis was performed for S2 and S3. The results identified plant utilization as the dominant determinant of unit pre-treatment cost, followed by membrane lifespan, the Lang factor, and the discount rate (Fig. 13.A). Varying the operating period from 250 to 365 days/year produced a cost range of 0.793–1.082 USD/m³, corresponding to a 34% deviation from the baseline value. Extending the membrane lifespan from 2 to 15 years shifted the cost to 0.776–1.046 USD/m³ (31%), whereas varying the Lang factor from 3.0 to 4.8 and the discount rate from 6% to 15% resulted in ranges of 0.771–1.036 USD/m³ (31%) and 0.750–1.012 USD/m³ (30%), respectively. By comparison, membrane price (±30%), plant lifespan (10–25 years), and electricity price (±30%) exerted substantially smaller effects, yielding cost ranges of 0.810–0.909 USD/m³ (12%), 0.848–0.929 USD/m³ (9%), and 0.847–0.872 USD/m³ (3%), respectively. Within the TEA framework, these results indicate that project-level and financial assumptions exert a stronger influence on pre-treatment economics than energy-price fluctuations. The limited sensitivity to electricity price further reflects the relatively low specific energy demand of the UF process, confirming that the estimated process economics are comparatively robust against variations in electricity tariffs.

Conversely, Fig. 13.B presents the pathway scenarios required for the MMM-based UF system (S3) to achieve cost competitiveness with the commercial UF system (S2). Under baseline conditions, S3 exhibits a unit pre-treatment cost of 0.859 USD/m³, approximately 10% higher than that of S2 at 0.780 USD/m³. Under the conservative pathway, extending membrane lifespan to 7 years, reducing chemical consumption by 30%, and increasing plant utilization to 345 days/year lowers

the cost to 0.786 USD/m³. The moderate pathway, which combines a 10-year membrane lifespan with a 50% reduction in chemical consumption and a 20% decrease in electricity cost, further reduces the cost to 0.774 USD/m³, corresponding to a 0.8% cost saving relative to S2. Under the optimistic pathway, extending membrane lifespan to 12 years, reducing membrane price by 20%, chemical consumption by 50%, and electricity cost by 30%, together with 345 operating days/year, decreases the cost to 0.714 USD/m³, yielding an 8.4% cost advantage over the commercial UF benchmark.

3.5. Environmental impact assessment

An environmental impact evaluation was conducted to examine the implications of geothermal brine pretreatment under the defined scenarios and capacities. The assessment used 1 m³ of treated brine as the functional unit and followed the general ISO 14040/14044 framework. The system boundary included the cradle-to-gate production of chemicals and membrane materials, together with the operational phase of pretreatment, while equipment manufacturing, downstream lithium recovery, and end-of-life management were excluded. A GREET/ EverBatt-based approach was used to quantify three process-relevant indicators: total energy demand, water consumption, and greenhouse gas (GHG) emissions [76–78]. These indicators were selected because they directly reflect the energy–water–carbon trade-off in geothermal brine pretreatment. The overall environmental impact assessment framework applied in this study is illustrated in Fig. 14.

The impact calculation was performed by multiplying the life-cycle inventory of each input by its corresponding impact intensity factor (e_i). For example, Indonesian grid electricity was assigned 9 MJ-eq/kWh, 2 L/kWh, and 0.87 kg CO₂-eq/kWh [79], while UiO-66-NH₂ carried a much higher embodied burden of 600 MJ-eq/kg, 12.5 L/kg, and 43 kg CO₂-eq/kg [80]. Therefore, the environmental profile of S3 must be interpreted carefully because the antifouling benefit of MMMs is accompanied by additional material burdens from MOF synthesis and membrane fabrication.

At C1, S1 exhibited the highest energy demand and GHG emissions, reaching 40.91 MJ-eq/m³ and 3.31 kg CO₂-eq/m³, respectively. Both membrane scenarios performed substantially better: S2 → 19.24 MJ-eq/m³ and 1.53 kg CO₂-eq/m³, S3 → 19.26 MJ-eq/m³ and 1.53 kg CO₂-eq/m³, corresponding to reductions of 53.0% in energy demand and 53.9% in GHG emissions relative to S1. Water consumption showed the opposite pattern: S1 → 20.01 L/m³ < S2 → 29.47 L/m³ ≈ S3 → 29.58 L/m³, indicating a 47–48% water penalty for the membrane-based routes.

At C2, the category-specific ranking observed at C1 was preserved, while all environmental impacts decreased with increasing treatment capacity. S1 decreased to 36.96 MJ-eq/m³, 19.13 L/m³, and 2.93 kg

