



# Combining Electrified Membranes and Electrochemical Disinfection for Virus Demobilization

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## Abstract

Electrified membranes (EMs) possess the capacity to handle the intrinsic restrictions of traditional membrane techniques. EMs show improved functions beyond separation. Electrification can increase the efficacy and sustainability of membrane techniques and encourage novel utilizations in water and wastewater treatment. As a process in which chemical oxidants are produced *in situ* via redox reactions on the surface of an electrode, electrochemical disinfection (ED) has recently magnetized increased interest as an option to conventional chemical dosing disinfection techniques. In this review, we focus on fresh improvements in EMs, especially on water and wastewater disinfection. A brief description is accorded to materials categories, synthesis procedures, and electrified filtration operating modes. A discussion is dedicated to applications of EMs, especially water disinfection via bacterial and viral inactivation. Future challenges and promising applications for EMs are underlined. On the other hand, a brief description of ED concepts and perspectives is given. ED does not demand the transport and storage of hazardous materials and could be scaled across centralized and distributed treatment contexts; it shows promise for use both in resource-limited settings and as a supplement for aging centralized systems. This discussion suggests that EMs would be merged with ED as an intensified process.

## Subject Areas

Chemical Engineering & Technology

## Keywords

Electrified Membranes (EMs), Electrochemical Disinfection (ED), Membrane Processes, Water Treatment, Reactive Oxygen Species (ROs), Reactive Chlorine Species (RCSs)

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## 1. Introduction

Dealing with the outstanding dares in global water scarcity and water pollution needs water treatment techniques that are multifunctional, modular, scalable, resilient, chemical-free, and energy-efficient [1]. Mature membrane techniques for potable water purification, wastewater treatment and reuse, and saline water desalination have a great contribution to furnishing clean and safe water, decreasing harmful ecological effects, and increasing water supply [2]. Such membrane-based separation methods, comprising microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO), separate solutes and pollutants from the water via physicochemical routes that depend on the characteristics of the membrane materials [1]. As an illustration, the elimination of emerging toxic contaminants with low molecular weight and the neutral charge remains very difficult employing most membrane materials, because of the restrictions of traditional size- and charge-exclusion separation routes [3]. Furthermore, interactions of organic matter, inorganic salts, and microbes with membrane surface unavoidably lead to membrane fouling that declines technique efficiency and lowers membrane lifetime [4] [5] [6]. Such dares have motivated the development of new membranes with more features and functionalities beyond those of traditional membranes [1].

Electrified membranes (EMs) possess the capacity to deal with such dares via introducing electroactivity as an added membrane function [1] [7]. Precisely, in addition to the conventional membrane functions of solute separation by steric hindrance and charge exclusion, EMs spread the function of membranes beyond pure separation by exploiting a variety of electron-based phenomena, comprising electrochemical oxidation and reduction, electrostatic adsorption and rejection, electrophoresis, and electroporation [8] [9] [10] [11]. Thus, EMs may efficiently decompose and/or alter pollutants and boost the rejection of charged species through filtration methods [1].

To reach such these roles, EMs have been used as porous flow-through electrodes in a two-electrode configuration (working and counter electrodes) by applying an electric potential difference across the electrodes [1]. Juxtaposed to conventional flow-by electrochemical configurations [12], the electrochemical flow-through filtration setup may enhance reaction kinetics [13], electrode stability [14], and mass transfer of pollutants to electrode active sites [15]. The increased efficiency of EMs with porous flow-through electrodes has been assigned to co-occurring field effects (such as thermal, fluid, and electric fields (EFs)) under spatial confinement within nanoscale intra-porous structures [15] participating

in fast pollutant elimination. EMs may target various pollutants, comprising heavy metals [16], emerging organic contaminants [17], pharmaceuticals and personal care products [18], microorganisms [19] [20], and uncharged molecules [1].

Besides pollutant elimination, EMs have the potential to reduce membrane fouling and scaling via different electrochemical strategies [1] [21]. As an illustration, organic and biological foulants may be decomposed through electrochemical self-cleaning founded on *in situ* formations of powerful oxidizing species (such as reactive oxygen species (ROSs) and reactive chlorine species (RCSs)) [14]. In addition, the chemical and hydrodynamic surroundings close to the membrane surface may be adjusted by controlling pH and bubbling with electrolysis to decrease inorganic scaling [8]. In addition to employing electrochemical reactions for fouling control, tailoring membrane surface features like charge [8], hydrophilicity [22], and interfacial nanostructure [1] via applied voltage can furnish an extra way to lessen membrane fouling by natural organic matter, bacteria, and inorganic scalants.

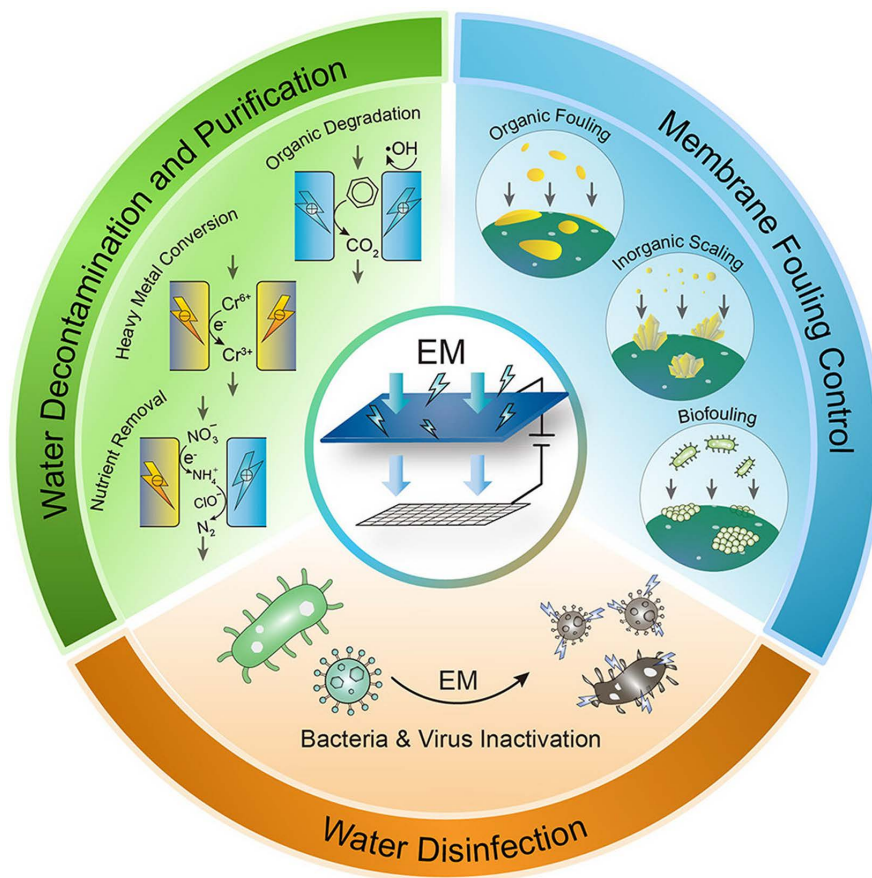
Until now, large research attempts have proved the capacity of EMs to deal with actual issues in water treatment [1]. Nonetheless, most investigations on EMs stay at the laboratory scale below well-manipulated circumstances, whilst disregarding examining their efficiency in real wastewater. In addition, the application of pollutant elimination and fouling control is scalable and modular EMs remains demanding, because these techniques stay largely affected by the electrofiltration method, module geometry, and filtration residence time. As a consequence, transferring EM techniques to real-world applications will need sizeable amelioration in pollutant reduction performance and selectivity, material stability, and process sustainability as well as optimization of module design. Considering the vacuum between pilot investigations and real usages, estimating EMs, from membrane materials synthesis and working principles to process-limiting factors and energy consumption, remains crucial for leading the expansion of EMs for environmental usages [1].

