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PERSPECTIVES

Intensifying electrified flow-through water treatment technologies via local environment modification

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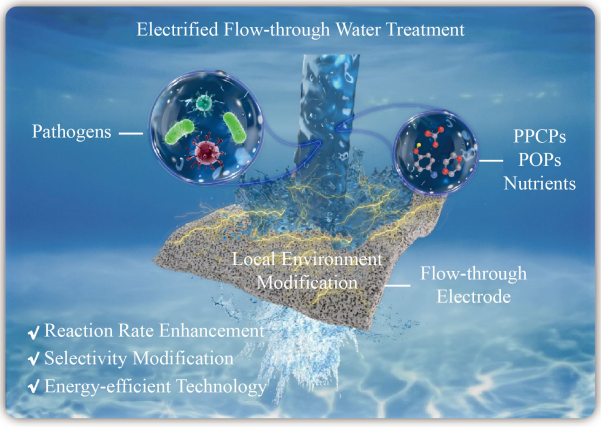
HIGHLIGHTS

- Modifying local environment can intensify the performance of flow-through electrodes.
- Reaction rate and selectivity can be improved by local environment modification.
- Modifications include spatial confinement, enhanced local field, and periodic vortex.
- Near-complete removal of low-concentration emerging contaminants can be realized.
- Electrified flow-through systems are promising for fit-for-purpose water treatment.

GRAPHIC ABSTRACT



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ABSTRACT

Removing high-risk and persistent contaminants from water is challenging, because they typically exist at low concentrations in complex water matrices. Electrified flow-through technologies are viable to overcome the limitations induced by mass transport for efficient contaminant removal. Modifying the local environment of the flow-through electrodes offers opportunities to further improve the reaction kinetics and selectivity for achieving near-complete removal of these contaminants from water. Here, we present state-of-the-art local environment modification approaches that can be incorporated into electrified flow-through technologies to intensify water treatment. We first show methods of nanospace incorporation, local geometry adjustment, and microporous structure optimization that can induce spatial confinement, enhanced local electric field, and microperiodic vortex, respectively, for local environment modification. We then discuss why local environment modification can complement the flow-through electrodes for improving the reaction rate and selectivity. Finally, we outline appropriate scenarios of intensifying electrified flow-through technologies through local environment modification for fit-for-purpose water treatment applications.

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Special issue—Towards a pollution-free planet

1 Introduction

Addressing the lack of clean and safe water is among the most serious challenges of our time for humanity's sustainability. The rapid expansion of urbanization and

industrialization inevitably leads to the generation of a large number of high-risk and persistent contaminants, such as pharmaceuticals and personal care products (PPCPs), persistent organic pollutants (POPs), antibiotic-resistant pathogens, nutrients, and heavy metals. The release of these high-risk contaminants threatens water resources and aquatic ecosystems (Chaplin et al., 2019; Zuo et al., 2023). However, conventional water treatment technologies generally suffer from limited treatment efficiency and selectivity as well as high energy consumption for realizing near-complete removal of these trace contaminants from water (Hodges et al., 2018).

Electrified flow-through technologies offer an efficient and energy-saving alternative for fit-for-purpose water treatment with high throughput (Sun et al., 2021). Flow-through electrodes with porous structures enable highly exposed electrocatalysts inside the pores and intensify reactants approaching the internal surfaces to enhance contaminant removal (Chen et al., 2024). Notably, a flow-through electrode can also serve as an electrified membrane with both electroactivity and separation properties when its pore size is on the micro/nanoscale (Chaplin et al., 2019; Sun et al., 2021). The compact and modular construction enables electrified flow-through technologies to be readily incorporated into existing water treatment systems for pre- and/or post-treatment, such as portable drinking water treatment and wastewater reclamation (Hodges et al., 2018; Zuo et al., 2023), or to serve as point-of-use devices for fit-for-purpose applications (Sun et al., 2021). Additionally, electrified flow-through systems can be powered by renewable energy (e.g., solar and wind energy) and even off-grid energy (e.g., triboelectricity), considering their energy-efficient, chemical-free, and modular features (Huo et al., 2020).

