Membrane Technology for Pathogenic Virus Removal: A Comprehensive Review of Centralized and Decentralized Water Treatment Processes

Rui Wang[1,*]

[1]College of Environmental Science and Engineering, Tongji University, Shanghai 200092, PR China.

*E-mail: uiwangr@gmail.com

Abstract

The recent COVID-19 pandemic has once again drawn attention to the risks of pathogenic viruses, as viruses can be transmitted through water environments, posing a threat to human health. Membrane technology, as a novel, green, and effective water treatment process, has been widely applied in the removal of pathogenic viruses from wastewater and drinking water. However, compared to refined centralized water treatment processes in urban areas, rural areas still lack effective water treatment strategies for decentralized water supply and drainage systems. Therefore, this review provides a comprehensive analysis of the hazards and transmission pathways of viruses, the mechanisms and influencing factors of virus removal by membrane processes. Based on the characteristics of centralized and decentralized water treatment systems, it further summarizes the application scenarios and effectiveness of the most

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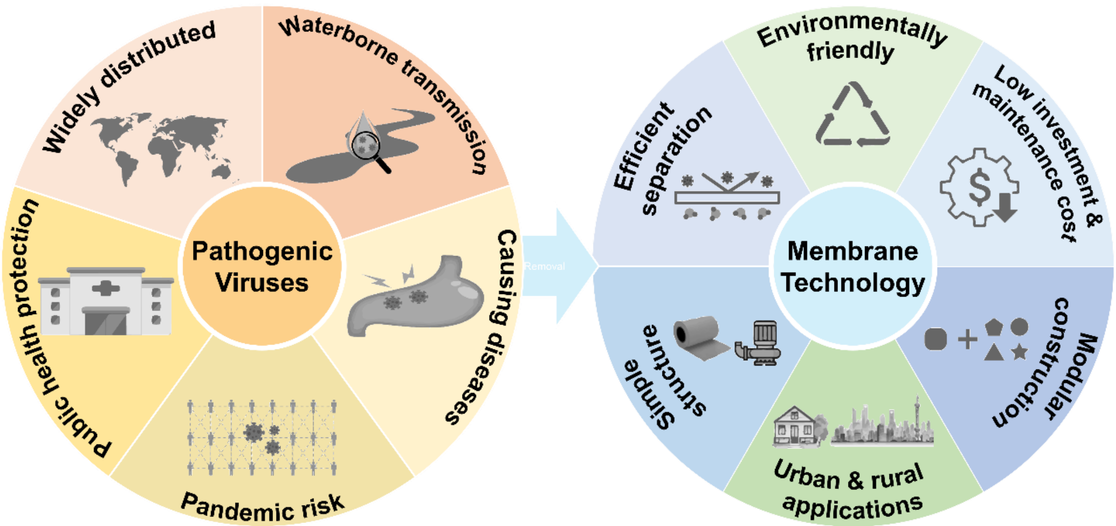
widely used membrane processes. We emphasize the need for context-specific development of membrane-based water treatment processes tailored to urban or rural areas, addressing both water supply and wastewater treatment processes, to ensure comprehensive public health safety in water environments. In addition, this review also discusses several challenges that membrane technology faces in virus removal, providing new insights for further research in membrane processes.

Keywords: pathogenic viruses, membrane technology, drinking water treatment, wastewater treatment, centralized, decentralized

Graphical Abstract



1. Introduction

Pathogenic viruses in the water environment are a serious public health and safety concern, because they can enter drinking water sources through surface runoff or municipal pipelines.[1, 2] When humans are exposed to pathogenic viruses, their physical well-being is compromised. For example, enterovirus and norovirus can induce gastrointestinal illnesses, poliovirus and zoster virus can lead to neurological disorders, and the recent COVID-19 virus has claimed over a million lives.[3-5] These viruses are widely distributed across the globe and can be transmitted through the water environment, having caused numerous pandemics and endangered the safety of billions of individuals worldwide.[6] Today, in the centralized water supply and drainage processes, disinfection has become an indispensable part of water treatment technologies. The main strategies employed include: (1) direct killing of viruses by oxidants such as O_3, Cl_2, ClO_2, etc.; (2) coagulation and precipitation of viruses by flocculants such as $AlCl_3$, $FeCl_3$, polyacrylamide (PAM), etc.; (3) destruction of the protein structure of viruses by ultraviolet irradiation, etc.[7-9] However, these disinfection strategies still come with limitations, as they are restricted by (1) the secondary contamination of disinfection by-products, (2) the economic costs of chemical agents, and (3) the long-term reliability of virus inactivation during water treatment processes.[9-11]

Membrane separation processes, as an emerging water treatment technology, with the advantages such as efficient separation, energy savings, and low costs.[12] They hold promising prospects in the removal of viruses from water environment.[13, 14] Among them, microfiltration

(MF, 0.1~10μm), ultrafiltration (UF, 10~100nm), and nanofiltration (NF, 1~10nm) are currently the most widely used membrane technologies.[13-15] These processes retain viruses larger than the membrane pore size on the influent side through the size exclusion effect, and can further enhance the removal efficiency by forming a gelatinous cake layer.[16-18] This significantly reduces the quantity of viruses in the permeate, ensuring a low viral risk in the water environment. Furthermore, membrane technology can serve as an independent water treatment module, coupled with other processes to meet specific water quality requirements. For instance, when there is a high quantity of microorganisms in the water environment (e.g., algae, bacteria, etc.), membrane fouling can be mitigated through pre-treatment processes such as coagulation and sedimentation.[19, 20] Similarly, when the salinity of the water source is excessively high, advanced treatment processes such as NF and reverse osmosis (RO) can be employed to improve the potability.[21, 22] In summary, the modular characteristics of membrane technology enable it to achieve highly efficient removal of viruses under complex water quality conditions.[22, 23]

However, despite its effective role in virus removal from water environment, membrane technology is currently primarily utilized in urban areas.[24] Due to the limitations imposed by factors such as regional economic development, population distribution and hydrological conditions, the deployment of membrane separation strategies in centralized water treatment in urban areas is challenging to extend to the decentralized water treatment processes in rural areas.[24, 25] Moreover, combined with the inadequate understanding of public hygiene among local residents, this often results in viral outbreaks in rural areas lacking water treatment facilities.[26] For example, recent pandemic reports from the World Health Organization (WHO) include poliovirus outbreaks in Tanzania, norovirus outbreaks in the Congo, and hepatitis E virus outbreaks in South Sudan, etc.[27] According to relevant research, approximately 1,800,000 deaths occur annually due to water quality safety and environmental sanitation, with over one-third of these fatalities involving children under the age of five.[28] Nearly all of these incidents are reported in developing countries. Therefore, researchers have also begun to develop and investigate membrane-based decentralized water treatment technologies, such as ceramic membrane (MF) and gravity-driven membrane (UF).[29, 30] These low-cost, low-maintenance, and easy-to-operate membrane processes can effectively prevent the harm of pathogenic viruses to human health.[31-33]

This critical review evaluates the contribution of membrane technology to the removal of pathogenic viruses in water environments for public health protection, focusing on (1) the transmission and hazards of viruses in the water environment, (2) the removal efficiency and mechanisms of membrane technologies for viruses, and (3) the application and promotion of membrane technologies under centralized and decentralized water treatment systems. Our primary aim is to discuss the removal of viruses from the water environment through membrane technology in both urban and rural areas, water supply and drainage processes, to ensure better and comprehensive protection of public water security.