In this review, we focus on fresh improvements in EMs, especially on water and wastewater treatment applications. A brief description is accorded to materials categories, synthesis procedures, and electrified filtration operating modes. A discussion is dedicated to applications of EMs, especially water disinfection via bacterial and viral inactivation (**Figure 1**). Restrictions to EM effectiveness like membrane material longevity, system compatibility, and process sustainability are as well examined. The energy consumption of EMs for pollutant reduction and fouling decrease is assessed. Future challenges and promising applications for EMs are underlined. On the other hand, a brief description of electrochemical disinfection (ED) concepts and perspectives is given.

## **2. Fabrication and Operation Modes of Electrified Membranes (EMs)**

### **2.1. Materials for Electrified Membranes (EMs)**

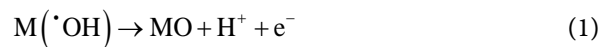
Fabricating EM preparation involves preparing membrane active layers with



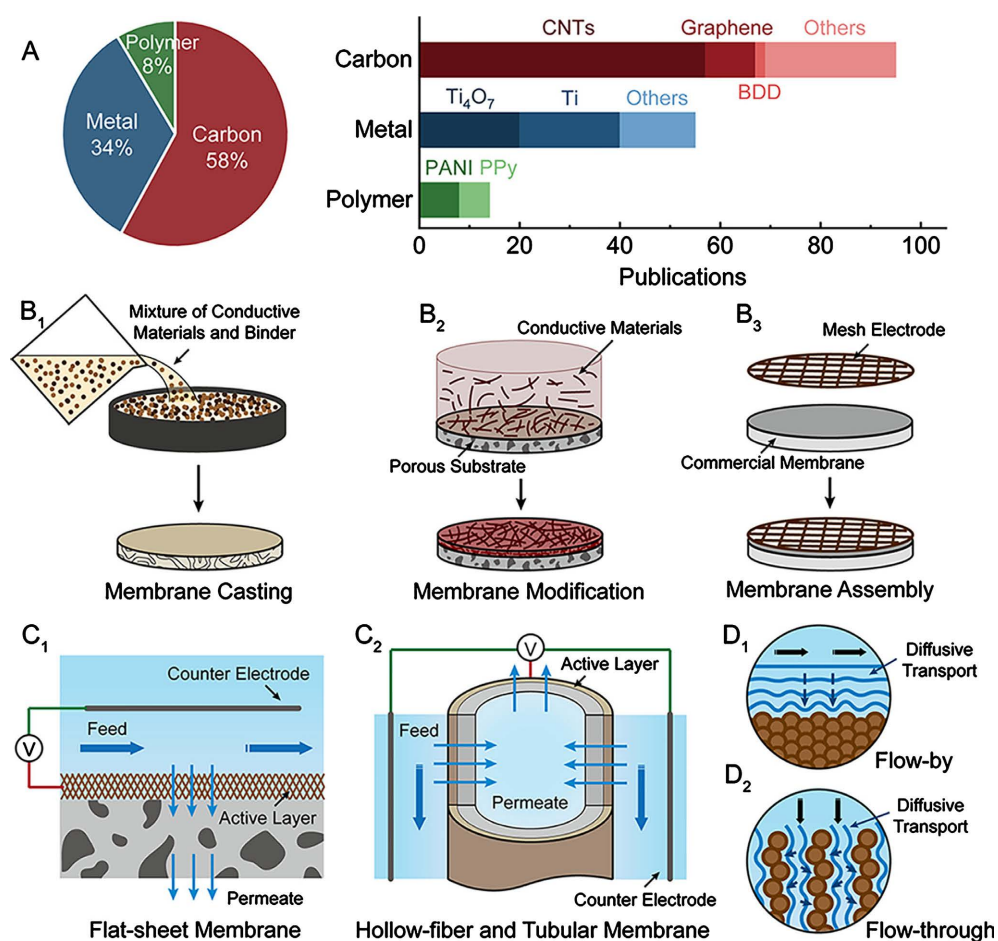
**Figure 1.** Possible ecological usages of electrified membranes (EMs): water decontamination and purification, water disinfection, and membrane fouling control [1].

elevated electrical conductivity and porosity. Sun *et al.* [1] revised the literature from the previous decade on conductive materials employed for EM fabrication, comprising carbonaceous materials, metals, metal oxides, and polymers.

The large part (about 60%) of the EMs investigated are fabricated employing carbonaceous materials (Figure 2(A)) [1]. In such materials, the carbon atoms are mainly  $sp^2$  hybridized with delocalized  $\pi$ - $\pi$  electrons, spreading elevated conductivity. Carbonaceous materials can work as either the anode or the cathode. Most carbonaceous anodes are viewed as “active” anodes with an overpotential for oxygen evolution reaction generally lower than 0.4 V [1]. The “active” anode surface (M) interacts highly with electrogenerated hydroxyl radicals ( $\cdot\text{OH}$ ) to produce a higher anodic oxide, as shown in Equation (1), which is only apt of partially oxidizing organic substances. Thus, a voltage below the oxygen evolution potential (OEP) of carbonaceous electrodes is usually applied for usages implying physicochemical-founded pathways (e.g., electrostatic repulsion, electrophoresis, and direct oxidation/reduction) [23].



