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REVIEW ARTICLE

Research progress of advanced oxidation technology for the removal of *Microcystis aeruginosa*: a review

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Abstract

In recent years, cyanobacteria blooms have continued to erupt frequently, seriously jeopardizing the safety of drinking water and human health. The safe, quick, and economical removal of cyanobacteria from water bodies, especially the dominant species of cyanobacteria, *Microcystis aeruginosa*, has captured a lot of scientists' attention. The application of advanced oxidation technology in water treatment is very promising, but it has not yet been used in production. To further promote the application of the advanced oxidation method in water treatment, this article combines the results of advanced research in China and abroad to review this emergent technology. Briefly, advanced oxidation process methods employ various mechanisms to remove the dominant species of cyanobacteria blooms *Microcystis aeruginosa*. This provides a theoretical reference and support for the efficient removal of harmful cyanobacteria from water.

Keywords Algae removal · *Microcystis aeruginosa* · Eutrophic water body · *Microcystins* · AOPs

Introduction

As more and more wastewater gets discharged into water bodies around the world, the content of nitrogen (N) and phosphorus (P) in the water has increased sharply, exceeding the self-purification level of the environment itself and resulting in the frequent occurrence of eutrophication. The outbreak of harmful algal blooms is an important feature of water eutrophication (Ma et al. 2016). The outbreak of cyanobacterial blooms is influenced by many factors, including temperature, light, pH, and nutrient concentration (Kong et al. 2020). Cyanobacteria cells have both the characteristics of particles and the characteristics of organic matter. Algal cyanobacteria are mostly negatively charged in water bodies, and because of their three-dimensionality, hydrophilicity, and electrostatic repulsion, they are stable in water bodies. In addition, studies have found that the surface of algae in natural water bodies can adsorb natural organic substances such as humic acid, making the cyanobacteria even more stable. *M. aeruginosa* is an algae with the ability to move freely in the vertical direction. It has a wide absorption range, fast growth rate, and the ability to release algal toxins, so it occupies a dominant position among other aquatic microorganisms. It is also the dominant species in cyanobacteria blooms (Yan et al. 2020).

Under normal circumstances, most of the algal organic matter (including algal toxins) exists in the cell and is called intracellular organic matter. When the cell is damaged, the intracellular organic matter is released into the extracellular environment and becomes extracellular organic matter (EOM). However, the removal of dissolved organic matter (including algal toxins) is much more difficult than the removal of *Microcystis aeruginosa* cells. Studies have found that EOM released by algal cells easily combines with inorganic substances in the water to become a source of organic colloids, thereby making raw water treatment more difficult (Ma 2017). At present, there is no quick method that can simultaneously remove algal cells and algal toxins without negatively impacting the environment. Advanced oxidation

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processes (AOPs) are currently used in various wastewater treatments and have achieved good results in the treatment of water bodies with harmful algal blooms. By generating reactive oxidant species (ROS) with strong oxidation capacity, AOPs can rapidly oxidize and degrade most organic compounds with less pollution to the environment. The specific advantages are as follows: (1) the refractory toxic organic pollutants are effectively decomposed by the free radicals generated by the reaction until they are completely converted into harmless inorganic substances without secondary pollution. (2) The reaction time is short, the reaction speed is fast, the process can be controlled, and all kinds of organic pollutants can be degraded (Yang et al. 2021a). To more effectively and safely remove *Microcystis aeruginosa* and repair water bodies, this article reviews the current common algae removal methods and provides suggestions for improving the technology.

Harm caused by *Microcystis aeruginosa* and its conventional removal method

Harm caused by *Microcystis aeruginosa*

The hazards caused by algal blooms include damage to human health, endangerment of aquatic organisms (Zhou et al. 2018), and severe damage to ecosystems (Fang et al. 2020). A reason why *Microcystins aeruginosa* is problematic is that it releases seven-ring mono-peptide compounds called *Microcystins* (MCs). At present, more than 100 types of *Microcystins* have been discovered, and the three main types are MC-LR, MC-RR, and MC-YR, wherein L represents leucine, R represents arginine, and Y represents tyrosine (Song 2020). These substances are acute liver toxins that can cause liver diseases in animals and humans by altering lipid metabolism and glucose metabolism. Of these, MC-LR is the most toxic and abundant microcystin. MC-LR prevents dephosphorylation in liver cells by inhibiting the activity of protein phosphatase-2A (PP2) and serine/threonine protein phosphatase-1 (PP1). The imbalance of protein phosphorylation causes cell physiological and biochemical reactions to become disordered, resulting in hepatocyte hemorrhage, liver failure, and hepatocellular carcinoma. In mouse experiments, its LD50 is 50 μg/kg. It is also the strongest known inducer of liver tumor cancer. Furthermore, it is a chronic kidney and brain toxin that can induce physiological changes in the respiratory and reproductive systems, and may even cause genetic mutations (Shi et al. 2020). MC-LR accumulates in aquatic animals or plants, thereby reducing not only the production of fish, shrimp, and other aquaculture industries, but also causing harm to the humans that eat them. Animals and humans may experience vomiting, diarrhea, fatigue, difficulty breathing, stomach pain and bleeding, and even death after contacting or eating microcystin (Song 2020). In addition, MCs and other toxins often coexist and influence each other in nature, and a combination of MCs and certain risk factors can significantly increase the risk of liver damage and liver cancer (Qian et al. 2021).

The status of the waters where the verdigris microcapsules erupted

Algal blooms caused by the eutrophication of water bodies have become a major challenge for water resources in recent years. Based on the total phosphorus eutrophication threshold (0.035 mg/L) established by the Organization for Economic Cooperation and Development, the severity of eutrophication now varies greatly from place to place, and eutrophication in some areas is extremely serious. Lakes and reservoirs have the worst eutrophication levels due to their low fluidity. For example, a third of the more than 800 reservoirs in Spain are experiencing severe eutrophication; out of 318 thousand lakes in Canada, a quarter of them are in a state of eutrophication; and the 1996 water quality survey report in the USA showed that eutrophication is threatening 51% of lakes and reservoirs (Liu et al. 2021a) According to a survey conducted by the United Nations Environment Programme, about 30–40% of reservoirs and lakes around the world have varying degrees of eutrophication (Cao 2015). Lake Aboka in Florida, USA, has experienced extreme eutrophication due to pollution by organic pesticides in recent years. The ecological environment of Lake Constance, the largest lake in Western Europe, began deteriorating in the 1950s, then started rapidly deteriorating in the 1970s. Lake Biwa, the largest lake in Japan, was once known as an oligotrophic lake. In the 1930s, the transparency of the center of North Lake Biwa could reach about 10 m. However, with rapid economic development, the transparency dropped to 6 m in the 1950s, and to an average of 4 m in the 1970s. Water blooms occurred in North Lake in 1997, as well, followed by frequent water blooms over the next 10 years (Cao 2015).

