

Review Article

Strategies for Solving the Issue of Malachite Green Residues in Aquatic Products: A Review

Jiang He ^{1,2,3} Ping Mo ^{1,2,3} Yu-Shuang Luo ^{1,2,3} and Pin-Hong Yang ^{1,2,3}

¹College of Life and Environmental Science, Hunan University of Arts and Science, Changde 415000, Hunan, China

²Collaborative Innovation Centre for Efficient and Health Production of Fisheries in Hunan Province, Changde 415000, Hunan, China

³Key Laboratory of Health Aquaculture and Product Processing in Dongting Lake Area of Hunan Province, Changde 415000, Hunan, China

Correspondence should be addressed to Jiang He; hejiang@huas.edu.cn

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Malachite green (MG) residue in aquatic products is a widely concerning issue, and the possible source of MG contamination includes its illegal usage and environmental pollution. A variety of strategies for solving such a problem have been proposed, and the research about them is summarized in this review. The MG contamination in aquaculture environments can be eliminated by adsorption, degraded by advanced oxidation processes (AOPs), or biodegraded by microbes or enzymes. The illegal usage of MG can be prevented by screening novel anti-*Saprolegnia* sp. agents from current available agricultural antibiotics, plant extracts, or antagonistic microbes. Nevertheless, deficiencies also existed in these proposed solving strategies. Therefore, further research opportunities in such areas were provided. This includes developing effective combinatorial methods (adsorption + AOPs or biodegradation) for eliminating MG from the aquaculture environment; systematically considering the impact of practical conditions on the efficiency of MG elimination; screening more efficient anti-*Saprolegnia* sp. agents; and systematically evaluating both the in vivo activities and safety of these agents.

1. Introduction

Aquatic products are high-quality protein sources in the human diet; therefore, the safety of them, especially the issue of chemical contaminants, is widely concerning. Familiar chemical contaminants in aquatic products include inorganic toxic elements, persistent environmental pollutants, and drug residues [1, 2]. Although numerous chemicals might contaminate farmed and wild fishes, malachite green (MG) seems to be the most concerning. MG has been used as a therapeutic agent for *Saprolegnia* sp. infection in aquaculture for a long time. However, the use of MG in food fishes has been banned worldwide since the 2000s due to its possible carcinogenicity in mammals. Even so, MG and its metabolite, leucomalachite green (LMG), are still frequently detected in aquatic products from all around the world. For example, the previous survey result of our group indicated

that 10.6% of the analysed live aquatic products were MG-positive [3]. Some published results of MG residue detection in fishes from different countries are listed in Table 1.

One of the possible reasons for the high positive ratio of MG contamination in aquatic products is the illegal use of this substance as a therapeutic agent in the aquaculture process. However, the research results of Schuetze *et al.* [4] revealed that MG residues were also detected in wild eels (*Anguilla anguilla*). Therefore, another source of MG residues in aquatic products may be the contamination of the aquaculture environment, i.e., water. Although its application in food fishes is illegal, MG is a multiple-use substance with several legal applications. For example, using it as a therapeutic agent for ornamental fish is still legal. In addition, MG is also a dye widely used in industrial fields such as textiles and lab research areas such as microbial staining. All of these legal applications, with the addition of the

production sites of MG, are potential sources of MG contamination in the aquaculture environment. The possible sources of MG contamination in aquatic products are summarized in Figure 1.

Global scientific researchers have put effort into solving the issue of MG residues in aquatic products. The effect of the cooking process [11, 12] and treatments such as photo degradation [13] and depuration [14] on the elimination of MG residues from fish tissues is very limited; therefore, it is more reasonable to prevent MG contamination during the aquaculture process. For the contamination of MG in aquaculture environments, strategies for purifying MG-contaminated wastewater have been proposed, which include adsorption by various adsorbents, degradation by advanced oxidation processes (AOPs), and biodegradation by microbes or enzymes. For the illegal use of MG during the aquaculture process, strategies that screen MG alternatives for the treatment of fish and fish eggs were proposed (Figure 1). To provide the current situation of scientific research on these strategies and therefore determine the future research opportunities, related literature searching from the “web of sciences” was summarized, and our perceptions were also provided in this review.

2. Elimination of MG from the Aquaculture Environment by Adsorption

Adsorption is a common method for eliminating dyestuffs in the water system, and the selected adsorbent is the most critical factor determining its effectiveness. A wide variety of adsorbents, for example, natural or modified clayey soil [15, 16], anaerobic granular sludge [17], perlite [18], natural zeolite [19], silica gel [20, 21], and alumina [20], have been used to remove MG from aqueous solutions. However, bio-waste-derived materials and artificially synthesized materials are two kinds of adsorbents with more attention (Figure 2).

Nonplant bio-wastes such as hen feathers [22], snail shells [23], sea shells [24], and eggshells [25] have been reported with well MG adsorption capacity. However, waste of fruits, vegetables, grain crops, and woody plants are more common bio-wastes that are used as sources of adsorbents for MG elimination. Generally, these bio-wastes were only physically treated by steps such as washing, drying, and smashing, and then directly used as adsorbents. In our group, wastes such as sugarcane rinds, orange peel, and fish scale were treated by these steps and successfully used as adsorbents for eliminating MG from water (data unpublished). However, various modifications can be applied to improve the adsorption capacity further. For example, rice straw was thermos-chemically modified with citric acid as the esterifying agent by Gong *et al.* [26]; and the maximum adsorption capacity for MG was improved from 94.34 mg/g to 256.41 mg/g. It is estimated that more than 1.1 billion tons of rice straw are left behind annually, which indicates the economic benefit of using rice straw in this process [27]. Similarly, when methanol was used as a solvent and triethylamine as a modification agent for sawdust treatment, the maximum adsorption capacity for MG was improved from 94.0 mg/g to 697.9 mg/g [28]. Bio-wastes can also be

prepared as activated carbon (AC), which is a well-known adsorbent for dye-stuff elimination. In this situation, the preparation method is critical in determining the adsorption capacity of AC. Generally, AC can be prepared by thermal treatment, chemical treatment, or a chemical-thermal process. Rajgopal *et al.* [29] showed that the adsorption capacity for MG was thermal activated carbon > acid + thermal activated carbon > commercial activated carbon > acid activated carbon.