Carbon nanotubes (CNTs) remain the most frequent conductive materials for EM preparation thanks to their applicability for facile generation of an interwoven



**Figure 2.** Electrified membrane (EM) materials, fabrication, and electrified modules. (A) Summary of publications on conductive materials for EM fabrication for ecological usages, founded on 150 papers published during the previous decade (2011-2020). Details may be found in [1]. The conductive materials are categorized into three classes: carbon, metals (comprising metals and metal oxides), and polymers. Such materials comprise carbon nanotubes (CNTs), graphene, and boron-doped diamond (BDD); substoichiometric titanium oxide (Ti<sub>4</sub>O<sub>7</sub>) and porous titanium (Ti); and polyaniline (PANI) and polypyrrole (PPy). Schematic illustration of EM fabrication through (B<sub>1</sub>) free-standing membrane casting, (B<sub>2</sub>) membrane modification, and (B<sub>3</sub>) composite membrane assembly. Schematic illustration of EM modules and cross-flow filtration mode for (C<sub>1</sub>) flat-sheet and (C<sub>2</sub>) hollow-fiber and tubular membranes. Thick blue arrows show feedwater flow, and thin blue arrows show water permeation across the membrane surface. Schematic depicting the impact of water convection on diffusion in (D<sub>1</sub>) flow-by and (D<sub>2</sub>) flow-through operation modes. Thick black arrows show the direction of convective transport, and thin blue arrows and lines illustrate the direction and changes of the diffusive transport, respectively [1].

and compact structure with high electrical conductivity and porosity [24]. Such structure authorizes CNTs to bond with each other via van der Waals interactions, that way keeping their stability throughout EM process [1]. Graphene-based materials, comprising graphene oxide and reduced graphene oxide, as well show useful electrical features. The conductive layer may be synthesized by deposition, conducting to an elevated specific surface area and controllable pore size distribution [25]. Functionalization with oxygen-containing groups authorizes CNT- and graphene-based materials to bind with other metal catalysts (e.g., Fe and Pd)

[26]. Boron-doped diamond (BDD), one of the most encouraging flat plate “nonactive” anodes for  $\cdot\text{OH}$  formation (*i.e.*, anodes with elevated OEPs) [27], stays scarcely mentioned for EM preparation, since the techniques for preparing the BDD layer with elevated specific surface area and porosity remain complicated and expensive [28]. Further, mesh electrodes (like carbon fiber cloth and carbon paper) may be utilized as conductive substrates for the modification of membrane structures [1]. The usage of high specific surface area coal and graphite has as well been noted [29].

Metals and metal oxides are an additional class of conductive materials that remain encouraging for EM fabrication. Among such materials, several studies have been dedicated to the employment of Magnéli phase  $\text{Ti}_4\text{O}_7$  [30] [31] [32], the most conductive phase of the substoichiometric titanium oxides ( $\text{Ti}_n\text{O}_{2n-1}$ ,  $4 \leq n \leq 10$ ) [33]. When used as a “nonactive” anode, the OEPs of  $\text{Ti}_4\text{O}_7$  (2.2 - 2.7 V *vs* SHE) are comparable to those of BDD electrodes (2.3 V *vs* SHE) [34], showing its superior efficiency for  $\cdot\text{OH}$  formation through water oxidization. In addition,  $\text{Ti}_4\text{O}_7$ , having a porous, stable monolithic structure, may be prepared via anodic polarization [1]. Among metal materials, Ti mesh remains one of the most encouraging metal substrates employed for functionalization with electrocatalysts (like  $\text{IrO}_2$ ,  $\text{RuO}_2$ , and doped  $\text{SnO}_2$ ) [35]. Further, steel mesh substrates may act as cathodes for electrochemical reduction or for catalyzing electro-Fenton reactions [36].

Conductive polymers have a conjugated backbone that generates a series of alternating single and double carbon bonds [37] [38]. The overlap of p-orbitals in the  $\pi$ -bonds authorizes the electrons to delocalize and move between atoms. Polypyrrole and polyaniline have been employed largely for EM preparation thanks to their stability under harsh conditions (like high pressure and chemical exposure) [1]. Nonetheless, several polymer-based EMs suffer from low conductivity and water flux [38]. Integrating conductive polymers with carbonaceous materials (e.g., CNTs and graphene) could ameliorate the electrical efficiency of EMs.

## 2.2. Electrified Membrane (EM) Fabrication

EMs are usually fabricated utilizing free-standing membrane casting, membrane modification, or composite membrane assembly [1]. Free-standing  $\text{Ti}_4\text{O}_7$  and carbonaceous EMs, having both separation and electrical features inside a porous monolithic structure, may be fabricated through the membrane casting method [39], as depicted in **Figure 2(B<sub>1</sub>)**. Frequently, a mixture of the conductive materials and binders (e.g., paraffin oil and polymers) [1] is first synthesized. The free-standing EMs are prepared through mechanical pressing, gel casting, or wet-spinning methods to shape the mixture, followed by thermal treatment to remove the binder.

The most usual technique for EM fabrication consists of modifying a porous substrate with conductive materials (**Figure 2(B<sub>2</sub>)**). Conductive substrates (e.g.,

carbon filter cloth, Ti mesh, stainless steel, and PANI) remain largely utilized. Functionalization of electrocatalytic materials (e.g., CNTs, graphene, and doped SnO<sub>2</sub> on the substrates) ameliorates their electrochemical efficiency and/or controls the membrane pore size. Cross-linked polymers with conductive materials like CNTs could authorize accurate control of the EM pore sizes. Further, using nonconductive substrates, comprising ceramic (Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub>) and polymeric (polyvinylidene difluoride and polytetrafluoroethylene) membranes is as well frequent [1].

A straightforward procedure to fabricate EMs consists of assembling mesh electrodes on commercial membranes (**Figure 2(B<sub>3</sub>)**). In such situation, flat plate electrodes with big pore sizes can be employed as the active layer. A mesh electrode could be pasted onto the membrane substrate or assembled into an electrofiltration module with a membrane [40]. Sun *et al.* [1] noticed that the present performance of these EMs carrying conductive layers with large pore sizes stays much lower than that of EMs fabricated by the other two procedures, even if their specific surface areas can be comparable, as discussed later.

### 2.3. Electrified Membrane (EM) Modules and Filtration Modes

Various types of flat-sheet, hollow-fiber, and tubular EMs could be employed for water treatment [41]. EM modules and filtration modes are shown in **Figure 2(C<sub>1</sub>)**, **Figure 2(C<sub>2</sub>)**. In usual usage, the EM frequently works as one of the electrodes, whilst the counter electrode is placed either on the feed or on the permeate side. The conductive layer of the EM usually faces the feed side [1].

