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Review

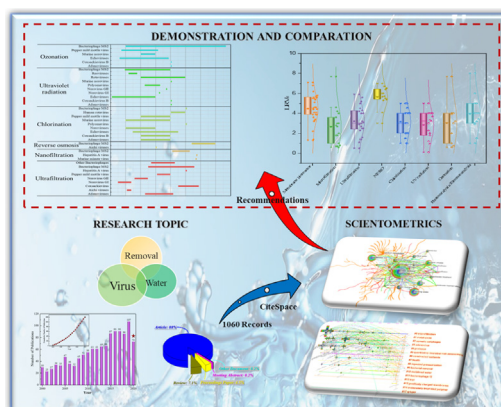
Comparative effectiveness of membrane technologies and disinfection methods for virus elimination in water: A review

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HIGHLIGHTS

- Research hotspots about virus removal in water are demonstrated.
- In-depth mechanisms on membrane and disinfection for virus removal are discussed.
- Virus removal efficiency in membrane and disinfection treatments is compared.
- Some perspectives and suggestions to eliminate viruses are proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

The pandemic of the 2019 novel coronavirus disease (COVID-19) has brought viruses into the public horizon. Since viruses can pose a threat to human health in a low concentration range, seeking efficient virus removal methods has been the research hotspots in the past few years. Herein, a total of 1060 research papers were collected from the Web of Science database to identify technological trends as well as the research status. Based on the analysis results, this review elaborates on the state-of-the-art of membrane filtration and disinfection technologies for the treatment of virus-containing wastewater and drinking water. The results evince that membrane and disinfection methods achieve a broad range of virus removal efficiency (0.5–7 log reduction values (LRVs) and 0.09–8 LRVs, respectively) that is attributable to the various interactions between membranes or disinfectants and viruses having different susceptibility in viral capsid protein and nucleic acid. Moreover, this review discusses the related challenges and potential of membrane and disinfection technologies for customized virus removal in order to prevent the dissemination of the waterborne diseases.

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Abbreviations of viruses

AdV	Adenovirus
HAdV	Human adenovirus
AiV	Aichi virus
AsV	Astrovirus
CV	Coxsackievirus
CVB	coxsackievirus B
EV	Enterovirus
ECHO	Echovirus
HA(E)V	Hepatitis A(E) virus
NoV	Norovirus
hNoV	Human norovirus
MNV	Murine norovirus
NV GI/II/IV	Norovirus genotype (I/II/IV)
PMMoV	Pepper mild mottle virus
PyV	Polyomavirus
JC/BK PyV	JC/BK polyomavirus
RV	Rotavirus
HRV	Human rotavirus
SaV	Sapovirus

1. Introduction

Global outbreaks of infectious diseases induced by viruses are escalating and seriously threatening human health. Viruses are noncellular biological entities with a simple structure composed of proteins and nucleic acids. Compared with bacteria and fungi, viruses have the characteristics of a small size, special structure, wide distribution, low infectious dose and strong pathogenicity (Xiao et al., 2013). It is well-documented that the global number of phages is estimated to be 4.80×10^{31} (Gumes et al., 2016), which is much higher than the number of organisms with a cellular structure.

According to epidemiological studies, human and animal excreta often contain numerous virus particles, which can enter the water environment via sewage discharge, septic-tank system filtrate, and runoff from agricultural areas (Rzezutka and Cook, 2004). For instance, human enteroviruses can be excreted at high concentrations (10^{11}

viruses/g-feces) from infected individuals and disseminated through the fecal-oral route (Haramoto et al., 2018). It is estimated that 2 to 12 million people die from waterborne diseases every year (US EPA, 2006a), and waterborne diseases can pose a substantial threat to the world public health security. The novel coronavirus (SARS-CoV-2) causing symptoms called COVID-19, causes respiratory infection, thus the major route of transmission is believed to be the inhalation of the virus. Still, the SARS-CoV-2 is also typified by fecal-oral transmission (Arslan et al., 2020), with important pollution sources from urban sewage and sewer overflow (Zhu et al., 2020). Wastewater treatment plants (WWTPs) with conventional procedures such as conventional activated sludge process serving as a significant barrier can essentially impede the spread of waterborne viruses to a certain degree (Sano et al., 2016), but more enhanced methods should be conducted to fulfill more rigorous standards.

At present, there is extensive research on pathogenic bacteria in wastewater treatment processes but relatively fewer studies on viruses. The occurrence, states of existence and decay of viruses are distinct from those of pathogenic bacteria. Hence, in this paper, we analyzed the research hotspots and development about virus removal in virus-containing wastewater/drinking water treatment in recent years. Subsequently, the membrane and disinfection technologies were reviewed along with a comprehensive evaluation and comparative analysis. Finally, we proposed some detailed conclusions regarding the removal effects and mechanisms of several common viruses by membrane and disinfection processes, along with the related challenges and comprehensive perspectives, to provide guidance for the further efficient elimination of waterborne viruses in future practical applications.

2. Research development of virus removal in water bodies

2.1. Overview of virus removal in water from Web of Science

To track the latest research progresses and hotspots, publications about virus removal in water were analyzed. A total of 1321 results were selected between 1st of Jan. 2000 and 16th of July 2021 from the core collection in web of science database with the topic “virus”, and the search terms were designed as (TS = (virus removal AND “water”)). Fig. 1(b) portrays that the field has received increasing attention over the past two decades, as the number of scientific articles has nearly tripled from 2007 to 2016 (Fig. 1(a)).

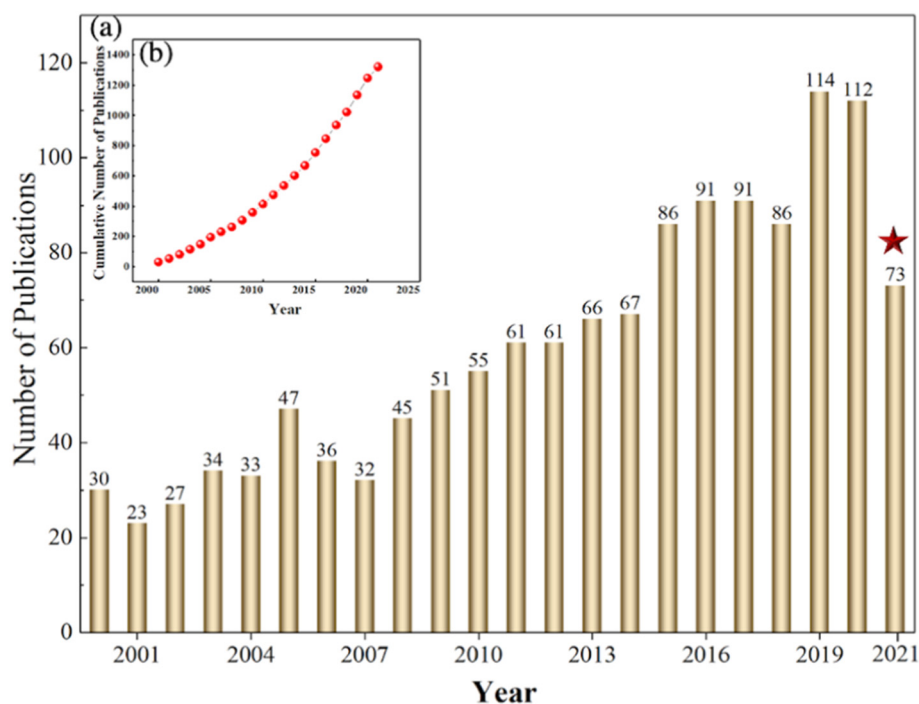


Fig. 1. (a) Annual distribution and (b) cumulative quantities of the studies on virus removal from water from the Web of Science. (All the documents were collected until July 16, 2021).

The global geographic distribution map of research production is visualized using ArcGis (as shown in Fig. 2) to identify the distribution of countries and institutions performing virus removal research in water, and the specific publication number in some countries as a function of year is displayed in Fig. S1. The research units are mainly distributed in North America (the United States and Canada), Europe (the Netherlands, France, Spain, etc.) and Asia (China, Japan, Korea, etc.). Typically, the USA has played a leading role during this period, generating 29.8% of the total publication number, followed by China and Japan, accounting for 7.2% and 6.02%, respectively, and more detailed data are shown in Table S1. In general, this field has been appealing to increasing attention.

2.2. Categories and elimination of viruses in water treatment

Viruses are tiny (10–100 nm) noncellular particles with nucleic acids (DNA and RNA) wrapped in a protein shell (Hata et al., 2011). Compared with other pathogens (i.e., bacteria and protozoa), viruses survive in the host for its lifetime in their natural state and retain their infectivity, making them difficult to be eliminated (Brooks et al., 2020). For example, AdVs, which have double-stranded DNA and are the abundant human viruses in WWTPs (Hata et al., 2013), are resistant to UV disinfection. Additionally, EVs, RVs, HAV and NoVs, etc., can survive in natural water with strong infectivity for more than one month (Brooks et al., 2020; WHO, 2011). Wastewater treatment is one of effective approaches to remove viruses, and the systematic diagram of virus control in water treatment is exhibited in Fig. 3. In the influent of WWTPs, most viruses are negatively charged because their isoelectric point (IEP) is lower than the pH of the influent (Michen and Graule, 2010), and the seasonal differences can also indirectly affect the virus distribution and their treatment effects (Pang et al., 2019; Nordgren et al., 2009), notably, viruses in untreated wastewater will contaminate surface water in periods of heavy rain and melting snow. Viruses usually exist in two states: one is exposed to various physical, chemical, and biological environments and the other is parasitic in host cells and coexists with microorganisms (Templeton et al., 2008). Some common viruses in water bodies and their elimination in WWTPs are tabulated

in Table 1. Virus removal efficiency is usually expressed as the log reduction values (LRVs).

From Table 1, we can observe that the removal efficiency of WWTPs for viruses exists discrepancies, and the overall removal (0.47–2.32 LRVs) is still incomplete when deploying conventional activated sludge or trickling filter as the treatment approach. Therefore, technical improvement of these processes or the introduction of some advanced treatment processes may be the effective measures to ensure the effluent quality in WWTPs as well as the safety of water quality, especially during the period of epidemic outbreak.

2.3. Keyword analysis of virus removal from water

Keyword analysis plays an important role in clarification of current hot topics in virus removal. The co-occurrence of the collected keywords in the integrated cluster view was analyzed using CiteSpace 5.6.R5 from the 1060 research articles (from 2000 to 2020) listed in Web of Science. After executing threshold configuration, the term in the Node Types function panel was selected, and Top 1.0% of most cited or occurred items from each slice as well as 100 of the maximum number of selected items per slice were designed. CiteSpace 5.6.R5's automatic clustering function was then utilized to picture cluster and timeline view figures after screening to remove duplicates. Fig. 4 depicts a co-occurring analysis of keywords and the trend of the evolution of keywords recommended by authors is illustrated in Fig. S2. For virus removal technologies and evaluation, the cluster titles are “microfiltration” and “quantitative microbial risk assessment”, manifesting that membrane filtration and the prevention of waterborne diseases have attracted more attention recently. Moreover, the title “inactivation” denotes that disinfection technology is a good choice for virus removal. According to the size of the nodes and the number of interleaving points, the keywords “virus removal”, “drinking water”, “wastewater treatment”, “membrane filtration”, “removal efficiency” and “disinfection” appear more frequently than others, with numbers of 171, 102, 73, 59, 44 and 39, respectively, and the relevant values are shown in Table S2. Additionally, the timeline shown in Fig. S2 embodies that research has been covering an increasing number of keywords over



Fig. 2. Global geographic distribution of research on virus removal from water (TS = (virus removal AND water)). The dots indicate the location of these 1060 literature research institutions, which are mapped by ArcGis according to the longitude and latitude of each research institution. The attribution is Source: Esri, Maxar, GeoEye, Earthstar Geographics, CNES/Airbus DS, USDA, USGS, AeroGRID, IGN, and the GIS User Community.

time and mainly rest with membrane filtration and disinfection technology, therefore, we picked these two methods as the review highlights.