Emerging contaminants are often present at relatively low concentrations (ng/L to μg/L for chemicals; < 100 CFU or PFU/mL for pathogens) in complex water conditions. This requires the flow-through electrodes to realize a high reaction rate and selectivity for removing the trace target contaminants from water (Hodges et al., 2018). To further intensify emerging contaminant removal, a viable option is to modify the local environment of the electrodes through methods such as nanospace incorporation, local geometry adjustment, and microporous structure optimization, to induce specific functions that can be triggered during flow-through operation (Grommet et al., 2020; Chen et al., 2024).

Here, we propose potential approaches, focusing on modifying the local environment of the electrodes, to address the challenges of electrified flow-through technologies for removing contaminants with low concentrations from water (Fig. 1). We first present methods and mechanisms that fit the local environment modification, including inducing spatial confinement, enhancing local electric field, and introducing microperiodic vortex. We then discuss why modifying the local environment can complement electrified flow-through water treatment and how flow-through operation excites the confinement effect to improve the reaction rate and selectivity. Finally, we exemplify scenarios of intensifying electrified flow-through technologies through local environment modification for fit-for-purpose water treatment applications.

2 Methods and mechanisms of local environment modification

Electrified flow-through technologies offer a viable alternative for efficient emerging contaminant removal from water (Fig. 1(a)). Modifying the local environment of the flow-through electrode could further enhance the removal efficiency under low concentrations. The local environment, particularly the reaction conditions at or near the water-electrode interface, significantly influences the reaction pathways by modulating electron transfer (Chen et al., 2024). Thus, modifying the local environment can regulate the thermodynamic and kinetic properties of the reactions to improve reaction rate and selectivity (Liu et al., 2020). In this section, we present strategies for local environment modification by incorporating micro/nanostructures on the flow-through electrode inner surface to promote electrochemical reactions (Fig. 1(b)).

2.1 Confinement effect

The confinement effect can result in a higher local concentration compared with the bulk solution by accumulating the reactants within a range of nanospaces, such as cavities, pores, and channels (Grommet et al., 2020). For example, modifying carbon nanotubes (CNTs) by Co_3O_4 nanoparticles can accumulate nonpolar organic pollutants, such as bisphenol A, on the hydrophobic CNT surface (Liu et al., 2023). The accumulated bisphenol A can easily react with the Co_3O_4 nanoparticles loaded at the local region to reduce the mass transport distance to enhance the utilization of the short-lived reactive species, such as singlet oxygen ($^1O_2$). By increasing the local catalyst loading in the nanospaces, the interactions between catalysts and reagents can also be intensified by the confinement effect to increase reaction rates (Grommet et al., 2020; Qiao et al., 2020). Therefore, a viable way to achieve this goal is to incorporate atomic-scaled nanoclusters and single atoms into the pores or hollows of nanomaterials, such as metal-organic frameworks (MOFs), zeolites, and CNTs.

Conducting reactions in a confined space can also increase the selectivity of a specific reaction over others. By modifying the local environment in nanospaces via exposing the active crystal facet, adjusting hydrophobicity, and forcing reactant orientation, the target contaminants can be selectively adsorbed and degraded through a controlled pathway (Chen et al., 2024). For example, Mn-doped Fe_2O_3 nanoparticles exhibit facet-dependent adsorption: [001]- and [116]-faceted nano-