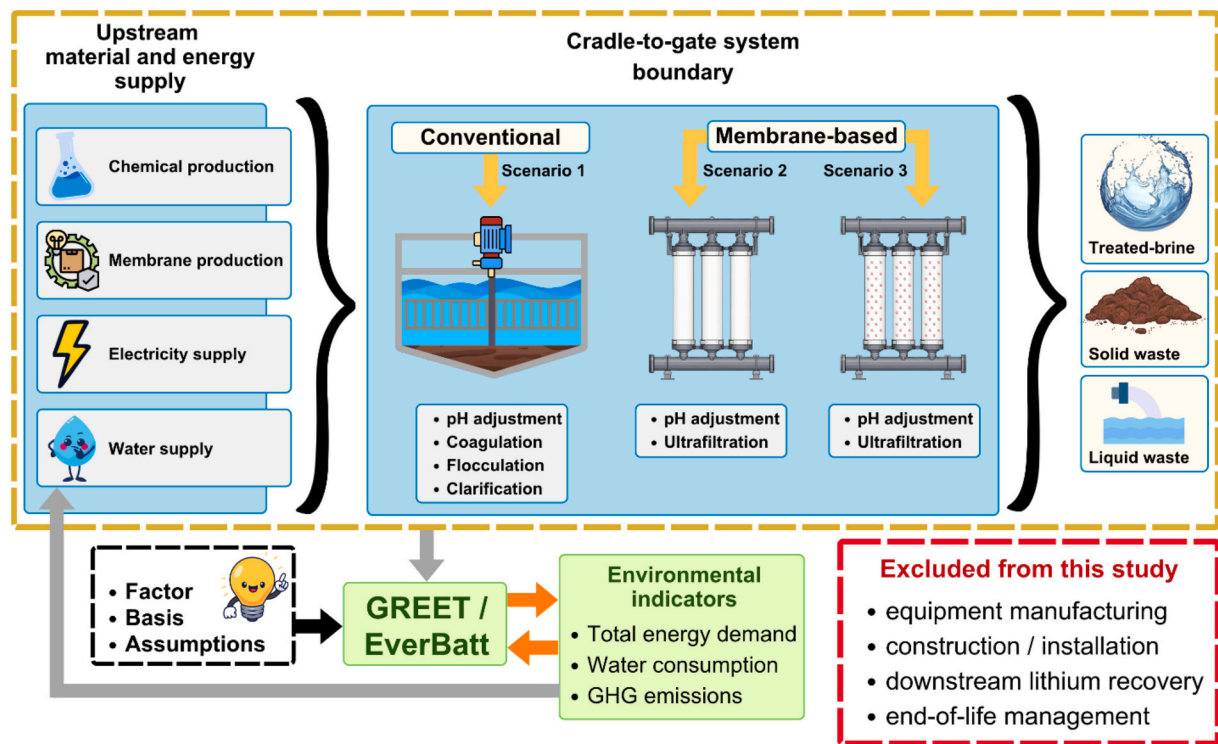


Fig. 14. System boundary and environmental assessment framework for geothermal brine pretreatment using GREET/EverBatt.

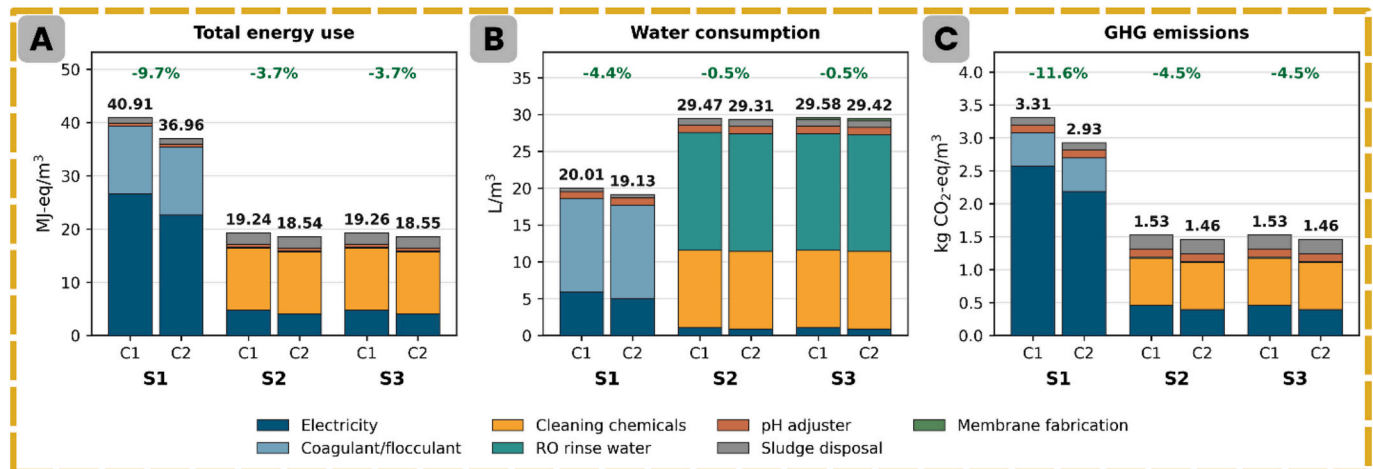


Fig. 15. Contribution of individual process components to the environmental impacts of geothermal brine pre-treatment scenarios at two capacities: (a) total energy use, (b) water consumption, and (c) GHG emissions.

CO₂-eq/m³ for total energy use, water consumption, and GHG emissions, respectively. The corresponding values for S2 were 18.54 MJ-eq/m³, 29.31 L/m³, and 1.46 kg CO₂-eq/m³, whereas S3 reached 18.55 MJ-eq/m³, 29.42 L/m³, and 1.46 kg CO₂-eq/m³. Thus, S2 remained marginally less burdensome than S3 for all three indicators, although the differences between the two membrane-based scenarios were negligible.

The scale effect was modest but strongly category- and scenario-dependent, ranging from 0.5% to 11.6%. S1 exhibited the greatest reductions, with total energy use, water consumption, and GHG emissions decreasing by 9.7%, 4.4%, and 11.6%, respectively, from C1 to C2. In comparison, S2 and S3 showed identical reductions of 3.7% in energy demand and 4.5% in GHG emissions, while their water consumption decreased by only 0.5%. Consequently, the energy and GHG advantages of the membrane-based systems relative to S1 narrowed slightly with increasing capacity: from approximately 53–54% at C1 to ~50% at C2.

Conversely, their water-consumption penalty increased from approximately 47–48% to 53–54%. These results indicate that increasing plant capacity provides measurable environmental benefits, particularly for the more electricity-intensive S1, but does not alter the overall comparative ranking of the treatment scenarios. A detailed comparison is presented in Fig. 15.