3. Membrane and disinfection technologies for virus elimination

3.1. MBR process for virus removal

Secondary treatment covers microbial metabolism, dissolution of organic matter, and precipitation and separation of biological metabolites. Virus removal in the conventional activated sludge (CAS) process is at 0.3–3.1 LRVs (Taboada-Santos et al., 2020), which is 1 LRVs more removal than in primary treatment. Membrane bioreactors (MBRs), which integrate CAS with membrane filtration, have a relatively high virus removal capacity, reaching 1.4 to 6.8 LRVs (Simmons and Xagorarakis, 2011; Sano et al., 2016) so that they are widely used as an advanced treatment technology.

In general, the removal efficacy of the MBR mainly relies on the following four mechanisms (Chaudhry et al., 2015b; Hai et al., 2014): i) the

adsorption of viruses to sludge particles. Viruses existing as aggregates can attach to sludge particles which consist of many bacteria and organic compounds that are larger than the pore size of membranes, rendering smaller probability of viruses' passing; ii) membrane interception. During membrane separation process, viruses that are larger than membrane pores or have the same electric charge as membrane can be validly intercepted; iii) adsorption and retention of the cake layer and gel layer on the membrane surface. Membrane fouling, including reversible and irreversible fouling, plays an important role in virus removal (Marti et al., 2011). The fouling caused by activated sludge on the membrane surface forms a dynamic filter layer composed of a cake layer and a gel layer that changes the separation performance of the membrane (Marti et al., 2011); iv) decay and inactivation. Proteases in enzymatic catalysis will break the viral capsid protein to inhibit viability of viruses. Besides, the virus decay by the predation by other microorganisms will also increase the virus removal. Normally, the membrane interception together with the adsorption and retention caused by the cake/gel layer accounts for over 50% of the total removal when the 0.04 μm nominal pore size of membrane module was used in MBR, otherwise the

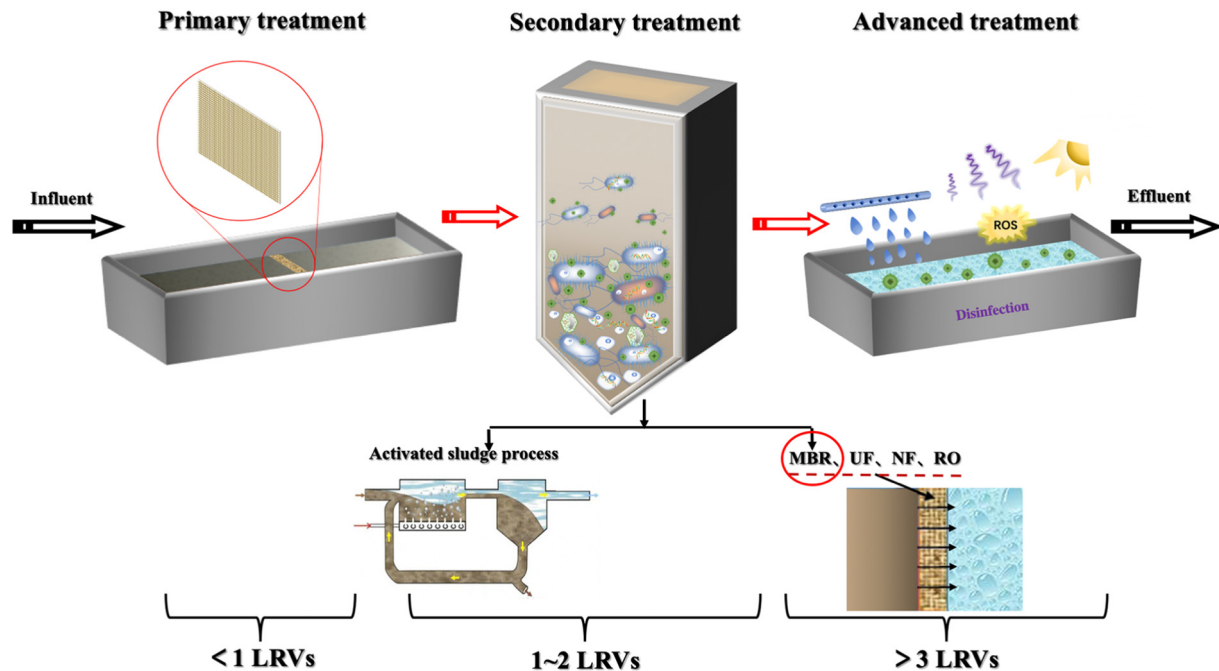


Fig. 3. Schematic diagram and relative removal efficiency of the system containing primary treatment, secondary treatment and advanced treatment used for virus control in wastewater treatment.

virus removal was mainly caused by cake layer and biomass considering the membrane module is 0.4 μm nominal pore size (Chaudhry et al., 2015b; Shang et al., 2005; Wu et al., 2010). The scheme of the MBR removal mechanisms is revealed in Fig. S3.

Membrane modules of MBRs with different pore sizes show no obvious variation in retention of the same virus (Hirani et al., 2010), while the removal efficiency of MBR systems varies for different viruses (Purnell et al., 2016). Presumably, besides mechanical sieving action, the biofilm and sludge particles attached to the membrane surface also play a certain role in virus removal in MBRs (O'Brien and Xagorarakis, 2020). These mechanisms are meanwhile influenced by the surface properties of viruses, such as the electrostatic charge and hydrophobicity of the viral capsid protein (Armanious et al., 2016). Miura et al. (2015) evaluated the removal performance of a MBR system and found that the contents of NV GII and parvovirus in solid phase were equal to or higher than in liquid phase, while EVs could not be detected in the solid phase, indicating that the adhesion between EVs and sludge was not strong. A similar conclusion was also drawn in the research of Sima et al. (2011), they discovered that NoVs were more efficiently removed than SaVs in full- and pilot-scale MBR plants, although they have a similar structure, morphology and size.

In addition, water quality parameters, including pH, temperature, suspended solids concentration, particle size distribution, dissolved organic concentration, viscosity, etc., can impact viruses' behavior is decided by viral protein (such as hydrophobic and charged regions) in aqueous medium (Prado et al., 2019a). Operational changes in MBRs have affected virus elimination owing to changing the composition of microbial community and membrane surfaces. For example, a longer hydraulic retention time (HRT) can enhance the adsorption of viruses by sludge particles, thereby improving the virus removal efficiency (Prado et al., 2019a). High sludge retention times (SRTs) can endow MBR facilities with high mixed liquor suspended solids (MLSS) concentration and abundant microbial community to produce nitrified effluents so that increasing removal performance (Hirani et al., 2014). The addition of powdered activated carbon or polymer flocculant has been verified to effectively alleviate the blockage of MBR membrane pores and improve the removal of bacteria, antibiotic resistance genes and viruses (Ravindran et al., 2009; Nnadozie et al., 2017). Besides, Delanka-

Pedige et al. (2020) proposed a low-energy algae wastewater treatment system to substitute for MBRs that can be translated to lower disinfection by-products (DBPs) formation while reducing pathogens via virus inactivation by algae. Therefore, it is more reasonable to optimize the design of MBR for further applications.

The pore size of membrane module used in MBRs normally ranges from 0.1 to 0.4 μm which is far beyond the size of most viruses (usually at nanometer), so the virus removal presented in MBRs predominantly rests on adsorption and aggregation processes by biofilm and cake layer instead of size exclusion. Herein, it would be prudent to elucidate the law of virus diffusion and adsorption and explore the interaction between viruses and different membrane foulant layers (Zhu et al., 2021).

3.2. Virus separation by membrane technology

Membrane technology, including mainly microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), is often used as an advanced treatment. The membrane virus retention mechanisms (illuminated pictorially in Fig. 5) are usually divided into four types (Duek et al., 2012; Gentile et al., 2018; Goswami and Pugazhenth, 2020): mechanical screening, electrostatic interactions, adsorption retention, and hydrophilic and hydrophobic interactions. Mechanical sieving, i.e., size exclusion, basically occurs on the membrane surface; electrostatic interactions are associated with the charge of viruses and the membrane; adsorption retention as well as hydrophilic-hydrophobic interactions are ascribed to physicochemical properties of the viruses and membranes themselves, which allow viruses to penetrate the membrane surface to be deposited on the membrane internal matrix. The size of most viruses is normally smaller than the pore size of MF membranes but larger than that of NF membranes and RO membranes, in this condition, size exclusion plays a decisive role in virus removal. While the pore size of UF membranes is comparable to virus particle size, the virus removal efficiency depends on both the surface properties of viruses and membrane. Specifically, the removal effect of MF for viruses is approximately 0.3–2.2 LRVs (Qiu et al., 2015). In UF, removal of NoV, AdV and RV is greater than 3 LRVs (Qiu et al., 2015), while NF and RO can normally achieve greater than 5 or 6 LRVs, respectively (Cetlin et al., 2018). Furthermore, the external

Table 1
Categories and the corresponding diseases caused by waterborne viruses as well as quantitative virus reduction in WWTPs.

Viruses	Genome	Dimension (nm)	Major diseases	Influent	Effluent	Virus reduction (\log_{10})	Technologies	Detection methods	References
Enteroviruses	ssRNA	20–100 (Guo and Hu, 2011)	Minor febrile illness, gastroenteritis, aseptic meningitis, paralysis, myocarditis (Iaconelli et al., 2017)	3.3×10^7 GC/mL	7.6×10^6 GC/mL	0.63	Italy; grid separation, primary sedimentation, secondary bio-logical treatment and disinfection	RT-PCR, Real-time qPCR	(Rosa et al., 2010)
Coxsackieviruses				3.24×10^5 copies/L	1.54×10^3 copies/L	2.32	Arizona, United States; activated sludge and trickling filter	RT-PCR	(Kitajima et al., 2015)
Astroviruses	ssRNA	25–35 (Jacukowicz and Domanska-Blicharz, 2017)	Gastroenteritis (Vu et al., 2019)	NG	2.69×10^3 copies/L	–	France; primary decantation and biological secondary treatment From May 2013 to May 2014	RT-qPCR	(Prevost et al., 2015)
Pepper mild mottle virus	ssRNA	–	Infections to solanaceous plants, mottled or yellow and green floral leaves on plants, malformation or bump spots on fruits	$3.7\text{--}4.4 \times 10^6/3.2\text{--}9.4 \times 10^6$ copies/L	$4.6\text{--}6.3 \times 10^5$ copies/L	$0.76\text{--}0.99/1.8 \pm 0.2$	Southern Arizona, USA; Plant A (conventional activated sludge process); Plant B (biological trickling filter process)	TaqMan-based qPCR	(Kitajima et al., 2014)
Norovirus genotypes GI/GII	ssRNA	25–40 (Cheetham et al., 2006)	Acute gastroenteritis (evacuation, vomiting, fever, abdominal pain) (Teixeira et al., 2016)	8.8×10^4 GC/L	3×10^4 GC/L	0.47	North Wales, UK; WWTP with filter beds for secondary treatment and serves ca. 4000 inhabitants	RT-qPCR	(Farkas et al., 2018)
Sapoviruses	ssRNA	25–40 (Cheetham et al., 2006)		7.8×10^6 GC/L	NG	–	New Caledonia; sample collected from April 2012 to March 2013	RT-qPCR	(Kaas et al., 2016)
Rotaviruses	dsRNA	55 (double-capsid) 70 (single-capsid) (Saif et al., 1980)	Gastroenteritis, diarrhea (especially for young children) (Banyai et al., 2018)	1.2×10^5 GC/L	2.6×10^4 GC/L	0.66	Eastern Cape, South Africa; activated sludge system with 40,000 m ³ /day flow rate	Quantitative TaqMan real-time PCR	(Osuolale and Okoh, 2017)
Adenoviruses	dsDNA	75–90 (Needle et al., 2019; San Martin and Burnett, 2003)	Respiratory disease, gastroenteritis, pneumonia, urinary disease, conjunctivitis, hepatitis, myocarditis, encephalitis (Iaconelli et al., 2017)	$4.3 \times 10^5\text{--}8.7 \times 10^6$ GC/mL	$1.22 \times 10^4\text{--}3.7 \times 10^6$ GC/mL	–	Egypt; 330,000 m ³ /day capacity	Real time PCR	(Elmahdy et al., 2019)
Aichi viruses	ssRNA	30 (Burutaran et al., 2015)	Acute gastroenteritis	$9.7 \times 10^4/2.0 \times 10^6$ copies/L	$1.1 \times 10^4/2.0 \times 10^5$ copies/L	0.94–0.99	Southern Arizona, USA; conventional activated sludge process/biological trickling filter process	TaqMan-based qPCR	(Cheetham et al., 2006)
Hepatitis A virus	ssRNA	27–30 (Feinstone et al., 1973)	Sporadic hepatitis (Iaconelli et al., 2017)	$2.01 \times 10^3\text{--}8.39 \times 10^3$ copies/L	$1.93 \times 10^3\text{--}8.70 \times 10^3$ copies/L	–	Kampala, Uganda; conventional activated sludge method, in summer 2016	qPCR and quantitative RT-PCR	(O'Brien et al., 2017)
Polyomaviruses	dsDNA	40 (Wen et al., 2004)	Malignancies, cancer (skin, prostate, colorectal) (Ugo, 2018)	3.9×10^5 GC/L	4.51×10^3 GC/L	1.93	Greater Cairo, Egypt; activated sludge as secondary treatment process with 600,000 m ³ /day	Real time PCR	(Hamza and Hamza, 2018)
SARS-CoV	ssRNA	80–120	Respiratory disease, lung/liver/kidney injury, multiorgan dysfunction, shock, metabolic acidosis (Li et al., 2020)	NG	2.4×10^3 copies/L	–	Japan; Conventional activated sludge process	RT-qPCR	(Haramoto et al., 2020)