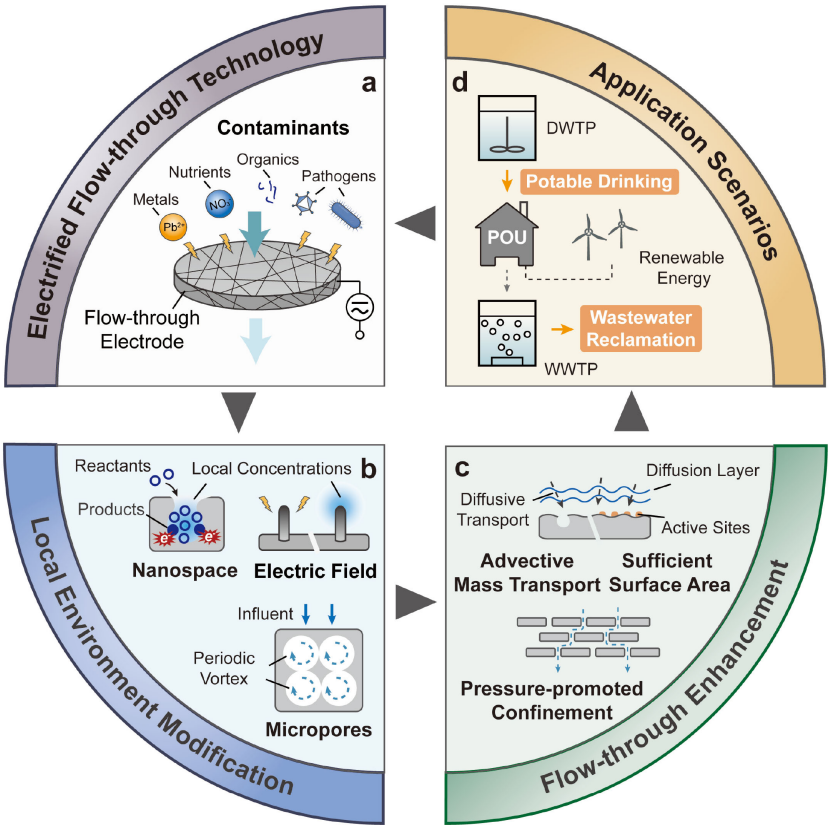

Fig. 1 Schematics of local environment modification for intensifying electrified flow-through water treatment. (a) Electrified flow-through technologies for removing high-risk and persistent contaminants from water. (b) Methods for local environment modification, including nanospace incorporation, local geometry adjustment, and microporous structure optimization. (c) Intensifying electrified flow-through water treatment by local environment modification, including incorporating advective mass transport, sufficient surface area, and pressure-promoted confinement. (d) Application scenarios of electrified flow-through water treatment, including potable water treatment at drinking water treatment plants (DWTP), wastewater reclamation at wastewater treatment plants (WWTP), and point-of-use (POU) applications powered by renewable energy.

particles show selective adsorption of Pb^{2+} and Hg^{2+}, respectively (Yuan et al., 2017). Metal ions can be selectively adsorbed with high capacity by controlling the Mn doping level to adjust the crystal facet, followed by depositing these controlled nanoparticles in the porous materials with nanospaces. These absorbed metal ions can be selectively reduced to metal particles by subsequent electrified reduction and removed from water. Additionally, reaction pathways can also be regulated by controlling the adsorption of the intermediate products in confined spaces to modify the selectivity. Pt single atoms anchored in CNT nanochannels have different neighboring dopants, including C, N, and S. The doping affects the adsorption of the intermediates during $2e^-$ electrochemical oxygen reduction (Zhao et al., 2022). The N- and S-doped Pt single-atom catalysts allow stronger adsorption of the intermediates (*OOH) within the nanoconfined spaces, resulting in much higher selectivity for hydrogen peroxide (H_2O_2) production (81.4%) compared with the Pt single-atom catalysts without doping (23.3%).

The confinement effect can also be applied for pathogen disinfection in water. Nanospaces created by molecular imprinting can provide specific binding sites of complementary size and shape to the biomolecular templates to adsorb target molecules (Yuan et al., 2020). High-risk pathogens or harmful genes can be adsorbed selectively when applying molecular imprinting with negative copies of the oligopeptides of microbial outer structures or DNA fragments to the electrodes for subsequent inactivation. Considering that harmful pathogens typically account for a small percentage of the total microorganisms in the environment, selectively inactivating the target pathogens can avoid the overuse of disinfectants to eliminate the formation of disinfection by-products. In addition, the selective removal of harmful genes, such as antibiotic resistance genes, holds promise to reduce the spread of antibiotic resistance in the environment.

