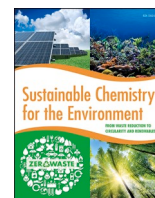




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Ozonation, electrochemical, and biological methods for the remediation of malachite green dye wastewaters: A mini review

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ABSTRACT

The extensive usage of malachite green (MG) dyes in the fish and dye industries cause serious water pollution, leading to catastrophic effects on living organisms and the environment. In addressing the challenges of health risks related to the use of MG, different removal strategies have been studied and developed. This review presents the recent advances and mechanistic pathways involved in ozonation, electrochemical advanced oxidation (electrooxidation), and biological removal techniques for MG. The successful applications of these techniques in decontaminating water from MG are discussed. Furthermore, combined removal techniques for enhancing MG removal and for the reduction of operation costs are presented as it was shown in some cases to be more effective than individual remediation methods.

1. Introduction

Malachite green, a triphenylmethane dye (shown in Fig. 1), originally found its wide applications in the textile industry [27,56] Dating back to 1933, it was extensively used in aquaculture for the prevention and treatment of protozoal and fungal infections on fish eggs, fingerlings, and adult fish [10]. However, MG became a highly controversial compound when used in the topical treatment in aquaculture because it accumulated systematically in the internal anatomy of the fish and produced significant internal effects on the treated fish [10]. The side effects are also felt on the consumers of treated fish as it posed a serious threat on the immune and reproductive systems. Hence, it is concluded that MG is genotoxic and carcinogenic because it interferes with the survival of living entities [63]. Therefore, it is expedient to remove MG from the aquatic systems to safeguard human health, environment, and aquatic species.

A wide variety of removal strategies have been developed for the decontamination of wastewater and aqueous environments from MG

such as photodegradation [43,45,68], adsorption [25,30,37], membrane filtration [26,31,49]. In this paper, we emphasized on ozonation, electrochemical advanced oxidation (electrooxidation), and the biological treatment methods for MG removal due to their high effectiveness in organic pollutants removal. Ozonation process is the first method discussed, which is a cleaner process for wastewater treatment without sludge generation and residual ozone decomposes to water and oxygen [21]. The second method discussed was electrochemical advanced oxidation, which makes use of electrolytically generated hydroxyl radicals for the abatement of dyes [48]. It is operationally safe, amenable to automation and environmentally compatible because it utilizes electron as the main reagent, which is an inbuilt clean species [52]. In a similar vein, biological removal methods are eco-friendly, versatile in nature, cost effective, and occur under mild reaction condition which completely mineralize toxic pollutants into non-toxic end product [69].

In this review, we presented a critical discussion of reports of recent investigations that were conducted to improve the efficiency of ozonation, electrooxidation, and biological methods for the degradation of MG

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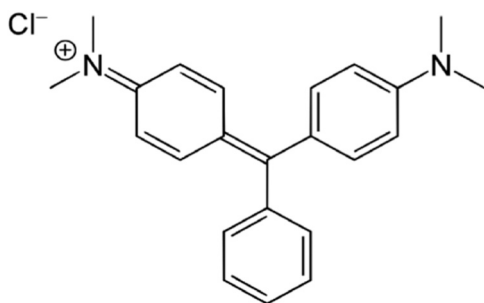


Fig. 1. Structural formula of MG.

in (waste)water, with detailed discussion on mechanistic pathways proposed for the elimination of MG. Also, this mini review provided an overview of the coupling of individual technologies for MG removal and mentioned the differences existing between the combined techniques and the ordinary techniques adopted for degradation of MG under similar optimum conditions.

2. Malachite green

2.1. Uses of MG

MG dye works as ectoparasiticide for eliminating gill and skin flukes [64]. In aquaculture industry, it is extensively used as a biocide as it is very effective against fungal and protozoal infections [53]. Also, it is applied as a topical treatment agent using flush or bath methods, whereby it can be systemically accumulated and give significant internal effects [64]. Sequel to the controversial risk MG dye can pose to the consumers of (treated) fish, several countries have banned MG dye and its usage has been disapproved by US Food and Drug Administration. In the food industries, MG is applied as food coloring agent or as food additive [17]. On the other hand, the major application of MG is in textile industries for dyeing fabrics where it accounts for 66.7% of dye containing wastewater generated [46]. Therefore, MG is widely applied in different areas/fields.