Notes for abbreviations: NG: not given, GC/L: genome copies/L, ssRNA: single-stranded RNA, dsRNA: double-stranded RNA, ssDNA: single-stranded DNA, dsDNA: double-stranded DNA, qPCR: quantitative polymerase chain reaction, RT-(q)PCR: reverse transcriptase-(quantitative) polymerase chain reaction.

3.2.1. Membrane modification

Membranes have different removal effects on different viruses. Fig. S4 shows the discrepancy in virus removal between MF and UF. This distinction depends on the pore size of the membrane, size and structure of the virus and electrostatic interactions (Gentile et al., 2018). The pore size of the membrane displays a distribution range. Different membrane modules with similar average pore sizes may have different rejection rates for the same virus, and the more uneven the pore size distribution of the membrane, the lower the retention rate of viruses (Fallahianbijan et al., 2017). Madaeni (1999) used a 0.2 μm MF membrane with a retention efficiency of 2 LRVs for poliovirus. However, Herath et al. (1999) used a hydrophilic membrane with a smaller pore size of 0.05 μm but found only a 0.2 to 0.7 LRVs removal of the bacteriophage Q β , which is almost same size as poliovirus.

3.2.1.1. Reinforcing interaction. Viruses can easily be adsorbed on the membrane surface or within its pores (Michen and Graule, 2010), and the adsorption behavior varies according to membrane material (Goswami and Pugazhenth, 2020). Meanwhile, electrostatic and hydrophobic interactions between viruses and membrane surfaces can increase the removal efficiency, which often happens in modified membranes by introducing functionalized materials, such as the functionalization of the yttria-stabilized zirconia capillary membranes with n-hexyltriethoxysilane and n-octyltriethoxysilane, for virus removal based on hydrophobic and electrostatic adsorption (Larson et al., 2018; Bartels et al., 2019) (as shown in Fig. 6(a)). The structure and removal effects of the modified composite membranes used in existing research for virus separation are tabulated in Table 2. UF membrane grafted with a zwitterionic polymer hydrogel enhances the electrostatic repulsion between viruses and the membrane surface to remarkably facilitate the removal of viruses, which can increase 4 LRV in HAdV and 3 LRV in MS2 higher removal than the unmodified membrane (Lu et al., 2017) (as portrayed in Fig. 6(b)).

Using terephthalaldehyde (TA) as a cross-linking agent, polyethyleneimine (PEI) was assembled layer by layer on the surface of a polyethersulfone (PES) MF membrane to form a positively charged membrane that can trap and inactivate viruses and eventually achieved 4.5–5 LRVs of virus reduction (Sinclair et al., 2019) (as shown in Fig. 7(a)). What's more, self-assembled block polymer membranes functionalized with macromolecular template capacitate highly selective separations (as shown in Fig. 7(b)). Gu et al. (2018) fabricated a highly porous and permeable functionalized monoliths with well-defined pore structures for the separation of large molecular virus particles, and a self-standing composite membrane with regular mesoporous

has been designed, which realized highly size-selective permeation of biomolecules at the nanometer-level (Yamauchi and Kimura, 2013).

The current materials used in fabrication methods for membranes are primarily based on empirical approaches and lack molecular-level design (Werber et al., 2016). The introduction of nanoparticles has been shown to reinforce the structural integrity and hydrophilicity of the membrane as well as the electrostatic interaction between the membrane and viruses to increase the water flux while improving the virus removal efficiency (Khin et al., 2012). Introducing metal nanoparticles (Liu et al., 2019), carbon nanotubes (Németh et al., 2019) and liquid crystals (Kuo et al., 2020) into the membrane or onto the membrane surface is currently an effective strategy. Y₂O₃ nanoparticles have the merits of a high isoelectric point (Zhao et al., 2018), large specific surface area (Zhang et al., 2019b), and multiple active surface sites (Meng et al., 2016). The positively charged plum-shaped SiO₂-Y₂O₃ composite nanofiber membrane shown in Fig. 7(c) has a high adsorption capacity of 87.96 mg/cm³ for viruses and can reach 5 LRVs (Liu et al., 2019). Loading AgNPs onto a polysulfone membrane also leads to a significant improvement in virus removal (Zodrow et al., 2009). In addition, a novel microporous nano-MgO-diatomite ceramic membrane has been prepared, which may provide a feasible method based on the strongly positive charge for virus removal (Meng et al., 2016). Moreover, a new type of composite film, in which nanoparticles of copper oxide, titanium oxide, and iron oxide are coated on a polytetrafluoroethylene membrane with an inorganic composite-based multiwalled carbon nanotube (MWCNT) carrier, has been introduced to remove viruses efficiently by increasing the electrostatic interaction between the membrane and viruses (Németh et al., 2019).

3.2.1.2. Enhancing precise screening. Meanwhile, a nanostructured liquid-crystalline (LC) membrane has received widespread attention due to its ability to form ordered nanostructures via a self-organization process (Marets et al., 2017). As portrayed in Fig. S5, the nanostructures can be classified as cubic, columnar and layered shapes (Marets et al., 2017; Hamaguchi et al., 2019; Kuo et al., 2020). Marets et al. (2017) demonstrated for the first time the possibility of using an LC-structured membrane with precise nanochannels for virus removal in water purification. These channels can supply pathways larger enough for water molecules to permeate but small for virus to pass through. Here, a nanostructured bicontinuous cubic LC thin film was photopolymerized onto a polysulfone support layer to make a regular and controllable pore structure, and its removal of bacteriophage Q β reached 6 LRVs. To further increase the water flux while efficiently rejecting viruses, a smectic A liquid-crystalline (SmA-LC) material with layered nanochannels and two-component columnar LC

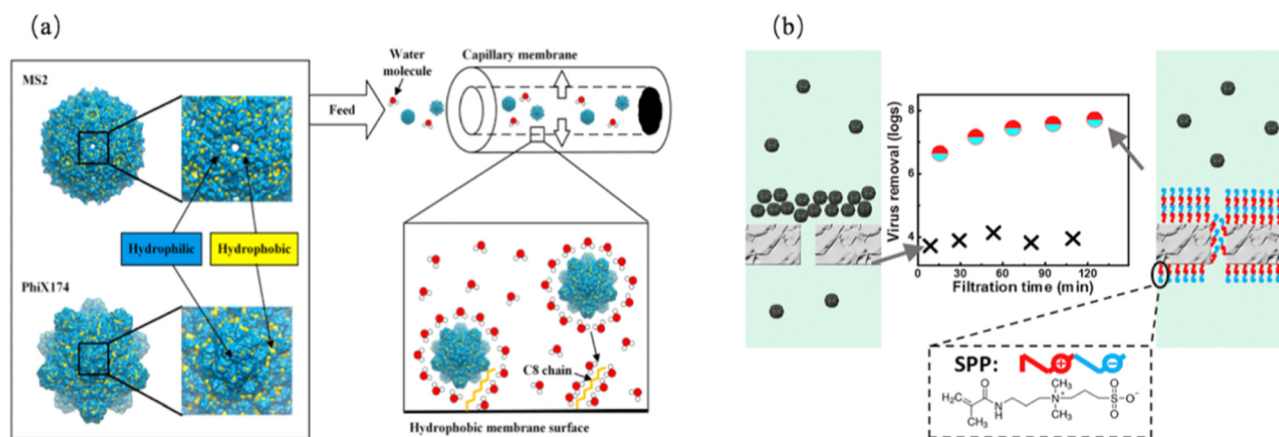


Fig. 6. Schematic diagram pertaining to modified membranes. (a) Hydrophobic yttria-stabilized zirconia capillary membranes are hydrophobized with n-hexyltriethoxysilane and n-octyltriethoxysilane, which can be beneficially utilized for virus retention as a result of hydrophobic interaction (Bartels et al., 2019); (b) graft-polymerization functionalization of 150 kDa ultrafiltration polyether-sulfone membrane with zwitterionic ([β -(methacryloylamino) propyl] dimethyl (3-sulfopropyl) ammonium hydroxide), achieving a higher virus removal on account of the weakened virus accumulation upon the modified membrane surface (Lu et al., 2017).

Table 2
Separation of viruses from wastewater using a polymer-modified membrane.