Cross-flow filtration remains the most frequent mode for practical water treatment usages. In such filtration mode, the feed flow passes along the membrane surface and the EF is perpendicular to feed flow (**Figure 2(C<sub>1</sub>)**, **Figure 2(C<sub>2</sub>)**). Dead-end filtration, in which feed flow is perpendicular to the membrane, suffers from rapid formation of a cake layer, which hinders EM efficiency [1].

Benefiting from the flow-through configuration, EMs may reach enough elimination of pollutants via convection-enhanced mass transport [1]. Traditional nonmembrane electrified techniques, run in batch or flow-by modes utilizing flat plate electrodes, conduct to the generation of thick diffusion boundary layers (~100 μm). Consequently, such techniques suffer from mass transport restrictions, since the overall reaction rate is governed by the diffusion rate of the pollutants to the electrode surface (**Figure 2(D<sub>1</sub>)**) [42]. By contrast, in flow-through mode, the usage of porous electrodes like EMs diminishes the thickness of the diffusion boundary layer to a length scale that is comparable to the membrane pore radius (**Figure 2(D<sub>2</sub>)**), therefore considerably ameliorating the mass transport rate. Usually, the smaller the EM pore size and the higher the water flux, the lower is the mass transport restriction [1].

When the membrane pore size is on the scale of nanometers (*i.e.*, UF scale), the pollutant elimination may be more elevated via nanoconfinement [43]. The

main pathways induced by nanoconfinement involve the enrichment of reactant species, like  $\cdot\text{OH}$ , in nanoconfined spaces, as well as the decrease of activation energy barrier or change of reaction pathways and kinetics [44]. Carbonaceous materials (like CNTs), which are frequently employed to produce porous structures with nanoconfinement, are as well efficacious for the adsorption of target pollutants within the membrane pores, augmenting the residence time and reduction performance [1]. The adsorption rate and capacity could be enhanced considerably via electrochemical assistance [45]. Nanoconfinement as well enriches the local level of pollutant molecules, encouraging adsorption and enhancing the following reactions [46].

The pathways examined in this Section could authorize EMs with outstanding features to attain a single-pass elimination of pollutants in seconds [1]. In particular, the elimination rates are several orders of degree greater than those attained by traditional batch and flow-by modes employing flat plate electrodes [47].

### 3. Virus Demobilization by Electrified Membranes (EMs)

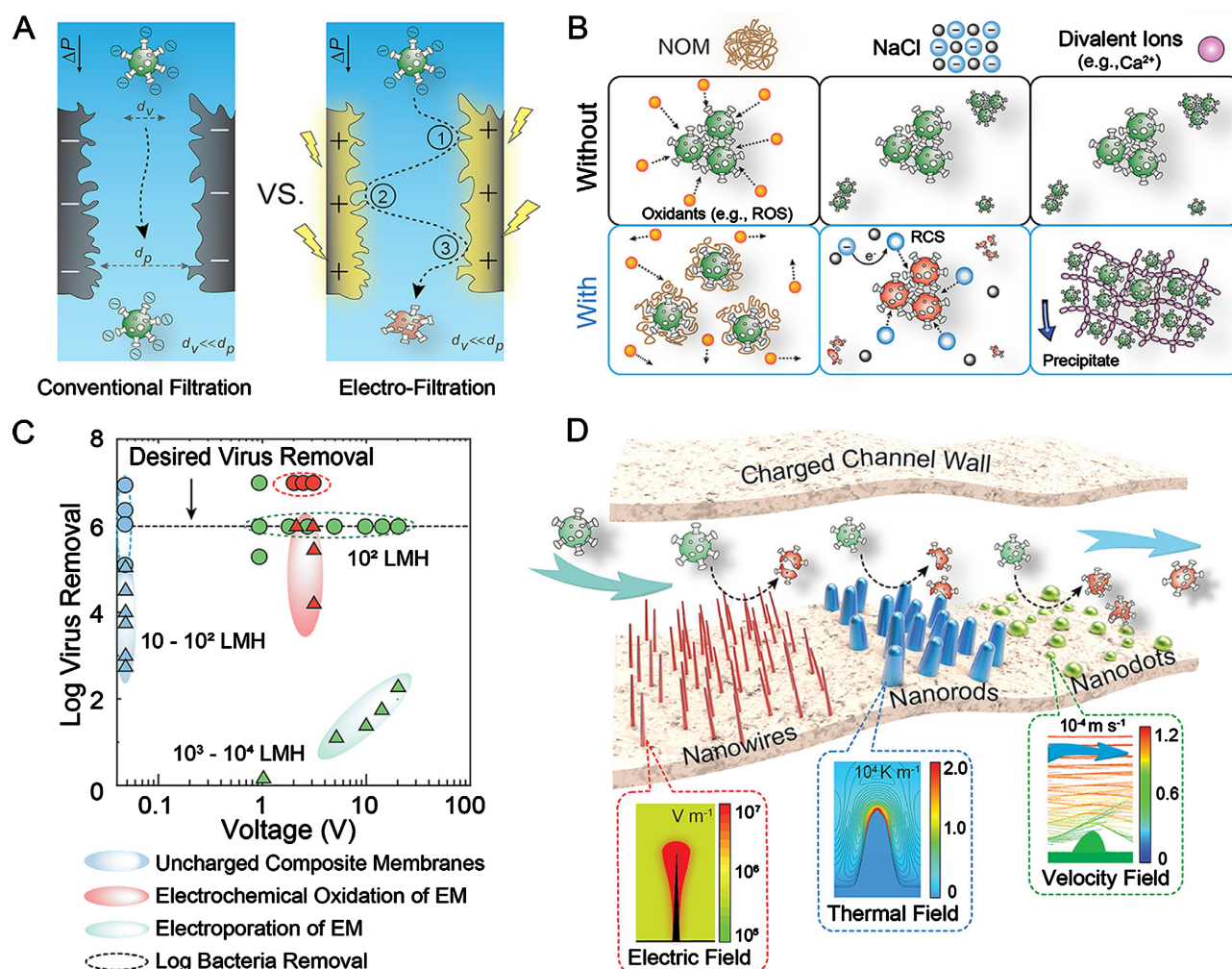
Since the outbreak of the global COVID-19 pandemic, there is a worldwide worry about the danger of pathogenic microorganisms to human health [48] [49] [50]. Wastewater reservoirs remain an elevated risk of pathogens infection, pushing investigation of techniques for eliminating and demobilizing pathogens [51] [52] [53] [54] [55]. Conventional MF/UF membrane filtration remains a performant process for retaining most protozoa and bacteria; nonetheless, MF/UF is unable to totally retain viruses that are much smaller than bacteria [56] [57]. Consequently, the request for improving membrane efficacy in dealing with viruses is expanding. As EMs present concurrent membrane filtration and *in situ* electrochemical reactivity, they have great benefits in rejecting and killing viruses [1].