2.2 Nanogeometry-enhanced local electric field

Free charges tend to accumulate in regions with the smallest radius of curvature in the electrode. One-

dimensional (1D) nanomaterials with high aspect ratios, including nanowires, nanorods, and nanotubes, can effectively accumulate charges at the tips and form intensified local electric fields (Huo et al., 2020). When modifying the electrodes with the 1D nanomaterials, the electroporation effect can be readily induced at the tips to damage bacterial membranes and viral capsids (Huo et al., 2016). This 1D nanomaterial-assisted electroporation enables reaction-free, physics-based disinfection that requires insignificant energy consumption (10^{-4} to 10^{-2} kWh/(m^3 × order)) and short contact time (< 10 s) (Huo et al., 2022).

Local electric fields can also promote the adsorption of charged ions, allowing increased local interfacial reactant concentrations to accelerate reactions (Chen et al., 2024). For example, by incorporating gold nanorods into electrodes, the local electric field can effectively absorb K^+ at the cathode surface (Liu et al., 2016). These concentrated K^+ can stabilize carbon dioxide at the tip region of the gold nanorods by non-covalent adsorption, enabling an enhanced local concentration for accelerating further reduction. Additionally, the high voltage under the local electric fields can promote reactions that require high potentials, such as generating hydroxyl radicals (•OH) through water oxidization.

2.3 Microporous flow pattern control

should diffuse to the electrode surface for degradation through concentration gradient. Optimizing local flow patterns by incorporating micro/nanoscale structures, such as cavities and channels, to generate vortices could overcome the limitation induced by diffusive transport. For example, the introduction of gyroid microstructures to a porous electrode can effectively promote the formation of strong periodic vortices in the diffuse layer to enhance mass transport (Yu et al., 2023). This gyroid porous electrode enables an enhanced nitrate reduction, 83% higher than that of the random porous electrode. Additionally, adjusting the pore size of the electrode porous structure can also optimize the mass transport of the target containments (Zhou et al., 2018). When functionalized with lamellar-structured graphene hydrogels, the densely packed porous electrode with nanoscale interlayer spacing allows water to flow along the narrow nanochannels and avoid the formation of dead volume zones, resulting in enhanced Pb^{2+} deposition.

3 Intensifying electrified flow-through treatment by local environment modification

Local environment modification typically involves confinement, nanogeometry optimization, and local flow-pattern regulation within the diffusion boundary layers of the water-electrode interface. Reactions under flow-by operation are limited by mass transport because reaction kinetics are dominated by reagent diffusion through the boundary layer. Flow-through operation allows water to flow through the porous electrodes to induce advection, which significantly reduces the thickness of the diffusion boundary layer to accelerate mass transport (Sun et al., 2021). Therefore, the performance of local environment modification can be maximized with sufficient mass transport during flow-through operation. In this section, we discuss the strategies of flow-through operation to excite the local environment modification for enhancing the removal of low-concentration contaminants (Fig. 1(c)).

3.1 Advection-enhanced mass transport

Applying electrified flow-through operation can effectively compress the diffusion layer at the water-electrode interface to a scale comparable to the pore radius. The induced advection-enhanced mass transport can accelerate the target contaminants approaching the inner electrode surface. The functions of local environment modification near the surface can effectively play a role in improving the reaction rate and/or selectivity, realizing near-complete contaminant removal at low concentrations in complex water matrices. For example, 1D nanomaterial-assisted electroporation for disinfection is only effective when the microorganisms approach the tips with sufficient proximity, because the local electric field exists only near the tip regions (Huo et al., 2016). By applying a flow-through electrode whose surface is vertically decorated with 1D nanomaterial, the microorganisms can effectively approach the electrode surface through advective transport, realizing complete disinfection within 10 s. In contrast, the disinfection performance decreased by more than five orders of magnitude under the same operating conditions without mass transport acceleration (Huo et al., 2021).