The outlook on the eutrophication situation in China is not optimistic. From 2000 to 2016, 1194 harmful algal blooms occurred in coastal areas of China, or about 70 per year on average (Yang et al. 2021b). The results of the second lake status survey in China showed that, of the 138 lakes with an area greater than 10 km^2, 85.4% of the lakes exceeded the eutrophication standard, 40.1% of the lakes reached the severe eutrophication standard, and Lugu Lake was entirely eutrophicated. The number of lakes that are oligotrophic increases each year. Additionally, the average number of lakes with high mesotrophic and oligotrophic levels throughout the whole lake has reached 14.6%. The highest regional proportion of eutrophication is found in the Northeast Plains and mountainous lakes at 96.0% followed by the lakes in the middle and lower reaches of the Yangtze River in the Eastern Plain at 85.9%. The lowest regional proportion is found in the Yunnan-Guizhou Plateau Lakes at 61.5% (Cao 2015). The water quality of the six major lakes in China (Taihu Lake, Chaohu Lake, Dian Lake, Poyang Lake, Dongting Lake, and Hongze Lake) was evaluated in 2011 according to the surface water environmental quality standards (Developed by the State Environmental Protection Administration of China), and the six lakes are all either in states of mild or moderate eutrophication. The absolute majority of the remaining small- and medium-sized lakes or reservoirs also have varying degrees of eutrophication (Hu and Zhu 2014).

Since the "Ninth Five-Year Plan" (1996–2000), Taihu Lake, Chaohu Lake, and Dian Lake (i.e., the "Three Lakes") have been listed as Chinese key governance areas (Fig. 1). Among the "Three Lakes," the water bloom duration of Taihu Lake fluctuates the most and has been decreasing year by year since reaching its peak in 2005. After 2010, the average duration of the water bloom in Taihu Lake was about 210 days per year. Until 2018, most of Taihu Lake water was still in moderate eutrophication (Taihu Basin Authority 2019). The range of the bloom durations of Chaohu Lake is smaller than that of Taihu Lake. Since its peak in 2007, it has fluctuated around 245 days per year. Except for a 356-daybloom that occurred in 2005, the bloom duration of Dian has fluctuated around 290 days per year. Dian Lake has the highest average water bloom severity of the "Three Lakes" because the meteorological and water quality characteristics of the area where it is located favor water bloom more than those of Taihu Lake and Chaohu Lake (Wang et al. 2018).

The impact of *Microcystis aeruginosa* outbreaks on water quality

When there is eutrophication in a water body, the color of the water surface can appear blue, green, red, brown, milky white, etc. depending on the color(s) of the dominant algae species. When cyanobacteria bloom in large numbers, it emits an odor that affects the normal physiological and biochemical functions of the water body and makes the physical and chemical indicators of the water body exceed its standard values. These planktonic algae also absorb the short-wave portion of visible light strongly, causing the water temperature to rise (Cao 2015).

The impact of Microcystis aeruginosa outbreaks on water ecology

Algae bloom in large numbers, reducing the transparency and dissolved oxygen level of the water, causing the pH of the water body to change, the water viscosity to increase, and the community structure of the native plankton to change. Blooms cause water bodies to become hypoxic because algae float on the surface of the water. This blocks the exchange of water and gas, thereby reducing the level of dissolved oxygen in the water. Also, when the algae die, it will be decomposed by microorganisms, which will consume a large amount of dissolved oxygen. Either way, the lower dissolved oxygen levels cause various aquatic organisms to die due to lack of oxygen, destroying the diversity of aquatic ecosystems (Wang 2018b). Since the large-scale bloom of cyanobacteria in Taihu Lake, the number of aquatic plants in West Taihu Lake has decreased by at least 50%, and the trend of population simplification has become increasingly obvious. As the lake eutrophication levels have intensified, *Ceratophyllum demerasum L.*, *Myriophyllum spicatum*,

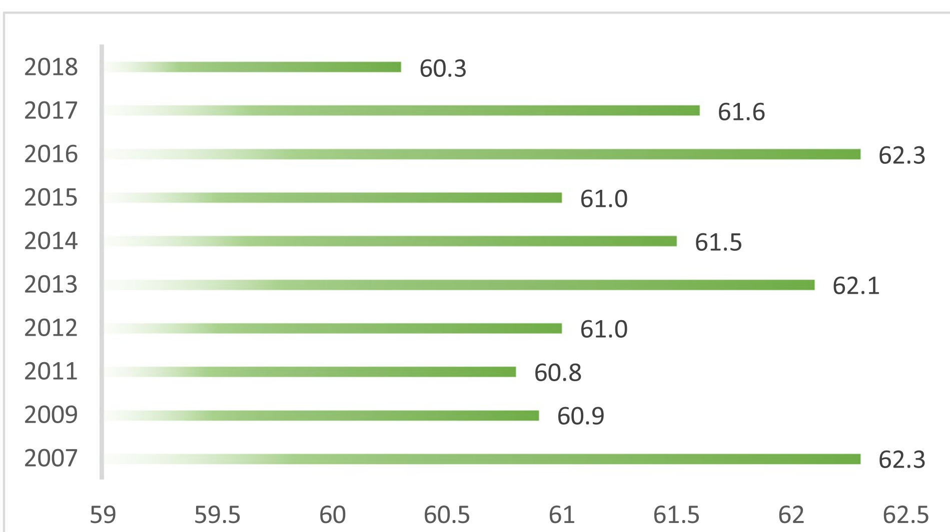

Fig. 1 Nutrition index of Taihu from 2007 to 2018 (Taihu Basin Authority 2019)

Hydrilla verticillata, and other submerged plants have become increasingly endangered. Dian Lake had 42 species of submerged plants in 1950. In 1980, there were 13 species. In 1990, there were 12 species. Based on the second national survey, there were only 8 species.