Contribution analysis further revealed distinct environmental burden profiles among the scenarios. For S1, electricity was the dominant contributor to total energy use (~65%) and GHG emissions (~78%), whereas coagulant/flocculant production accounted for approximately one-third of the energy demand and represented the largest contributor to water consumption (~64%). In contrast, the environmental profile of S2 and S3 was governed primarily by operational consumables. Cleaning chemicals contributed approximately 60% of total energy use and nearly half of GHG emissions, while RO rinse

water became the dominant source of water consumption, accounting for approximately 53–54%, followed by cleaning chemicals at ~36%. Electricity remained a secondary but non-negligible contributor, particularly to energy demand and GHG emissions. Membrane fabrication contributed only a marginal fraction of the overall impacts, explaining why S2 and S3 remained nearly indistinguishable despite the additional fabrication burden associated with the MMM system.

4. Conclusion

This study demonstrates that polysulfone/UiO-66-NH₂ MMMs are technically promising for UF-based pretreatment of silica-rich geothermal brine. UiO-66-NH₂ incorporation improved membrane hydrophilicity, reducing the water contact angle from 80.8° to 47.6°, and increased permeability from 158.94 to 376.03 L m⁻² h⁻¹ bar⁻¹. The optimized MMM achieved a maximum FRR of 80.85%, while Hermia-based fouling analysis indicated that inorganic fouling was mainly controlled by internal pore narrowing, rather than surface cake formation. The scale-up assessment shows that membrane-based pretreatment offers a lower capital burden than conventional coagulation–floculation–clarification. At C1, TCI decreased from USD 194,040 for S1 to USD 108,360 for S2 and USD 127,080 for S3. Scaling from C1 to C2 further reduced the unit pretreatment cost from 1.29 to 0.68 USD/m³ for S1, 0.80 to 0.43 USD/m³ for S2, and 0.91 to 0.47 USD/m³ for S3. However, S3 was not yet economically superior to commercial UF due to higher membrane-related costs. The GREET/EverBatt-based assessment confirmed that membrane-based pretreatment substantially reduced energy demand and GHG emissions compared with S1. At C1, S1 showed 40.91 MJ-eq/m³ and 3.31 kg CO₂-eq/m³, whereas S2 and S3 were much lower at 19.24–19.26 MJ-eq/m³ and 1.53–1.53 kg CO₂-eq/m³. However, membrane-based scenarios increased water consumption from 20.01 L/m³ for S1 to 29.47–29.58 L/m³. Overall, commercial UF remains the most attractive base-case option, whereas UiO-66-NH₂ MMMs involve a trade-off among performance, cost, and environmental impact. Achieving competitiveness will require three parallel improvements: (i) increasing membrane durability to prolong service life; (ii) reducing material losses and simplifying the casting process to lower fabrication costs; and (iii) improving antifouling performance to decrease water use, cleaning chemical consumption, and energy demand.

CRedit authorship contribution statement

Pra Cipta Buana Wahyu Mustika: Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Conceptualization. **Cornelius Satria Yudha:** Writing – review & editing, Resources. **Melisa Christine Prayogo:** Writing – review & editing, Software, Investigation, Formal analysis. **Putu Duddy Sutrisna:** Writing – review & editing, Supervision, Resources. **Sutijan Sutijan:** Writing – review & editing, Supervision. **Widi Astuti:** Writing – review & editing, Supervision, Resources, Formal analysis. **Deliana Dahnum:** Writing – review & editing. **Amir Razmjou:** Writing – review & editing. **Himawan Tri Bayu Murti Petrus:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition.

Declaration of Generative AI and AI-assisted technologies in the writing process

In the preparation of this work, the author(s) utilized ChatGPT to enhance readability and language clarity. Following its use, the author(s) thoroughly reviewed and revised the content as necessary, assuming full responsibility for the final version of the publication.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jwpe.2026.111022>.

Data availability

Data will be made available on request.

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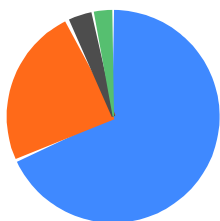
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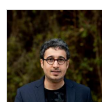
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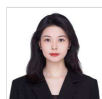
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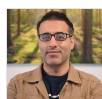
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Yingchao Dong, PhD

The Chinese University of Hong Kong, School of Science and Engineering, Shenzhen, China

Environmental engineering, Membrane separation technology, Water treatment, Chemical separation engineering, Artificial intelligence, Data-driven process modelling, Process safety, Water–energy–environment nexus, Desalination, Green sustainable technology

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**Xiaoguang Duan, PhD**

The University of Adelaide, Adelaide, Australia

Advanced oxidation processes, Wastewater treatment, Catalysis, Plastics conversion, Nanomaterials

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**Benny Freeman, PhD**

The University of Texas at Austin, Austin, Texas, United States of America (the)

membranes, polymer science

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**Ralph Rolly Gonzales, PhD**

Bioeconomy Science Institute Scion Group, Rotorua, New Zealand

Membrane, Wastewater treatment, Resource recovery, Desalination, Surface and interfacial science, Reverse osmosis/Nanofiltration, Circular economy, Minimal liquid discharge, Process intensification, Membrane fabrication, Fouling-resistant surfaces, Biomass, Valorization, Circular bioeconomy

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**Lei (Leah) Guo, PhD**

University of Arkansas College of Engineering, Fayetteville, Arkansas, United States of America (the)