2.2. Toxicity of MG

Malachite green has been reported to be a controversial chemical substance owing to its extreme toxicity to the human system. Previous studies have confirmed the carcinogenic nature of MG. ([12], 2005) acknowledged that Syrian hamster embryo cells treatment with MG could cause damage of DNA by oxidation as a result of production of free radical that might also be the cause of malignant transformation [12]. This ultimately makes MG an extremely toxic chemical substances to mammalian cells [18]. Similarly, (Rao, 1995) confirmed that MG can inhibit synthesis of basal DNA while it also enhances hepatocytes alteration to induce liver cancer in rats. These findings imply ingestion of MG could cause human DNA damage.

Unfortunately, the toxicity of the dye increases with time, concentration, and temperature [64]. The implication of this is that non-removal of MG in wastewater streams will increase the level of MG gradually in the and its associated toxicity. The adverse effects exerted by MG dye include but are not limited to teratogenicity, mutagenesis, carcinogenesis, respiratory toxicity, and chromosomal fractures [64]. Srivastava and team members documented the toxicity of MG dye in selected mammals. Prior to the research work of Srivastava and co-authors, Culp and Beland [17] revealed the toxicological effects of MG dye in their published review article. These studies, with some other ones [14,15], imply that MG dye toxicity has long been a subject matter. The effects of MG were also investigated to be cytotoxic to two human cell lines. The finding further revealed that the dye reduced proliferation capability and impaired mitochondrial activity [65].

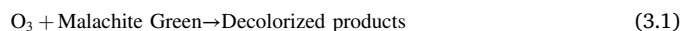
Further, an attempt was also made to study the adverse impacts of MG on soil biota using some model microorganisms [23]. The study confirmed toxicity in bacteria and fungi, showing an elevated level of reactive oxygen species. Moreover, DNA polymorphism and fragmentation ascertained genotoxicity in the microorganisms. Recently, Shukla et al. [59] explored the feasibility of sandalwood-derived carbon quantum dots as bioimaging tools to investigate the toxic effects of MG in selected bioindicators. In this study, animal model systems, MG-63 osteoblast cell line, golden hamster, were used. Consequently, a dose-dependent decrease in cell viability was observed with MG dye [59].

In mammalian body system, MG can be biochemically reduced to a chemical substance known as leucomalachite (LMG) that can accrue in the body for greater than 5 months [47,50]. LMG has been found to increase lung adenomas in control Fischer 344 (F344) male rats, facilitating liver tumor when appropriately initiated [16]. Similarly, another research group has ascertained the carcinogenicity and genotoxicity of LMG in F344 and B6C3F1 rats [42]. Therefore, LMG and MG are toxic substances, even though LMG has histopathologically been shown to demonstrate higher toxicity than MG during exposure to humans. Hence, it can be safely concluded that malachite dye interferes with the survival of living entities. It is worth mentioning that studies on the toxicity of MG have only been sparsely explored and/or reported in recent times. This area requires urgent attention as there is an advancement in technology, which increases the probability of the relative use of MG dye.

3. Removal strategies of MG

3.1. Ozonation approach

Ozonation is one of the efficient methods for the degradation of dyes [66]. It functions by destroying the chromophore of dyes and any other chemical bonds in dyes via a process known as ozonolysis [5,76]. There are two known ozonolytic degradation pathways, namely: the free radical mechanism in which ozone generates radicals that attacks the dyes while the second one is the direct attack by molecular ozone on the dyes. The high oxidization potential of ozone (2.07 V) makes it so powerful at destroying organic pollutants while its counterpart (hydroxyl radical) possesses even more oxidization potential (2.8 V) [76]. Due to the highly oxidizing nature of ozone, the design of reactor for ozonation of MG is always carried out with this in mind. In some of the ozonation reactor designs, there is provision of potassium iodide for trapping residual ozone while the teflon lid and cooling water system to control the temperature of the entire reactor (Fig. 2). A generalized equation for ozonolysis process for MG degradation is shown in the following equation.