2. The current state of contamination by pathogenic viruses in the water environment

2.1 Sources, transmission routes and hazards of pathogenic viruses

2.1.2 Sources and contamination levels

Typically, in natural rivers and lakes, the background level of pathogenic viruses in the water environment is relatively low. For enteric viruses, it is approximately 10^2 to 10^4 copies L^{-1}.[34, 35] However, the water environment serves not only as the primary carrier for the survival of pathogenic viruses but also as a harbor for the accumulation of various viruses.[36] When virus-containing pollutants enter the water environment, the number of viruses begins to increase rapidly. For example, through the urban sewage networks, a multitude of pathogenic viruses, such as norovirus, echoviruses, adenovirus, polioviruses, coxsackieviruses, and HIV, can rapidly proliferate and damage the water environment, sourced from various origins, including medical wastewater from hospitals, domestic sewage from communities, and leachate from landfills.[36-38] Although the numbers of these pathogenic viruses are significantly reduced after passing through the wastewater or drinking water treatment plants (WTPs), taking several WTPs in **Table 1** as an example, they can still be detected in large quantities (exceeds the WHO requirements, 10 viruses per liter).[39] This suggests that such effluent water as an important source of viral contamination, can pose a significant hidden risk for disease transmission, it may mix with drinking water sources through surface runoff, and allowing viruses to return to the community via the pipe network (as shown in **Figure 1**).

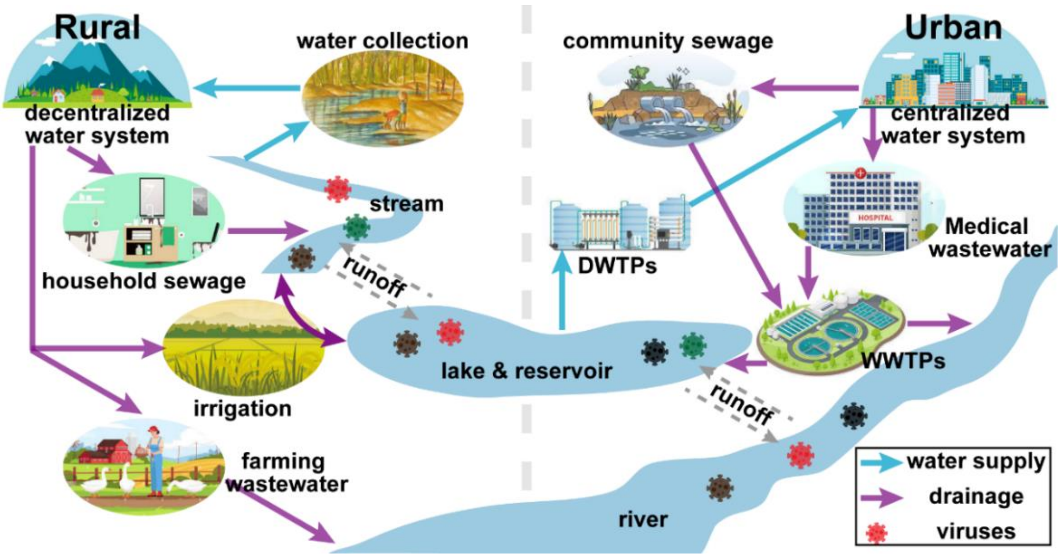


Figure 1. Sources and transmission routes of pathogenic viruses in the water environment (draw according to the content of references).[2, 36, 37, 40]

For rural areas, the issue of viral contamination of drinking water sources is even more concerning.[41, 42] Due to differences in lifestyle and customs, untreated sewage containing pathogenic viruses, including human, livestock, and wildlife feces, as well as agricultural irrigation, is commonly discharged in rural areas.[24] This not only directly contaminates drinking water sources but also indirectly exacerbates the transmission of vector-borne or water-borne diseases, such as encephalitis, yellow fever, dengue fever, Lassa fever, Ebola hemorrhagic fever, and Marburg virus disease.[43] Moreover, in rural areas, there is often a lack of effective water treatment facilities for water supply and drainage. Residents typically directly consume water from surface water sources such as lakes, reservoirs, or groundwater sources like wells, without undergoing reliable virus inactivation processes,[24] and the number of enteric viruses can even reach 10^7 copies L^{-1}.[44] This makes it easier for viruses to be transmitted to humans via the fecal-oral route, leading to illness or even death.[41]

2.1.2 Transmission routes and hazards

Pathogenic viruses in water environments directly or indirectly pose serious and far-reaching threats to human health. According to researchers, globally, there are approximately 1.2 to 2.4 million cases of diarrhea each year caused by viruses like norovirus, rotavirus, sapovirus, astrovirus, and enterovirus, resulting in 180,000 to 290,000 deaths.[3] Meanwhile, diseases caused by mosquito and fly bites, such as yellow fever virus and Zika virus, result in approximately 84,000 to 170,000 severe cases and 29,000 to 60,000 deaths each year.[26] In addition to these acute diseases, the mortality resulting from chronic diseases caused by viruses, such as the hepatitis virus, is also significantly alarming, with approximately 248 million people worldwide infected with the HBV.[45] All the cases mentioned above represent only a fraction of the diseases caused by pathogenic viruses. As of 2001, the number of known infectious viruses that can cause diseases in humans had already exceeded 217.[6, 43]

Unfortunately, when pathogenic viruses break out in urban areas, they can quickly spread through the pipeline network to affect the majority of the population.[1, 46] For example, an outbreak of sapovirus and norovirus in Finland led to over 400 cases of gastroenteritis in approximately a week, affecting about 4,000 people.[46] In contrast, when viruses break out in rural areas, the lack of effective treatment measures can result in even more serious casualties.[41, 42] For instance, an outbreak of hepatitis E virus in the Kashmir region of India resulted in 52,000 cases of jaundice and over 1,700 deaths, affecting around 600,000 people.[41] In conclusion, whether viruses harm human health through water-borne or vector-borne routes, direct consumption, or indirect contact, water supply and drainage processes are the primary medium for their transmission (**Figure 1**). Therefore, significantly reducing viral contamination in the water environment through WTPs can greatly enhance the health and safety of residents.

161 **Table 1.** The number of viruses in the influent and effluent of several typical water treatment plants worldwide.