#### 3.1. Virus Demobilization Using Anodic Electrified Membranes (EMs)

EMs eliminate viruses via combining physical retention with EF engendered electroadsorption and electrooxidation (EO) [58] [59] [60] (**Figure 3(A)**). Using EMs as the anode electrically adsorbs negatively charged viruses onto the membrane surface throughout filtration [61] [62] [63], while concurrently demobilizing viruses via producing oxidative stress by direct and indirect EO [64] [65] [66]. Also, reactive oxidants like ROSs and RCSs are diffuse in the permeate, stopping viral diffusion and securing water safety [1]. Oxidant production rates are crucial to virus demobilization in EM filtration, which is considerably affected by membrane materials, applied voltage, and feedwater chemistry [67] [68].

CNTs are recommended for preparing anodic EMs over a set of conductive materials like titanium suboxide and silver nanowires [69]. Without using an external voltage, a CNT-EM has attained 4 log virus elimination (with an initial virus concentration of  $10^7$  plaque-forming units (PFU)/mL) via aromatic  $\pi$ -band





**Figure 3.** Virus elimination and demobilization in electrified membrane (EM) filtration. (A) Schematic showing viruses passing through a traditional, nonconductive membrane and demobilization by anodic EM filtration via direct oxidation and reactive-oxygen or -chlorine-species (ROS/RCS)-mediated indirect oxidation. (B) Schematic depicting the effect of solution chemistry on virus demobilization in EM filtration. Natural organic matter (NOM) competes with viruses for ROS/RCS consumption, which decreases demobilization efficiency. Solutions possessing an elevated NaCl concentration promote RCS formation in anodic filtration and facilitate virus demobilization. Divalent ions (like  $\text{Ca}^{2+}$ ) induce virus aggregation that improves virus elimination via physical retention and then EO by EMs. (C) Comparison of applied voltage, water flux, and corresponding virus elimination efficacy in various categories of flow-through filtration membranes. The water flux of membranes throughout virus demobilization is presented as liters per square meter per hour (LMH). Solid and dashed ovals show log virus (triangle) and bacteria (circle) demobilization below similar circumstances. Electrified membrane electrooxidation (EM EO) greatly attains wanted virus demobilization with comparatively low voltage and moderate water flux. Data are obtained from selected literature. The model bacteria mostly utilized are *Escherichia coli* and *Bacillus subtilis*. Data depicted are mostly for MS2 and UZ1 viruses. (D) Increasing virus elimination in anodic EM filtration with optimized membrane electrode geometry. Engineered nanowire, nanorod, or nanodot additions to membrane geometries potentially increase nanoscale mass transport by locally amplifying the EF, concentrating reactants, increasing temperature, and enhancing flow convection [1].

induced adsorption. When an external potential (2 and 3 V) was used with the EMs, no culturable viruses ( $>6$  log elimination) could be observed in the permeate [70]. Viruses are demobilized in electrofiltration with a residence time (in CNT apparatus) of just 30 s [70], probably due to 1) the destruction of viral morphology under EF [70] and 2) the produced oxidative stress on viral envelope,

capsid protein, and nucleic acids provoked by ROSs/RCSs [71]. Juxtaposed with traditional electrochemical techniques for point-of-use virus demobilization [72] [73] [74], the energy consumption of the CNT-EM is lower: just 8.3 Wh/m<sup>3</sup> of permeate (with applied voltage of 2 V) [1].

Practical solution chemistries of feed waters influence not only electrochemical reactions (like oxidant formation) but also viral migration and aggregation behavior, as a result of that affecting the viral demobilization efficacy of EMs [1]. Three well-examined solution components influencing viral demobilization are natural organic matter (NOM) [75] [76] [77], chloride ions [78], and divalent cations (**Figure 3(B)**) [79] [80]. Co-occurring NOM in feedwater probably influences disinfection performance of EMs via 1) competing with viruses for oxidant consumption, 2) stopping the agglomeration of viral particles through electrostatic stabilization, and 3) averting virus-membrane contact via blocking EM reactive sites or binding with viruses to form a shielding layer [70]. Chloride ions considerably enhance virus demobilization effectiveness of a CNT filter at an applied voltage over 2.3 V, since such potential is apt to initiating the one-electron oxidation of Cl<sup>-</sup> to RCS [70]. Even if the RCS production is beneficial for viral elimination in EM process, the hazard of disinfection by-products (DBPs) formation remains an issue [81] [82] [83]. As a result, DBPs level has to be constantly observed when high-level chloride ions coexist with viruses in the feedwater [84]. Divalent cations (e.g., Ca<sup>2+</sup>) conduct to virus agglomeration via charge neutralization [85] or by bridging between viral particles and NOM (or CNTs), and then improve virus elimination via EMs by physicochemical filtration [80].

### 3.2. Efficacy of Viral Removal by Electrified Membranes (EMs)

In addition to EMs, additional types of membranes (like nanocomposite membranes and flow-through electroporation) have been tried for disinfecting viruses [1]. Nanocomposite membranes functionalized with metal nanoparticles (NPs) (e.g., AgNPs, Cu<sub>2</sub>O, TiO<sub>2</sub>NPs, Ag-/Cu-TiO<sub>2</sub> hybrid NPs) manifest considerable viral elimination (3 - 5 log) via synergy among physical retention by the membrane and the biocidal property of metal NPs (**Figure 3(C)**) [86]. Since the effectiveness of nanocomposite membranes mostly depends on contact among viruses and anchored NPs, low permeate flow rate (permeate flow rates of 10<sup>1</sup> - 10<sup>2</sup> L/(m<sup>2</sup>h) are needed for viral disinfection [1].

Electroporation uses sharp tip structures (like metal nanowires) on a membrane electrode to locally improve the EF [1]; this creates a transmembrane potential that conducts to pore generation in the lipid bilayer of enveloped viruses and following viral inactivation [87] [88]. Even if electroporation offers superior bacteria disinfection (>6 log bacterial removal under a flow rate of 10<sup>3</sup> - 10<sup>4</sup> L/(m<sup>2</sup>h) (**Figure 3(C)**) [89], its virus elimination (1 - 2 log) is notably lower under identical circumstances. Further, disinfection through electroporation of nonenveloped viruses has not yet been studied. Among all types of membrane, EM filtration attains similar efficiencies for both virus and bacteria disinfection

under a flow rate of near 100 L/(m<sup>2</sup>h) with moderate applied potential (2 - 3 V) [1]. The comparatively long retention times required are a great obstacle, since these cannot be attained in practical membrane filtration without recirculation.