Bio-waste-derived materials possess adequate MG adsorption capacity, with the adsorption capacity varying from dozens to hundreds of mg/g. However, researchers are focusing on preparing artificially synthesized materials and using them as more efficient adsorbents for MG removal, recently. Magnetic nanoparticles (MNPs) are typical examples of such kinds of adsorbents. Although MNPs based on cobalt ferrite (CoFe_2O_4) were reported [30], most MNPs were based on Fe_3O_4 that were prepared by the precipitation method using ferrous chloride and ferric chloride as raw materials. The prepared MNPs were then stabilized by special reagents or further functionalized by coating with special materials [31–36]. For example, biopolymers and Fe_3O_4 MNPs-based nanocomposites were synthesized by Mittal *et al.* [33], and the maximum adsorption efficiency for MG was up to 497.15 mg/g. Carbon nanotubes (CNTs) are another kind of synthesized nanoabsorbent that is widely used for MG removal. Similar to MNPs, CNTs were firstly prepared and then further treated by special procedures as stated by [37–39]. For example, N-doped CNT stabilized Cu_2O nanoparticles were fabricated by Li *et al.* [39]; and the maximum adsorption efficiency for MG was up to 1495.46 mg/g. Cellulose-based nanoadsorbents, which are derived from natural cellulose but with artificial modifications, were also successfully used to remove MG with high efficiency. For example, the reported maximum adsorption efficiency of cellulose nanocrystals (CNCs) [40] and cellulose nanofibril aerogels [41] to MG was 212.7 mg/g and 243.9 mg/g, respectively. Apart from MNPs, CNTs, and cellulose-based nanomaterials, several other nanocomposites also showed high adsorption efficiency to MG, such as nanoiron oxide-loaded alginate microspheres [35], zeolite nanostructures [42], iron nanoparticles loaded on ash [43], bimetallic Fe–Zn nanoparticles [44], kappa-carrageenan-g-polyacrylic acid/ TiO_2 - NH_2 hydrogel nanocomposite [45], gum xanthan/psyllium-based nanocomposite [46], and hematite-reduced graphene oxide composites [47]. In addition, reports using graft copolymer derived from amylopectin and poly(acrylic acid) [48], graphite oxide/polyurethane foam material [49], chitosan beads [50], and reinforced hybrid porous beads that are prepared with alginate, aluminum-pillared montmorillonite, polyvinyl alcohol, and CaCO_3 [51] for MG removal also exist in literature. However, adsorbents that possess the best adsorption efficiency to MG were reported by Zheng *et al.* [52] and Lin and Chang [53]. In the work of Zheng *et al.* [52], granular composite hydrogels were prepared using chitosan as the grafting backbone, acrylic acid and itaconic acid as the monomers, and attapulgite as the inorganic component, and the saturation adsorption capacity of MG can be achieved at 2433 mg/g. While zeolitic

TABLE 1: Some survey results of malachite green (MG) contamination in aquatic products.

No.	Country (state or province)	Aquatic products	Number of samples	Detection method	Ratio of positive samples (%)	Total of MG and LMG (mg kg ⁻¹)	References
1	Germany (Berlin)	Eels caught downstream from municipal sewage treatment plants	45	LC-MS/MS	55.56	0.04–0.77	[4]
2	Croatia	Carp and rainbow trout	72	ELISA	68.10	0.1–0.3	[5]
3	Iran	Farmed rainbow trout	144	HPLC-DAD	48.60	0.30–146.1	[6]
4	China (Hunan)	Chinese soft-shelled turtle, Mandarin fish, and grass carp	180	HPLC-FLD	10.56	0.6–7.0	[3]
5	Iran	Farmed rainbow trout	120	HPLC-DAD	58.40	0.02–0.89	[7]
6	Malaysia	Red tilapia, African catfish, barramundi, hybrid grouper, and striped catfish	37	LC-MS/MS	43.20	0.53–4.10	[8]
7	Armenia	Sevan trout, rainbow trout, and sturgeon	29	LC-MS/MS	34.50	1.1–4.8	[9]
8	United States (Northeast)	Retail catfish fillets	362	ELISA	22.10	n.a.	[10]

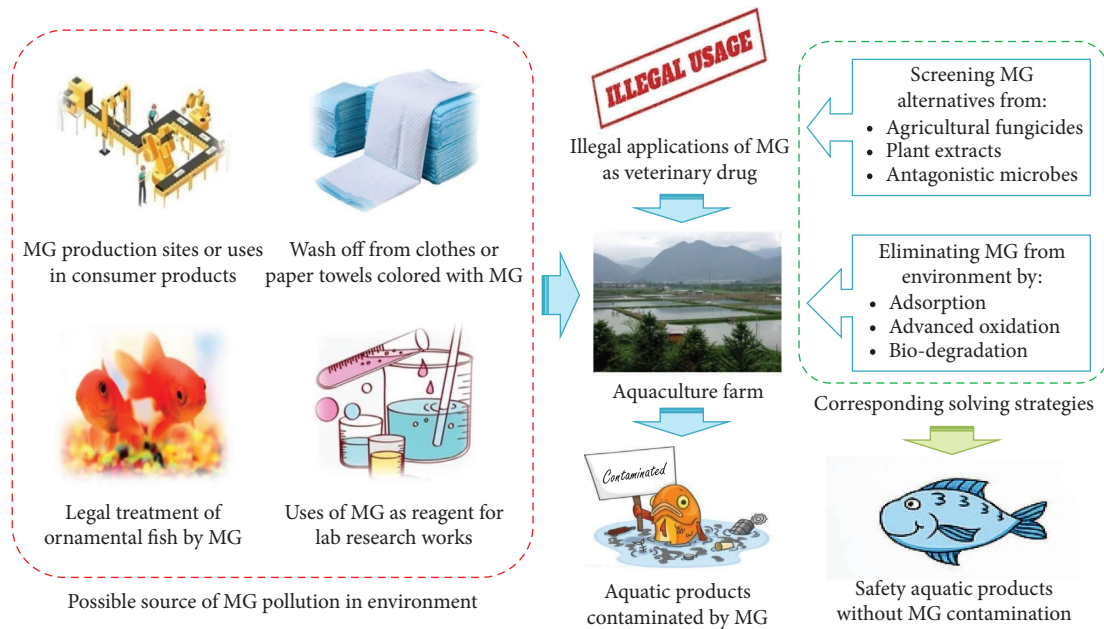


FIGURE 1: Possible sources of malachite green (MG) contamination in aquatic products and corresponding solving strategies.

imidazole framework-67 was synthesized by Lin and Chang [53], its adsorption capacity can be achieved at about 3000 mg/g in optimum conditions.