Region	Process of WTPs	Virus[(a)]	Number of viruses	Hazards	Function of WTPs [(b)]	Ref.
Calgary, Canada	Activated sludge + Ultraviolet (UV)	ReoVs	Influent: 1.7 log MPN L^{-1} Effluent: 0.2 log MPN L^{-1}	Causes symptoms such as diarrhea and vomiting.	Municipal wastewater treatment (centralized)	7
Hokkaido, Japan	MnO_x filtration + microfiltration (MF)	EVs	Influent: 3.5 log copies mL^{-1} Effluent: 1.0 log copies mL^{-1}	Causing symptoms such as fever, pharyngitis, cough, and muscle pain.	Drinking water treatment (centralized, underground water)	8
Pursat, Cambodia	Silver-impregnated ceramic filter	MS2 phage	Influent: 10^5~10^8 PFU mL^{-1} LRV: 1.3~2.4 log	Uncertain risk, typically used as fecal contamination indicators.	Drinking water treatment (decentralized, rainwater)	47
Kampala, Uganda	Conventional activated sludge process (CASP)	HA(A)Vs	Influent: 7.7 $\times 10^3$ copies L^{-1} Effluent: 6.1 $\times 10^3$ copies L^{-1}	Causing symptoms such as fatigue, nausea, and abdominal discomfort.	Municipal wastewater treatment (centralized)	37
California, USA	Membrane bioreactor (MBR)	NoVs GII	Influent: 10^5 copies mL-1 LRV: 4.6~5.7 log	Causing symptoms such as vomiting, diarrhea, abdominal pain, and nausea.	Municipal wastewater treatment (centralized)	48
London, UK	MBR + GAC filtration	FRNAPH	Influent: 2.7 $\times 10^4$ PFU 100 mL^{-1} LRV: 3.8 log	Uncertain risk, typically used as fecal contamination indicators.	Municipal wastewater treatment (centralized)	49
Michigan, USA	MBR	AdVs	Influent: 10^8~10^9 viruses L^{-1} Effluent: 10^3~10^4 viruses L^{-1}	Causing symptoms such as the common cold, and pharyngitis.	Municipal wastewater treatment (centralized)	50
Tokyo, Japan	Slow sand filtration (SSF)	PMMoV	Influent: 2.4 log copies L^{-1} LRV: 1.8~2.8 log	Uncertain risk, typically used as fecal contamination indicators.	Drinking water treatment (centralized, lake water)	51
Ouerdanine, Tunisia	Conventional activated sludge process (CASP)	HA(A)Vs	Influent: 5.3 $\times 10^3$ copies mL^{-1} Effluent: 2.8 $\times 10^3$ copies mL^{-1}	Causing symptoms such as fatigue, nausea, and abdominal discomfort.	Municipal wastewater treatment (centralized)	52

Bologna, Italy	Conventional activated sludge process (CASP)	SOMCPH	Influent: 6.8 log PFU 100 mL^{-1} Effluent: 3.9 log PFU 100 mL^{-1}	Uncertain risk, typically used as fecal contamination indicators.	Municipal wastewater treatment (centralized)	53
Giza, Egypt	UASB + BAF + IPS	NoVs GGI	Influent: 3.5 $\times 10^4$ copies L^{-1} LRV: 3.3 log	Causing symptoms such as vomiting, diarrhea, abdominal pain, and nausea.	Pilot wastewater treatment (decentralized)	54
Trondheim, Norway	Lake bank filtration (LBF)	FRNAPH	Influent: 0.3~13 log PFU L^{-1} LRV: 2.1~3.2 log	Uncertain risk, typically used as fecal contamination indicators.	Drinking water treatment (centralized, lake water)	55
Ohio, USA	MBR	EVs	Influent: 4.7 log copies mL^{-1} Effluent: 0.3 log copies mL^{-1}	Causing symptoms such as fever, pharyngitis, cough, and muscle pain.	Municipal wastewater treatment (centralized)	56
Ouro Preto, Brazil	Biodigester septic tank (BST)	AdVs	Influent: 10^7~10^8 UFC mL^{-1} LRV: 3.0 log	Causing symptoms such as the common cold, and pharyngitis.	Black wastewater treatment (decentralized)	57
Nantes, France	Anoxic/oxic MBR (A/O-MBR)	SaVs	Influent: 3.0 $\times 10^5$ copies L^{-1} Effluent: 4.5 $\times 10^2$ copies L^{-1}	Causing symptoms such as vomiting, diarrhea, nausea, and abdominal pain.	Municipal wastewater treatment (centralized)	58
Sud Yungas, Bolivia	Upflow anaerobic sludge blanket (UASB)	RVs	Influent: 10~100 copies mL^{-1} LRV: 0.8 log	Causing symptoms such as vomiting, diarrhea, and fever.	Domestic wastewater treatment (decentralized)	59
New South Wales, Australia	MBR	FRNAPH	Influent: 4.7 log PFU 100 mL^{-1} Effluent: 0.5 log PFU 100 mL^{-1}	Uncertain risk, typically used as fecal contamination indicators.	Municipal wastewater treatment (centralized)	60
Groningen, Netherlands	Aerobic granular sludge (AGS)	FRNAPH	Influent: 10^6 PFU 100 mL^{-1} LRV: 1.5~2.8 log	Uncertain risk, typically used as fecal contamination indicators.	Municipal wastewater treatment (centralized)	61
Edmonton, Canada	Activated sludge + Ultraviolet (UV)	EVs	Influent: 4.61 GE copies L^{-1} Effluent: 2.87 GE copies L^{-1}	Causing symptoms such as fever, pharyngitis, cough, and muscle pain.	Municipal wastewater treatment (centralized)	62

Dhaka, Bangladesh	Ultrafiltration (UF)	AdVs	Influent: 10^3~10^4 copies mL^{-1} Effluent: ND~10^2 copies mL^{-1}	Causing symptoms such as the common cold, and pharyngitis.	Drinking water treatment (decentralized, river water)	63
North-Rhine Westphalia, Germany	Activated sludge	SARS-CoV-2	Influent: 25 copies mL^{-1} Effluent: 13 copies mL^{-1}	Cause pneumonia, along with fever, cough, and dyspnea.	Municipal wastewater treatment (centralized)	64
Jerusalem, Israel	Activated sludge + Chlorination (Cl_2)	SARS-CoV-2	Influent: 10^4 copies mL^{-1} Effluent: 10^2 copies mL^{-1}	Cause pneumonia, along with fever, cough, and dyspnea.	Municipal wastewater treatment (centralized)	65
Rome, Italy	Conventional activated sludge process (CASP)	EVs	Influent: 3.3×10^7 GC L^{-1} LRV: 0.63 log	Causing symptoms such as fever, pharyngitis, cough, and muscle pain.	Municipal wastewater treatment (centralized)	66
Arizona, USA	Activated sludge + Chlorination (Cl_2)	PMMoV	Influent: 3.7×10^6 copies L^{-1} LRV: 0.76 ± 0.5 log	Uncertain risk, typically used as fecal contamination indicators.	Municipal wastewater treatment (centralized)	67
Eastern Cape, South Africa	Activated sludge	RVs	Influent: 1.2×10^5 GC L^{-1} Effluent: 2.6×10^4 GC L^{-1}	Cause diarrhea, vomiting, fever, abdominal pain, and can lead to dehydration.	Municipal wastewater treatment (centralized)	68
Yamanashi, Japan	Activated sludge	SARS-CoV-2	Influent: 10^3~10^4 copies L^{-1} Effluent: 10^2~10^3 copies L^{-1}	Cause pneumonia, along with fever, cough, and dyspnea.	Municipal wastewater treatment (centralized)	69

(a). Abbreviations for viruses are shown in the *Supporting Information*.

(b). The functions of the WTPs are determined by the source of the influent and the destination of the effluent.