Changing membrane-electrode geometry could increase viral disinfection performance. Narrowing membrane channels creates nano/micro-scale spatial confinement in EM filtration, encouraging electrochemical reactions via augmenting the frequency of collisions among viral particles and electrically produced oxidants [1]. Exploratory and computational investigations have established that patterned nanostructures ameliorate mass transport in the electrode-solution interfacial region, probably via the next pathways (Figure 3(D)): 1) nanostructures locally boost the EF that grows oxidant production on electrodes [90]; 2) patterned nano-geometry leads to nano/microscale turbulence, augmenting convection and local mixing of reactants [1]; 3) the geometric morphology conducts to spatially confined redox reactions at electrode/solution interfaces that produce temperature gradients along the nanostructures, which then speed up local convection and improve reactant mobility. Nonetheless, the feasible usage of such approach faces dares about the stability of patterned nanostructures under complex water matrices and the preparation of the sophisticated nanostructures inside porous EMs. Novel materials and techniques to manufacture nanostructure engineered EMs, which could furnish a large surface area, elevated conductivity, and increased corrosion resistance, deserve more study. Moreover, detailed techno-economic studies must be performed to evaluate the economic feasibility of such techniques contrasted to traditional desalination processes.

### 3.3. Dares and Restrictions for Electrified Membranes (EMs)

Even if numerous investigations have been dedicated to EMs for virus demobilization, EMs engineered just for such objective are far to attain economic feasibility as techniques implying chemical disinfection or membranes with smaller pore sizes (such as “tight” UF, NF, and low-pressure RO) could already attain virus disinfection/removal. In MF/UF techniques, virus demobilization can be simultaneously reached when engineering EMs for water treatment through electrochemical oxidation. Therefore, a large number of the mentioned dares for electrochemical oxidation are as well applicable to dares particular to virus demobilization. As an illustration, examining parameters of EMs for virus demobilization should extend beyond a single model virus and ideal water matrices because of the influences of species variations and the occurrence of NOM integrated with low virus levels. Also, nanostructure-enabled EM techniques for virus demobilization mostly lack practicality, due to the sophisticated processes for material preparation and the unpredictable operational stability under an EF and harsh water flow circumstances [1].

## 4. Electrochemical Disinfection (ED)

As aforesaid, EMs are firstly founded on the function of membranes and secondly

on separation by profiting from a set of electro-based phenomena, comprising electrostatic adsorption and rejection, electrophoresis, electroporation, and electrochemical oxidation and reduction [1]. Indeed, electrochemical oxidation has been considered as an encouraging technique to disinfection that could be leveraged in both centralized and distributed treatment contexts [91] [92] [93]. In ED, an oxidant is produced *in situ* through redox reactions on the surface of an electrode [94] [95] [96]. As with conventional disinfection technologies, several electrochemically produced oxidants may be employed in disinfection comprising  $\text{Cl}_2$  [97] [98] [99],  $\text{O}_3$  [100] [101] [102],  $\text{SO}_4^{\cdot-}$  [103] and  $\cdot\text{OH}$  [104] [105]. Because of modular cell design, electrochemical oxidation may potentially be scaled across centralized and distributed treatment contexts [106]. Besides, electrochemical oxidant formation may be run in resource restricted settings that usually lack transportation and electrical power distribution infrastructure via utilizing electricity produced onsite by renewable energy sources like photovoltaic cells with minimal external chemical additions [107]. There is a small interest dedicated to contextualizing the inherent dares of utilizing ED setups in both water and wastewater treatment [91].

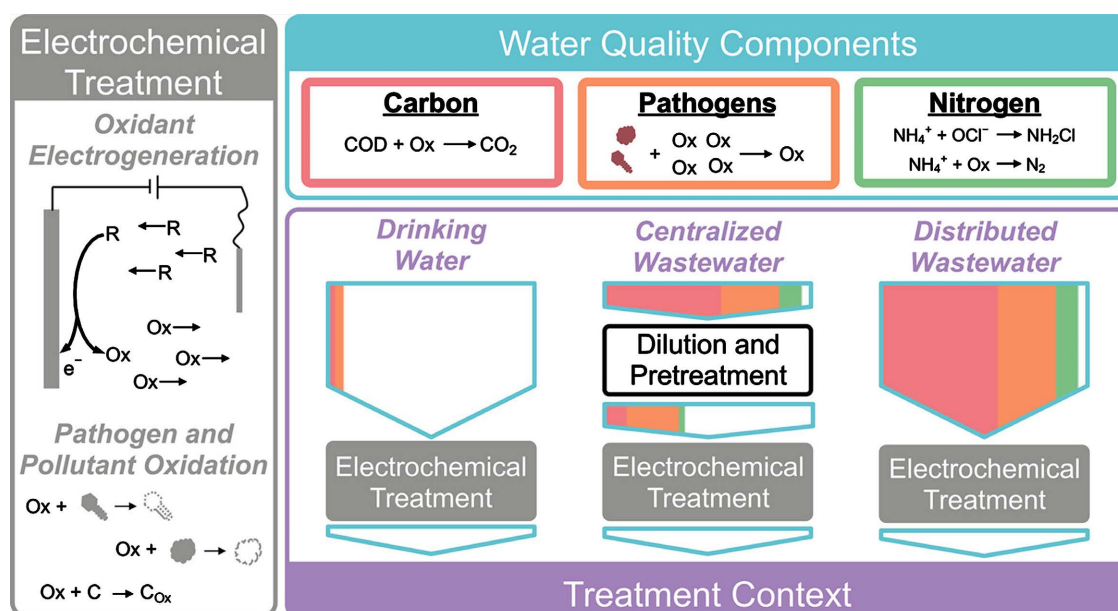
In this Section, the attention is accorded to discuss the effects of treatment context, oxidant selection, and operating practice on the reported and inherent efficacy restrictions of ED setups in terms of oxidant dose and electrical energy consumption. Finally, reporting standards for future ED studies is suggested and pathways for future development are proposed.