In the abovementioned works, the adsorption mechanism was investigated and the adsorption conditions were optimized by researchers. For mechanism investigation, the adsorbents were generally characterized by techniques such as scanning electron microscopy (SEM), Fourier-transform infrared spectroscopy (FTIR), energy dispersive X-ray spectroscope (EDS), Brunauer-Emmette-Teller (BET) surface area analysis, and Raman spectroscopy [30, 33]. Therefore, information about the surface structure, surface area, surface charge, and the presence of functional groups on the adsorbents can be obtained to illustrate the

adsorption mechanism. Generally, porous structures and carboxylic groups have been proven to be beneficial for MG adsorption. In addition, adsorption kinetic studies, equilibrium studies, and thermodynamic analysis were also generally conducted [54, 55]. The obtained data were further fitted with specific models to investigate the inherent mechanism of the adsorption process. The adsorption process can be performed under the batch model or the column model. No matter which model was applied, the impact of a series of factors on the adsorption was investigated, and the adsorption conditions were optimized. When the batch model was used, the considered factors generally included initial dye concentration, contact time, pH, temperature, adsorbent dose, ionic strength, and

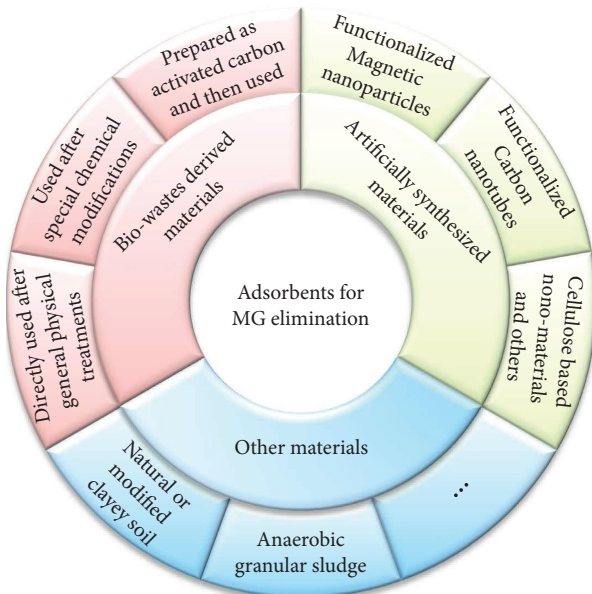


FIGURE 2: Typical adsorbents for eliminating malachite green (MG) from contaminated water.

agitation [56, 57]. While in the column model, the impact of flow rate and bed depth should be carefully considered [35]. According to the literature, pH conditions have a significant effect on the adsorption process of MG. Generally, alkaline ambience favors MG adsorption because MG might be competing with the H^+ ion in the solution so that more negligible adsorption is found [58]. In addition, ultrasound irradiation and microwave can significantly enhance the mass transfer from adsorbate to the adsorbent; therefore, ultrasound-assisted [59, 60] and microwave-assisted [61] adsorption approaches were also developed for MG elimination and improved performances were achieved.

3. Degradation of MG in the Aquaculture Environment by Advanced Oxidation Processes (AOPs)

AOPs, such as chemical oxidation and photochemical oxidation, were another kind of method that was widely applied in the area of eliminating MG in the water environment (Figure 3). In this process, reactive oxygen species (ROSs) are generated in situ, and then the organic pollutants are oxidized. The advantages of AOPs include rapid oxidation reaction rate, high mineralization efficiency, and environmental-friendly.

Fenton oxidation is a typical chemical oxidation technique successfully applied for MG degradation. The general principle of such practice is based on the chain reaction of Fe^{2+} and H_2O_2 to form the OH radical. Hameed and Lee [62] investigated the degradation of MG in aqueous solution by the Fenton process, and 99.25% degradation was achieved after 60 min of treatment under optimized conditions. According to the principle of the Fenton process, Fe^{2+} is oxidized to Fe^{3+} and then regenerated by the reaction with H_2O_2 [63, 64]. However, the rate of Fe^{2+} regeneration is

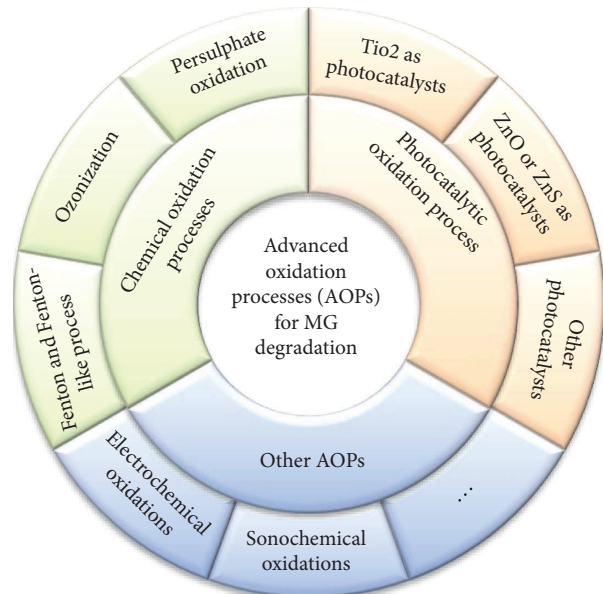


FIGURE 3: Typical advanced oxidation processes (AOPs) for degrading malachite green (MG) in contaminated water.

much slower than its oxidation, and therefore the Fenton oxidation efficiency is mainly determined by the rate of Fe^{2+} regeneration [65]. In addition, the optimum pH condition for traditional Fenton oxidation is in a narrow range (near 3), which greatly limits the practical application of such a technique [66]. Therefore, the conventional Fenton reaction was modified to solve these two issues. For example, the [Fe(III)-salen]Cl complex Fenton system was established by Bai et al. [65] to improve the rate of Fe^{2+} regeneration, and the EDTA-Fe(III) Fenton-like oxidation system was established by Hu et al. [66] to facilitate the degradation under neutral conditions. Satisfactory results were obtained when these two modified Fenton systems were applied for MG degradation in contaminated water. Apart from Fenton oxidation, other chemical oxidation techniques used for MG degradation include ozonisation [67] and persulphate oxidation [68], and satisfactory results were also obtained in these processes.

Photochemical oxidation is a simple, clean, and relatively inexpensive AOP method that has aroused a wide interest in applying it in MG degradation. Although photo degradation of MG was observed when there was a direct exposure of its solution to natural sunlight [69] or UV-Vis light with a specific wavelength [70, 71], photo-oxidation processes coupled with a specific photocatalyst were more commonly used. The theory of the photocatalytic oxidation process has been widely illustrated in the literature, in which the properties of the photocatalysts are critical. Many photocatalysts have been prepared and applied in the photocatalytic degradation of MG in contaminated water, but TiO_2 is the most broadly used one. In the published works, TiO_2 was often prepared as nanotubes [72] or nanoparticles [73, 74], and sometimes it was immobilized on glass materials [75]. To further improve the photodegradation efficiency, diverse TiO_2 -composite materials were proposed.