2.2 Common technologies for pathogenic virus removal in the water environment

Regarding the issue of virus removal, under different water treatment modes, the main strategies can be classified into two categories: (1) centralized treatment, such as drinking water treatment plants (DWTPs) and wastewater treatment plants (WWTPs), and (2) decentralized treatment, such as household water purifiers and biogas digester.

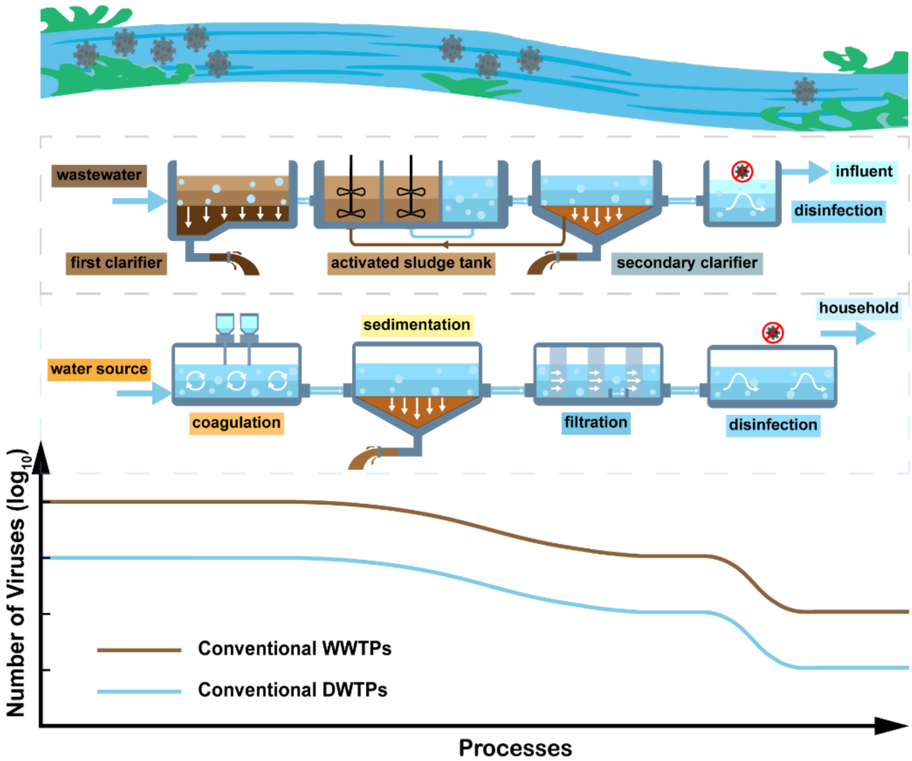


Figure 2. Schematic diagram and pathogenic virus removal efficiency at various stages of conventional centralized WWTPs and DWTPs (draw according to Table 1 and Table S2&S3).

In urban centralized water treatment, the most widely used wastewater treatment strategy is the activated sludge process, while the drinking water treatment strategy involves coagulation, sedimentation, filtration, and disinfection.[70] Unfortunately, a large amount of data shows that the activated sludge process has a very limited effect on virus removal, with primary treatment almost unable to remove viruses, and secondary treatment achieving a removal of only about 1~2 log.[9, 61] Similarly, coagulation-precipitation removes about 0.4~1.7 log of viruses from drinking water (**Figure 2**).[8, 71] Therefore, for the effluent of DWTPs and WWTPs, disinfection processes are always necessary, with chemical agents such as Cl_2, ClO_2 and NaClO being used for oxidative inactivation of viruses.[70]

However, this process easily generates disinfection by-products like trihalomethanes (THMs), haloacetic acid (HAA), and bromide precursors, causing secondary harm to the human.[11] In contrast, although the use of ultraviolet radiation is environmentally friendly, the photoreactivation makes its viral inactivation effect unstable and susceptible to the influence of turbidity.[10] In decentralized household wastewater treatment, rural residents often collect domestic wastewater in biogas digesters for fermentation or discharge it without treatment, both of which have limited disinfection effects.[59] For household drinking water treatment, boiling is used for disinfection.[72] Although boiling water can effectively inactivate viruses, it directly accelerates the depletion of forests and grasslands, indirectly exacerbating greenhouse effects and soil erosion.[24] Therefore, the challenge of removing viruses through sustainable technologies to protect human health remains persistent.

3. The mechanisms, applications, and influencing factors of pathogenic virus removal by membrane technology

Membrane separation technology provides a new approach to virus removal in water treatment processes (**Figure 3**). As a physical barrier based separation process that strictly adheres to the size exclusion effect, membrane technology offers stable and consistent results in virus removal.[13] Additionally, compared to conventional technology (**Table S3**), membrane filtration does not require the addition of any chemical agents, which not only saves costs but also makes it environmentally friendly.[15] Currently, with the development of membrane technology, membrane filters have become more affordable, and their operational procedures have simplified. Therefore, the application of membrane technology in virus removal processes is gradually being promoted worldwide.

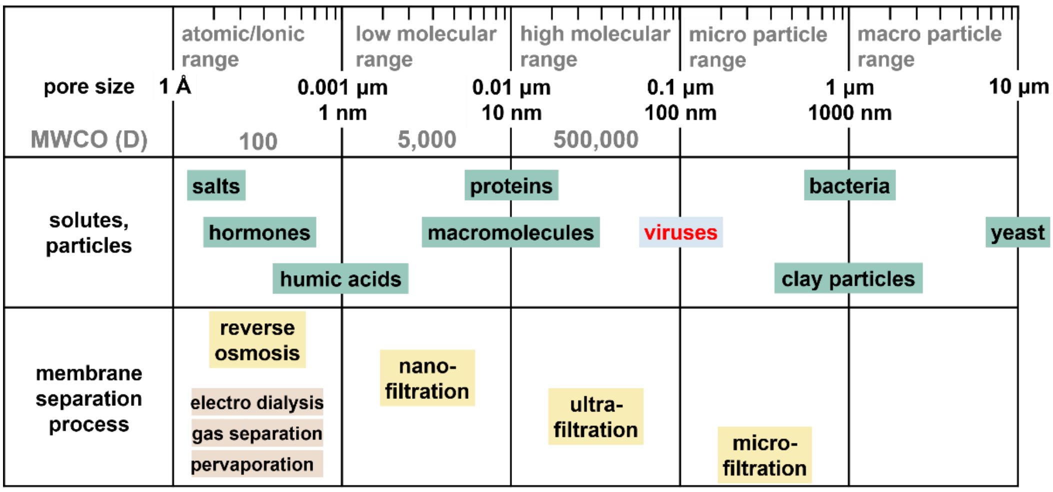


Figure 3. The pore size of membrane separation processes, the molecular weight cutoff (MWCO), the size of solutes and particles, and examples of their virus removal capabilities.[24]

3.1 Microfiltration and Ultrafiltration

In general, the effluent water quality requirements for wastewater treatment and drinking water treatment are different, which leads engineers to employ various membrane processes to address virus contamination when designing water treatment plants. For centralized drinking water treatment, considering the water quantity and stability in the water supply, membrane units typically utilize UF membranes with pore sizes ranging from 10 to 100 nm, which appropriately cover the size range of most viruses.[70] And combined with MF as pre-treatment to control membrane fouling.