#### 4.1. Electrochemical Disinfection (ED) Concepts

Even if the working concept of ED through *in situ* formation of oxidants does not change, the specific oxidant formed may influence the needed dose across treatment contexts and introduce chemical reaction pathways that compete with pathogen demobilization [108] [109] [110]. Besides, the water composition across treatment contexts, and then the oxidant demand for disinfection, may change greatly (Figure 4). For ED, the three frequent treatment contexts are water treatment, centralized wastewater treatment, and distributed wastewater treatment [91]. In the water treatment context, ED may take place following coagulation/flocculation, sedimentation, and filtration stages where relevant, analogous to conventional chemical disinfection methods [91]. Whereas the scale of centralized water treatment systems could change, the composition and quality of the water to be treated depend on source and season. Within the centralized wastewater treatment context, ED would also proceed analogously to conventional chemical disinfection methods following primary treatment and secondary biological treatment at a centralized treatment plant.

#### 4.2. Electrochemical Disinfection (ED) Perspectives

Novel investigations focused on augmenting sanitation access throughout the world and ameliorating present sanitation system resiliency has conducted to



**Figure 4.** Electrochemical disinfection (ED) produces oxidants *in situ* to demobilize pathogens and chemically transform reduced carbon and nitrogen molecules. Differences in water quality (most importantly carbonaceous chemical oxygen demand (cCOD), ammonia, and pathogen levels) observed across the water treatment contexts such as drinking water and wastewater directly impact required oxidant dose [91].

refreshed attention in ED setups for treating and disinfecting water [111] [112] [113]. Nonetheless, many investigations have tested ED device efficacy, reporting metrics have varied largely [114] [115]. When assessing ED devices both internally (like across oxidant types, working parameters, and setup arrangements) and externally (such as conventional chemical disinfection, filtration techniques [116] [117] [118], and thermal demobilization [119] [120]), it is crucial to evaluate both the disinfection performance and the energy consumed throughout process [91].

As a consequence, next researches with electrochemical treatment must directly detail both the dose produced and the energy consumption throughout production to let for juxtaposition across working parameters and procedures [91] [121]. If dose cannot be directly estimated, as in the instance of several  $\cdot\text{OH}$  founded devices, then the concentration of oxidant must be evaluated and detailed utilizing regularly adopted practices [122] [123]. Also, the charge efficacy of oxidant formation must be detailed where possible to contrast running features across the range of treatment contexts [124].

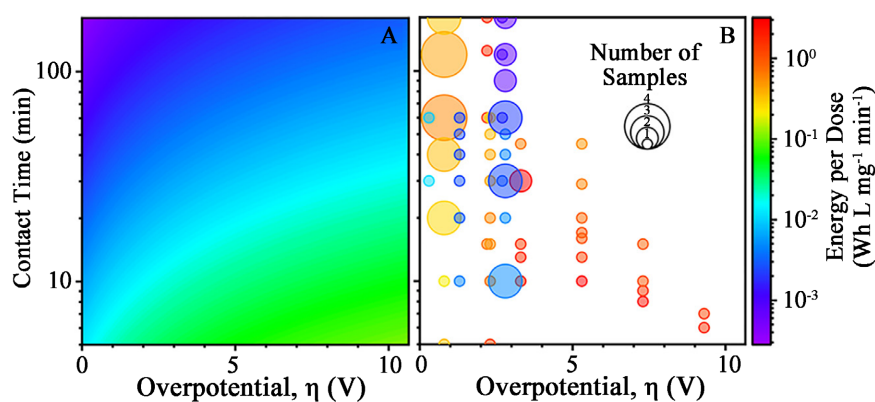
Hydroxyl radicals seem to be an encouraging oxidant for application in electrochemical treatment thanks to their high oxidant strength and possibility for demobilizing highly recalcitrant pathogens like *Cryptosporidium* spp. [91]. Nevertheless, their efficiency has not been as easily assessed comparatively to more usual oxidants (e.g.,  $\text{Cl}_2$ ) [125] [126] [127]. Thus, more investigations remain required to both describe the efficacy and navigate trade-offs among energy consumption and oxidant strength in  $\cdot\text{OH}$  founded setups, especially at adequate doses for killing viruses and *Giardia*. By more comprehending disinfection effectiveness and energetics in  $\cdot\text{OH}$  founded devices, the comparative strengths

and demerits of various oxidants could be defined for applicability across treatment contexts. Within resource limited settings, merging renewable energy sources (like photovoltaic cells) constitutes a significant route for sustainable ED [128] [129] [130].

Finally, substitutional arrangements and functioning procedures stay to be explored particularly targeting energy per dose lowering [91]. Both empirical investigations and simulated efficiency indicate that using lower cell voltages through prolonged residence periods conducts to appreciably lowered energy per dose (Figure 5). In both centralized and distributed treatment contexts, this will probably need engineering vessels that maximize the residence period over disinfection. More investigations stay requested to determine and navigate expected trade-offs among effectiveness and economic viability of batch, semi-batch, and continuous operation when used at high residence periods (especially for larger, centralized systems that usually work at very high throughput and can need continuous treatment setups). Even if such modifications could constitute an unsuitable handicap for present centralized treatment systems, prolonged contact periods are habitual in the batch or semi-batch treatment systems largely employed for distributed or semi-centralized treatment in resource-limited settings [131] [132]. Considerable dares endure for the diffuse deployment of robust ED setups. Nonetheless, when employed inside treatment contexts that boost intrinsic advantages, ED offers real promise for increasing worldwide access to sanitation and safe drinking water (Table 1).

## 5. Combining Electrified Membranes (EMs) and Electrochemical Disinfection (ED) for Virus Demobilization

As seen in Section 4, in the field of disinfecting water, if there is a technology



**Figure 5.** (A) Theoretical minimum energy per dose as a function of overpotential (horizontal axis) and contact time (vertical axis) for a simulated 1 L,  $\text{Cl}_2$  producing apparatus. All cell voltage is assumed to be attributable to the working electrode. (B) Calculated energy per dose from  $\text{Cl}_2$  generating systems as reported in the literature plotted at corresponding overpotentials (horizontal axis) and contact times (vertical axis). Data reported for identical overpotential and contact times were averaged to a single energy per dose value with the bubble size indicating the number of samples averaged under those operating conditions [91].