As blooms progress, the nutrients in the water body increase, allowing small planktonic cyanobacteria to dominate the ecosystem and large aquatic plant communities to be degraded to varying degrees. The large number of phytoplankton, mainly cyanobacteria, that floats on the surface of the water body seriously affects the photosynthesis of aquatic plants. On one hand, this is good because it causes many floating aquatic plants to die, and the resulting debris to decompose at the bottom of the lake, providing nutrients for submerged plants. The rapid "steppization" of the water bottom, on the other hand, will also cause submerged plants to die and fall incompletely decomposed on the bottom of the lake, accelerating the swamping process and destroying the original ecological structure (Cao 2015).

The impact of *Microcystis aeruginosa* outbreaks on water treatment facilities

After a bloom occurs, algal cells accumulate and become denser. As a result, they can easily block pipelines and affect the flow rate of the water. Some of the algae entering the pipe network create odors, which increase the cost of the treatment process. Algae can also be a substrate for proliferating microorganisms, resulting in the accelerated growth of microorganisms and the deterioration of water quality. Bacterial propagation may also accelerate the corrosion of the pipeline network and shorten the service life of water treatment pipeline facilities (Wang 2018b).

Traditional algae removal methods

At present, the more commonly used methods to remove *Microcystis aeruginosa* include the following: chemical, physical, and biological methods (Fang et al. 2020). This section will introduce the classification of these common algae removal methods and explain their advantages and disadvantages.

Physical removal method of *Microcystis aeruginosa*

Physical methods that remove algae include clay, artificial salvage, activated carbon adsorption, filtration, ultrasonic waves, air flotation (Yang et al. 2021b), etc. Filtration removes algae using micro-mesh filtration. This method can not only effectively remove dinoflagellate from the water, but also remove soluble carbon, total inorganic and organic nitrogen, total inorganic and organic phosphorus, and chlorophyll-a. The ultrasonic method uses the cavitation, acoustic current, mechanical, and thermal properties of ultrasonic waves to denature algal protein so that it loses its activity. Research has found that low-intensity ultrasonic waves (power 40 W, action time 10 s) can accelerate the sedimentation of *Microcystis* cells (Li et al. 2021b). The physical method takes effect quickly, has little impact on the environment, and is easy to operate, but it cannot completely eradicate algae blooms. Moreover, it requires a lot of manpower, material resources, maintenance, and special equipment or technology to implement it (Yang et al. 2021b).

Chemical removal method of *Microcystis aeruginosa*

The chemical method inhibits the growth of algae using algaecides. Algaecides are compounds that can kill cyanobacteria in water bodies by reacting with the functional proteins in cyanobacteria cells and destroying the structure of the algae (Ma 2017). Commonly used chemical agents include persulfate, permanganate, and polyaluminum chloride. At the same time, the surface of the sediment is covered with a flocculant to prevent the release of phosphorous and to adsorb or precipitate nutrients. Commonly used flocculants include oxidizing agents, insoluble iron-containing compounds, zeolite, leaded earth ore extraction residue, fully modified bentonite, clay particles, and calcite (Ma 2017). The advantages of the chemical method are its low cost, simple operation, efficiency, and efficacy (as shown in Table 1), but the shortcomings like having a greater impact on the environment, potentially causing damage to non-target organisms, possibly leading to the development of drug resistance genes, and causing secondary pollution are equally obvious (Li et al. 2021b). For example, copper sulfate was once the most widely used algaecide, but this heavy metal chemical was found to not only severely lyse cells and produce a large amount of algal toxins, but also allow residual copper ions to accumulate in fish and shrimps. As it had many adverse effects on the environment and human health, it is currently banned from use (Song 2020).

Biological removal method of *Microcystis aeruginosa*

Biological methods either use organisms (silver carp, bighead carp, algae-eating insects, algae-eating protozoa, and microorganisms (Xiong et al. 2020), etc.) or the allelochemicals that these organisms produce like simple phenol, flavonoids, fatty acids, and pyrogallic acid (made by *Myriophyllum Verticillatum* L. (Shi et al. 2020)) to remove algae. Algaelytic compounds like BSO_2 and BSO_3 produced by marine-efficient *Vibrio alginolyticus* can also be used for algae treatment. Specific allelopathic substances and allelopathic plants are shown in Table 2. Microorganisms probably remove algae using the following mechanisms: (1) By damaging the cell membrane of *Microcystis*;

Table 1 Comparison of the effect of different flocculants on the removal of *Microcystis aeruginosa* (Song 2020)

Flocculant	Optimum addition rate	Removal rate	Cost[a]	MCs released during stacking	Shortcomings
$AlCl_3$ (Sun et al. 2013)	15 mg/L	99%	0.288	Release after 6 days	Residual metal ions are toxic
PACl (Pei et al. 2014)	4 mg/L	90%	0.0169	Mass release after 2 days	Increases the lysis of algae cells
Chitosan (Li et al. 2014)	7.31 mg/L	94.5%	0.967	Mass release after 4 days	Not soluble in water
$FeCl_3$ (Sun et al. 2012)	100 mg/L	89%	0.422	Mass release after 10 days	Increases water chroma
CTS/AC (Jin et al. 2017)	2.6 mg/L CTS and 7.5 mg/L AC	95%	0.501	No obvious release within 4 days	High cost
HTCC (Wang et al. 2017)	1.5 mg/L	99%	0.695	Mass release after 8 days	High cost
TXC (Hou et al. 2018)	12 mg Ti/L	90%	1.24	Slowly increased at first, then decreased after 6 days	High cost and not suitable for actual water bodies
Tanfloc (Xiong et al. 2020)	10.42 mg/L	98.9%	0.153	Mass release after 8 days	Not suitable for actual water bodies

[a]Price (Chinese Yuan) of flocculant needed to treat one ton of wastewater

(2) by destroying the physiological and ecological functions of the algae cells; (3) by inducing the synthesis of reactive oxygen species in the algal cells; (4) by hindering gene expression in the algal cells (Xiong et al. 2020), etc. The advantages of this method are that it is relatively safe and has little impact on the environment. However, it is difficult to implement and slow to take effect. It is also not suitable for the treatment of sudden blooms and may introduce invasive species.