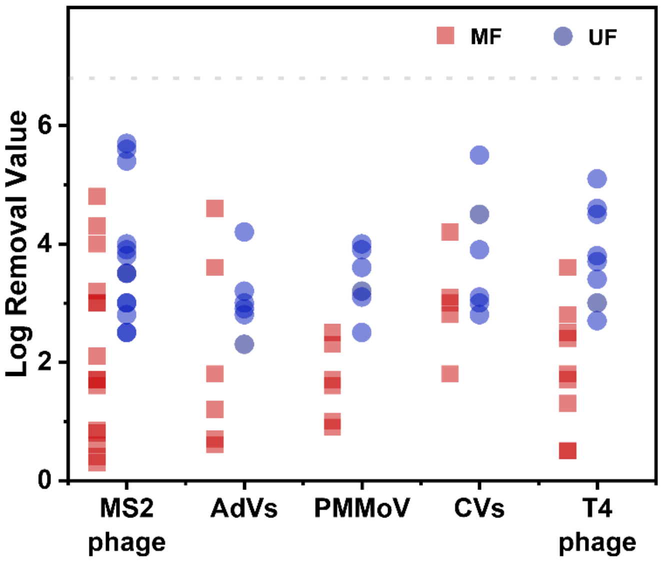


Figure 4. The removal efficiency of different viruses by MF and UF (specific data is extracted from Table S2).

To directly understand the effectiveness of MF or UF as viral barriers in centralized DWTPs, we compiled their removal performances for different viruses in the DWTPs and the laboratory pilot. As shown in **Figure 4**, the removal efficiency of MS2 phage is significant, which were 2.0±1.3 log for MF and 3.8±1.1 log for UF, respectively.

The membrane unit, as the core of the filtration system, is considered to achieve separation through strict size exclusion effects (**Figure 5a**). Therefore, it is generally assumed that membranes cannot retain substances smaller than their pore size. Based on this, previous studies have proposed estimation formulas for the theoretical retention rates of viruses by membrane processes, i.e., Equation 1,[73] and the logarithmic removal value determined by experimental is calculated using Equation 2:

$$\mathrm{R} = 1 - (1-\lambda)^2 \cdot [2 - (1-\lambda)^2] \cdot (K_s/2K_t) \tag{1}$$

$$\mathrm{LRV} = -\log_{10}(C_{\mathrm{effluent}}/C_{\mathrm{influent}}) \tag{2}$$

Where R is the theoretical retention rate (0 < R < 1) of the membrane for viruses within the diameter range of the pore size; λ is the ratio of virus diameter to membrane pore size (i.e. $d_{\mathrm{virus}}/d_{\mathrm{pore}}$); K_s and K_t are the hydrodynamic coefficients, specific calculations was described in the reference;[74, 75] LRV is the logarithmic retention value; C_{influent} and C_{effluent} are the virus concentration in the influent and effluent water, respectively.

However, numerous research cases indicate that the actual removal rate of viruses by membrane processes is significantly higher than the theoretical retention rate based on size exclusion effects. Specifically, for membranes with pore sizes of 50 nm and 100 nm, the theoretical retention rates for MS2 phages are 0.44 log and 0.11 log, respectively (specific calculations are shown in *Supporting Information*). However, during the laboratory membrane separation process, the observed removal rates are as high as 3.54 log and 1.79 log, respectively.[76] This phenomenon suggests the possible existence of alternative mechanisms for virus removal via membrane processes.

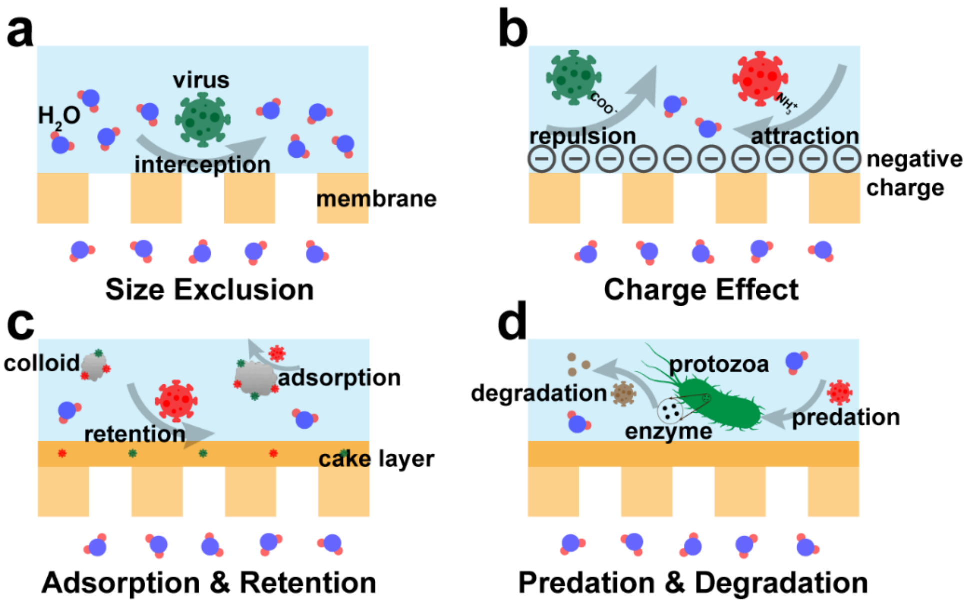


Figure 5. Mechanisms of pathogenic virus removal by membrane processes under various utilization situations (draw according to the content of references).[48, 77, 78]

The charge characteristics of both the virus and the membrane surface play a significant role in enhancing the removal rate.[16, 17] It is well known that the majority of viruses carry a negative charge, such as the MS2 phages.[79] And the properties of materials determine the charge of membrane surface, for instance, polyvinylidene difluoride membranes exhibit a significant negative charge.[77] The attraction or repulsion forces between the charges on viruses and membrane surfaces can significantly prevent viruses from entering the permeate, thereby enhancing the virus removal capacity of the membrane (**Figure 5b**). Furthermore, the colloidal-like properties exhibited by proteins enable effective adsorption of viruses within the membrane pores, making an indispensable contribution to virus removal by the membrane.

3.2 Membrane Bioreactor

When membrane units are employed in WWTPs, the typical influent for MBR is the effluent from sedimentation tanks or a mixture of activated sludge. In this situation, due to the presence of a significant amount of colloids, particulates, organic matter, protozoa, and metazoa in the influent, a relatively thick gelatinous cake layer tends to form on the membrane surface.[80] Despite the cake layer may weaken the contribution of the charge effect, the overall virus removal capacity of the MBR is not reduced. As shown in **Figure 6**, the removal capacity of MBR is even superior to that of MF and UF with the same pore size.

Although MBR exhibits significant virus removal performance, it is highly dependent on the formation of a cake layer. For instance, at a WWTP in Michigan, the membrane unit (hollow fiber, PVDF, 450 nm) initially exhibited a removal efficiency of only 0.2 log for adenovirus (~75 nm) during the early filtration stage. However, approximately one week later, this efficiency increased to around 6.3 log.[81] During this process, the dynamic cake layer formed by colloids, particulates and microorganisms in the wastewater provides two distinct pathways for the removal of viruses, i.e., (1) adsorption and retention by the cake layer, and (2) predation and degradation by microorganisms (**Figure 5c&5d**).