Water and Wastewater Treatment, Materials Development, PFAS, Resource Recovery

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**Liang Guo, D.E., PhD**

Ocean University of China, Qingdao, Shandong, China

Mariculture pollution control, wastewater biological treatment and resource utilization, waste sludge treatment and disposal

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Aydin Hassani, PhD

Near East University, Nicosia, Cyprus

Advanced Oxidation Processes, Electrochemical Advanced Oxidation Processes, Water and Wastewater Treatment, Nanostructured Catalysts, Photocatalysis, Materials Chemistry, Adsorption, Environmental Remediation

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**Peiying Hong, PhD**

King Abdullah University of Science and Technology, Division of Biological and Environmental Science and Engineering, Thuwal, Saudi Arabia

Microbial contaminants, Water quality, Water reuse, Wastewater, Anaerobic biotechnologies

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**Weiwei Huang, PhD**

Hainan University, Haikou, Hainan, China

Wastewater treatment, Anaerobic digestion, Reclamation and reuse of biomass, Granular sludge, Aquaculture

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**Arpita Iddya, PhD**

Nanyang Technological University, Singapore, Singapore

Membranes, Resource Recovery, Electrochemistry

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**Rencun Jin, PhD**

Hangzhou Normal University, Hangzhou, China

environmental engineering

**Priyanka Kumari, PhD**

Khalifa University, Abu Dhabi, United Arab Emirates

Membrane technology, wastewater treatment, photoelectrocatalysis, CO₂ capture, resource valorization

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**Zhen Lei, PhD**

Xi'an University of Architecture and Technology, Xi'an, China

Environmental engineering, Wastewater treatment, Anaerobic digestion, Membrane bioreactor, Membrane fouling, Resource recovery

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**Zhongfang Lei, PhD**

University of Tsukuba, Tsukuba, Japan

Wastewater treatment, Granular sludge, Resources recovery, Anaerobic digestion, Reclamation and reuse of biomass

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**Jianxin Li, PhD**

Tiangong University State Key Laboratory of Separation Membranes and Membrane Processes, Tianjin, China

Membrane materials synthesis, Membrane processes for water treatment and purification, Electrochemical membrane reactor, Industrial effluent processing and resource recovery/recycle

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**Frank Lipnizki**

Lund University, Lund, Sweden

**Rui Martins, PhD**

University of Coimbra, Coimbra, Portugal

Chemical Engineering; Advanced Oxidation Processes; Water Reclamation; Drinking Water; DBPs

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Kerry McPhedran, PhD (EnvEng), MSc (Bio), PEng

University of Saskatchewan, Department of Civil Geological and Environmental Engineering, Saskatoon, Saskatchewan, Canada
municipal wastewater treatment, industrial wastewater treatment, stormwater treatment

**SangYong Nam, PhD**

Gyeongsang National University, Jinju-si, Korea, Republic of

Membrane Fabrication, Water Treatment Membrane Organic Solvent Nanofiltration, Reverse osmosis, Gas Separation Membrane, Anion exchange membrane, Water Desalination, ion exchange membrane, hollow fiber membranes, water electrolysis using membrane, hydrogen production

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**Oded Nir, PhD**

Ben-Gurion University of the Negev, Be'er Sheva, Israel

Physico-chemical separation technologies, Water and wastewater treatment, Desalination, Resource recovery from waste streams, Nutrient recovery from waste streams, Water chemistry, Membrane separation technology, Hybrid and integrated processes, Reactive-transport process modeling

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**Ranwen Ou, PhD**

Xiamen University, Xiamen, Fujian, China

Water treatment, adsorption, membrane separation, porous materials, resources recovery

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**Mariam Ouda, PhD**

Khalifa University, Abu Dhabi, United Arab Emirates

Water treatment, Wastewater treatment, Materials Science, Membrane Technology

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**Juan Manuel Peralta-Hernandez, PhD**

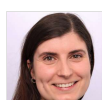
University of Guanajuato, Guanajuato, Mexico

Advance oxidation processes, Electrochemical Advance Oxidation Processes, Water treatment

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Vicenç Puig

Polytechnic University of Catalonia - Barcelona Tech, Barcelona, Spain

**Cejna Quist-Jensen, PhD**

Aalborg University, Aalborg, Denmark

Water and Wastewater treatment, membrane techonology, resource recovery

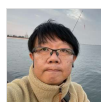
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**Heidi Richards, PhD Chemistry**

University of the Witwatersrand Johannesburg, Johannesburg, South Africa

membranes, wastewater treatment, emerging contaminants, industrial effluent, remediation, adsorbents

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Nagahiro Saito, PhD

Nagoya University, Nagoya, Japan

Electrocatalysis, Oxygen Reduction Reaction (ORR), Platinum Nanoparticles, Quantum Size Effects, Fuel Cells, Biomass-Derived Hydrogen, Solid-State Batteries, MoS₂ Monolayers, 2D Materials Synthesis, Solution Plasma Processing, Carbon Nanotubes, Defect Engineering, Materials Informatics, Topological carbon

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Wei Song, PhD

Guangdong University of Technology, Guangzhou, China

Advanced oxidation processes, Water and Wastewater Treatment, Membrane water treatment, Urban Water Systems Evaluation

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Vassilis Stathopoulos

National and Kapodistrian University of Athens, Athens, Greece



Bihu Suchetana, PhD

Indian Institute of Technology Roorkee, Roorkee, India

Water quality, wastewater treatment, machine learning, environmental regulations & policy, climate change



Ajay Sujan

Madan Mohan Malaviya University of Technology, Gorakhpur, India

Hydrodynamics, Modeling and Simulation, Wastewater Treatment, Machine Learning

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Miao Tian, PhD

Northwestern Polytechnical University, Xi'an, Shaanxi, China

Polymer composite, Membrane distillation, Solar-driven interfacial evaporation, Forward osmosis, Interfacial Polymerization, Bipolar membrane, Reverse osmosis