Other removal methods of *Microcystis aeruginosa*

Other emerging methods include magnetic flocculation and floating algae removal (Fang et al. 2020). Another method called flocculation-air flotation uses the dissolved air of high-efficiency shallow air flotation equipment to pass dissolved gas in the water into the wastewater. As a result, tiny bubbles get released and bring suspended solids in the wastewater to the surface, wherein flocculants are added. This method comprehensively utilizes electric neutralization, bridging, net catching, sweeping, and adsorption of flocculants to coagulate and flocculate the dispersed phase in the wastewater, which removes pollutants (Li et al. 2019).

Using advanced oxidation technology to remove water pollutants

Advanced oxidation technology is an oxidation process proposed by Glaze et al. in 1987 that can generate hydroxyl radicals (·OH). Essentially, a large number of free radicals with strong activity is generated through sound waves, light, electricity, catalysts, etc., and these free radicals act on pollutants by causing them to undergo chemical reactions such as breakage of organic chemical bonds, ring opening, and electron transfer. As a result, the pollutants eventually become small molecules or inorganic substances that are easily degraded. The specific advantages and disadvantages of advanced oxidation processes (AOPs) are shown in Table 3. AOPs can also contain Fendon oxidation technology, activated persulfate oxidation technology, photocatalytic oxidation technology, ozone oxidation technology, and some cooperative oxidation technologies (Yang et al. 2021b).

AOPs can rapidly oxidize and degrade most organic substances (i.e., organic dyes, surfactants, hydrocarbons, phenols, active pharmaceutical ingredients, pesticides, etc.) by generating reactive oxidant species (ROS) that have a strong oxidizing ability. One study found that by simultaneously using hydrodynamic or sonic cavitation and either the Fenton method, ozonation, hydrogen peroxide, ultraviolet radiation, or catalysts and persulfate, etc., organic pollutants that are difficult to degrade (including drugs, organic dyes, pesticides, phenol, and their derivatives) can be completely oxidized (Gągol et al. 2018). The ROS that play a major role in this process are either hydroxyl radicals (·OH) or sulfate radicals ($SO4^{2-}$) (Liu et al. 2021b).

AOPs have been widely used in applications such as air treatment, organic pollutant treatment, and plastic treatment (Yang et al. 2021b). Its application in water treatment mostly consists of the treatment of wastewater that is produced during printing and dyeing (Hu and Guo 2021), papermaking, and aquaculture (Qiu et al. 2021). It is an emerging technology in algal removal that has attracted the attention of researchers.

This article mainly elaborates on the following aspects of the research progress of common advanced oxidation methods that are used to remove *Microcystis aeruginosa* in China and abroad: (1) Research methods of advanced oxidation used to remove *Microcystis aeruginosa*; (2) research

Table 2 Aquatic plants with allelopathic effects on *Microcystis aeruginosa* (Pei et al. 2014)

Plant type	Plant name	Experimental method	Main allelochemicals	Effective inhibition rate	References
Submerged macrophyte	*Myriophyllum spicatum*	Lixivium	Tannin, fatty acids	58.5%	Li et al. (2014)
	Myriophyllum aquaticum	Planting water	–	89.61%	Ma et al. (2016); Jin et al. (2017)
	Myriophyllum verticillatum	Co-culture	–	> 90%	Wang et al. (2017)
	Elodea canadensis	Lixivium	Organic acids	47.7%	Hou et al. (2018)
	Vallisneria natans	Planting water, lixivium	Phenol acids	52.9%	Li et al. (2014); Xiong et al. (2020)
	Hydrilla verticillata	Planting water	Organic acids	> 80%	Wang and Liu (2017); Zhang and Wang (2015)
	Antirrhinum majus	Planting water, lixivium	–	100%	Wu et al. (2008)
	Hippuris vulgaris	Planting water	–	< 50%	Gao (2018)
	Najas minor	Co-culture	–	94.1%	Wang et al. (2011)
	Potamogeton malaianus	Lixivium	Fatty acids, sterol compound	–	Jia et al. (2019)
	Potamogeton maackianus	Co-culture	–	100%	Wu et al. (2008)
	Potamogeton pectinatus	Co-culture	–	93.6%	Wang and Cai (2011); Zhang and Tian (2018)
	Potamogeton crispus	Co-culture, lixivium	–	94.92%	Xian et al. (2005)
Emergent macrophyte	*Acorus calamus*	Lixivium	Tannin	94.6%	Tang et al. (2007); Zhang et al. (2014)
	Iris pseudacorus	Culture filtrate	Fatty acids	70%	Wang and Liu (2017); Hu et al. (2010)
	Typha orientalis Presl	Lixivium	Tannin	16.5%	Tang et al. (2007)
	Pontederia cordata	Co-culture, planting water, lixivium	Tannin	98.5%	Li et al. (2014)
	Alternanthera philoxeroides	Lixivium	Tannin	17.6%	Tang et al. (2007)
	Sagittaria trifolia	Planting water, lixivium	Sterol compound, flavones, organic acids, ester	89.3%	Zhang et al. (2016)
	Eleocharis dulcis	Planting water, lixivium	Sterol compound, flavones, organic acids, ester	91.4%	Zhang et al. (2016)
	Arundo donax	Lixivium, pyrolytic fluid	Phenols	95.31%	Tan et al. (2013)
	Nelumbo nucifera	Planting water, plant tissue fragment	–	> 90%	Wang et al. (2017)
	Phragmites communis	Allelochemicals extract	Ester	–	Qian et al. (2017)
	Iris tectorum	Co-culture	–	70.4%	Li (2016)
Floating macrophyte	*Eichhornia crassipes*	Co-culture	Organic acids	85%	Wang (2014)
	Nymphoides peltatum	Co-culture	–	94.68%	Hong et al. (2011)
	Nymphaea tetragona	Planting water, lixivium	–	50.7%	Li et al. (2007)
	Hydrocleys nymphoides	Planting water, lixivium	–	74.59%	Gao (2018)
	Pistia stratiotes	Lixivium	–	> 80%	Zhang and Tian (2018)
	Salvinia natans	Co-culture	Fatty acids, ester, benzene homologs	100%	Yang et al. (2016)
	Lemna minor	Co-culture	–	35.77%	Wang et al. (2010)