Enhancing mass transport also contributes to the regulation of local flow pattern that affects adsorption between the reagents and electrocatalysts to modify selectivity. In electrochemical nitrate reduction, a CNT electrified membrane incorporating Cu single atoms enhances the adsorption of the reduction intermediates during flow-through operation, enabling a near-complete conversion of low-concentration nitrate (10 mg-N/L) with high selectivity of 86% for nitrogen generation in 10 s (Wang et al., 2023). This is a significant enhancement over the flow-by operation, which achieves only 30% nitrate removal with a limited nitrogen selectivity of 7% under the same operating conditions.

3.2 Pressure-promoted confinement

Pressure is applied to drive water flow through the porous electrode. This external force enables reactants in water to

be advected into nanospaces such as the interlayer of 2D nanosheets (e.g., graphene, MXene, and MoS_2 nanosheets) to induce the confinement effects. For example, a laminate flow electrode consisting of monolayer Co-doped TiO_2 nanosheets has sub-nanometer (several angstroms) interlayer channels (Meng et al., 2022). The contaminants in water advected into the layered confined channels can react *in situ* with the decorated atomic catalysts inside the channels. Fast degradation of ranitidine is achieved with a reaction rate of 1.06 m/s, five orders of magnitude higher than that under the flow-by mode.

Selectivity can also be adjusted when water flows through confined nanochannels (Yang et al., 2019). By flowing H_2O_2 into a nanochannel CNT (diameter of 7 nm), the decorated Fe_2O_3 nanoparticles (particle size of 2 nm) inside the CNT convert H_2O_2 to $^1O_2$. This pathway is different from the typical Fenton reactions in bulk solutions that generate $^{\bullet}OH$. The Fe_2O_3 nanoparticles exhibit stronger electronic interaction in the confined nanochannel to selectively convert the H_2O_2 to $HO_2^{\bullet}/O_2^{\bullet-}$ for further $^1O_2$ generation. In contrast to the short-lived and non-selective $^{\bullet}OH$, the meta-stable $^1O_2$ in the CNT nanochannels shows higher selectivity toward electron-rich compounds for a 22.5-fold faster degradation of methylene blue. Additionally, the flow direction between the fluid and electrode interface can also be regulated during flow-through operation, which can include periodic vertexing to further enhance mass transport (Yu et al., 2023).

3.3 Sufficient exposure on active sites

Flow-through electrodes with micro/nanochannels offer more surface area for catalyst decoration and exposure compared with flat electrodes (Yang et al., 2023). The high surface area provides sufficient active sites for efficient contaminant degradation during flow-through operation. The active surface area of a flow-through RuO_2-doped porous electrode (pore size of 25 μm) is 400 times higher than that of a flow-by plate electrode (Chen et al., 2022). Reducing the pore size of porous electrodes can increase the utilization of active sites under the same electrode porosity. A 25-μm pore size electrode enabled higher efficiency to reduce the energy consumption to 60.4 kWh/kg-N, 39.9% less than that of the 90-μm pore size electrode, when achieving complete ammonia removal (Chen et al., 2022).

4 Perspectives, applications, and outlook

Electrified flow-through operation can effectively decrease the thickness of the diffusion boundary layer to a length scale comparable to the pore radius. Advection in the porous electrodes significantly accelerates the transport of the target contaminants to the confined region. This enables local environment modification that primarily dominates within the diffusion boundary layer of the water-electrode interface to effectively play a role in increasing local concentrations, modifying adsorption, and/or optimizing flow patterns to further improve the reaction rates and selectivity. Therefore, electrified flow-through water treatment can be significantly enhanced via local environment modification to achieve near-complete removal of high-risk, persistent contaminants with low concentrations. Some related examples of removing low-concentration PPCPs, POPs, harmful pathogens, nutrients (N and P), and heavy metals from water are shown in Table 1.