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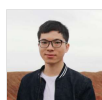


Vinay Tyagi, PhD

National Institute of Hydrology, Roorkee, India

Biological wastewater treatment, Anaerobic Digestion, Waste-to-energy, Energy & , Resources Recovery, Advanced Biomass Pretreatment Technologies, Sludge management, Solid Waste Management

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Zhuyuan Wang, PhD

The University of Queensland, Brisbane, Australia

Membrane separation, ion exchange membrane, 2D membrane

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Steven Weinman, PhD

The University of Alabama, Department of Chemical and Biological Engineering, Huntsville, Alabama, United States of America (the)

Water Treatment Membranes, Wastewater Treatment, Liquid-Liquid Extractions

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Filicia Wicaksana, PhD

The University of Auckland, Auckland Central, New Zealand

Membrane technology, Membrane fouling studies, Membrane fouling mitigation strategies, Membrane process applications for wastewater treatment, food processing, and resource recovery

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Sumith Wickramasinghe, PhD, Dr

University of Arkansas, Fayetteville, United States of America (the)

membrane separations, water treatment, water reuse

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Bing Wu, PhD

University of Iceland, Reykjavík, Iceland

Membrane technology, Water &, , Wastewater Treatment, Water Reuse &, , Resource Recovery, Biological wastewater treatment technology

**Baoshan Xing**

Tohoku University, Sendai, Japan

Anaerobic digestion, Anammox, Wastewater treatment, Resource recovery, Membrane technology, Anaerobic membrane bioreactor AnMBR, Dynamic membrane bioreactor DMBR, Nitrogen removal, Biogas production, Volatile fatty acid VFA recovery, Rumen microbiota, Lignocellulosic biomass

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**Shouliang Yi, PhD, FRSC**

Sichuan University, Chengdu, Sichuan, China

Membrane separations for advanced water purification, Carbon capture and utilization, Clean energy production, CO2 capture, Gas separations, Functional materials

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Tsinghua University, Beijing, China

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Khalifa University, Abu Dhabi, United Arab Emirates

Chemical Engineering, Water technologies, Material science, Separation, Resource recovery, Atmospheric Water Generation

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**Maria Cristina de Almeida Silva**

Federal University of Rio Grande do Sul, Porto Alegre, Brazil

water treatment, wastewater treatment, biological treatment process, anaerobic treatment processes, biorefinery, bioenergy, waste and effluent recovery, biogas generation

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Wei Lun Ang

The National University of Malaysia, Bangi, Malaysia

Membrane Processes, Nanotechnology, Water/Wastewater Treatment, Electro-Oxidation

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Payam Arabkhani

Iran University of Science and Technology, Tehran, Iran (Islamic Republic of)

Nanostructured Materials Synthesis and Characterization, Adsorption and Photodegradation, Detection and Removal of Emerging Contaminants from Polluted Water, Process Modeling and Optimization, Physicochemical Water Treatment Processes

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Muhammad Bilal Asif

University of Cambridge, Cambridge, England, United Kingdom

Water treatment, Membrane Technology, Advanced Oxidation Processes, Emerging contaminants, Catalysis, Nanomaterials

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Syed Asim Ali

King Fahd University of Petroleum & Minerals, Dhahran, Saudi Arabia

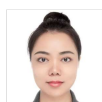
Photocatalysis, Electrocatalysis, Materials Science, Water Splitting, CO₂ reduction, Nitrogen reduction reaction, Ammonia decomposition, 2D materials

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Aziz Ur Rahim Bacha

University of Gujrat, Gujrat, Pakistan

Advanced Oxidation Processes, Bismuth-based Materials, Catalyst Characterization, Environmental Chemistry, Environmental Photochemistry, Environmental Pollutants Removal, Microplastics Degradation, Nano Materials Synthesis, Photocatalysis and Photoelectrocatalysis, Photodegradation, TiO₂-based Materials, Wastewater Treatment

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Yueping Bao

Nankai University, Tianjin, China

water treatment, catalysis



Abdelmjid Bouazizi

Moulay Ismail University, Meknes, Morocco

Ceramic Membranes, Wastewater Treatment, Environmental Remediation, Low-Cost Materials, Adsorption, Photocatalysis, Nanomaterials, Industrial Effluents, Water Purification, Sustainable Materials

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Ying Shi Chang

Nagasaki University, Nagasaki, Japan

Membrane technology, Water treatment, Sustainable process engineering



Aydin Cihanoglu

Ege University, Bornova, Türkiye

Ion exchange membranes synthesis and characterization, Membrane filtration processes, Membrane fouling and biofouling processes, Resource recovery

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Raffaele Cucciniello

University of Salerno, Fisciano, Italy

environmental catalysis, environmental chemistry, biomass valorization, life cycle assessment, waste conversion in value-added chemicals, green chemistry, wastewater treatment, biorefineries.

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Nicola Di Costanzo

University of Basilicata, Potenza, Italy

sewage sludge, anaerobic digestion, static magnetic field, biomethane, IFAS-OSA, MBBR, intermittent aeration, wastewater

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Angelo Fenti

University of Campania Luigi Vanvitelli, Department of Advanced Medical and Surgical Sciences, Naples, Italy

Environmental chemistry, Analytical chemistry, Wastewater pollution, Contaminants of emerging concern (CECs), Advanced oxidation processes (AOPs), Adsorption processes, Microplastics and nanoplastics, Pyrolysis-gas chromatography/mass spectrometry (Py-GC/MS), Laser direct infrared (LDIR) analysis

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Jiaxin GUO

Xi'an Jiaotong University School of Chemical Engineering and Technology, Xi'an, China

Desalination and water/wastewater treatment technologies, Advanced membrane fabrication by electrospinning technology, Membrane distillation, and Conveyance system for water/wastewater.