Membrane specification/charge (pH = 7.4)	Average pore size	Viruses	Operating conditions	Removal efficiency (LRVs)	References
Microfiltration/0.1–1 μm Polyethersulfone membrane coated with PEI/positive	0.45 μm	Bacteriophage MS2	0.2 bars (4 ± 0.9) × 10 ⁹ PFU/mL	>3	Sinclair et al., 2018
TiO ₂ tubular ceramic microfilters	0.8 μm	Bacteriophage P22	5 × 10 ⁹ PFU/mL	5	Guo et al., 2015
Nano-composite electrospun nanofiber membrane (PAN-ATTM, PAN-TEOS)	0.8 μm	Semliki Forest virus	10 ⁶ PFU/mL	1.96	Al-Attabi et al., 2019
Chitosan membranes modified by pyromellitic dianhydride	–	Bacteriophage MS2	10 ⁹ PFU/mL	3	Majiya et al., 2019
PAN/PET-cellulose nanofibers/positive	0.73 μm	Bacteriophage MS2	10 ⁶ PFU/mL	4	Wang et al., 2013
Microporous ceramics with ZrO ₂ and Y ₂ O ₃ coatings/positive	0.2–2 μm	Bacteriophage MS2	3 bars 10 ⁷ PFU/mL	4 7	Wegmann et al., 2008 Wegmann et al., 2009
SiO ₂ -Y ₂ O ₃ composite nanofiber membrane/positive	0.1 μm	Bacteriophage MS2	–	4	Liu et al., 2019
PEI-TA-PES membrane (LBL); PEI-Ag/CuNPs-PES membrane	0.45 μm	Bacteriophage MS2	4 × 10 ⁸ PFU/mL	4.5–5	Sinclair et al., 2019
Coating of the ceramic membrane with HTS and OTS	0.15 μm	Bacteriophage MS2	2.5 bars	0.3 ± 0.1	Bartels et al., 2019
Nano-TiO ₂ -PVDF flat membrane	0.2 μm	Bacteriophage PhiX174 Phage F2	10 ⁹ PFU/mL 1.35 × 10 ⁷ PFU/mL	3.4 ± 0.2 3.88	Zheng et al., 2013
Coating of fiber filter with MWCNTs–copper hydroxide precipitate	0.4 μm	Bacteriophage MS2	10 ⁸ PFU/mL pH = 5	>5	Domagala et al., 2020
Spray-dried alumina granules modified with copper (oxide) nanoparticles on ceramic filter	1–2 μm	Bacteriophage MS2	10 ⁴ PFU/mL	3.1; 3.2	Mazurkow et al., 2020.
Ultrafiltration/2–100 nm Polysulfone (capillary)	20 nm	Bacteriophage MS2	10 ⁶ –10 ⁷ PFU/mL	2.5–6	Kreißel et al., 2012
Graft-polymerized zwitterionic SPP on polyethersulfone membrane	50 nm	Bacteriophage MS2, Human adenovirus	0.69 bar 2.3–3.0 × 10 ⁹ PFU/mL	>6, 6.6–7.8	Lu et al., 2017
Polysulfone membrane coated with Columnar LC-PET film	3.5 nm	Bacteriophage Qβ, Bacteriophage MS2, Aichi virus	0.3 MPa 10 ⁷ –10 ⁸ PFU/mL	>6.7, 6.3, 7.6	Kuo et al., 2020
Coating of the polysulfone ultrafiltration membrane with nAg/negative	–	Bacteriophage MS2	5 ± 0.2 × 10 ⁵ PFU/mL	4	Zodrow et al., 2009
Nano-TiO ₂ -PAN flat membrane	50 nm	Phage F2	1.35 × 10 ⁷ PFU/mL	6.4	Zheng et al., 2013
Nanofiltration/1–2 nm Polysulfone membrane coated with Two-Component Columnar LC-PET film	1.6 nm	Bacteriophage Qβ	0.3 MPa 10 ⁸ PFU/mL	4.4 ± 0.3	Kreißel et al., 2012
Polysulfone membrane coated with Columnar LC/PET film	1.8 nm	Bacteriophage Qβ	0.3 MPa 10 ⁸ PFU/mL	4.7 ± 0.3	Gupta et al., 2019
Reverse osmosis/<1 nm Polysulfone membrane coated with Cub _{bi} LC	0.6 nm	Bacteriophage Qβ	0.8 MPa NaCl 500 mg/L 10 ⁸ PFU/mL	>6.3	Marets et al., 2017

Notes: PEI: polyethyleneimine; PAN: polyacrylonitrile; ATTm: ammonium tetrathiomolybdate; TEOS: tetraethyl orthosilicate; PET: poly(ethylene terephthalate); TA: terephthalaldehyde; PES: polyethersulfone; LBL: layer-by-layer; HTS: n-hexyltriethoxysilane; OTS: n-octyltriethoxysilane; PVDF: polyvinylidene fluoride; SPP: ([3-(methacryloylamino) propyl]dimethyl (3-sulfopropyl) ammonium hydroxide); LC: liquid-crystalline; Cub_{bi}: bicontinuous cubic. MWCNTs: multi-walled carbon nanotubes.

membranes was prepared, providing prewetted channels with hydrophilic conditions, which were more conducive to water molecules to penetration. The removal of these two membranes on viruses reached 4.4 ± 0.3 and 7.3 LRVs, and the water fluxes were 20 ± 6 and 15 L m⁻² h⁻¹, respectively, 40 times and 30 times the flux of the cubic LC membrane (0.54 ± 0.05 L m⁻² h⁻¹) (Hamaguchi et al., 2019; Kuo et al., 2020). In addition, Lee et al. (2018) employed a bioconjugated method to load feline calicivirus (FCV), a model virus with a structure similar to NoVs, onto the membrane surface, creating a dual functional membrane capable of both easily detecting and blocking NoVs. This concept of using 2D materials could likely endow the increase of the regular size and the transport nanochannels of pores, and the minimum pinholes for the fabrication of membranes to achieve precise sieving.

3.2.2. Membrane fouling

The presence of membrane fouling improves the retention of the membrane by either clogging breach or forming a cake layer on the membrane surface, which can impact the composition of membrane surfaces to remove viruses as a secondary barrier. As shown in Fig. S6, one study used four membranes to investigate their separation performance and found that the LRVs for MS2 ranked as follows: fouled

membrane > backwashed membrane > chemically cleaned membrane ≈ pristine membrane (Lu et al., 2013), which implies that membrane fouling plays a positive role in virus retention. An analogous conclusion was also reflected in another investigation in which the LRVs gradually increased when the time after the membrane wash increased from 0 to 24 h (Chaudhry et al., 2015b), hence grasping the influence of fouling on virus reduction is a pivotal consideration.

Organic fouling can alter the nature and location of nanoparticle capture by competitive adsorption to change the capacity of the membrane for virus retention, as suggested in the studies of Fallahianbijan et al. and Nazem-Bokae et al. (Fallahianbijan et al., 2019; Nazem-Bokae et al., 2020). One study has shown that different membrane fouling layer can either improve or attenuate the removal of adenovirus relying on layer properties, as an illustration, humic acid can increase the removal since humic acid containing carboxylic and functional groups can adsorb onto the membrane surface, thereby altering the electrostatic properties and surface hydrophobicity of the membrane and enhancing the interaction between adenovirus and membrane surface, while SiO₂ fouling can decrease the removal as the fouling layer caused by larger SiO₂ particles is too porous to effectively reject adenovirus yet may lead to the accumulation of virus near the cake-membrane interface

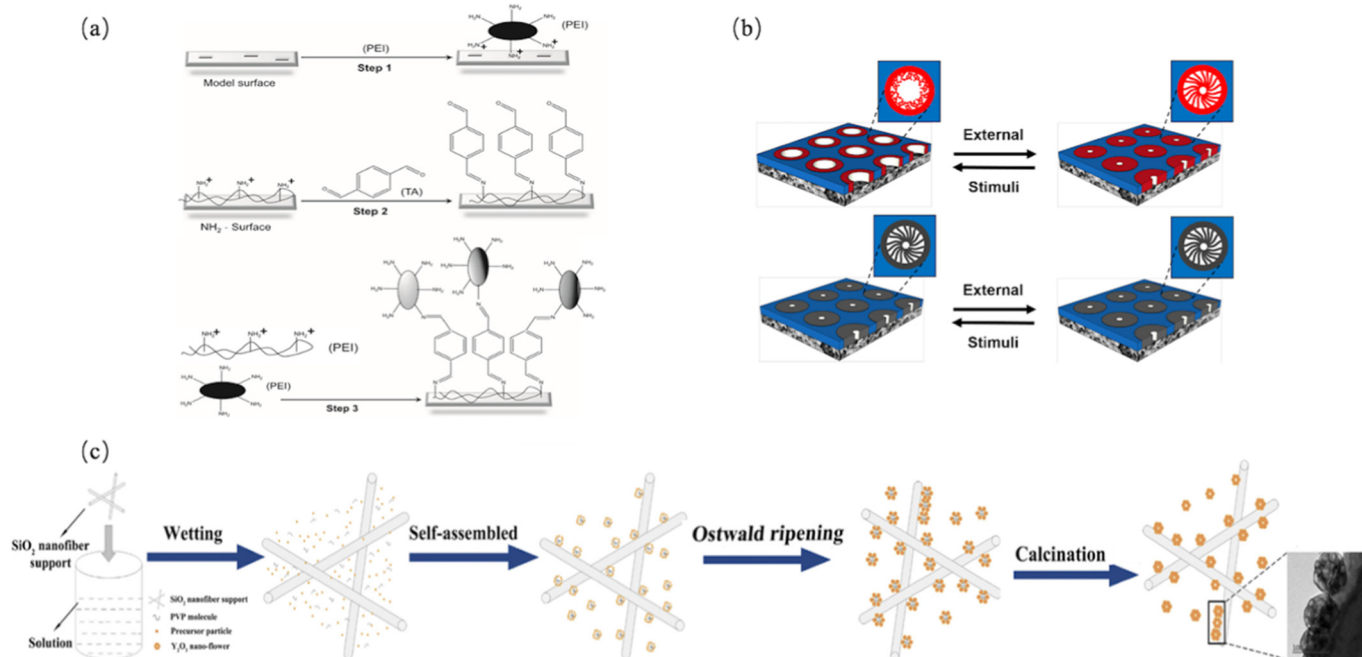


Fig. 7. Schematic diagram pertaining to modified membranes. (a) A stable covalent layer-by-layer strategy used to fabricate ultrathin polyelectrolyte/polyethyleneimine (PEI) multilayers by chemical-crosslinking with terephthalaldehyde (TA) (Sinclair et al., 2019); (b) self-assembled block polymer membrane with tailor-made functionality designed by the macromolecular template, which possessed a precise Molecular Weight Cut Off (MWCO) with 8 Å selective separation (Zhang et al., 2017); (c) self-assembly of the positively charged SiO₂-Y₂O₃ composite nanofiber membrane with a plum-flower-like structure, rendering an excellent virus retention as a result of high adsorption capacity (Liu et al., 2019).

(Yin et al., 2015). The increase in virus removal owing to irreversible fouling is higher than reversible fouling (ElHadidy et al., 2014). Membrane biofouling, generally regarded as an irreversible fouling (Nagaraj et al., 2018), will help to achieve more than 7 LRVs of virus removal during long-term filtration in various membrane processes, including the coagulation-MF system, MBR, UF and coagulation-ceramic MF, etc. (Shirasaki et al., 2008; Chaudhry et al., 2015a; Alansari et al., 2015; Michalsky et al., 2009). During these processes, the effect on virus removal by different mechanisms, such as physical sieving, adsorption, cake and gel layer formation that could be expounded by biofouling surface characteristics and structural properties.

Meanwhile, some studies have demonstrated that irreversible fouling is one of the main factors that causes membrane aging (Antony et al., 2016), and the effect of aged RO membranes on virus removal shows certain improvements (Chesters et al., 2013; Pype et al., 2016). Intermittent operation can retain some microbes and organics in the interval of the RO element and increase their propagation during shutdown, leading to irreversible fouling, which can partially enhance virus retention (Torii et al., 2019b). However, intermittent operation can also cause deterioration of the surface of the membrane, which would lead to an increase in permeability but with compromised virus retention (Wang et al., 2017). A study has shown that virus retention is reduced to 2 LRVs after repeated pressurization for 10,000 times (Torii et al., 2019a).

Although membrane fouling is one of major drawbacks of the membrane treatment procedure, fouling will increase virus separation to some degree. By performing an in-depth study of the different types and degrees of membrane fouling and of the mechanism of virus retention to find the best equilibrium point, the operation cost of the membrane treatment can be greatly reduced to provide a theoretical proposal for membrane cleaning and strengthened virus separation during long-term operation.

3.2.3. Hybrid processes

The application of membrane-based hybrid processes can also further enhance inactivation and removal of viruses (Matsushita et al.,

2011). A graph of the integration of coagulation and membrane filtration as well as of practical treatment effects is displayed in Fig. S7.

Adding an iron electrocoagulants to an MF system or the addition of glycine before NF can aggregate viruses into larger particles, making virus removal up to 6.5 LRVs (pH = 6.4) (Tanneru and Chellam, 2012). By adding polyaluminum chloride (PACL) as pretreatment coagulant for ceramic membrane filtration, the floc size was increased to form a looser and more porous filter cake layer so as to render more MS2 transferring to the floc phase and reduce the number of small particles that can lead to pore plugging, so the virus removal and membrane anti-fouling performance were all promoted (Im et al., 2019). One study that utilized UF process combined with coagulation and sedimentation to treat virus-containing wastewater showed removal of 1.8–4.2 LRVs more than UF alone (Lee et al., 2017), the introduction of sedimentation could limit the load of coagulation floc on membrane, which achieve a continuous operation in practical use.