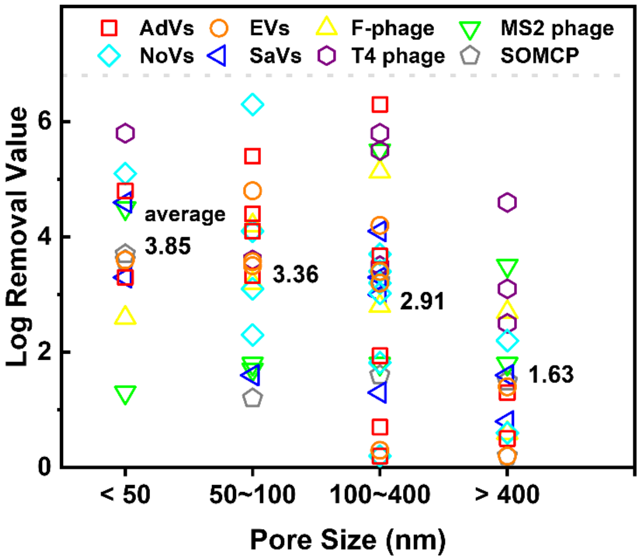


Figure 6. The virus removal efficiency of membrane units with different pore sizes in pilot and full-scale MBR processes (specific data is extracted from Table 1 and Table S2).

This indicates that the contribution of the cake layer (including colloids and microbes) in the MBR system to virus removal is crucial. Especially when the size of the virus is smaller than the membrane pore, the contribution of the cake layer becomes even more significant. Specifically, in a study conducted by Rabia Chaudhry, the removal rate of φX174 phage increased gradually in three MBR experiments: a clean membrane, a membrane with a cake layer, and a membrane system containing sludge, measuring 0.4 log, 1.1 log, and 2.3 log, respectively.[80] Therefore, parameters related to the cake layer and sludge in wastewater treatment systems can influence the efficiency of virus removal, such as sludge retention time (SRT), hydraulic retention time (HRT), concentration of mixed liquor suspended solids (MLSS), aeration intensity, and influent carbon-to-nitrogen ratio.[14, 18] Meanwhile, the contribution of the membrane unit cannot be ignored either. As shown in **Figure 6**, as the pore size of the membrane unit decreases, the average virus removal efficiency of the MBR system significantly increases. The membrane unit provides the MBR system with a strong size exclusion effect, including (1) retention of viruses and (2) retention of sludge. This ensures that the MBR system can achieve a stable and efficient removal state more quickly, thereby keeping the virus concentration in the effluent within a safe range.

3.3 Nanofiltration and Reverse Osmosis

As environmental hygiene deteriorates, the requirements for addressing viral risks in drinking water have become more stringent. Taking rotavirus as an example, the WHO recommends in its drinking water quality guidelines that the number of viruses per liter of drinking water must not exceed 10.[39] Therefore, even though the virus content in the permeate from MF and UF is already very low, further treatment with NF and RO is not redundant. For these two membrane processes, where the MWCO is much smaller than molecular of virus particles, their virus removal efficiencies range from 2.8~6.1 log for NF and 3.0~7.9 log for RO, respectively (**Figure 7**).

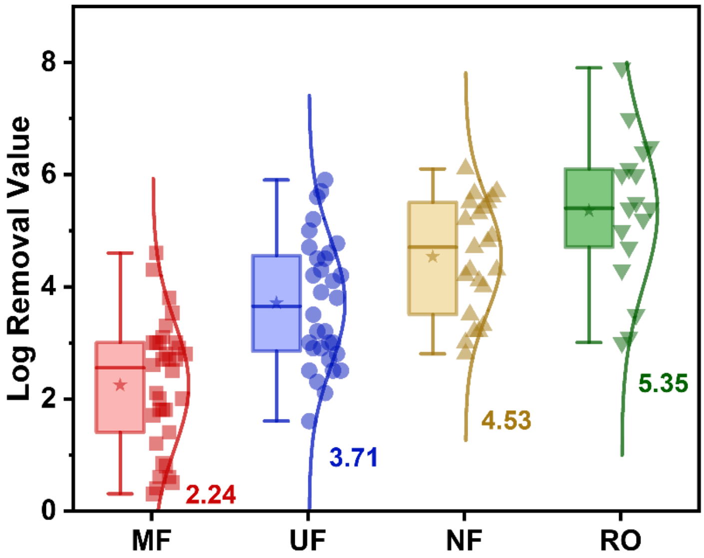


Figure 7. The virus removal efficiency by MF, UF, NF, and RO (colored numbers represent the averages, and specific data is extracted from Table S2).

Certainly, as we have observed, membrane processes always exhibit a range of fluctuating values for virus removal, even though NF and RO can theoretically retain viruses completely. Researchers believe that the primary reason for viruses passing through NF and RO membranes is defects in the membrane structure caused by unexpected issues during preparation and utilization.[13] The most crucial aspects among these are the pore size, the distribution of membrane pores, and its integrity, as the size exclusion effect represents the fundamental characteristic of membrane separation processes.

In practical engineering, defects in the installation of membrane units can directly affect the integrity of the filtration system, such as compromised O-rings and broken mechanical seals.[13] The worse the integrity of membrane system, the higher the probability of viruses passing through it. And operation conditions can also significantly influence the efficacy of virus removal, including crossflow velocity, transmembrane pressure, and cleaning strategies. In a membrane system, crossflow velocity can mitigate the negative impact of concentration polarization, thereby reducing the probability of viruses passing through the membrane pores.[82, 83] Conversely, increased transmembrane pressure accelerates the accumulation of viruses on the membrane surface and may potentially lead to membrane stretching, which can damage its structure. Gemunu Herath conducted ceramic membrane filtration experiments to quantify the impact of crossflow velocity and transmembrane pressure on virus removal rates.[84] Under constant pressure (40 kPa), as the velocity increased from 0.85 m s^{-1} to 2.45 m s^{-1}, the removal rate of Qβ phage increased from 25% to 75%. In contrast, under constant velocity (1 m s^{-1}), as the pressure increased from 40 kPa to 120 kPa, the removal rate decreased from 30% to 5%.[84] Inappropriate cleaning strategies can result in membrane aging, such as those involving acid, alkali, and chlorine cleaning.[85] During the cleaning process, the hydrophobicity and surface charge of the membrane may be altered. Oxidative cleaning can disrupt the composite layer on the membrane surface, leading to a loss of filtration effectiveness and deterioration of the permeate water quality.[86, 87] Furthermore, repeated pressurization can also lead to a decline in the performance of membrane separation. For instance, a study indicated that after undergoing ten thousand pressurization cycles, the removal efficiency of φX174 phage by RO membranes decreased from approximately 4.0 log to 1.8 log.[83]

4. Membrane technology for decentralized water treatment systems

Although MBR systems can be applied to both centralized and decentralized WWTPs, it is regrettable that effective technologies such as MF, UF, NF, and RO used in urban centralized DWTPs are challenging to apply in rural decentralized DWTPs. The primary issues are economic costs. Due to the long distances between rural settlements, the cost of installing water supply pipelines is extremely high. And the complex geological environments in rural areas intensify the difficulties in constructing pre-treatment facilities.[24, 25] Additionally, the operation and maintenance of membrane processes also pose challenges for residents. All these factors limit the promotion of centralized drinking water treatment processes in rural areas. In other words, decentralized DWTPs require a low-cost, easy-to-operate process with effective virus removal. In the past decade, with continuous improvements in MF and UF technologies by researchers and engineers, low-cost, low-maintenance membrane processes have gradually entered the household water purification phase. And typical point-of-use processes include ceramic membrane (MF) and gravity-driven membrane (UF).