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Jemi Gandhi

Ahmedabad, India

Water treatment, Photolysis, Photocatalysis, Microbiology, Environmental Science, Ultraviolet Light Emitting Diode, Advanced Oxidation Processes, Toxicity



Haftu Gebrekiros Alemayehu

Technion Israel Institute of Technology, Haifa, Israel

Membrane filtration processes (nanifiltration and reverse osmosis), desalination, water and wastewater treatment, ion-ion selectivity, Adsorption, advanced porous materials, 2D materials, graphene-based, polyamide membranes.

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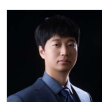


Oliver Iorhemen

University of Northern British Columbia, Prince George, Canada

Biological wastewater treatment, Biofilm processes for wastewater treatment, Process Intensification of biofiltration for drinking water treatment, Wastewater Biorefinery

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Jiantao Ji

Zhengzhou University, Zhengzhou, China

Biological sewage treatment, Anammox, Nitrogen and phosphorus removal, Partial denitrification, Emerging contaminants

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**Xin Jin**

Xi'an Jiaotong University School of Human Settlements and Civil Engineering, Xi'an, China

Advanced oxidation processes, Coagulation, Flotation, Advanced wastewater treatment and reuse, Industrial wastewater treatment

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**Jiana Jing**

Beijing University of Technology, Beijing, China

Energy-Efficient Electroactive Membranes, Advanced Oxidation Processes for Wastewater Treatment, Environmental Functional Materials, Emerging Contaminants Control

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**Sankha Karmakar**

Indian Institute of Technology Madras, Chennai, India

Separation and Extraction Processes, Membranes, Novel Materials, Processes for Resource Recovery and Valorisation

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**Shimelis Kassahun**

Addis Ababa University, Addis Ababa, Ethiopia

Heterogeneous Photocatalysis, Adsorption, Advanced Oxidation Processes, Emerging pollutants removal, Catalysis

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**Anamika Kushwaha**

Korea University, Seongbuk District, Korea, Republic of

Emerging pollutants, PET degradation, Resource recovery, Circular bioeconomy, Microbiome studies, Biogeochemical, Biopolymer

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Hu Li

Ningxia University, Yinchuan, China

Advanced Oxidation Processes for Water Pollution Control; Environmental Behavior of Emerging Contaminants in Water, microbial risk and control, disinfection, Environmental microbiology



Meng Li

Guangzhou University, Guangzhou, China

Advanced Oxidation Process, Electrochemical Oxidation, Electrochemical reduction, Heavy metal extraction, PFAS, Bioelectrochemical Remediation

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Dachao Lin

Guangdong University of Technology, Guangzhou, China

Water treatment by membrane separation, Membrane fouling mechanism, Short-path membrane filtration, Electrochemical membrane separation, Functional membrane material, Membrane bioreactor

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Weichen Lin

Beijing Normal University School of Environment, Beijing, China

Wastewater treatment, Membrane-based technology, Reverse osmosis, Nanofiltration, Membrane fouling, Computational fluid dynamics, 3D printing, Desalination

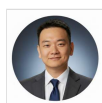
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Xiao Liu

Chinese Academy of Agricultural Sciences Institute of Agricultural Resources and Regional Planning, Beijing, China
Environmental Sciences

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Zekun Liu

Claros Technologies Inc, Minneapolis, United States of America (the)

PFAS degradation, UV-based reduction

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Zhe Liu

Xi'an University of Architecture and Technology, Xi'an, China

Aerobic granular sludge, Industrial wastewater, Environmental functional materials, Artificial intelligence

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Chenghai Lu

Chinese Academy of Sciences Research Centre for Eco-Environmental Sciences, Beijing, China

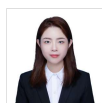
Drinking water treatment, Membrane, Nanofiltration, Membrane process



Jie Mao

Chinese Academy of Sciences Research Centre for Eco-Environmental Sciences, Beijing, China

Advanced oxidation, Chemical and Catalytic Conversion Processes, Water disinfection



Yueshuang Mao

Shanxi University, Taiyuan, China

water pollution control;, Pollutant removal and resource conversion;, environmental chemistry;

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Fan Mo

Northeastern University School of Resources and Civil Engineering, Shenyang, China

Environmental Functional Materials for Pollution Utilization & Resource Recovery

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Gotore Obey

Kyushu University International Institute for Carbon-neutral Energy Research, Fukuoka, Japan

Environmental biotechnology, Carbon capture and storage, Microwave assisted CO₂ regeneration, Amine functionalized sorbents, Biochar technology, Acid and mine drainage remediation, Environmental microbiology, Environmental remediation, Water and wastewater treatment, Adsorption processes.