Furthermore, the incorporation of two membrane systems can provide a double guarantee of effluent quality. UF integrated with RO can achieve 7.7 LRVs for high norovirus concentrations (Yasui et al., 2017). The MBR combined with the RO process, which takes advantage of size exclusion, electrostatic repulsion, hydrophilic and hydrophobic interaction, sorption of the membrane, and the adsorption of attached and suspended biomass, demonstrated excellent performance for the removal of viruses, emerging pathogens and micropollutants and was suitable for reclaimed water production (Prado et al., 2019b). In short, both the addition of coagulants prior to membrane filtration and the combination of different membrane process systems are capable of increasing virus retention to ensure favorable effluent conditions.

As for membranes, owing to the limitations of membrane materials and membrane fabrication methods, traditional separation membranes have an uneven pore size distribution and thick selective separation layers, which restrict improvements of water flux and separation selectivity (Werber et al., 2016). Therefore, overcoming the “trade-off” between flux and retention by designing and preparing high-performance membranes with a uniform pore size and nanometer thickness is an advisable direction for progress. It is important to effectively control virus transmission in water by understanding how virus

removal is affected by interactions between viruses and membranes instead of only relying on membrane retention.

3.3. Disinfection for virus inactivation

To date, the major disinfection methods include chlorine (including free chlorine, chlorine dioxide, monochloramine), ozone and ultraviolet radiation. Various strains of the same virus usually have different susceptibilities to these disinfectants. For example, some mutant NoV strains were less sensitive to disinfectants than others and can still be alive even after wastewater treatment (Rachmadi et al., 2018). Meister et al. (2018) also found that the inactivation kinetics of EVs with diverse serotypes isolated in the laboratory or outdoors vary considerably. Therefore, it is necessary to select an appropriate disinfection strategy under specific conditions.

3.3.1. Chlorination

At present, the dominant disinfection is still chlorination. Free chlorine commonly damages the proteins and nucleic acids of viruses, while chlorine dioxide (ClO_2) and monochloramine (NH_2Cl) mainly damage the viral capsid (Wigginton et al., 2012). The corresponding inactivation mechanism is shown in Fig. 8(a). During the disinfection process, the CT value, which is obtained by multiplying the disinfectant concentration by the contact time and is affected by temperature and pH, can be adjusted to achieve satisfying disinfection outcomes. The US EPA notes that CT value needs to be 3-, 4-, and 6- $\text{mg} \times \text{min/L}$ to attain 2, 3, and 4 LRVs, respectively, at a temperature of 10 °C and pH of 6.0-9.0 when using chlorination disinfection (US EPA, 1991).

Generally, the disinfectant type and its initial concentration as well as the virus category have a certain impact on the disinfection efficiency. Rachmadi et al. (2020) utilized Tobit and simple linear regression analysis to calculate the CT values required to reach 4 LRVs of virus removal in chlorine and NH_2Cl disinfections (as shown in Fig. 8(b)). It shows that the CT values in free chlorine disinfection are much lower than those in NH_2Cl disinfection for the same virus, which probably means that the subcellular components (e.g., viral DNA, RNA, capsid) are more susceptible to free chlorine than NH_2Cl and the activity of free chlorine still remains outstanding. A study showed that HRV inactivation reached up to 5 LRVs by increasing the initial concentration of the disinfectant, whereas the inactivation could not be further increased by only prolonging the contact time (Xue et al., 2013), this phenomenon demonstrates that the initial concentration of the disinfectant is a necessary factor for disinfection. In addition, the CT values for CVB and ECHO were higher than those for AdVs and MNV with free chlorine and the AdVs demanded higher CT values than others with monochloramine, which may be due to the different chemical reactivities of the viral proteins and genomes to these disinfectants or because of the enhanced

resistance of the viruses to the disinfectants from the high rate of mutation in the evolutionary process (Rachmadi et al., 2020).

However, chlorination disinfectants can often react with organics or nitrogen in wastewater, producing toxic and harmful DBPs, such as trihalomethanes (THMs) or haloacetic acid (HAA) carcinogens, haloacetonitrile (HANs) and *N*-nitrosodimethylamine (NDMA) (Zhang et al., 2020b). Therefore, we can take full advantage of some natural or synthetic compounds to replace chlorination disinfectants. Park et al. (2018) and Dunkin et al. (2017) used a micrometer-sized silica hybrid composite decorated with silver nanoparticles (AgNP-SiO_2) and peroxyacetic acid (PAA) to successfully remove hNoV, MNV and bacteriophage MS2 from different water media. In addition, ferrate, as a newly emerging disinfectant, has a better disinfection efficiency than other materials due to its higher redox potential. Some research results adopting pseudo-second-order kinetics and Chick-Watson inactivation dynamic models expounded that H_2FeO_4 and HFeO_4^- had far higher MS2 MNV inactivation than FeO_4^{2-} and the increase of pH decreased the amount of H_2FeO_4 to reduce the inactivation rate constant (k_d) (Wu et al., 2019; Manoli et al., 2020). On the other side, the intracellular algal organic matter (IAOM) that exhibited a stronger reaction with ferrate has a stronger inhibitory effect on MS2 inactivation than extracellular algal organic matter (EAOM). Therefore, it is significant to explore the influence of disinfectant types and their initial concentration, virus category and sensitivity, DBP generation, pH and temperature, coexisting substances, etc. in detail when using chlorination disinfection.

3.3.2. Ultraviolet radiation

The radiation in the wavelength range of ultraviolet (UV) has relatively high energy, and microorganisms absorb photons in high absorption coefficient between 200 and 300 nm. UV radiation disinfection does not produce detrimental byproducts that appear in chlorination disinfection. Employing UV to irradiate viruses can destroy the viral genome (including the phosphate bond between DNA/RNA) and the cross-link between capsid proteins (Wigginton et al., 2012). Different wavelengths of UV have distinguishable inactivation mechanisms for viruses. Bravo et al. (2018) found that UV_{254} and UV_{280} can cause mutations in the viral genome and thus inhibit DNA replication, while UV_{224} irradiation barely affects the integrity of the HAdV-2 genome, but the changes it causes in the capsid structure may restrain the translocation of the viral genome into the nucleus in host cells (As depicted in Fig. 8(c)).

However, traditional UV mercury vapor lamps are fragile and pose a risk associated with mercury. Therefore, some believe that UV light-emitting diodes (UV-LEDs) are expected to substitute for traditional UV lamps for virus disinfection (Li et al., 2019; Nguyen et al., 2019). The development of UV-LEDs will evidently reduce the need of energy and the price of electricity ascribed to the higher use of wind power and sun power. Song et al. (2019) used UV-LEDs to irradiate *E. coli* and

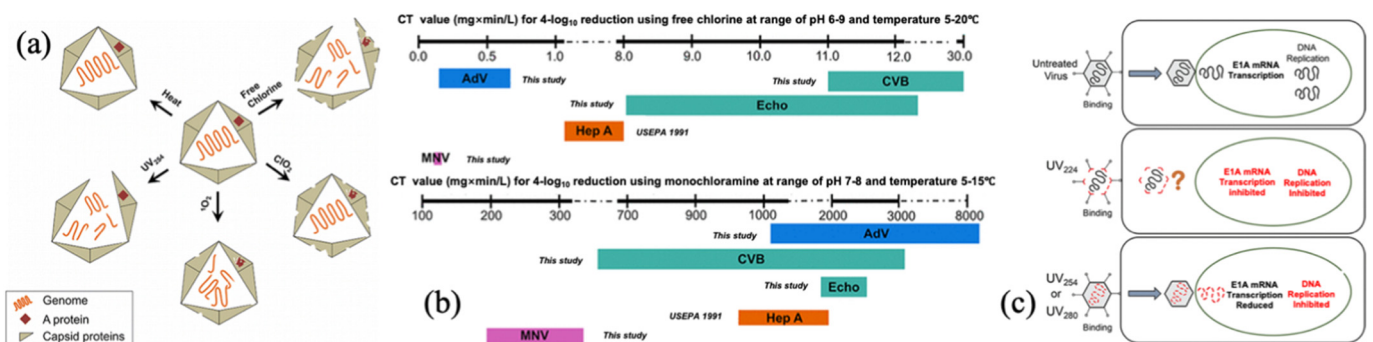


Fig. 8. (a) Principle of different disinfections for damaging the virus structure. UV irradiation and free chlorine caused inactivation primarily by damaging both viral genome and protein, $^1\text{O}_2$ caused inactivation by impairing genome replication and ClO_2 by the degradation of proteins (Wigginton et al., 2012); (b) CT values needed to achieve 4 LRVs of the removal of AdV, CVB, ECHO and MNV using free chlorine at pH 6–9 and temperature 5–20 °C (above) and using monochloramine at pH 7–8 and temperature 5–15 °C (below) (Rachmadi et al., 2020); (c) distinctive inactivation mechanism of UV radiation at different wavelengths. UV_{224} mainly affected the integrity of viral capsid to inhibit the delivery of viral genome into the host cell, while UV_{254} and UV_{280} mainly restrained the DNA replication (Bravo et al., 2018).

bacteriophage MS2 and found that compared with *E. coli* removal, the damaged RNA of MS2 could not be repaired owing to the lack of repair enzyme. The application of UV-LEDs to virus disinfection has only attracted attention in recent years (Keshavarzfathy et al., 2021; Rattanukul and Oguma, 2018), so deeper studies need to be performed.

To further improve the disinfection efficiency, altering work pattern or adding other substances including sodium hypochlorite, chlorine dioxide, ozone or Fenton's reagent can be employed. For some insensitive viruses, Zyara et al. (2016) combined UV with chlorine to successfully reduce the chlorine-resistant strains by 3–5 LRVs with reduced DBP production. Additionally, Schijven et al. (2019) conducted a microbiological quantitative risk assessment (QMRA) and used somatic coliphages and bacteriophage MS2 as indicators of AdVs to explore the inactivation efficiency of UV and ClO₂, proving that the predetermined target could be realized if low-concentration ClO₂ (0.05–0.1 mg/L) was added during UV (40 mJ/cm² or 73 mJ/cm²) disinfection.

The adsorption and aggregation of viruses must be considered in UV disinfection. Feng et al. (2016) discovered that MS2 formed aggregates and was less inactivated if cations such as sodium (at pH 3) or calcium (at pH 7) were introduced, as the virions situated inside the aggregates were prevented from the UV irradiation, but MS2 was more likely to adsorb to particles and inactivated in the presence of organic particles such as microcystis aeruginosa. As for WWTPs that adopt UV for disinfection, ultraviolet wavelengths and intensity should be considered, and 100, 143, and 186 mJ/cm² are theoretically required for 2-, 3-, and 4-log virus inactivation, respectively (US EPA, 2006b). In the dynamic process of wastewater treatment, it is inevitable to encounter matrix objects that have complicated interactions with viruses, so researchers should use qualitative or quantitative methods to explore the relationships in depth.

3.3.3. Ozonation

Ozone is a strong oxidant and can interact with water to generate free radicals to destroy the protein and nucleic acids of viruses (Wigginton and Kohn, 2012; Zhang et al., 2016). Ozonation can reduce viruses beyond conventional treatments or even make viruses undetectable, so it may be used to reduce the spread of human viruses (Wang et al., 2018). Cai et al. (2014) determined that with a pH increase and temperature decrease and in the presence of particles, organics and coexisting ions, inactivation would be reduced. Concretely, first, pH affects the oxidation ability and attenuation rate of ozone, which were respectively weaker and rose in alkaline conditions; second, a temperature increase would augment the energy of ozone molecules to facilitate the movement of ozone; third, particles would bind to the virus to a certain extent, thereby shielding the virus and influencing inactivation; lastly, the presence of dissolved organic matters would consume a large amount of ozone, thus compromising the ozone inactivation efficiency.