The time taken by the ozone to degrade MG increased with the increase in the concentration of MG while there was approximately 90% MG removal from wastewater in the presence of ozone within the first 5 min [7]. The need to reduce the amount of oxidants used in ozonation has led to the use of 'assisted-ozonation'. In an investigation, sound energy was used to assist ozonation and it saved 11% of the oxidant's amount compared to similar investigations without the assistance of sound energy. It also led to the reduction of the reaction time for the degradation of MG [76]. The degradation of MG in "sound-assisted" ozonation was described as a mass transfer process that involves some chemical reactions. In a simple term, the absorption of gaseous ozone takes place first and it then reacts with molecules of dissolved MG [74]. An enhancement in the stoichiometric ratio (number of moles of MG that can easily be oxidized by a mole of ozone : Z_{app}), due to aiding of the ozonation process by ultrasonication compared to ultrasonication alone is given in Fig. 3.

Another effort aimed at enhancing the degradation of MG via

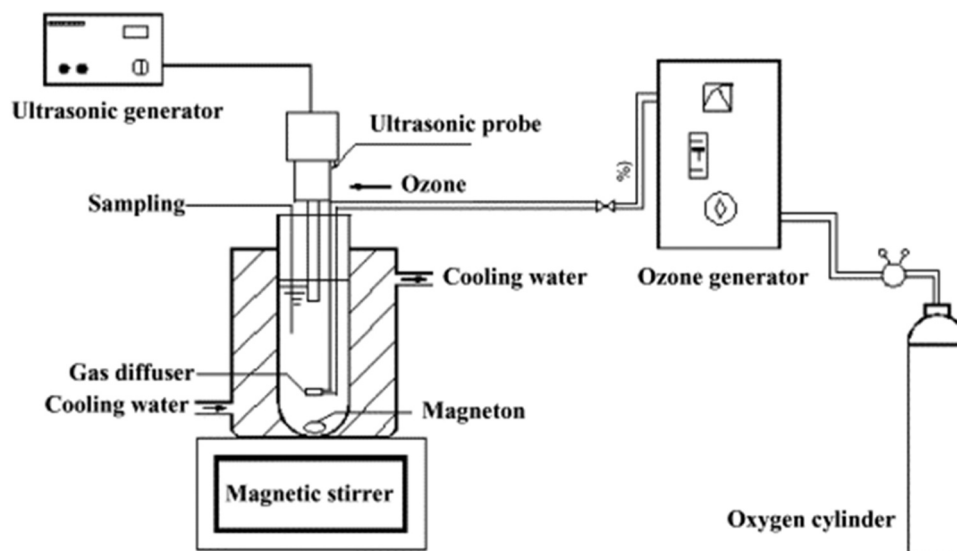


Fig. 2. Ozonation reactor for the degradation of dyes. Reproduced with permission from Elsevier [74].

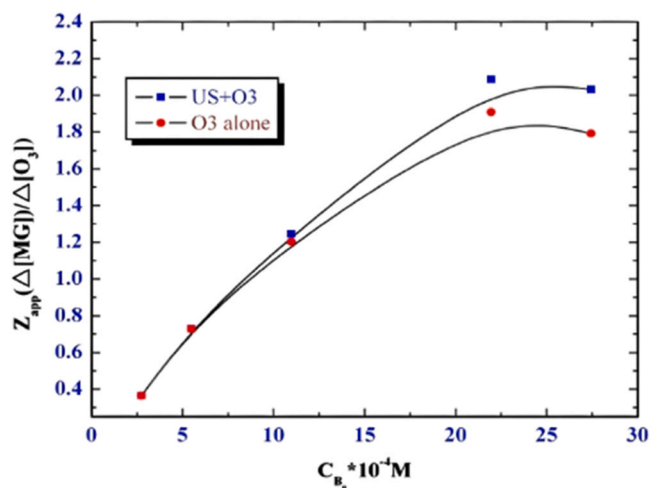


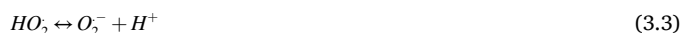
Fig. 3. Comparison of Z_{app} in sound-assisted and non-sound assisted ozonation. Reproduced with permission from Elsevier [76].

ozonation is the introduction of catalyst. Parvarideh et al. compared the efficiencies of ozonation with and without catalyst [51]. Their findings showed that complete decolorization of 50 mg L^{-1} of MG was achieved in 15 mins compared to 30 mins that was required without the use of the catalyst with ozone. In similar research, there was 96.74% MG removal in 110 mins of ordinary ozone degradation but when chemically modified clay was introduced as the photocatalyst the same degradation efficiency was achieved within some seconds. This confirmed that the presence of (photo)-catalyst in ozonation process enhances the degradation of MG [41].