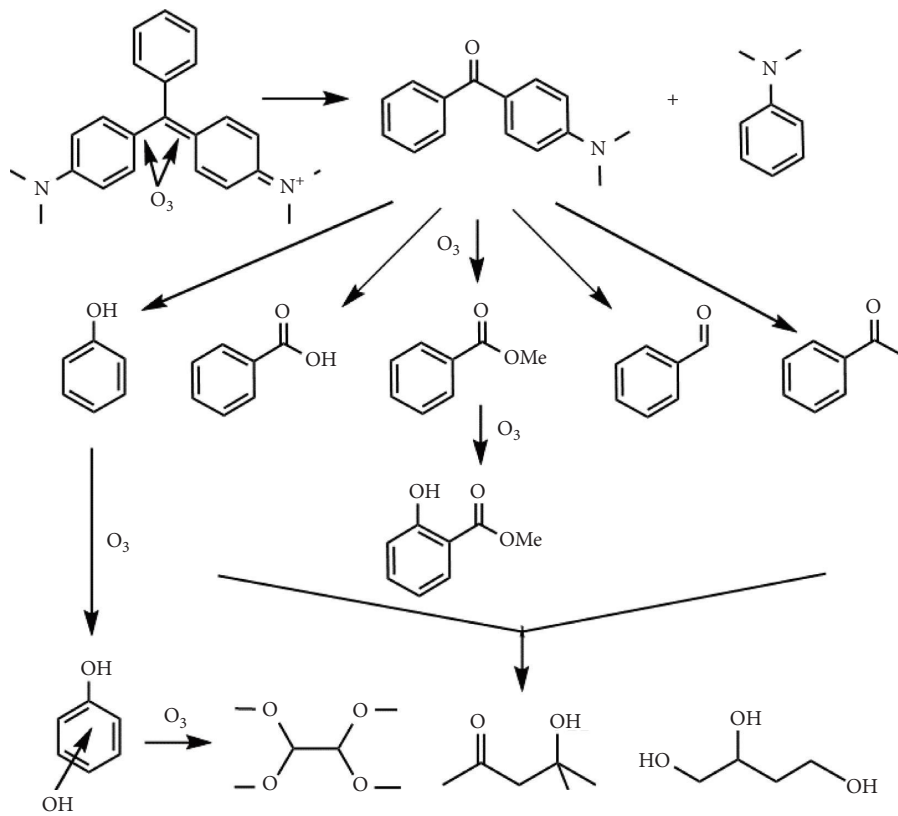
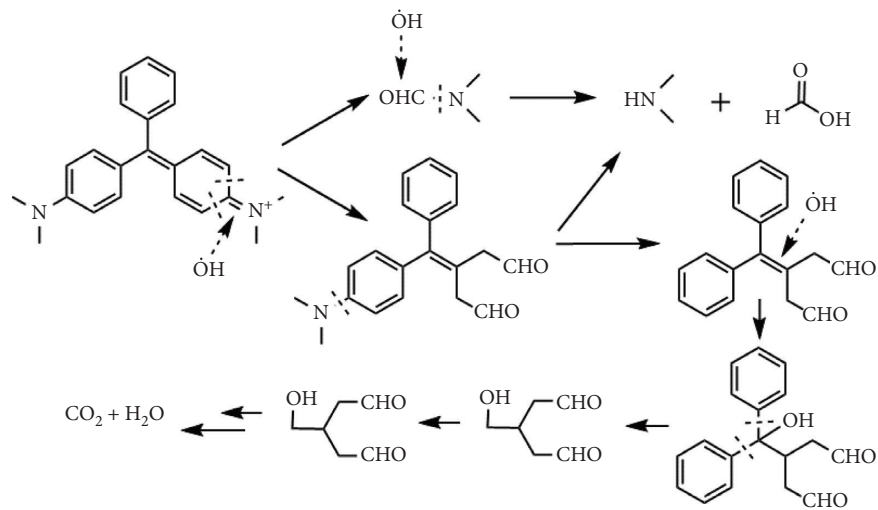


FIGURE 4: Continued.