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Umapathi Reddicherla

Inha University, Michuhol-gu, Korea, Republic of

Energy Harvesting, Colorimetric Sensors, Electrochemical Sensors, Energy Storage, Nanogenerator, Optical Sensors

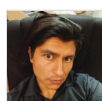
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Gengbo Ren

Hebei University of Technology School of Energy and Environmental Engineering, Tianjin, China

Electrochemical Water Treatment Technology, Electrokinetic Technology, Environmental Functional Materials



Odín Reyes Vallejo

Cinvestav, Mexico City, Mexico

water treatment, photocatalysis, adsorption, Photocatalytic hydrogen production and CO₂ photoreduction, Green synthesis of functional materials, Biomass valorisation and waste-derived materials

[View full biography](#)



Prangya Rout

Thapar Institute of Engineering & Technology Deemed-to-be-University, Indra Puri Colony, India

Advanced wastewater treatment, Anaerobic treatment, Bioreactor design, Emerging contaminants, Environmental Biotechnology, Environmental microbiology, Microplastics, Membrane technology, Nutrient removal, and Resource recovery

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Hamidreza Sharifan

Florida Atlantic University, Department of Chemistry and Biochemistry, Boca Raton, United States of America (the)

Water quality, Nanoparticle, Remediation, Plant, Algae, PFAS

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José Leandro da Silva Duarte

Complutense University of Madrid, Department of Chemical and Materials Engineering, Madrid, Spain

Electrochemical water treatment, Advanced oxidation processes AOPs, Electroanalysis and voltammetry, Environmental electrochemistry, Water–energy nexus in arid regions, Biomass-derived materials for remediation, Active chlorine generation and

monitoring, Industrial wastewater management

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Chaoqun Tan

Southeast University, Nanjing, China

Advanced oxidation processes, Disinfection, Nanomaterials, Drinking water treatment, persulfate, emerging contaminants, radicals, chlorine

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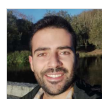


Jie Teng

Changzhou University, Changzhou, China

water treatment and resource recovery, adsorption, catalysis, functional materials design

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André Torres-Pinto

University of Porto, Faculty of Engineering, Porto, Portugal

Carbon nanomaterials, Photocatalysis, Electrocatalysis, Water and wastewater treatment, Advanced oxidation processes

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Akshaya Verma

Siksha O Anusandhan, Bhubaneswar, India

Water and wastewater treatment, Ion Exchange Membrane Bioreactor, Coagulation-Flocculation and Electro-coagulation, Adsorption, Advanced oxidation processes

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Weigang Wang

Donghua University, Shanghai, China

Bioprocess, Nutrient removal, Wastewater treatment, Anammox



Xianze Wang

Northeast Normal University School of Environment, Changchun, China

Water pollution control, Resource recovery, Membrane-based separations, Environmental functional materials, Microplastic pollutants, Advanced water-treatment processes

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Yongtao Xue

Chalmers University of Technology, Gothenburg, Sweden

Advanced oxidation processes, Adsorption, Photocatalysis, Membrane separation, Wastewater treatment, Machine learning, Sustainable development, Functional nanomaterials

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Linyan Yang

East China University of Science and Technology, Shanghai, China

Membrane Filtration for Water Treatment, Formation Mechanism and/or Minimization of Micropollutants, Domestic and Industrial Wastewater Treatment

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Bin Yao

Hunan Agricultural University, Changsha, China

Plasma, Ionizing radiation, Advanced oxidation processes (AOPs), Heterogeneous catalysis, Adsorption, Photochemistry, PFAS remediation, Reactive species, Selective oxidation, Advanced reduction processes (ARPs)

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Chengsong Ye

Xiamen University of Technology, Xiamen, China

drinking water, microbial risk and control, disinfection, viable but non-culturable bacteria, advanced oxidation process

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Qi Zhou

University of Science and Technology Beijing, Beijing, China

Nitrogen and phosphorus removal, Denitrification, Biological wastewater treatment, Pyrite-based denitrification, Sulfur-based denitrification, Micropollutants removal

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Ze Zhu

Purdue University, West Lafayette, United States of America (the)

Agricultural System, Food-Energy-Water Nexus, Waste management, Aquaculture, Modeling, Aquaponics

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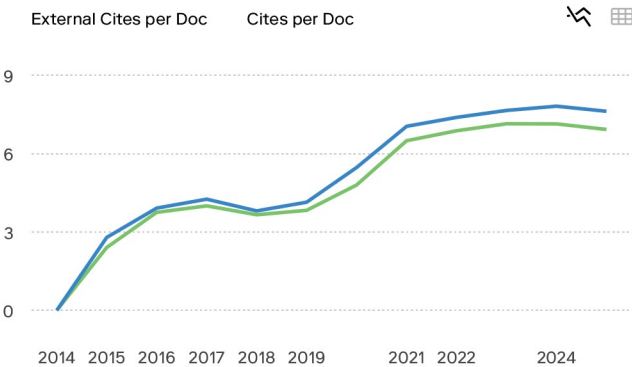