By improving the reaction kinetics and selectivity through local environment modification, the electrified flow-through technologies can realize fit-for-purpose removal of target contaminants with high water treatment throughput. For example, a metal-deposited ceramic membrane can selectively produce $^1O_2$ to remove electron-rich contaminants of PPCPs and POPs in confined micropores (Zhao et al., 2020). CNT electrified membranes can enhance the adsorption of the reactants in nanopores for achieving near-complete removal of nutrients (N and P) with high selectivity (Jin et al., 2023; Wang et al., 2023). Additionally, 1D nanomaterial-assisted electroporation can effectively induce sub-lethal damage on bacteria under a high water flux of 10^4 L/(m^2·h) during flow-through operation. The low-concentration chlorine that typically exists in tap water (~0.5 mg-Cl_2/L) is sufficient to aggravate such damages for complete bacterial inactivation (Huo et al., 2022). Notably, the energy consumption of the electrified flow-through water treatment is low: 10^{-3} to 10^1 kWh/(m^3 × order) for contaminant degradation and 10^{-4} to 10^1 kWh/(m^3 × order) for disinfection (Table 1; Miklos et al., 2018; Huo et al., 2020; Sun et al., 2021). Rational module configuration, such as a tubular concentric structure, also facilitates the scale-up of the electrified flow-through systems (Pei et al., 2022).

Electrified flow-through technologies can also be integrated into existing water treatment processes for pre- and/or post-treatment, ideally suitable for high-standard portable drinking water treatment and wastewater reclamation (Fig. 1d). For example, 1D nanomaterial-modified flow-through electrodes can achieve efficient and energy-saving disinfection under high water flux. Such electrodes can be applied for treating drinking water after storage and piped distribution to eliminate the impacts of microbial regrowth (Huo et al., 2016). Additionally, carbonaceous electrified membranes enable near-complete and selective removal of low-concentration nutrients from water (Jin et al., 2023; Wang et al., 2023). The application of such membranes may address the critical need for nutrient control for wastewater reclamation, because eutrophication can be triggered by

Table 1 Summary of applying electrified flow-through technologies for removing contaminants with low concentrations from water

Flow-through electrode	Contaminant	Mechanism	Performance (concentration; efficiency; Residence time)	Energy consumption per order (kWh/m)	Ref.
TiO_2-doped porous Ti electrode	4-Chlorophenol	Nanoconfinement-enabled reaction pathway control	13 mg/L; 94%; 14 s	10^0	Kang et al. (2023)
Pd/Pt-coated ceramic membrane	Sulfamethoxazole	Enhanced adsorption of intermediates	2.5 mg/L; 83%; 23 s	10^{-2}	Zhao et al. (2020)
SnO_2-Sb-doped Ti foam	Sulfamethoxazole, norfloxacin, and ibuprofen	Sufficient exposure on active sites	2–5 mg/L; 90%; 12 min	10^0–10^{-2}	Yang et al. (2023)
Nanodefect-doped nanofiber membrane	Propranolol	Sufficient exposure on active sites	20 mg/L; 99%; 2.5 s	10^{-2}	Gao et al. (2023)
Fe_2O_3 nanoparticles-doped CNTs	Tetracycline	Nanoconfinement-enhanced $^1O_2$ generation	18 mg/L; 98.4%; 3 h	10^{-2}	Guo et al. (2021)
Cu_3P nanowire-modified Cu mesh	Bacteria and viruses	Local electric field-enhanced electroporation	10^6 CFU or PFU/mL; 6.0-log; 10 s	10^{-4}	Huo et al. (2022)
Cu single atoms-modified CNT membrane	Nitrate	Enhanced adsorption of intermediates	10 mg-N/L; 97% (86% N_2 selectivity); 10 s	10^0	Wang et al. (2023)
Microgyroid-modified electrode	Nitrate	Microporous-enabled flow pattern control	50 mg-N/L; 95%; 2 h	10^1	Yu et al. (2023)
Hydroxyl-terminated MXene filter	Phosphate	Sufficient exposure on active sites	5 mg-P/L; 99%; 30 min	10^{-3}	Jin et al. (2023)
Graphene hydrogel	Pb ion	Microporous-enabled flow pattern control	20 mg/L; 86%; 40 min	10^{-2}	Zhou et al. (2018)

the low-concentration nutrients if the reclaimed water is reused as a supplemental source for surface water.