Generally, commercial ceramic membranes are manufactured using alumina, and the primary configurations currently include tubular filters (**Figure 8a**) and pot filters (**Figure S1**). While the cost of ceramic materials may be slightly higher than that of polymer materials, the thermal stability, chemical stability, high mechanical strength, and low membrane fouling characteristics of ceramic membranes contribute to a longer life cycle, reducing the need for frequent replacements.[78] The advantage of low-maintenance makes ceramic membranes highly suitable for decentralized drinking water treatment. In theory, ceramic membranes, as a form of MF-level process, have limited virus retention capabilities. However, based on numerous practical cases, ceramic membranes have demonstrated significant effectiveness in virus removal. During the production process, raw materials often incorporate dopants such as silver nitrate, iron oxide, and copper nitrate.[88] These dopants provide additional virus inactivation pathways, thereby enhancing the performance of ceramic membranes in virus removal. For example, a pot ceramic membrane filter widely used in Cambodia can achieve a removal rate of 3.5 to 4.8 log for MS2 phage in the first 100 liters of water during challenge testing, with an average subsequent removal rate of approximately 1.2 log.[47] Moreover, in laboratory-scale tests, tubular ceramic membranes achieved a removal rate of 6.7 log or even higher for MS2 phage.[29] It is praiseworthy that these ceramic filters, priced at less than $10, provide a robust barrier for the health of local residents.[47]

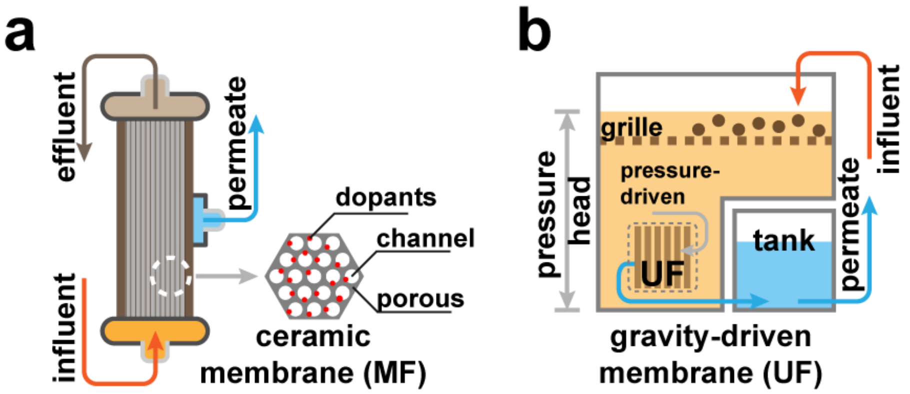


Figure 8. The schematic diagrams of typical low-maintenance processes suitable for decentralized drinking water treatment in rural areas; **(a)** ceramic membrane microfiltration, **(b)** gravity-driven membrane ultrafiltration.

Similar to ceramic membrane microfiltration, the gravity-driven membrane (GDM) developed by the Eawag team also offers the advantages of low-cost and low-maintenance.[33] In principle, even pot ceramic filters are membrane separation systems driven by gravity. The GDM process using UF membranes as the filtration unit was first reported in 2010.[89] It can also be described as a biofilm-controlled UF process, because the decomposition of organic fouling by microorganisms in the cake layer maintains a dynamic balance between the resistance from the influent and the total resistance of the membrane system.[33] Due to the dead-end filtration approach in GDM process, its permeate flux is slightly lower than that of other membrane processes, usually around 3 to 8 L m^{-2} h^{-1}.[90] For a GDM process with a stable permeate flux of 5 L m^{-2} h^{-1}, only 0.5 m^2 of filtration area is needed to produce 60 L of suitable drinking water daily (**Figure S2**). This system is well-suited to meet the daily water needs of households with lower water demand. Most importantly, regardless of the influent water quality, the GDM process can always achieve a threshold of permeate flux and maintain stability over the long term without the need for cleaning. The low investment cost, low maintenance requirements, and low energy consumption of the GDM process perfectly address the challenges of decentralized drinking water supply. As a novel technology, although there are not many reported cases regarding the virus removal effectiveness of the GDM process, there is no doubt that the dual retention effects of UF membranes combined with the cake layer can provide a relatively effective virus removal performance. For example, in a study by Peter-Varbanets, the GDM process equipped with flat-sheet UF membranes achieved an approximately 4 log removal rate of MS2 phage after the formation of the cake layer.[90] And another GDM process using hollow fiber UF membranes also demonstrated a stable 4 log removal efficiency.[30] In other words, the GDM process is reliable in addressing pathogenic virus risks.

Up to now, these two simple and effective membrane processes have been applied in several countries, including Kenya, Nepal, and Uganda, among others.[31, 32, 90] Both ceramic membrane and gravity-driven membrane are recommended by the WHO for point-of-use drinking water treatment, especially when the quality of the water source is poor.

5. Advanced membrane technologies for pathogenic virus removal

In contrast to MF, UF, NF, and RO, which primarily rely on smaller pore sizes or lower MWCOs to intercept and retain viruses as physical barriers, several new membrane technologies have shown excellent potential for virus removal. These can be categorized into (1) membrane distillation, which inactivates viruses through high temperatures; (2) membrane-based photocatalytic, which inactivates viruses through oxidation reaction of free radicals, and (3) membrane-based electrosorption, which capture viruses through Coulombic force. All of these provide a more direct pathway for virus removal based on the size exclusion effect of the membrane unit.

For membrane distillation (MD), it is a membrane process driven by the vapor pressure gradient. In the operation of the MD system, water from the high-temperature impure feed stream passes through the membrane pores in the form of water vapor, liquefies in the lower-temperature distillate stream, and flows out to achieve the separation of water and solutes (**Figure 9a**). This dictates that MD systems require the use of hydrophobic membrane units.[91] Hydrophobic membranes have a stronger virus retention capacity compared to hydrophilic ones used in conventional membrane filtration processes, allowing MD system to retain viruses on the feed stream side. Moreover, the high-temperature conditions during MD system operation lead to irreversible structural damage to viruses, providing dual assurance for virus removal through a combination of membrane retention and thermal inactivation. According to the research by Mukta Hardikar, the MD system equipped with a nominal pore size of 450 nm PTFE membrane demonstrates an increased removal rate of MS2 phages with the rise of feed stream temperature. At 45°C, 55°C, and 65°C, the removal rates can reach 2 log, 4 log, and 6 log, respectively.[91] This implies that at typical MD operating temperatures (greater than 65 °C), the virus removal rate is appreciable, even when the size of membrane pores is much larger than that of the virus. In addition, compared to boiling, the energy consumption of MD system is slightly lower, as it can utilize low-grade waste heat to address viral risks in water environments.