Table 3 Advantages and disadvantages of advanced oxidation technology at this stage (Zhang and Wang 2015)

AOPs	Advantages	Disadvantages
Fenton	The ratio of oxidant to pollutant is easy to adjust; can mineralize organic matter in the dark	Produces a lot of sludge; corrosion reactor; harsh reaction conditions; affected by pH (Wu et al. 2008; Wang and Liu 2017)
Photocatalytic oxidation	Relatively friendly to the environment without introducing impurities; mild reaction conditions, strong oxidation	Low light energy utilization efficiency; limited by the light transmittance of the reaction environment (Gao 2018); electron–hole complexes easily (Wang et al. 2011)
Electrochemical catalytic oxidation	Uses oxidation–reduction reactions to achieve pollutant removal; avoids secondary pollution; has strong controllability; flocculation and sterilization effects	High electrode material cost; limited by reaction conductivity, electrode is easy to passivate, low current efficiency (Jia et al. 2019)
Persulfate oxidation	Persulfate radicals are highly oxidizing; easy to control the response; the free radical generation process is relatively gentle; can be used for groundwater repair	Increases the salinity of wastewater (Zhang 2018)
Ozone oxidation	Friendly to the environment; no secondary pollution	Incomplete mineralization of organic matter and low ozone utilization (Xian et al. 2005; Wang and Cai 2011)

progress on the removal of *Microcystis aeruginosa* achieved using collaborative methods. The latter was done to aid follow-up research on algal removal technology.

Research progress of advanced oxidation methods for removal of water pollutants

AOPs mainly use its strong oxidation ability to act on chlorophyll of algal cells. The structure of chlorophyll mainly consists of porphine which is a complex conjugated system composed of four pyrrole rings. DNA damage and protein damage are the main pathways of ·OH-induced cell death. When a large amount of ·OH is produced, the cell membrane can rupture, leading to cell lysis and death, while a moderate amount of ·OH can damage DNA and proteins, leading to cell death (Zheng 2019). The abundant hydroxyl radicals (·OH) and sulfate radicals ($SO_4^{-}\cdot$) with strong oxidizability produced by AOPs make the pyrrole ring break, and then destroy the structure of chlorophyll. This process leads to the termination of intracellular photosynthesis and metabolism; simultaneously, the protein synthesis was obstructed which contributes to the death of *Microcystis aeruginosa* (Dong 2015).

Electrochemical catalytic oxidation

At present, there are several electrochemical oxidation schemes. Their killing effect on algae in eutrophic water happens as a result of the combined effect of direct oxidation (electrodes) and indirect oxidation (production of oxidizing substances). When algae that is suspended in the water touches the surface of an electrode, they can directly exchange electrons with the surface of the electrode. This causes changes in the structure of the cell membrane surface that inactivates the algae. Under the catalyzing power of electricity, the electrode material will also produce a large amount of strong oxidizing substances that attack the phospholipid bilayer of the cell membrane. The damages to the phospholipid bilayer cause a large amount of cytoplasm to leak, and make the cell enter the decline phase in advance, directly killing the cell. The oxidation reaction is as follows (1) Bai (2017),

$$H_2O \rightarrow \cdot OH + H^+ + e^- \quad (1)$$

A variation on this process removes *Microcystis aeruginosa* through an electric reactor. When the current density is 10 mA/cm^2 for 30 min, the number of *Microcystis aeruginosa* drops rapidly, and the treated algae have almost no possibility of growing again. Another method uses a strong ionization discharge method to efficiently prepare an ·OH solution under atmospheric pressure capable of killing *Microcystis aeruginosa*, *Ceratophyllum*, and *Scenedesmus tetracera* within 4.5 s (Ding et al. 2017).

Ozone oxidation

Ozone oxidation mainly degrades organic pollutants in two ways: direct and indirect. The direct method involves the direct reaction of pollutants with ozone (O_3) molecules, and the indirect method involves the ozone-catalyzed reaction of pollutants with hydroxyl radicals (Dong 2015). The direct reaction takes advantage of the oxidizing property of O_3 to oxidatively degrade pollutants. Although O_3 has a higher oxidation potential of 2.07 eV, it is selective in degrading organic pollutants and the reaction rate is relatively slow. The indirect reaction uses O_3 as a catalyst to decompose hydroxyl radicals into eitherhydroxy radicals (·OH) or superoxide radicals ($\cdot O_2^-$). Hydroxy radicals (·OH) have strong oxidation characteristics (2.8 eV), and low selectivity, and can degrade or even mineralize most organic pollutants.

Photocatalytic oxidation

Photocatalytic oxidation technology uses ultraviolet light to excite electron–hole pairs on the surface of the photocatalyst and then act on H_2O or OH^- to generate ·OH, $\cdot O_2^-$ (Dong 2015), etc. The e^- can also react with O_2 to generate $\cdot O_2^-$, and the $\cdot O_2^-$ can go on to generate hydrogen peroxide (H_2O_2) under acidic conditions. The H_2O_2 can produce ·OH under ultraviolet light irradiation, and the ·OH can degrade organic pollutants into water, carbon dioxide, and harmless salt in a chain reaction. The reaction formula is as follows (Li et al. 2021a):