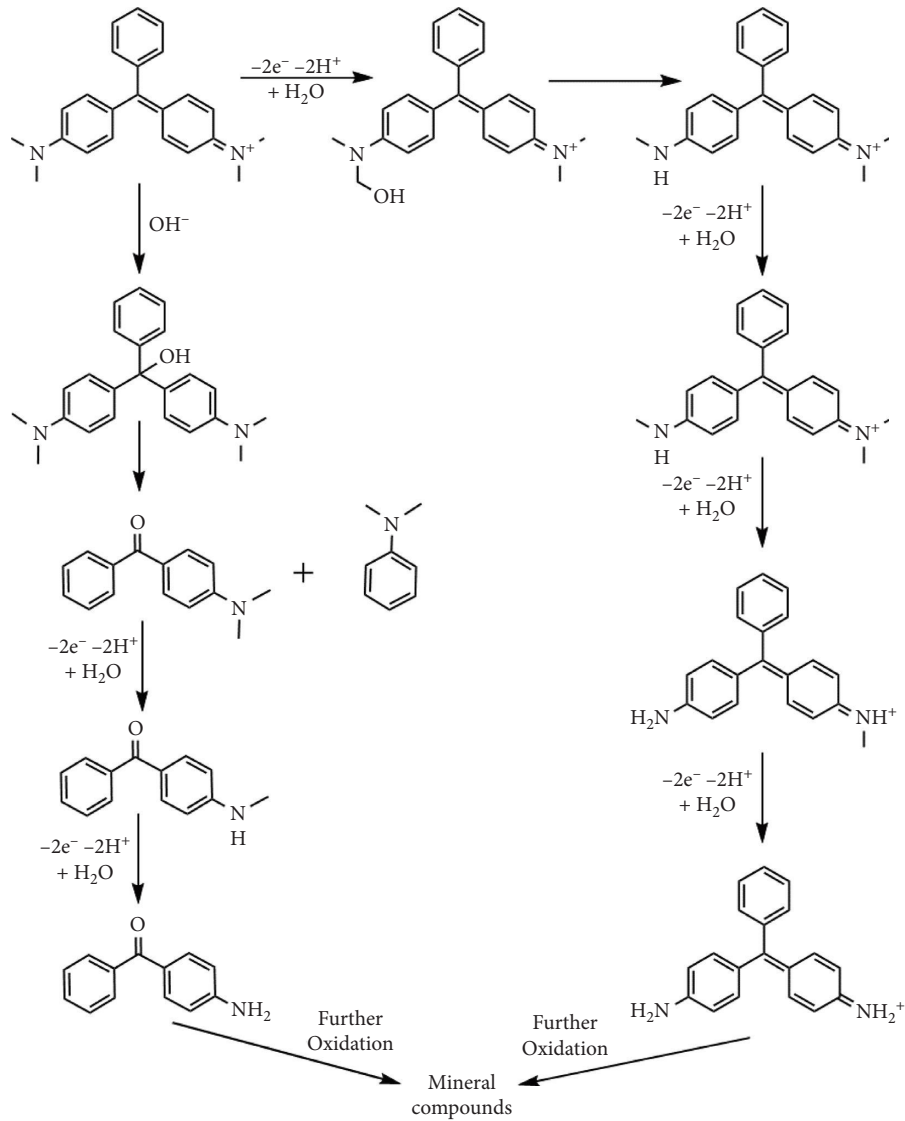


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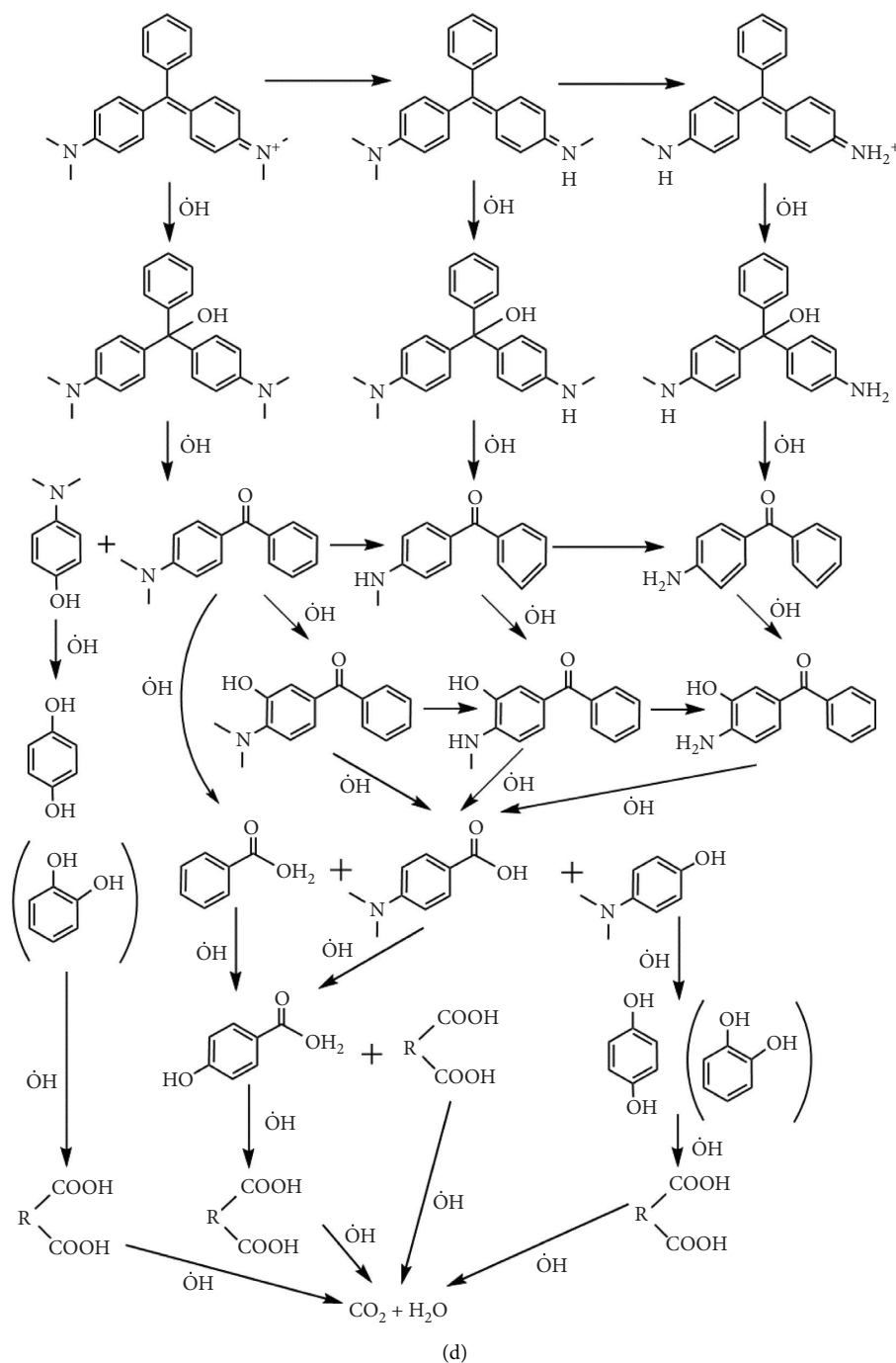


FIGURE 4: Proposed degradation pathways of malachite green (MG) under different advanced oxidation processes (AOPs). (a) Fenton oxidation [65]; (b) ozonization [67]; (c) electrochemical oxidation [88]; and (d) photochemical oxidation [91].

For example, TiO₂ nanotubes functionalized with CuS quantum dots [76], nanosized hybrids of TiO₂/SiO₂ [77], Sn-doped TiO₂ nanostructured thin films [78, 79], bidoped TiO₂-ZrO₂ ferromagnetic nanocomposites [80], and Ni_x:TiO₂ thin films [81]. ZnO or ZnS is another category of photocatalyst that has been applied in the photodegradation process of MG. As the same to TiO₂, it has been used alone in the form of quantum dots [82], thin films [83], nanoparticles [84] and powders [85], or prepared into composites such as

Pt-doped ZnO [86], Mn-doped ZnS [84], Fe³⁺-doped ZnS [82], and a hybrid of ZnO nanorods grown on 3D reduced graphene oxide/Ni foam [87]. Novel photocatalysts are constantly being synthesized and successfully applied for the photodegradation of MG, which illustrates the great interest of researchers in such a category of AOPs. Apart from the properties of the photocatalyst, the pH has also been confirmed to significantly impact the photodegradation velocity of MG in the simulated water samples. In addition,

TABLE 2: Isolation source and activity of some typical malachite green (MG) degradation bacteria that have been published since 2017.