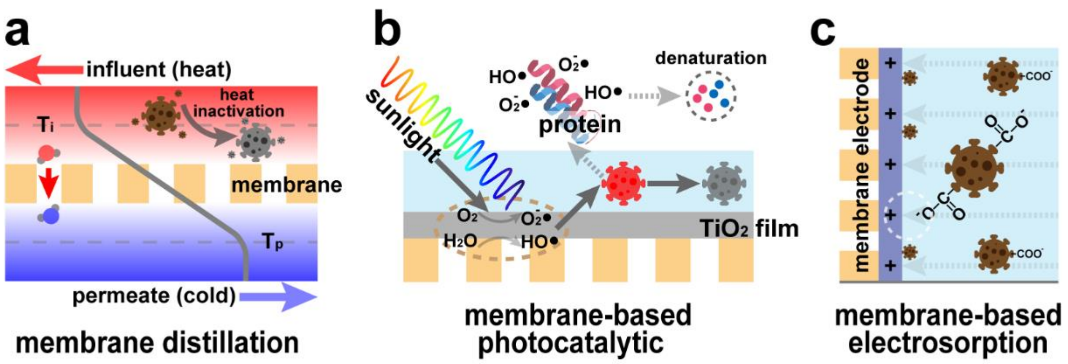


Figure 10. The schematic diagrams of advanced membrane technologies; **(a)** membrane distillation, **(b)** membrane-based photocatalytic, **(c)** membrane-based electrosorption.

Among various novel material composite membranes, photocatalytic membranes are one of the outstanding designs. In the context of frequent virus outbreaks, photocatalytic materials serve as an effective alternative to traditional chlorine disinfection, as they do not produce disinfection by-products and can be reused.[92] Photocatalytic membranes possess the advantages of being environmentally friendly, efficient, and sustainable. Currently, titanium dioxide (TiO_2) is the most extensively studied photocatalytic material. The composite membranes can be easily produced by loading TiO_2 nanoparticles onto the surface of commercial membranes to form a coated film (**Figure 9b**). Researchers believe that virus removal mechanisms of TiO_2 composite membranes include (1) size exclusion of membrane pores, (2) adsorption of TiO_2 nanoparticles, and (3) oxidation reactions of free radicals.[92, 93] It is note that TiO_2 undergoes a photocatalytic reaction under sunlight irradiation, generating reactive oxygen species (ROS) such as hydroxyl radicals ($OH-$) and superoxide radicals ($O_2\cdot$). These ROS can induce protein denaturation, capsid degradation, genome damage in viruses, leading to their complete inactivation.[93] Specifically, in a study conducted by Inna Horovitz, under simulated sunlight irradiation, a nominal 800 nm pore size flat Al_2O_3 membrane with TiO_2 coating could achieved a removal rate of approximately 4.9 log for MS2 phages.[94]

Membrane-based electrosorption also provides an alternative strategy for the removal of viruses in water environments. Although typical membrane electrode technologies, such as capacitive deionization (CDI), are commonly used for the removal of ionic contaminants, they also exhibit good capture capabilities for charged viruses.[95] As shown in **Figure 9c**, under the action of Coulombic force, the viruses are firmly adsorbed onto the electrode until saturation is reached. Compared to dead-end filtration, in the charging-discharging cycle of the membrane electrode, viruses are desorbed during the discharging phase and enter the concentrated stream, thereby completely leaving the membrane filtration system. The periodic desorption process prevents the accumulation of viruses on the membrane surface, reducing the possibility of viruses entering the permeate. Most importantly, electrosorption exhibits a strong ability to remove viruses. For instance, research conducted by Rebecca Gordon indicates that under low concentrations of PBS solution conditions, CDI achieves a removal rate of approximately 4.5 log for MS2 phages.[95]

In addition, there are numerous other membrane separation processes for virus removal, which can be roughly categorized as (1) improved processes based on MF, UF, NF, and RO; and (2) improved processes based on the advanced membrane technologies. For the former, it primarily involves enhancing virus adsorption and retention on membranes by loading materials with a high specific surface area on the membrane, such as activated carbon, graphene, carbon nanotubes, etc. For the latter, the focus is on addressing the challenges that these advanced membrane technologies face during operation. For example, wetting, scaling, and fouling can affect the performance of membrane distillation; turbidity and the wavelength of light can influence photocatalytic reactions; and the pH and ionic strength of the influent can impact the electrosorption process. In summary, although these advanced membrane processes still face a series of challenges in practical applications, their excellent virus removal capabilities provide more strategies for mitigating virus risks in water environments.

6. Conclusion and outlook

When addressing the hazards of pathogenic viruses in water environments, membrane separation technology stands out as a direct, efficient, and reliable treatment method for virus removal. It allows for flexible selection of MF, UF, NF, and RO modules as virus removal units, catering to the specific needs of centralized or decentralized, drinking water or wastewater treatment. Whether employed in centralized drinking water treatment through direct membrane filtration combined with pre-treatment, utilized in decentralized drinking water treatment through ceramic microfiltration or gravity-driven ultrafiltration, or applied in wastewater treatment through membrane bioreactor, these processes consistently achieve efficient virus removal. The mechanisms include size exclusion, charge effects, adsorption and retention by the cake layer, as well as predation and decomposition by microorganisms. All these works together to intercept viruses on the influent side.

Despite being one of the most effective virus removal strategies, these membrane processes still have limitations. Various factors can influence the actual virus retention efficiency of membrane processes, including the membrane integrity, pore size and distribution of the membrane, the pH and ionic strength of the influent, and the transmembrane pressure and stream flow rate during process operation. Furthermore, pressure-driven membrane processes can retain viruses but cannot completely inactivate them. This could result in the intrusion of viruses into bacteria or other microorganisms, releasing small molecular toxic substances and more viruses. Additionally, whether in MF and UF or NF and RO, the entry of viruses into the permeate through membrane units is inevitable. While these membranes can effectively retain viruses, they are not absolute barriers, even when the size of the viruses is significantly larger than the membrane pores. It is necessary to gain a deeper understanding of the process of virus transport through membrane pores and to clearly delineate improvement strategies for membrane processes.

Certainly, in the face of the threat posed by pathogenic viruses, there is much work to be done. Firstly, in developing countries where the impact of virus outbreaks is most severe, the design, application, and promotion of membrane processes are crucial. They require simpler, cheaper, and more effective membrane technologies to ensure the safety of drinking water. Since membrane processes typically only retain viruses and lack the capability to inactivate them, a significant number of live viruses may still persist in sludge and backwashing effluents. This could pose potential threats to the water environment, emphasizing the importance of comprehensive and harmless treatment throughout the entire process. Moreover, it is essential to prioritize the detection and monitoring of virus content in the influent and effluent of membrane processes. A robust early warning system can provide engineers with the enough time to maintain membrane process, residents to take emergency measures, and governments to make informed decisions. Ultimately, this ensures public health safety in water environments.

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