Meanwhile, integrating ozone with biologically activated carbon, free chlorine or other catalysts may lead to better virus inactivation. Im et al. (2018) combined ozone, coagulation and ceramic membrane filtration to remove viruses for water reclamation and found that the reduction of MS2 could be increased from 2.1 to 6.8 LRVs as the amount of ozone increases since this increase can support more MS2 removal by subsequent membranes. Moreover, ozone can act with catalysts to form hydroxyl radicals that can accelerate the oxidation and decomposition of viruses. For example, when utilizing volcanic rocks as catalysts, ozone could almost completely remove NV GI, GII and JC PyV, but if only ozone was used, JC PyV was still not removed after 150 min (Gomes et al., 2019). However, it is relatively difficult to detect virus inactivation in ozone disinfection because of the lack of real-time tracking methods and the high reactivity of ozone. Some investigators have established Bayesian power models and an experimental batch system to overcome this issue and use the second-order rate constant ($k_{03\text{-virus}}$) to quantify ozone exposure as well as to evaluate the results of ozonation for the inactivation of EVs and bacteriophages, finally, they

received a good disinfection efficiency (Wolf et al., 2019; Wolf et al., 2018). What's more, arising from the extraordinary instability of ozone, the bromide ion in water is easily oxidated to bromate that is mentioned as a kind of DBPs in the drinking water standard in many countries (US EPA, 2009; China, Standards for drinking water quality (GB5749-2006), 2006; Japan, Water Quality Standard, 2015), which is a remarkable barrier for its extensive application. As such, we should optimize treatment process to minimize the formation potential of DBP precursors and reasonably control the disinfection amount and time.

3.3.4. Catalytic oxidation

Light irradiation excites electrons in the valence band (VB) of semiconductors to the conduction band (CB), and the holes formed in the VB and the excited electrons can both participate in redox reactions. Photocatalysts include natural photocatalysts (García-Gil et al., 2020; Ryberg et al., 2018), semiconductor oxides (such as titanium dioxide) (Reddy et al., 2017; Liga et al., 2011; Zheng et al., 2018), plasma (Guo et al., 2018), metal oxides, and graphene-based photocatalysts (such as g-C₃N₄) (Zhang et al., 2019a; Cheng et al., 2018).

Semiconductor oxide-based photocatalysis has recently been one of the hottest research topics. Under light irradiation at ambient temperature, semiconductors will yield many kinds of reactive species (RS), including hydroxyl radical ($\bullet\text{OH}$), superoxide radical ($\bullet\text{O}_2^-$), singlet oxygen ($^1\text{O}_2$), hydrogen peroxide (H₂O₂), etc. (Li et al., 2008; Lee et al., 2011; Wu et al., 1998), to destruct viral genome and protein for virus inactivation. Among these photocatalysts, titanium dioxide (TiO₂) is widely applied in the field of drinking water and wastewater treatment since it can generate reactive oxygen species (ROS), including a surface-produced $^1\text{O}_2$ and free metal ions to damage the capsid protein (Reddy et al., 2017). To overcome inherent limitation of the slow reaction kinetics and reduce the electron-hole recombination rate in TiO₂ catalyze, Liga et al. (2011) and Liga et al. (2013) studied the ability of photocatalytic silver-doped and SiO₂-doped titanium dioxide nanoparticles (nAg-TiO₂ and SiO₂-TiO₂) to inactivate bacteriophage MS2 and found that compared to TiO₂ alone, the MS2 inactivation rates induced by nAg-TiO₂ and SiO₂-TiO₂ increased by more than 5 and 2.7 times, respectively, mainly attributed to the increased generation of free $\bullet\text{OH}$ from silver and the larger adsorption surface area of the catalyst with the addition of silica (as shown in Fig. 9(a)). Zheng et al. (2018) mentioned that the addition of Cu-TiO₂ can provide a large active surface of catalysts and produce more free $\bullet\text{OH}$, but with its continuous increment, the increase in solution turbidity and the decrease of photon penetration ability as well as enhanced catalyst agglomeration led to a decline in catalytic performance. When the visible light intensity and temperature were increased, the catalysts were fully excited to generate more holes and electrons, intensifying the oxidation effect of the catalyst. In addition to TiO₂, Hu et al. (2010) also used Ag-AgI/Al₂O₃ as a plasma to achieve a 3.2 LRVs for RV removal under visible light, which mainly because the generated inorganic anion radicals caused by plasmon resonance of Ag nanoparticles not only boost the electron transfer but have high bactericidal activity. Sarkar et al. (2018) prepared nanopores by electrospray deposition of silver ions on a single-layer molybdenum disulfide (MoS₂) nanosheet, the caused molybdenum-rich defects can efficaciously generate ROS under visible light to remove 7 LRVs of bacteriophage MS2 in water.

For some nonmetallic photocatalysts, besides common graphene oxide-based catalysts (Akhavan et al., 2012), graphitic carbon nitride (g-C₃N₄), a visible-light-response semiconductor with a two-dimensional conjugated structure characterized by robust physicochemical stability, low cytotoxicity, facile synthesis and suitable electronic band structures (~2.7 eV), has been studied extensively (Zhang et al., 2019a; Lin et al., 2014). The mechanism of g-C₃N₄ for virus inactivation under visible-light irradiation is shown in Fig. 9(b) (Zhang et al., 2019a). Cheng et al. (2018) prepared the Ag₃PO₄-g-C₃N₄ (AgCN) photocatalytic composite and observed that this composite could completely

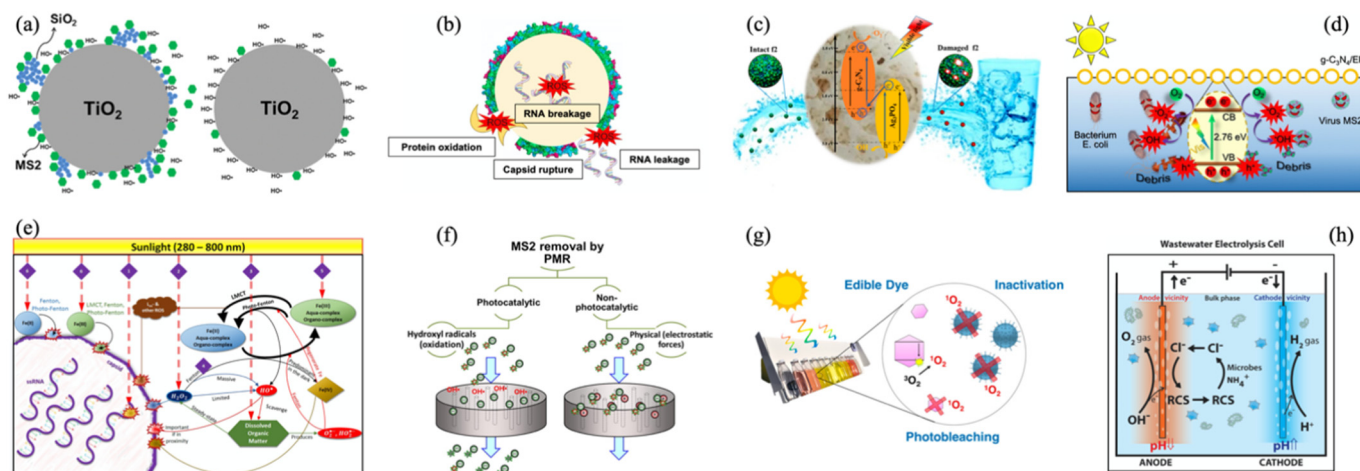


Fig. 9. (a) Photocatalytic inactivation of TiO_2 toward bacteriophage MS2 with (left) and without (right) the addition of SiO_2 nanoparticles (Liga et al., 2013); (b) response of viral death to $\text{g-C}_3\text{N}_4$ -based photocatalytic disinfection including protein oxidation, capsid rupture, RNA breakage and RNA leakage (Zhang et al., 2019a); (c) a possible Z-scheme inactivation mechanism of bacteriophage f2 by Ag_3PO_4 - $\text{g-C}_3\text{N}_4$ photocatalytic material. The f2 was oxidized by photogenerated holes (h^+) under visible light irradiation, together with hydroxyl radicals ($\bullet\text{OH}$) formed by reaction between h^+ and H_2O or OH^- near the surface of Ag_3PO_4 and superoxide radical ($\bullet\text{O}_2^-$) caused by trapping electrons by dissolved oxygen near the surface of $\text{g-C}_3\text{N}_4$ (Cheng et al., 2018); (d) proposed mechanism of bacteriophage MS2 inactivation by $\text{g-C}_3\text{N}_4$ -EP520 under visible-light irradiation (Zhang et al., 2018); (e) proposed MS2 inactivation route during the photo-Fenton process through (1) direct sunlight, (2) oxidative stress exerted by H_2O_2 , (3) irradiation of the DOM to generate H_2O_2 , O_2^- , $^1\text{O}_2$ and other ROS, (4) enhancement of the $\bullet\text{OH}$ production under solar light in Fenton reaction, (5) aquo-complexes by hydrolysis and organo-complexes in the presence of DOM for Fe(III) in the wastewater and (6) organo-complexes from the interaction of Fe(II) and Fe(III) with amino acids in MS2 capsid (Giannakis et al., 2017); (f) process of bacteriophage MS2 removal by PMR (including the oxidation of hydroxyl radicals in photocatalytic and electrostatic force in non-photocatalytic) (Horovitz et al., 2018); (g) enhanced solar disinfection method using an edible dye as a photosensitizer to generate $^1\text{O}_2$ for virus inactivation and signify the finish of solar disinfection by photobleaching (Ryberg et al., 2018); (h) an electrolysis cell for toilet wastewater disinfection in which the free reactive chlorine produced in situ instead of $\bullet\text{OH}$ and other reactive oxygen species was the main disinfection ingredient (Huang et al., 2016).

inactivate 3×10^6 PFU/mL bacteriophage f2 within 80 min and exhibited superior stability because of the production of photogenerated holes (h^+), $\bullet\text{OH}$ and $\bullet\text{O}_2^-$ (Fig. 9(c)). Others incorporated $\text{g-C}_3\text{N}_4$ with a low-density porous expanded perlite (EP) mineral (Fig. 9(d)) to achieve 8 LRVs of MS2 inactivation in 240 min under visible-light irradiation, in which the increments of dissolved oxygen (DO), proton concentration, salinity (Na^+) and hardness (Ca^{2+}) decreased the electrostatic repulsion between MS2 and the photocatalyst to facilitate MS2 inactivation (Zhang et al., 2018).

As a green and sustainable technology, advanced oxidation processes (AOPs), especially the photo-Fenton technique, have been increasingly utilized to achieve conspicuous treatment of chemical pollutants but have been less applied to microbes. Therefore, Giannakis et al. (2017) introduced H_2O_2 or Fenton's reagent along with light to inactivate viruses (Fig. 9(e)) and discovered that higher iron concentrations were able to improve virus inactivation and that Fe(II) could interact with key constituents in the viral capsid, generating intermediate ROS near viruses to inactivate them. Furthermore, the ROS that formed from the complexation reaction of iron and dissolved organic matter (DOM) also played a role in photocatalytic disinfection. Sun et al. (2016) also combined UV with peroxy chemicals, including H_2O_2 and peroxydisulfate (PDS), to achieve disinfection in which three reactive species, $\bullet\text{OH}$, sulfate radical ($\text{SO}_4\bullet^-$) and carbonate radical ($\text{CO}_3\bullet^-$), were responsible for MS2 inactivation.