Bacteria	Isolation source	Activity	Test sample	References
<i>Aeromonas hydrophila</i>	Activated sludge	96.8% within 12 h	200 mg/L in medium	[103]
<i>Aeromonas</i> sp.	Tanning mill	Almost 100% within 36 h	<1000 mg/L in medium	[104]
<i>Bacillus vietnamensis</i>	Sediments from the Eastern Arabian Sea	100% within 4 h	50 mg/L in medium	[105]
<i>Enterobacter</i> sp.	Textile effluents and sludge	100% within 78 h–144 h	15 mg/L in medium	[106]
<i>Klebsiella aerogenes</i>	Leaves of the wetland plant <i>Suaeda salsa</i>	100% within 8 h	100 mg/L in medium	[107]
<i>Klebsiella pneumoniae</i>	Sediments from MG-contaminated aquaculture ponds	100% within 12 h	1–10 mg/L aqueous solution	[108]
<i>Pseudomonas</i> sp.	Activated sludge	100% within 12 h	1000 mg/L in medium	[109]
<i>Pseudomonas veronii</i>	Activated sludge	93.5% within 7 days	50 mg/L in medium	[110]
<i>Tenacibaculum</i> sp.	Pacific deep-sea sediments	98.8% within 12 h	20 mg/L in medium	[111]

Escherichia coli and sucrose dissolved in contaminated water can suppress the photodegradation performance of MG.

Other AOPs that have been successfully applied for the degradation of MG in water include electrochemical oxidation and sonochemical oxidation. In terms of mechanism, electrochemical oxidation was performed in two ways, i.e., direct and indirect oxidation. In direct oxidation, MG was destroyed after direct electron transfer at the electrode surface, while in indirect oxidation, MG was degraded by electro-generated ROSs [63, 64]. The electrocatalytic degradation of MG in aqueous solution with various self-prepared electrodes was investigated by Amin and Davood [88], and their results indicate that the G/ β -PbO₂ electrode has the best degradation efficiency. Sonochemical oxidation also involves direct and indirect mechanisms. The direct mechanism means the formation of cavitation bubbles that grow and then collapse, creating strong breaking forces with extremely high temperatures and pressures. The indirect mechanism also means the oxidation by generated ROSs [63, 64]. The impact of operational parameters on the sonochemical degradation of MG was once investigated by Behnajady et al. [89]; and the results illustrated that OH has a vital role in the degradation process. An intensified sonochemical oxidation process was proposed by Moumeni and Hamdaoui [90] for MG degradation. In this process, bromide is added to the oxidation system and induces a considerable enhancement in MG degradation efficiency.

In some of the abovementioned works, the degradation products of MG were identified by methods such as GC-MS and (or) LC-MS, and the possible degradation pathway of MG was then proposed. Although the degradation pathway of MG in different AOPs was variant Figure 4, the high mineralization efficiency of these approaches was confirmed. The complete degradation of MG into CO₂ and H₂O illustrates the advantage of AOPs compared to other elimination methods. In addition, two or more AOPs can be combined to achieve higher MG mineralization efficiency. For example, sono-photocatalytic degradation [92], photo-fenton degradation [93], electro-fenton degradation [94], photo-electro-fenton degradation [94], and photo-electrochemical degradation [95], have been utilized.

4. Biodegradation of MG in the Aquaculture Environment by Microbes or Enzymes

Microbes can eliminate pollutants through adsorption or degradation. Typical examples of microbes with adsorption capacity of MG include the freshwater algae *Pithophora* sp. [96], bacteria *Bacillus cereus* [97], photoautotrophic cyanobacterium *Synechococcus elongates* [98], and microalgae *Desmodesmus* sp. [99]. However, biodegradation of chemical pollutants by a microbe or its enzyme is a more sustainable approach than the adsorption method, and several review articles have focused on this topic [100, 101]. Many scientists have been engaged in the isolating and identifying of microbes with MG degradation ability, and numerous relevant scientific papers have been published.

A wide range of prokaryotic and eukaryotic microbes have been reported to result in satisfying biodegradation activity of MG. Although the filamentous actinobacteria *Streptomyces chrestomyceticus* [102] was recently reported, most of the reported prokaryotic microbes with well MG degradation activity belong to bacteria. Some recently isolated MG-degradation bacteria are listed in Table 2, which include *Aeromonas* sp., *Bacillus* sp., *Enterobacter* sp., *Klebsidella* sp., *Pseudomonas* sp., and *Tenacibaculum* sp. In our group, a *Pseudomonas* sp. strain was also isolated from sediments collected from an aquaculture pond. Its MG degradation efficiency within 14 h was over 80% when the initial concentration of MG was 50 mg L⁻¹ (data unpublished). Microalgae *Cosmarium* sp., which belongs to eukaryotic microbes, is capable of degrading MG. However, the most reported MG-degrading eukaryotic microbes are fungi, especially filamentous fungi. Single-cell fungi *Saccharomyces cerevisiae* [112] and *Debaryomyces nepalensis* [113], nonbasidiomycete filamentous fungi *Aspergillus Niger* [114], *Aspergillus flavus* [115], *Penicillium* sp. [116–117], *Cunninghamella elegans* [119], and *Myrothecium roridum* [116, 120], and white-rot fungi *Pycnoporus cinnabarinus*, *Pleurotus pulmonarius*, and *Fomes fomentarius* [121] have been confirmed with different MG degradation activities.

For the abovementioned microbes, the time required to degrade the MG in a model sample completely ranged from several hours to several days. However, the initial content of MG in the model samples varied in different studies.

TABLE 3: The anti-*Saprolegnia* activity of some typical phytochemicals.

Plant extracts	MIC [#] ($\mu\text{g/mL}$)	Target <i>Saprolegnia</i> sp.	References
Carvacrol	40	<i>S. ferax</i>	[137]
Thymol	100	<i>S. ferax</i>	[137]
Thymoquinone	150	<i>S. parasitica</i>	[148]
Thymoquinone	125	<i>S. salmonis</i>	[148]
Hinokitiol	12.5	<i>S. parasitica</i> and <i>S. diclina</i>	[149]
Citral	200	<i>S. parasitica</i> and <i>S. diclina</i>	[149]
Allylisothiocyanate	100	<i>S. parasitica</i> and <i>S. diclina</i>	[149]
Rhein	20	<i>Saprolegnia</i> sp. JL (HM 637287)	[150]
Aloe-emodin	50	<i>Saprolegnia</i> sp. JL (HM 637287)	[150]
Honokiol	5–10	<i>S. parasitica</i>	[154]
Magnolol	5–10	<i>S. parasitica</i>	[154]
Tomatine	99.4	<i>S. parasitica</i>	[155]
Piperine	71.3	<i>S. parasitica</i>	[155]
Plumbagin	47.0	<i>S. parasitica</i>	[155]
Curcumin	16	<i>S. parasitica</i> and <i>S. australis</i>	[140]
Eugenol	25	<i>S. parasitica</i> and <i>S. australis</i>	[140]
Cinnamaldehyde	25	<i>S. parasitica</i> and <i>S. australis</i>	[140]

[#]Minimum inhibitory concentration for vegetative growth.