$$H_2O + h^+ \rightarrow \cdot OH + H^+ \quad (2)$$

$$OH^- + h^+ \rightarrow \cdot OH \quad (3)$$

$$O_2 + e^- \rightarrow \cdot O_2^- \quad (4)$$

$$\cdot O_2^- + e^- + 2H^+ \rightarrow H_2O_2 \quad (5)$$

$$H_2O_2 + hv \rightarrow 2 \cdot OH \quad (6)$$

Sono-chemical oxidation

Acoustic cavitation essentially generates sound waves in liquid using ultrasonic vibration. Cavitation refers to the rapid transition of a liquid to the gaseous state that occurs due to a pressure drop. During the process, so-called cavitation bubbles with an implosive force are formed in the liquid. In a flowing liquid stream, the increase of local static pressure will cause the boiling point of the liquid to drop, thereby increasing the rate of evaporation and the appearance of bubbles. During the subsequent pressure increase, the bubbles implode, producing a strong shock wave. During the 1 ns-long implosion process, the temperature inside the bubble can reach 4726.8 °C and the pressure can reach about 9869 atm. This effect is often used in sewage treatment technology because it enhances the removal of pollutants. It can also decompose water molecules into molecules with high oxidation potential, including HO·, $HO_2^-\cdot$, and H_2O_2 that can react with most organic pollutants. The treatment of wastewater today mainly employs hydrodynamic cavitation and acoustic cavitation. During hydrodynamic cavitation, bubbles are generated using a reactor or a flow system which forces the liquid to flow under reduced pressure (Gągol et al. 2018).

Fenton oxidation and Fenton-like oxidation

The Fenton oxidation method uses divalent iron ions to catalyze the formation of ·OH from H_2O_2 which has a strong oxidizing effect on organic pollutants. The specific process is shown in Fig. 2. The hydroxyl radical generated by the Fenton reagent (Fe^{2+}/H_2O_2) destroys the structure of the organic matter, which is then oxidized and degraded (Dong 2015). The reaction can be summarized in two major reactions. First, the iron is oxidized to form free hydroxyl groups, and second the iron is reduced toFe^{2+}. The latter reaction which is depicted by Eqs. (7)–(8) (Liu 2020) is critical to the process:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \cdot OH + OH^- \quad (7)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + \cdot HO_2 + H^+ \quad (8)$$

There will be a series of subsequent reactions that can be classified as transfer reactions (9)–(11) and termination reactions (12)–(14) based on their products. The chain transfer reaction:

$$Fe^{2+} + \cdot OH \rightarrow Fe^{3+} + OH^- \quad (9)$$

$$\cdot OH + H_2O_2 \rightarrow \cdot HO_2 + H_2O \quad (10)$$

$$Fe^{3+} + \cdot HO_2 \rightarrow Fe^{2+} + O_2 + H^+ \quad (11)$$

Chain termination reaction:

$$\cdot HO_2 + \cdot HO_2 \rightarrow H_2O_2 + O_2 \quad (12)$$

$$\cdot OH + \cdot OH \rightarrow H_2O_2 \quad (13)$$

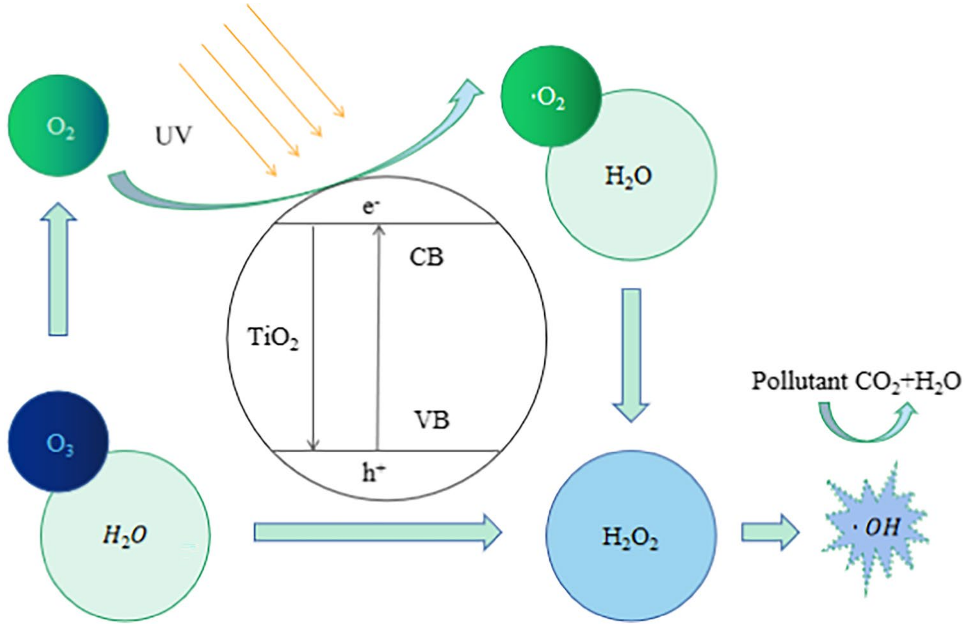

Fig. 2 The UV/O_3/TiO_2 coupling reaction mechanism (Zhu et al. 2021)

$$\cdot HO_2 + \cdot OH \rightarrow H_2O + O_2 \tag{14}$$

According to the above series of reactions, it can be concluded that generating of a large amount of hydroxyl radicals necessitates the consumption of the OH^- from the initial reaction (Fig. 3). Thus, the Fenton process is generally considered to be an efficient reaction at an acidic pH (Liu 2020). Studies have shown that the electro-Fenton method can remove up to 95% of metronidazole in wastewater at a pH range of 3–5 and reduce the number of intermediate degradation products. It has been successfully applied in commercial production (Sun 2020).