Photocatalysis has also been emerging in another promising technology, the Photocatalytic Membrane Reactor (PMR), to inhibit viruses and other microorganisms. The PMR is a hybrid reactor that combines photocatalysis and membranes. Zhang et al. (2020a) employed a PMR driven by visible light-emitting diodes (Vis-LEDs) and used a self-made metal-free heterojunction with the merits of efficient virucidal effects and easy recovery via microfiltration as a catalyst to totally inactivate HAdV under optimal conditions. Other researchers proposed a N-doped TiO_2 -coated Al_2O_3 photocatalytic membrane reactor to remove MS2 (as shown in Fig. 9(f)), and the results demonstrated that the removal of MS2 adsorbed to the coated membrane in the PMR throughout photocatalysis due to the $\bullet\text{OH}$ was 4.9 ± 0.1 LRVs. Meanwhile, natural organic matter (NOM) has obviously been shown to be a negatively charged photocatalytic inhibitor (Horovitz et al., 2018).

Solar water disinfection (SODIS) is inherent clean, simple, economical and space-saving and has therefore been implemented in developing countries. The water is placed into polyethylene terephthalate (PET) or polypropylene (PP) bottles which are later exposed to the direct sunlight for 6 h in clear days or for 48 h in cloudy weather in order to ensure the safety of drinking water (Luzi et al., 2016; Oates et al., 2003; García-Gil et al., 2020). The optical and thermal effects act synergistically for the inactivation of organisms, in the meantime, the aluminum foil reflectors, container volume and the turbidity of water can retard the radiation efficiency (Kehoe et al., 2001), so some avenues like filter prior to solar exposure are deployed (Reed, 1997). Viruses are generally inactivated through indirect sunlight-mediated inactivation induced by ROS, notably $^1\text{O}_2$, produced by exogenous photosensitizers (e.g., humic acids) in waters (Kohn and Nelson, 2007). SODIS is able to inactivate some ssRNA viruses but requires extra auxiliary measures when effectively reducing DNA viruses resistant to oxidation (Carratalà et al., 2016). Walker et al. (2004) reduced the F-specific RNA bacteriophage MS2 by 3.5 LRVs after 6 h of natural sunlight via applying a solar disinfection pouch. To improve the efficacy of solar disinfection, Ryberg et al. (2018) proposed an enhanced SODIS method using a food dye—erythrosine—as a photosensitizer and disinfection indicator, finally accomplishing more than 4 LRVs of bacteriophage MS2 inactivation within 5 min (as shown in Fig. 9(g)). It must be noticed that the working life span of plastic bottles is limited. On the other hand, SODIS might be inefficient in places with weak sunlight radiation and inadequate heat, such as places far from the equator or lacking sunshine.

For systems in which it is difficult to remove viruses with ordinary disinfectants, we can consider electrochemical treatment. Electrochemical oxidation adopts an electrolysis cell where the electrodes under redox reactions with an applied voltage to remove microbial pathogens. When using chlorine as the disinfectant, the generated reactive chlorine species (RCS, such as Cl_2 , HOCl , ClO^-) and chlorine free radicals (such as $\bullet\text{Cl}$, $\bullet\text{Cl}_2$) are the main disinfection agents, while water or other saline solutions are used as the electrolyte and the formed $\bullet\text{OH}$, H_2O_2 , ozone (O_3) and $\bullet\text{O}_2^-$ serve as the main roles in disinfection, as evinced in Fig. 9 (h) (Huang et al., 2016). Heffron et al. (2019) first applied sequential electrocoagulation-electrooxidation (using boron-doped diamond electrodes) to better mitigate bacteriophages MS2,

FX174 and human echovirus, which resulted principally from the positive stimulation effect in the physical reduction of coagulation-filtration, ferrous iron-based disinfection and electrooxidation disinfection. To date, there has been little research on (photo-) electrocatalytic oxidation for virus treatment, but as a developing technology, this kind of oxidation is worthy of more public attention.

On the whole, catalytic oxidation technology possesses the strengths of easy operation, mild reaction condition, toxicity-free, high permanent oxidative stability and environmental friendliness. Different catalyst loadings, light intensities and wavelengths, as well as different doped elements including metal ions, non-metal, sulfide, etc., can influence the disinfection effect by changing the type and quantity of reactive species. Generally, there is electrostatic repulsion between viruses and catalysts, which can be weakened by increasing DO, proton concentrations and cation amount as well as by reducing NOM to improve the catalytic effect. It should be noted that lower synthesis cost, superior catalytic efficiency and the large-scale availability ought to be the imperative goal for the following research.

3.4. Comparison of virus elimination processes

Numerous research studies are dedicated to finding a way to yield highly productive and high-quality water from wastewater. The removal efficiency varies for different technologies and viruses. The comparison shown in Fig. 10 indicates that NF/RO have relatively stable removal values of 4.1–7 LRVs, while the removal efficiency of MBR, MF and UF is more affected by the heterogenous infectivity of viruses and shows a removal range of 1.4–7.1, 0.7–4.7 and 0.5–5.9 LRVs, respectively. Among the disinfections, the deviation of chlorination is relatively small, which may be one of the reasons for its widespread application, and by adjusting the CT values can we achieve sufficient virus removal. As for UV radiation, ozonation and catalytic disinfection, different levels of virus inactivation can be achieved (with 0.09–5, 0.6–7.7 and 1–8 LRVs, respectively) through various disinfectant concentration, light intensity, photocatalyst type (for generating validly reactive substances) and the contact probability between viruses and disinfectants. On the whole, all the methods have acceptable effects

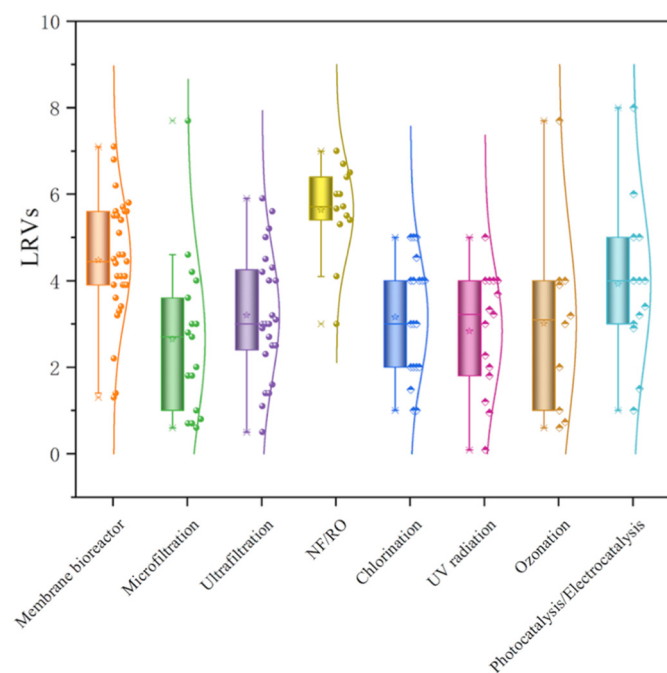


Fig. 10. Comparison of the removal efficiencies of viruses by each water/wastewater treatment process. The detailed quantitative data and the relevant operation settings are described in Table S3.

for virus removal, and we can opt for an appropriate method to increase selectively targeted virus elimination as much as possible.

Recent studies have revealed that various technologies have already been used for virus-containing wastewater/drinking water treatment. Fig. 11 summarizes the range of the various types of virus removal in each process, and all processes are compared in Table 3. Overall, virus elimination by means of membrane filtration and disinfection technology was within the range of 0.5–7 LRVs and 0.09–8 LRVs, respectively.

In the case of enteric viruses (such as Coxsackievirus and HAV), UF has a relatively higher retention efficiency for HAV than even for AdVs with a large size, which indicates that electrostatic and hydrophobic interactions may play crucial roles in determining virus retention. As for NoVs, more removal occurs in the MBR than in MF and UF, which may suggest stronger reactivity between NoVs and activated sludge or that NoVs may be prone to adsorb on sludge particles with subsequent separation by a membrane, and NoVs GI shows more resistant to remove than NoVs G II. The smaller removal values of PMMoV obtained in MBR denote that this virus is significantly resistant to biological treatment. Separation of viruses using a membrane cannot be regarded as a simple screening process, and the generalization of the behaviors of these viruses has not been unified. The influence of the characteristics of the viruses themselves must also be considered.

The inactivation of all the disinfection methods for the same virus is mostly between 1 and 4 LRVs, and the inactivation is sometimes over 5 LRVs. The data collected so far have indicated that chlorine or chlorine dioxide is sufficient for SARS-CoV reduction (Li et al., 2020); chlorination and ozonation show slightly better inactivation for PMMoV; ozonation and photocatalysis are better for AdVs (highly resistant to monochloramine and UV irradiation (Hu et al., 2010)) and CVB elimination; chlorination is better for RVs, JC PyV and ECHO removal; and UV radiation is better for NoVs reduction, and all the methods are acceptable for bacteriophage MS2 reduction. Some assumptions can be made: (1) since the phage MS2 is a non-pathogenic bacterial virus, its special nature may be more facily sensitive to external disinfection conditions, so its inactivation has adequate effects overall; (2) judging from the available results, UV relatively exhibits less preponderance for most virus inactivation, probably because of its non-durable disinfection efficiency and the adverse consequences from coexisting ions, or obtaining better inactivation would be costly and the different wavelengths of UV light needs to be optimized to promote virus inactivation; (3) during photocatalysis and other processes involving free radicals for oxidation, viruses have different resistances, and the activity of free radicals under different conditions is also an important factor.

4. Conclusions and future perspectives

Viruses are distinct from other pathogens and show varying reactions in treatment processes, leading to discrepancies in their fate and behavior in water. Better understanding the characteristics, behavior and migration laws of viruses in water treatment and improving the comprehensive assessment of virus contamination along will help guide future research and provide theoretical guidance for the control of waterborne diseases.

The removal efficiencies for AdVs, NoVs GII and Bacteriophage MS2 are comparable in the MBR process, which demonstrates that the MS2 can be regarded as a good indicator or surrogate for AdVs and NoVs GII in MBR. Nevertheless, it has not yet been clearly determined whether virus reduction is attributable to decomposition by activated sludge or retention by membranes. In the former case, determining which one of the microbial communities plays a leading role will be the focus, and the coexistence relationship and interactive mechanisms (like adsorption) between some component (such as particulate matter, other microorganisms, chemical substances and protozoa) in activated sludge and viruses need to be intentionally explored. While in the latter condition, the decisive function from membrane ought to come into people's sight.