Therefore, it is hard to accurately compare the MG degradation activities of microbes obtained from different scientific groups. Even so, the MG degradation efficiency of *Deinococcus radiodurans* was up to 97.2% within 30 min [122], and this strain seems to be the most efficient MG degradation microbe. In some of the published works referenced above, the impacts of environmental factors on the MG degradation activities were investigated, and the degradation conditions were also optimized [104, 108]. Furthermore, the degradation products, active enzymes, and related genes for some microbes were identified, and therefore the possible degradation pathway of MG for these microbes was proposed [105, 107]. Reduction to leuco form, N-demethylation and reductive splitting of the triphenyl rings were involved in the most proposed MG degradation pathways [105, 107]. However, mineralization, i.e., thorough degradation of MG molecules was hard to fulfill by microbes, and even the degradation products might still retain toxicity. Laccase (benzenediol: oxidoreductase, EC 1.10.3.2), and several other enzymes are related to the MG degradation process in microbes, and the addition of small molecules as redox mediators can improve the rate of laccase-mediated MG degradation [105]. Although most of the MG degradation microbes were utilized as free cells, the MG degradation efficiency of immobilized *Burkholderia cepacia* [125] and *Pseudomonas* sp. [117] was also investigated. Most of the reported MG degradation microbes only show activity in the culture medium. However, the *Klebsiella pneumonia* WA-1 isolated by Li et al. [108] could efficiently degrade MG in an aqueous solution without any nutrients, which illustrates the great potential for practical application of this strain.

Enzymatic degradation of MG was also widely concerned in recent years. Although manganese peroxidase isozyme H4 (MnPH₄)-mediated MG degradation was reported by Saravanakumar et al. [125], most of the publications in this area used laccase from different sources. For example, laccase from *Trichoderma asperellum* was applied for degradation of MG by Shanmugam et al. [126]; and

97.18% degradation was achieved within 98.58 min when the initial content of MG was 122.66 mg/L, the enzyme concentration was 1.50 U/mL, and pH was 6.75. In addition, the kinetics study indicated that the K_m and V_{max} of *Trichoderma asperellum* laccase were 87.50 mg/L and 1.28 mg/L/min, respectively [126]. A heterologous expressed laccase was prepared by Xu et al. [127] and successfully applied for MG degradation. Besides the free enzyme, immobilized laccase has also been utilized for MG degradation. For example, laccase from *Bacillus subtilis* was immobilized onto magnetic graphene oxide (MGO) and utilized for MG degradation by Qiao and Liu [128]. The results indicated that 99% MG degradation was achieved within five hours, and the storage stability of the immobilized enzyme was significantly improved. Furthermore, a novel laccase-mimicking multi-copper BSA-Cu complex was prepared and successfully utilized for MG degradation [129]. All of these works illustrate the diversity of laccase-mediated degradation of MG, i.e., the enzyme can be extracted from different microbes, heterologously expressed, or even artificially mimicked, and the enzyme can be utilized in free or immobilized form.

5. Screening MG Alternatives for the Treatment of Fish and Fish Eggs

Although it was once an effective treatment against protozoan ectoparasites, MG was most frequently used to treat *Saprolegnia* sp. infections of fish and fish eggs. The reasons for the persistent illegal use of MG include its superior effectiveness against *Saprolegnia* sp. and the susceptibility of saprolegniosis in the aquaculture industry. After the use of MG for food fishes was banned, scientists have committed to developing effective alternative strategies for treating *Saprolegnia* sp. infections and therefore preventing the continued illegal use of MG in the aquaculture industry.

Several chemical agents other than MG have been applied for the treatment of saprolegniosis, and some have

TABLE 4: The advantages and disadvantages of different strategies for solving the issue of malachite green (MG) residues.

Strategies	Detail description	Advantages	Disadvantages
Adsorption	By bio-wasters derived materials	Low-cost	MG-loaded adsorbents might cause seconding pollution
	By artificially synthesized materials	High efficiency	Difficult to recover the MG-loaded adsorbents
Degradation	By chemical oxidation process	High efficiency	Chemical agents might cause adverse impact to water system
	By photocatalytic oxidation process	High efficiency	Chemical agents might cause adverse impact to water system
Bio-degradation	By microbes or related enzymes	Eco-friendly	Low efficiency or sensitive to environment
Substitution	Screening and applying MG alternatives	Most efficient	Complicated process of research and development

been officially registered. Formaldehyde [130], potassium permanganate [131], and sodium chloride [132] are typical examples of such kinds of agents. However, the practical application of these three agents was limited for one reason or another. Pyceze™ (with bronopol as the active ingredient) was registered in some EU states for treatment baths for food fish eggs, and it is particularly recommended for the treatment of salmonid fish eggs and the treatment and control of *Saprolegnia* sp. infections in rainbow trout and Atlantic salmon [133]. Selecting appropriate MG replacement candidates from now available agricultural fungicides is also a feasible strategy. Seven fungicides that were widely used in agriculture were investigated in vitro by Mostafa et al. [134], and azoxystrobin and metalaxyl were proved to be effective and safe for saprolegniasis control in the fish industry. Metalaxyl is the active ingredient of “Mei Ting,” an officially registered drug for saprolegniasis control in China since 2017. In addition, an early study by Min et al. [135] indicated that both vegetative hyphae growth and zoospore germination of *Saprolegnia parasitica* could be inhibited by chitosan in vitro. Mycelial growth and zoospore germination were entirely inhibited by 0.05% and 0.06% chitosan, respectively. Muzzarelli et al. [136] further investigated the activity of modified chitosan against *Saprolegnia parasitica*. The results confirmed the effective fungistatic action of methylpyrrolidinone chitosan, N-carboxymethyl chitosan, and N-phosphonomethyl chitosan.