**Table 1.** Advantages and dares of electrochemical disinfection (ED) across oxidant types and treatment contexts [91].

|                          | Advantages   | Dares  |
|--------------------------|--|--|
| <b>Oxidants</b>          |  |  |
| Cl <sub>2</sub>          | Cl <sup>-</sup> is among the most abundant anions in most waters; lower standard potentials than most electrochemically generated oxidants; longer lifetimes allow for disinfection residual; can combine with ammonia to form chloramines with lower DBPs and more stable residual.                             | Cl <sup>-</sup> must be present in sufficient concentration or externally supplied; many oxidized Cl species are toxic (ClO <sub>2</sub> <sup>-</sup> , ClO <sub>3</sub> <sup>-</sup> , etc.); current efficiency depends on Cl <sup>-</sup> concentration, limiting performance in low salinity waters; can generate chlorinated DBPs (e.g., trihalomethanes and haloacetic acids). |
| •OH                      | Strongest disinfectant among common electrochemically generated oxidants; can be generated without specific precursor ions (e.g., Cl <sup>-</sup> ); can also oxidize total organic carbon (TOC) and many micropollutants; capable of treating <i>Cryptosporidium</i> at practical doses.                        | Highest standard potential of common electrochemically generated oxidants; radical species have very low lifetimes in solution; can generate BrO <sub>4</sub> <sup>-</sup> and brominated compounds in Br <sup>-</sup> containing waters.  |
| <b>Treatment context</b> |  |  |
| Drinking water           | Low organic carbon levels reduce scavenging and necessary dose, depending on placement in treatment train; does not require onsite hazardous chemical storage; lower overall dose required compared to wastewater.   | May require Cl <sup>-</sup> addition if Cl oxidant species are desired; most oxidants produced electrochemically have shorter lifetime, making residual generation difficult.  |
| Centralized wastewater   | Cl <sup>-</sup> concentration is usually sufficient to generate Cl oxidant; high NH <sub>4</sub> <sup>+</sup> levels can scavenge more oxidized Cl species (ClO <sub>2</sub> <sup>-</sup> , ClO <sub>3</sub> <sup>-</sup> , etc.) and produce chloramine; dose can be dynamically adjusted during severe events. | High carbonaceous chemical oxygen demand (cCOD) levels introduce competition for oxidant; electrochemical systems typically scale linearly for cost and energy demand.   |
| Distributed wastewater   | Electrochemical systems can be easily scaled down for portable units; a single unit can be used to electrochemically generate oxidants for simultaneous treatment of cCOD, NH <sub>4</sub> <sup>+</sup> , and pathogens.   | Higher energy inputs are required for treatment of multiple target species; electrochemical systems typically have high capital cost compared to other methods.  |

that has attracted considerable interest from water treatment engineers it is the ED method [122] [123]. The largely accepted tendency about the use of the ED technique is to apply it as a combined stage with more methods. For instance, as a form of ED, electrocoagulation (EC) technique is usually inserted as a pre-stage before EO method in the treatment train. For this integration, more important virus removal is likely attained by the collective actions of physical retention by coagulation/filtration, ferrous iron-based disinfection, and EO disinfection. However, additional research needs to be performed to (i) distinguish among the EF and cohesion contributions, and to assess the more and more likely generation of •OH during the EC technology. On the other hand, as in the chemical water disinfection, similar issues like DBPs formation have also appeared in the EC applications. More research needs to be pointed into such directions [123].

In a similar manner, this discussion suggests that EMs would be merged with

ED as an intensified process for disinfecting and treating water. The suggested device may involve the two processes combined in a single recipient. Such combination would be similar as done in the case of membrane bioreactor, in which membrane process is integrated into a biological reactor [116].

## 6. Conclusions

Electrified membranes (EMs) possess the capacity to handle the intrinsic restrictions of traditional membrane techniques and show improved functions beyond separation. Further, as a process in which chemical oxidants are produced *in situ* via redox reactions on the surface of an electrode, electrochemical disinfection (ED) has recently magnetized increased interest as an option to conventional chemical dosing disinfection techniques. In this review, we focused on fresh improvements in EMs, especially on water and wastewater disinfection. We presented a brief description of materials categories, synthesis procedures, and electrified filtration operating modes. Also, we discussed applications of EMs, especially water disinfection via bacterial and viral inactivation, and described concepts of ED and its perspectives. From this work, the main points drawn may be listed below:

1) EMs could be appropriate for progressing fit-for-purpose water and wastewater treatment, where pollutant elimination and organic fouling control constitute the most possible utilizations. Electrified conversion of pollutants like nitrate and organic micropollutants could efficiently eliminate such contaminants while eliminating the production of concentrated waste streams and decreasing the injection of chemicals. Flow-through EMs furnish a scalable platform for such electrochemical techniques thanks to intensifications in mass transfer, conversion efficiency, and stability. Besides pollutant elimination, EMs are encouraging for organic fouling control, enhancing membrane longevity while reducing pretreatment steps and the introduction of chemicals. EMs are most convenient for UF and MF utilizations thanks to the practicability of membrane surface amendment and flow-through filtration operation [1].

2) Even if EMs have revealed notable merits for a set of implementations, some obstacles last for performing their application. In general, actual implementations need the maturing of solutions for working EMs under real water/wastewater circumstances. To attain sustainable and energy-efficient EM techniques, additional studies must be dedicated to reducing the phenomenon of competing reactions and improving the stability of EMs throughout the long-term operation, particularly in complex water matrices. Moreover, the generation of poisonous DBPs (e.g., chlorinated organic compounds, and oxyhalides like  $\text{ClO}_4^-$ ) stays to be dominated. When treating water for potable use or municipal wastewater is considered, applying EMs to such waters that possess comparatively low salt levels must be estimated. EMs for pollutant elimination must reach ~100% elimination efficacy with high current efficiency and low energy consumption during single-pass flow-through filtration to eliminate disposal



hazards. The engineering of EM modules requires to be ameliorated to ease the combination with conventional membrane processes or implementations for decentralized water treatment. When such dares are handled, the improved running of EMs must force them feasible for real water treatment implementations [1].

3) This discussion suggests that EMs would be merged with ED as an intensified process for disinfecting and treating water. The suggested device may involve the two processes combined in a single recipient.

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## Conflicts of Interest

The authors declare no conflicts of interest.

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## Abbreviation

|      |                                     |
|------|-------------------------------------|
| BDD  | Boron-doped diamond                 |
| cCOD | Carbonaceous chemical oxygen demand |
| CNTs | Carbon nanotubes                    |
| DBPs | Disinfection by-products            |
| EC   | Electrocoagulation                  |
| EMs  | Electrified membranes               |
| ED   | Electrochemical disinfection        |
| EF   | Electric field                      |
| EO   | Electrooxidation                    |
| LMH  | Liters per square meter per hour    |
| MF   | Microfiltration                     |
| NF   | Nanofiltration                      |
| NOM  | Natural organic matter              |
| NPs  | Nanoparticles                       |
| OEP  | Oxygen evolution potential          |
| PANI | Polyaniline                         |
| PPy  | Polypyrrole                         |
| RCSs | Reactive chlorine species           |
| RO   | Reverse osmosis                     |
| ROs  | Reactive oxygen species             |
| UF   | Ultrafiltration                     |