The modular and compact construction facilitates electrified flow-through systems to be applied for decentralized, point-of-use applications, such as household and rural water treatment. For example, carbonaceous electroactive filters can effectively remove multiple organic pollutants, toxic metals, and pathogens simultaneously under a long operation time (several months) with low manufacturing cost (< 20 USD) (Liu et al., 2020). Further, low and/or intermittent energy inputs are sufficient to power the electrified flow-through systems for point-of-use water treatment in many cases (Huo et al., 2023). For this reason, renewable and even off-grid energy sources, such as wind, solar, and thermal energy, are available to power the flow-through systems instead of grid electricity, offering a promising alternative where centralized water treatment is not feasible (Rahimi et al., 2018).

Modifying the local environment of the flow-through electrodes to intensify water treatment should overcome the limitations related to the cost and stability of the functionalized nanomaterials, especially during long-term operation. Scalable system configurations with large surface area and stable structure of the electrodes can further lower the treatment costs when scaled up. Further, foulants contained in the complex water matrices may block the confined range during flow-through operation, requiring the electrodes to possess antifouling and/or self-cleaning properties for fouling control (Wang et al., 2020). These fouling control properties can improve the operational stability of the systems when treating water with complex matrices, therefore extending the lifespan of the flow-through electrodes and reducing the maintenance costs. Specifically, self-cleaning is more energy efficient than antifouling in real applications because of its intermittent operation (Sun et al., 2021). Additionally, techno-economic analysis and life cycle assessment are necessary to quantify economic and environmental impacts, including manufacturing costs, greenhouse gas emissions, and potential toxicity to humans and the ecosystems, before expanding the electrified flow-through systems for water treatment in large scales (Xia et al., 2021; Sun et al., 2023). A cradle-to-gate assessment helps to further improve the design of the confined nanospaces in the electrodes and the electrified flow-through systems to meet the needs of fit-for-purpose water treatment, especially when considering the potential scenarios of powering the systems with renewable energy.

Conflict of Interests Zheng-Yang Huo is a youth editorial board member, Xia Huang is an editorial board member, and Menachem Elimelech is an advisory board member of *Frontiers of Environmental Science & Engineering*. The authors declare that this research was

conducted without any commercial or financial relationships that could be construed as a potential conflict of interest.

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Authors Biography

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Xia Huang is a Professor at School of Environment of Tsinghua University, titled the National Science Fund for Distinguished Young Scholars and the Special Expert of Ministry of Education. Now she is a Director of State Key Joint Laboratory of Environment Simulation and Pollution Control. Her research interests focus on novel wastewater treatment processes coupled with biological, membrane and electrochemical technologies for water, energy and resource recovery. Till now, she has published 5 books, more than 400 journal papers. She is currently the Distinguished Fellow of the International Water Association (IWA) and was Ex-Chair of the IWA Specialist Group on Membrane Technology. She serves as an

Editor of *Water Research X*, and Executive Associate Editor-in-Chief of *Front. Environ. Sci. Eng.* She was awarded 2009 *Environ. Sci. Technol.* Best Paper and 2018 *Environ. Sci.: Wat. Res. & Technol.* Best Paper, the 2nd Class of the State Science and Technology Progress Award thrice from Chinese Government.

Menachem Elimelech is the Sterling Professor of Chemical and Environmental Engineering at Yale University. His research focuses on membrane-based technologies at the water-energy nexus, materials for next-generation desalination and water purification membranes, and environmental applications of nanomaterials. Professor Elimelech was the recipient of numerous awards in recognition of his research contributions. Notable among these awards are the 2005 Clarke Prize for excellence in water research; election to the US National Academy of Engineering in 2006; Eni Prize for 'Protection of the Environment' in 2015; and election to the Chinese Academy of Engineering in 2017, the Australian Academy of Technology and Engineering in 2021, and the Canadian Academy of Engineering in 2022. Professor Elimelech has advised 49 Ph.D. students and 45 postdoctoral researchers, many of whom hold leading positions in academia and industry. In recognition of his excellence in teaching and mentoring, he received the W.M. Keck Foundation Engineering Teaching Excellence Award in 1994, the Yale University Graduate Mentoring Award in 2004, and the Yale University Postdoctoral Mentoring Prize in 2012.