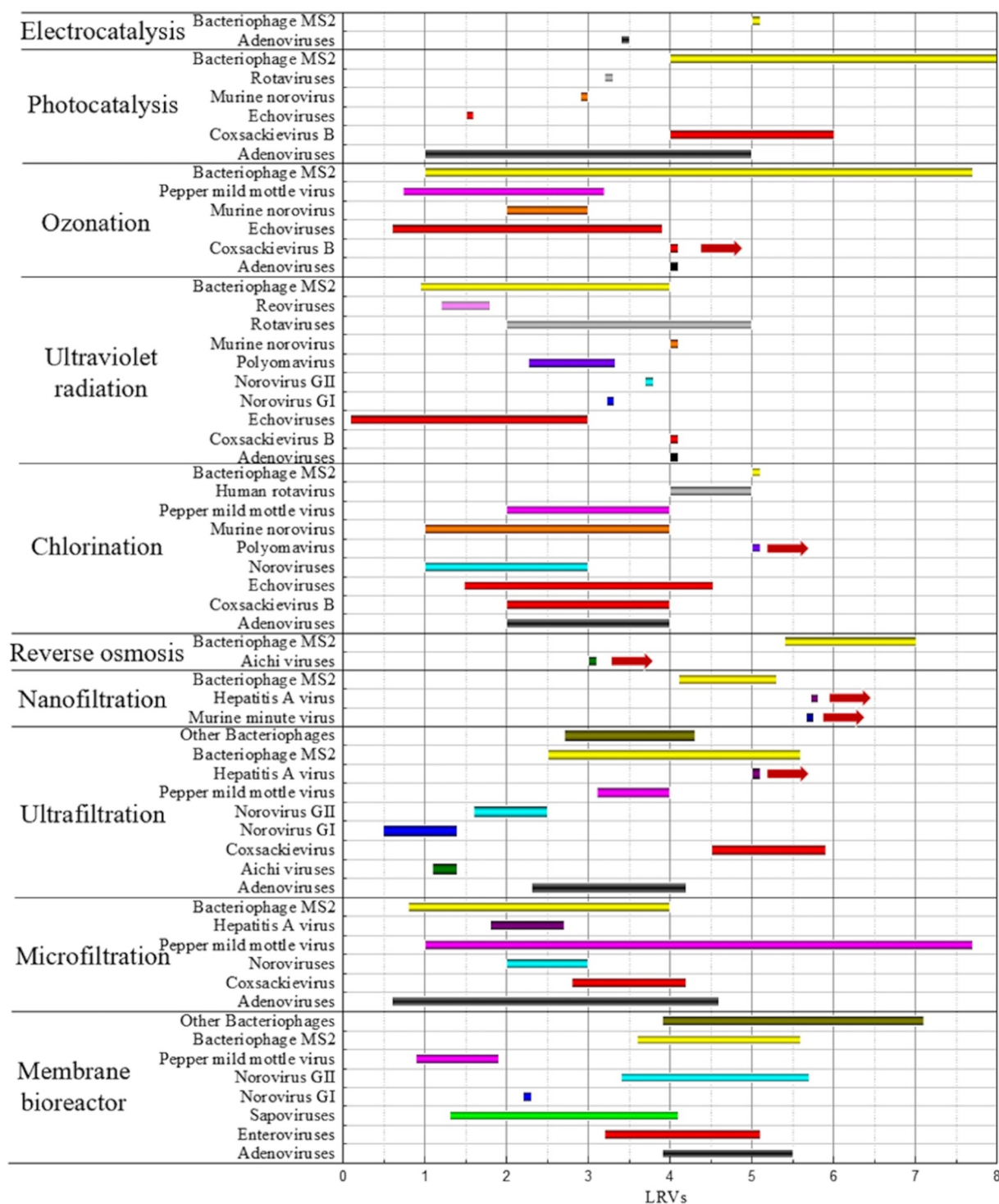


Fig. 11. Ranges of the removal efficiency of viruses by various water/wastewater processes originating form information in Table S3.

Even though the inherent nature of viruses, such as difference in nucleic acid, has a few influences over their removal from membrane, the removal effect is more closely dependent on the membrane properties like the pore size distribution and the interaction between the virus and membrane. In most of cases, the pore sizes in MF and UF membranes are often bigger than the diameter of viruses, thus the acquired propensities of virus removal are susceptibly affected by electrostatic and hydrophobic interactions. Introducing nanoparticles or nano-sheets including liquid crystals, carbon nanotubes, and block polymers (Kuo et al., 2020) that have uniform-sized nano-channels, large specific surface area and high reactivity can be treated as a measure to obtain

filtration membrane with high pore interconnectivity and long-term antiviral performance, where the extra removal attributed by the fouling layer is certainly worth to be concerned. A single antiviral strategy can only weaken typical interactions between viruses and the membrane surface, limiting the scope of pollutants that can be treated. However, the synergistic effect achieved by multiple antiviral methods is still vague, which makes it hard to be controlled with high accuracy.

Moreover, it is expected to develop integrated membrane processes to promote virus elimination such as coagulation, adsorption, precipitation and disinfection. Yet different combinations may provide more or less effective removal of the viruses, and place one technique in

Table 3
Virus removal range and the strength and weakness of each technology.

Process	Removal (LRVs)	Major function mechanism	Strength	Weakness
MBR	1.4–7.1	Attachment of virus to mixed liquor solids; retention by membrane; retention by membrane cake layer; inactivation of viruses by enzyme	High removal efficiency; high flux and less space demand	Incomplete removal of dissolved organic matters (<500 kDa); high cost for operation and maintenance
Microfiltration	0.7–4.6	Adsorption largely onto membrane surface or within its pores; follow by size exclusion	High permeability; low pressure-driven process	Low removal effect; health risk potential for humans
Ultrafiltration	0.5–5.9	Retention by membrane and attachment of virus onto membrane surface or sorption within its pores	High flux and permeability; low energy cost and effective removal of high molecular weight matter	High capital investment and operation; removal efficiency is unstable
Nanofiltration/reverse osmosis	4.1–7	Size exclusion; Electrostatic interactions	High performance, security and reliability, dedicated removal of enveloped and nonenveloped viruses based only on size-exclusion	High requirements for influent quality
Chlorination disinfection	1–>5	Damage in protein, nucleic acid and viral capsid	Easy to handle, economical, long residual	DBP formation, corrosive, residual toxicity
UV radiation disinfection	0.09–5	Formation of lesions in viral genome and destruction of the cross-link between genome and protein	No DBP formation, short contact time, less operating process and space, no extra chemicals, less susceptible to temperature and pH	No residual disinfection efficiency, relatively high level of energy consumption with a certain compromise in UV-LEDs
Ozonation disinfection	0.6–7.7	Free radical formation from reaction between ozone and water	Short contact time, inactivation of viruses	No residual disinfection efficiency, high energy consumption, relatively hard to detect
Photocatalysis disinfection	1–8	Redox reaction of some reactive species (h^+ , e^- , $\bullet OH$, $\bullet O_2$, 1O_2 , H_2O_2 , etc.) with visible or UV light	Facile preparation, favorable catalytic performance, low operation cost, good stability	low quantum yield for a few materials
Electrocatalysis disinfection	3.4–5	Redox reaction of some reactive species ($HOCl$, $\bullet Cl$, $\bullet OH$, $\bullet O_2$, O_3 , H_2O_2 , etc.) in electrolysis cell	Applicable to some hard-removing viruses	Electricity consumption

pretreatment or posttreatment would also produce discrepant results (Im et al., 2019). Furthermore, there is a need to judge the type of the treated water. Compared with domestic sewage and industrial wastewater, the medical wastewater contains more pathogens like viruses, as such, it deserves more exceptional cautions. On the other hand, maintaining good property stability during the long-term operation in large-scale application is of great importance, and it is also apparent that there is a large deviation between the laboratory simulation experiment which is more inclined to mechanism investigation and the large-scale application to verify actual treatment effects, so we must seek strategies to close the gap between them.

Major disinfection treatments include chlorination, UV radiation, ozonation and catalytic oxidation. Some alternatives or combined techniques, such as ferrate and novel $g\text{-C}_3\text{N}_4$ and TiO_2 -based photocatalytic composites, can be considered as beneficial measures to increase virus inactivation. Many factors should be considered for disinfection, such as the type and initial concentration of the disinfectant and virus and the pH, temperature and matrices (such as particles, DOM, coexisting ions, dissolved oxygen) of the treated water and others. If some coexisting substances cause flocculation to wrap viruses or compete with viruses to react with disinfectants, the disinfection efficiency will be compromised. Otherwise, the efficiency will increase if the adsorption between particles and viruses dominates.

Viruses have different disinfectant-resistance due to their unique genome structure and morphology (i.e., icosahedral or helical symmetry, whether containing envelope and spike). The disinfection mechanism for virus reduction and inactivation primarily lies in the interaction between the disinfectants and viruses to destroy the viral capsid protein and nucleic acid, thereby irreversibly inhibiting the transmission of viruses to host cells as well as their reproduction. Other biological factors such as bacterial compounds will influence the disinfection effect, for example, in drinking water production, adding lipopolysaccharide or peptidoglycan of bacterial origin to enterovirus defended the viral capsid from thermal breakage when the capsid was the prime disinfection target (Waldman et al., 2017), and mixture with *E. coli* 285 obviously affected the removal of bacteriophage $\phi 2$ (Zheng et al., 2018). Till now, enteric viruses with ssDNA genome are few. Albeit chlorination probably gives rise to secondary pollution, it displays relatively better

inactivation effects for JC PyV (dsDNA), PMMoV (ssRNA), and RVs (dsRNA) than other disinfections. UV radiation, less susceptible to temperature and pH than other disinfectants, is relatively effective when eliminating ssRNA (e.g., NoVs) through mutating viral genome or changing capsid protein under different wavelengths. Ozonation with short operation time has over 4 LRVs of reduction for dsDNA (e.g., AdVs) and ssRNA (e.g., CVB, bacteriophage MS2 and PMMoV) but less for dsRNA by generating hydroxyl radicals to oxidize and damage nucleic acid. Catalytic oxidation has less satisfactory removal for some dsRNA viruses such as RVs and ssRNA viruses including NoVs and ECHO but exhibits good results for other dsRNA viruses (e.g., AdVs) and ssRNA viruses like CVB by means of producing reactive species (including free radicals) for the purpose of protein oxidation, capsid rupture and genome breakage. In the near future, new emerging disinfection technology especially photocatalysis or photoelectrocatalysis with fantastic disinfection performance has great potential in virus inactivation in water. Of course, it is worth noting that during the disinfection process, the damaged protein may self-repair and regenerate under appropriate conditions (Nelson et al., 2018), so we are supposed to pay our attention to destroying viral genome so as to inactivate viruses thoroughly.

With regard to the quantitative virus removal, the evidence from this study suggests that membrane filtration and disinfection technologies can treat virus-containing wastewater/drinking water over a wide range (0.5–7 LRVs and 0.09–8 LRVs, respectively). From another perspective, the combination of membrane-based separation and other technologies may harness the strengths and circumvent their respective shortcomings. Tertiary treatment including membrane filtration and disinfection technology will be indispensable in the near future. The presence of organic matters and inorganic ions will impact the inactivation by disinfectant toward viruses (Cai et al., 2014), so membranes separation can be necessary prior to disinfection in some cases. UF and MF membranes are generally an effective barrier for protozoa and bacteria but have limited effects to remove viruses due to their small size. Studies have suggested that a hybrid MF-UV process with a photocatalytic membrane for the removal and inactivation of bacteriophage P22 was more effective (LRV = 5.0 ± 0.7) than stand-alone MF/UV disinfection or MF-UV with a non-photocatalytic membrane (Guo et al., 2015), and

the integrated coagulation–MF/UF filtration–UV obtained the highest removal of fluorescent-conjugated MS2 at 5 mg Al³⁺/L (Guo and Hu, 2014). Specifically, some inspiring results in our work can be proposed: for AIVs, it is suggested to use a combined UF-disinfection process; for AdVs, adopting an MBR system alone or using UF/MF with ozonation/photocatalysis (also suitable for CVB) can be considered; for NoVs, linking MF/UF and UV disinfection together may lead to superior virus removal outcomes; and for PMMoV reduction, simultaneously supplementing the UF/MBR unit with chlorination/ozonation would be a nice choice.

In all processes for eliminating viruses, we should avoid the secondary pollution that may be caused by building extra components such as disinfection by-products and removing viruses may encounter the low-cost and specialization issues in small-scale water supply plants. In the context of COVID-19 pandemic, it may be a smart strategy to combine disinfection and membrane separation with molecular imprinting technology to enhance the selective targeted removal of viruses, at the same time, some traditional and emerging monitoring equipment can be deployed to online real-time water quality monitoring of viruses to reduce the higher risk that may be caused by the leakage of the pipe network.

Finally, since there is a large number of viral strains in water, some indicators or surrogates that have behaviors similar to viral pathogens and are easy to detect and quantify should be used to assess water pollution, virus attenuation and virus migration in water. Importantly, bacteriophage MS2 can be adopted as a substitute for NoVs and RVs to explore the disinfection efficiency of UV, but MS2 is often detected with higher LRVs and usually does not have the strong correlation with other indigenous viruses regarding removal efficiency in chlorine and photocatalysis disinfection, which may lead to an overestimation of the effectiveness of water treatment. Thus, it is recommended to look for adequate potential indicators of indigenous viruses.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.149678>.

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