A possible source of replacement candidates for MG is plant extracts. The antimycotic activity of several plant essential oils and extracts against *Saprolegnia ferax* was screened in vitro by an Italian scientific group. The results showed that the essential oils of thyme (*Thymus vulgaris*) and the methanol extract of *Artemisia verlotorum* displayed suitable activities; their MIC (minimum inhibitory concentration) was 40 ng/mL [137] and 0.25% [138], respectively. After that, more plant essential oils and extracts were screened for their anti-*Saprolegnia* activities. Essential oils prepared from fifteen different plants were tested by Nardoni et al. [139], and their MIC values against *Saprolegnia parasitica* ranged from 0.5% (*Origanum vulgare* and *Thymus vulgaris*) to over 10%. Further GC-MS analysis indicated that high content of thymol and carvacrol were included in the essential oils of *Origanum vulgare* and *Thymus vulgaris*, respectively. Furthermore, ethanolic extract of *Piper betel* leaves [140], *Kaempferia galangal* roots [140], *Punica granatum* [143], *Thymus vulgaris* [143], *Thymus linearis* leaves [140], and *Sambucus ebulus* [143], petroleum ether extract of *Cnidium monnieri* [144], *Magnolia officinalis*

[144], and *Aucklandia lappa* [144], and aqueous extract of *Brassica nigra* [145] were also evaluated, and different anti-*Saprolegnia* activities were obtained. Similar work was also conducted by Adel et al. [146], and the results indicated that essential oils of *Cuminum cyminum*, *Eryngium campestre*, and *Mentha piperita* could be potential candidates for controlling *Saprolegnia parasitica* infection in aquaculture. One more exciting work in this area was recently published by Meneses et al. [147], where a nanocomposite based on aqueous extract of *Terminalia catappa* and silver nanoparticles (AgNPs) was prepared and its anti-*Saprolegnia* activity was evaluated both in vitro and in vivo. When the concentrations of aqueous extract and AgNPs were 125 ng/mL and 91.5 ng/mL, respectively, high viable rate of eggs, a low fungal infection rate, and excellent larval survival were obtained in prophylactic tests with eggs of angelfish (*Pterophyllum scalare*).

Apart from crude plant extracts, a series of purified phytochemicals have also been evaluated for their anti-*Saprolegnia* activities. The results of these works are summarized in Table 3. It indicated that the MICs of these phytochemicals against *Saprolegnia* sp. Ranged from several $\mu\text{g/mL}$ to over 100 $\mu\text{g/mL}$. In vitro tests of these phytochemicals include preventing vegetative growth and zoospore germination. Generally, the required concentration for preventing zoospore germination was lower than for preventing vegetative growth. In vivo tests were also conducted for some phytochemicals. For example, exposure of abraded grass carp to a rhein bath at 20 mg/L for 7 d could prevent 93.3% of *Saprolegnia* infections, while 67.7% of infected fish could be recovered by treatment with rhein [150]. When applying phytochemicals in the aquaculture industry, their safety should be carefully considered. The acute activity test indicated that the 48h-LC₅₀ of rhein to grass carp was 148.5 mg/L, which is about 7.4 times the effective dose [150]. Therefore, rhein might be an ideal candidate for treating *Saprolegnia* infection. However, the safety of other phytochemicals that are listed in Table 3 has not been evaluated yet.

Screening natural competitors of *Saprolegnia* and utilizing them for biocontrol of saprolegniasis in aquaculture are a strategy also being considered. In an early study conducted by Hussein and Hatai [154], the antifungal activities of 47 bacteria isolated from the lesions of salmonid fishes with saprolegniasis were investigated, and five strains that belong to the genera *Alteromonas*, *Pseudomonas*, and *Aeromonas* were proved to inhibit the growth of *Saprolegnia parasitica*. *Streptomyces* sp. can produce abundant bioactive

secondary metabolites, and it is an important source of medical and agricultural antibiotics. A Japanese scientific group led by Professor Nobutaka Imamura successfully isolated several effective anti-*Saprolegnia parasitica* antibiotics from the fermentation broth of *Streptomyces* sp. strains, including oridamycins A [155], saprolmycins A [156], and saprolmycins E [156]. The MIC values for them were 3.0 µg/mL, 3.9 ng/mL, and 7.8 ng/mL, respectively. Furthermore, a *Burkholderia* sp. strain that strongly inhibits both the hyphal growth and spore germination of *Saprolegnia* sp., was recently isolated by Zhang et al. [157], and 2-pyrrolidone-5-carboxylic acid was identified as the active substance. In our group, a strain that can specifically inhibit the growth of *Saprolegnia parasitica* was recently isolated, and it was identified as *Streptomyces collinus* [158]. The anti-*Saprolegnia parasitica* active compounds of this strain have not been identified yet; however, preliminary studies indicate that the active compounds are thermostable (data unpublished).

6. Conclusion and Future Outlook

Satisfactory results regarding the adsorption of MG by various adsorbents, the degradation of MG by numerous AOPs, and the biodegradation of MG by diverse microbes or enzymes have been widely reported. The advantages and disadvantages of these methods are listed in Table 4. The adsorption method cannot degrade MG; hence, the treatment of MG-loaded adsorbents should be carefully considered to prevent it from becoming a new MG contamination source. While for the AOPs and biodegradation methods, chemicals (Fenton agents, photocatalysts, etc.) and microbes (or enzymes) need to be input into the water system, respectively, which may also induce a negative impact on the aquaculture industry. In response to these dilemmas, the combinatorial method would be a reasonable choice. Methods such as “adsorption + photocatalytic degradation” [159] and “adsorption + biological degradation” [160] have been proposed. However, most works about the elimination of MG from the water system were conducted under laboratory conditions, and practical applications were rarely reported. Many obstacles must be overcome when transferring these MG elimination technologies from the laboratory to practical application sites. For example, complex practical conditions may be unsuitable for MG adsorption or degradation. As per the strategies of screening MG alternatives for fish and fish egg treatment, satisfactory results have also been widely reported. However, most of these works only carried out in vitro anti-*Saprolegnia* sp. tests, and research about the in vivo activities of these reported MG alternatives was rarely found in the literature. In addition, systematic safety evaluations for these reported MG alternatives were also infrequently carried out.

Therefore, the following opportunities should be considered when conducting further research toward solving the issue of MG residues in aquatic products: (1) developing effective combinatorial methods (adsorption + AOPs or biodegradation) for eliminating MG from the aquaculture environment; (2) systematically considering the impact of

practical conditions on the efficiency of MG elimination; (3) screening more efficient anti-*Saprolegnia* sp. chemicals either from plant extracts or antagonistic microbes; and (4) systematically evaluating both in vivo activities and safety of potential MG alternatives.

Data Availability

The data supporting this systematic review are from previously reported studies and datasets, which have been cited.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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