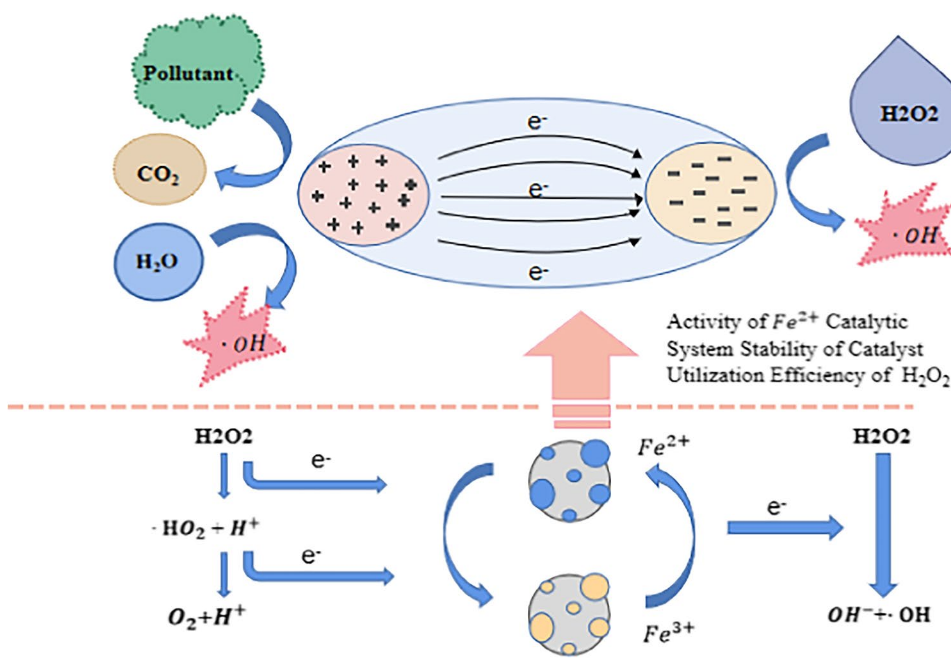

Fig. 3 The cyclical activity of the Fe^{2+} catalytic system maintains the stability of the catalyst and results in the utilization of H_2O_2 (Lv and Hu 2017)

Synergistic method to remove *Microcystis aeruginosa*

Photocatalysis/O_3 combined oxidation

The principle of this technology is as follows: First, the photocatalyst generates (e^-)-hole pairs (h^+), as shown in reaction formulas (15)–(18), which is a process that produces ·OH. At the same time, catalyzing O_3 provides more ·OH as shown in Formulas (19)–(21). It can be concluded that a molecule of O_3 can generate 2 ·OH if it receives a photon or undergoes catalysis, and the efficiency of the reaction is very high. Second, O_3 molecules will react with e^- to form $\cdot O_3^-$ (see reaction Formula (22)). The photogenerated e-reaction can reduce the recombination rate of e^- and h^+ which is conducive to the efficient reaction of h^+ and which generates more ·OH. At the same time, O_2 also reacts with e^-, as shown in Formulas (17)–(19). Moreover, $\cdot O_3^-$ in an acidic solution will react to form ·OH, as shown in Formulas (24)–(25). In addition to the above reactions, there are other reactions that occur, such as (26)–(29) (Li et al. 2021a). The overall process is shown in Fig. 4. These reactions produce hydroxyl radicals (·OH) needed for AOPs that can be used to treat sewage or harmful algal blooms. They illustrate the efficient part of the whole process; that is, not only ·OH but also O· and $\cdot O_3^-$ with strong oxidizing properties are produced. Therefore, AOP method is a promising way of water treatment.

$$H_2O + h^+ \rightarrow \cdot OH + H^+ \quad (15)$$

$$OH^- + h^+ \rightarrow \cdot OH \quad (16)$$

$$O_2 + e^- \rightarrow \cdot O_2^- \quad (17)$$

$$\cdot O_2^- + e^- + 2H^+ \rightarrow \cdot HO_2 \quad (18)$$

$$\cdot HO_2 + hv \rightarrow 2 \cdot OH \quad (19)$$

$$O_3 + h\nu \rightarrow O_2 + O \quad (20)$$

$$O_3 + \text{catalyst} \rightarrow O_2 + O \quad (21)$$

$$O \cdot + H_2O \rightarrow 2 \cdot OH \quad (22)$$

$$O_3 + e^- \rightarrow \cdot O_3^- \quad (23)$$

$$\cdot O_3^- + H^+ \rightarrow HO_3 \cdot \quad (24)$$

$$HO_3 \cdot \rightarrow O_2 + \cdot OH \quad (25)$$

$$O_3 + H^+ \rightarrow HO \cdot + O_2 \quad (26)$$

$$O_3 + OH^- \rightarrow HO_2 \cdot + O \cdot_2 \quad (27)$$

$$O_3 + OH_2- \rightarrow \cdot O_3^- + HO_2 \quad (28)$$

$$O_3 + H_2O \rightarrow O_2 + \cdot HO_2 \quad (29)$$

UV/H2O2 synergistic oxidation

UV/H_2O_2 synergistic oxidation is promising because it is inexpensive, the addition of H_2O_2 and UV irradiation operations are relatively simple, and it does not cause secondary pollution. Regardless of whether it uses H_2O_2 or UV irradiation,

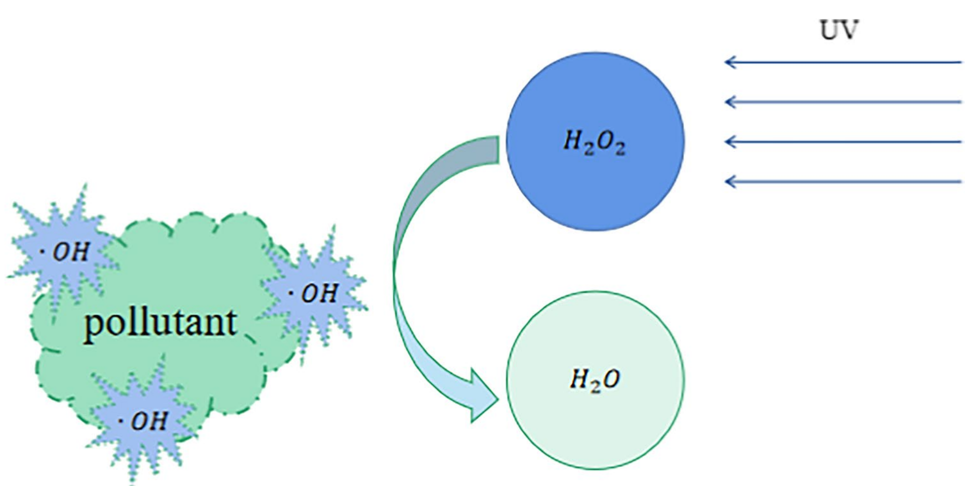

Fig. 4 The principles of UV/H_2O_2 combined technology (Tan et al. 2013)

studies have shown that blue-green algae have a more sensitive response to this treatment than eukaryotic algae and aquatic plants (Sun 2020). Studies have reported that the increase of H_2O_2 may cause oxidative stress by forcing the algae to produce ROS. Under certain oxidative conditions, algae can resist strong oxidation from the outside through their antioxidant defense system which is comprised of enzymes such as superoxide dismutase and catalase. However, when the oxidative level is relatively high, the enzymatic and non-enzymatic antioxidant systems are destroyed, leading to cell death. The principle mechanism is shown in Fig. 5 (Wang 2018a).