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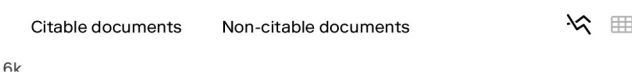
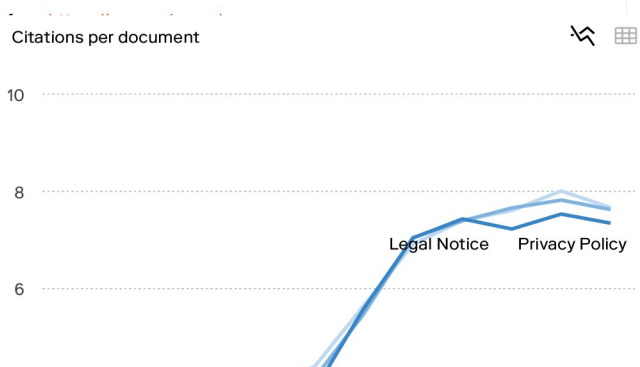
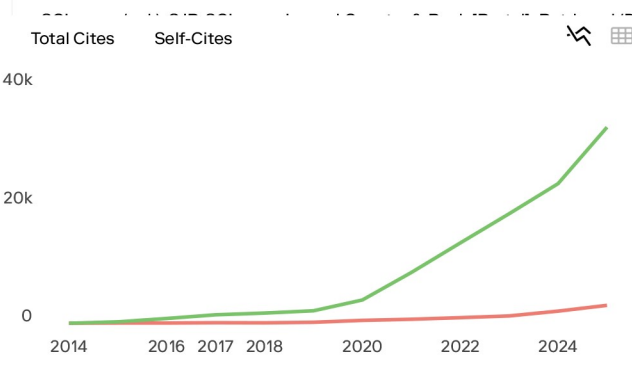
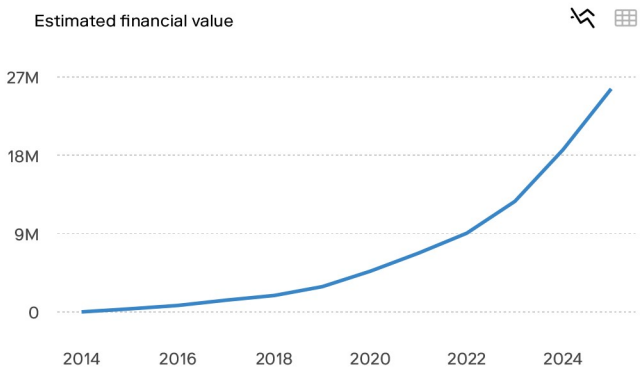
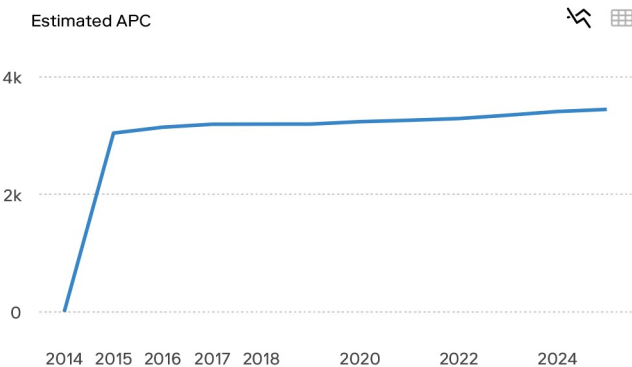
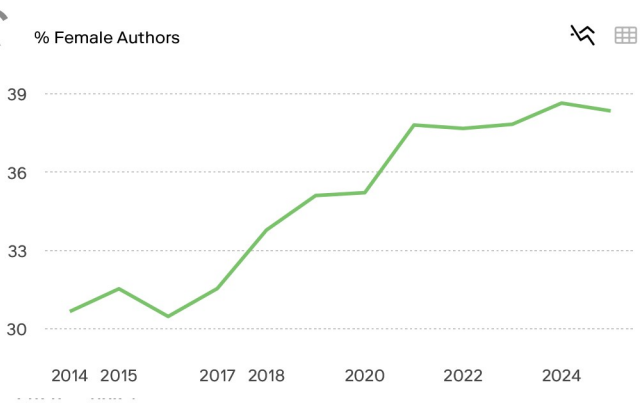
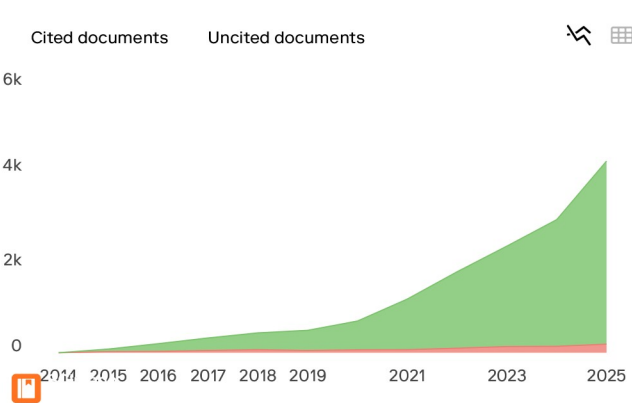
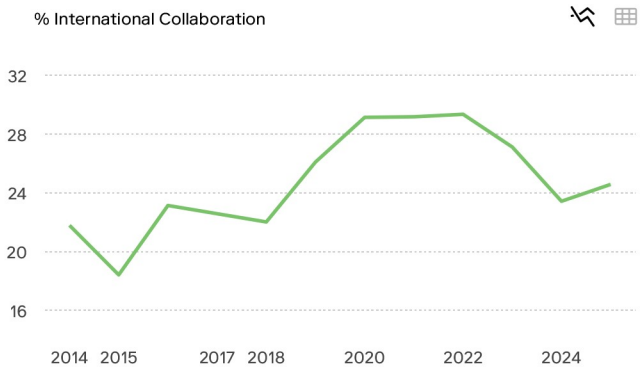
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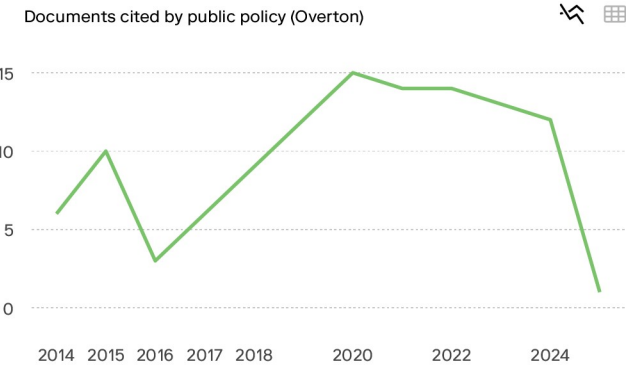
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Cites / Doc. (4 years)	2019	4.382
Cites / Doc. (4 years)	2020	5.642
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