UV/chloramine oxidation

During a process called photolysis which utilizes a low-pressure mercury lamp emitting 254-nm light, oxidants with a high molar absorption coefficient and quantum yield are produced. Thus, generating free radicals from HOCl and OCl^- could be 30–75% more efficient than generating free radicals from H_2O_2. For this reason, the UV/chloramine (UV/NH_2Cl) AOP has received some attention. In principle, UV photolysis of NH_2Cl produces ·Cl and amino radicals (·NH_2; Eq. (30)). Although ·NH_2 is relatively inactive, ·Cl can directly degrade pollutants or form ·OH as shown by the following Eqs. (30)–(33) (Chuang et al. 2017) and in Fig. 6.

$$NH_2Cl + hv \rightarrow \cdot NH_2 + \cdot Cl \quad (30)$$

$$\cdot Cl + \cdot OH^- \leftrightarrow ClOH \quad (31)$$

$$\cdot Cl + H_2O \leftrightarrow ClOH\cdot^- + H^+ \quad (32)$$

$$ClOH\cdot^- \leftrightarrow \cdot OH + Cl^- \quad (33)$$

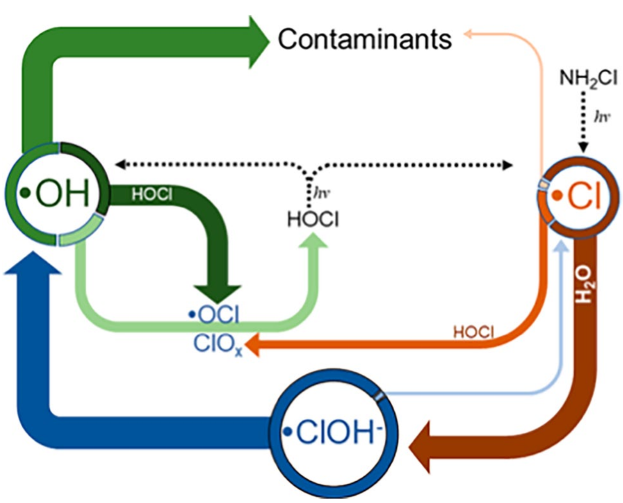

Fig. 5 The UV/NH_2Cl oxidation principle (Chuang et al. 2017)

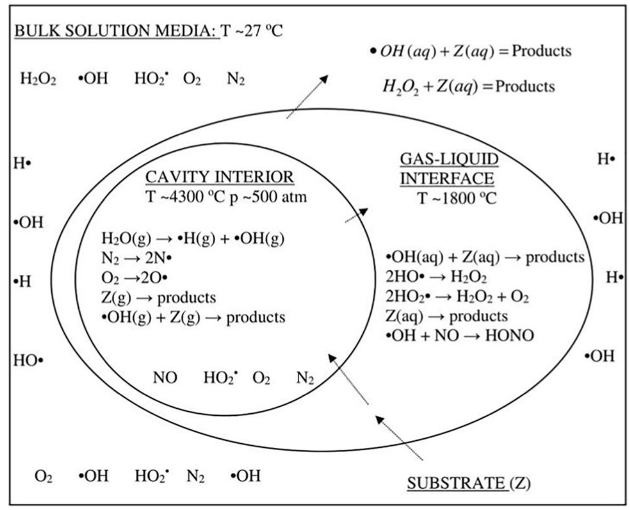

Fig. 6 The three reaction zones in the cavitation process (Gągol et al. 2018)

AOPs and cavitation synergy

Hydrodynamic cavitation has been widely used in water treatment to, for example, help biological, physical, and chemical methods remove organic contaminants. Cavitation can more effectively generate hydroxyl radicals responsible for oxidizing pollutants. It also creates local hot spots in the media being processed, wherein implosion bubbles cause a sudden increase in pressure and temperature and soon after separate contaminants by pyrolysis. In addition, volatile organic pollutants penetrating the cavity being formed coupled with a high-energy implosion help make the oxidation process more effective. The dissipated energy of cavitation bubbles is usually used to destroy the structure of microorganisms and bacteria present in the wastewater.

The use of hydrodynamic cavitation in sewage treatment ensures a significant increase in the oxidation efficiency of pollutants. As a result, when a chemical oxidant capable of generating hydroxyl radicals characterized by a high oxidation potential is introduced into the medium to be treated, the following phenomena occur: First, cavitation bubbles concentrate free radicals and homogenize the system, thereby improving the reduction efficiency. Second, cavitation enhances the chemical generation of highly active hydroxyl radicals that can oxidize various organic pollutants such as carboxylic acids, pesticides, alcohol, chlorinated solvents, or drug (Gągol et al. 2018).

Prospects

At present, advanced oxidation technology is still an emerging water treatment technology. Its advantages include high efficiency, strong universality, and thoroughness. However, due to its high processing cost and huge material consumption, it cannot be widely used, and it is still a long way from industrialization. The current issues that need to be overcome are as follows: (1) Research needs to be done on the production mechanism of ·OH or SO_4^{2-} to improve production efficiency; (2) these costs of processing methods should be reduced, perhaps by combining them with traditional processes.

Author contribution Qingyun Zhai: conceptualization, methodology, writing—original draft. Lili Song: validation, formal analysis, investigation, data curation. Xiyan Ji: supervision; conceptualization; review and editing; funding acquisition. Yueshu Yu: data curation, formal analysis. Jing Ye: conceptualization, review and editing. Wenwu Xu: visualization. Meifang Hou: review and editing.

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Availability of data and materials The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethical approval This research does not involve ethical issues.

Consent to participate This research does not involve ethical issues.

Consent to publish All authors confirm that this paper has not been published before in any form.

Competing interests The authors declare no competing interests.

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