Overall, the mechanism of MG degradation by ozone can be via direct mineralization of MG by ozone molecule (O_3) or its mineralization by in situ generated free radicals (like hydroxyl radicals). The former predominates in an ozonation reactor system that is not aided by ultrasonication or photocatalysis. However, for “sound-assisted” ozonation, radicals are formed. Hydrogen and hydroxyl radicals have been documented to be released from water vapor by ultrasonication. At low frequency of ultrasonication, however, free radical contribution can be regarded as minor as low levels of hydrogen and hydroxyl radicals would be generated [76].

Lastly, a quite amount of other reactive oxygen species can be generated, to aid the mineralization of MG, as presented in the following chemical equations:

Reaction of ozone with hydroxide ion:



Reaction of ozone with superoxide anion:



Formation and decomposition of hydrogen trioxide:



Reaction of hydroxyl radical with ozone to form HO_4 :



Subsequent decomposition of HO_4 :



Consequently, MG can be both oxidized by these strongly oxidizing reactive oxygen species by attacking or cleaving the molecule at the C=C segment. Other investigations involving the ozonation and assisted ozonation for the removal of the MG dyes are presented in Table 1.

3.2. Electrochemical advanced oxidation processes

Electrochemical advanced oxidation process is also known as electrooxidation. In this process, electrical energy is employed for the degradation of dyes. One of the common forms of this process is anodic oxidation and it is carried out via direct or indirect anodic reactions [39]. In both cases, oxygen is obtained from the solvent (usually water) to initiate the oxidation process. This method is advantageous because it is cheap and it does not require the addition of other chemicals [13,24]. Different electrodes have been investigated for the degradation of MG. One of the previous investigations employed stainless steel as the cathode and boron-doped diamond as the anode while the conditions were maintained as pH of 3 and current intensity of 32 mA cm^{-2} . Under these conditions when sodium sulphate (Na_2SO_4) was used as the electrolyte, there was 98% degradation within 1 h of electrolysis [24]. Brine

Table 1
MG removal by assisted-ozonation methods.

Ozonation method	Initial concentration of MG	Other conditions	Performance	References
Ozonation with cationic catalyst montmorillonite	10^{-4} M	pH = 3; 0.5 g/h of ozone concentration;	95% yield in less than 1 mins compared with over 2 h without the catalyst.	[41]
Alumina- catalytic ozonation	3000 mg L ⁻¹ and 50 mg L ⁻¹	pH = 3	While there was complete decolourization of the 50 mgL ⁻¹ within 30 mins, there was over 95% decolourization of 3000 mg L ⁻¹ within 15 mins of ozonation	[51]
Ozonation with 1,8-dimethyl-1,3,6,8,10,13-hexaazacyclotetradecane nickel(II) perchlorate	10 mg L ⁻¹	1 g/L potassium persulphate; pH 3 and pH 9	Complete decolourization of MG within 8 mins and loss of antimicrobial activities by MG	[22]
Al-Fe modified- and pure montmorillonite K10	5×10^{-5} M	20 mL of dyes; 5 mg of catalyst and 0.0166 g O ₃	88.54–97.50% of MG degradation in 40 s	[40]
Ozonation only	0.30–1.82 mM	pH = 3; T = 19 °C	86% of MG degradation in 10 mins with reduction in the antibacterial activities of the dye	[40]

has also been used as an electrolyte while silver and carbon/TiO₂ were used as cathode and anode, respectively. The conditions was maintained between pH of 7–8 and this led to complete degradation of 25 ppm MG to carbon dioxide within 30 mins of electrolysis [36]. In a bid to enhance the electrochemical degradation of MG, other methods have been combined. For instance, Fenton reagent has been introduced into the electrochemical system to degrade MG. The method was found to be effective at degrading MG and the ferrous ion (Fe²⁺) were regenerated after the process which implies that the process can be recycled without

introducing fresh Fe²⁺ into the system [9]. Another practice is to modify the electrodes by depositing precious metals such as silver and gold on it to enhance the overall performance of the electrochemical degradation process [38].

3.2.1. Ultrasonic-assisted electrochemical oxidation

The use of sound to enhance the electrochemical oxidation processes has been reported [62]. For instance, Ren et al. investigated ultrasonic assisted electrochemical elimination of MG from wastewater [55]. The

Table 2
Interpretative intermediate products of MG degradation from mass spectrometry [55].

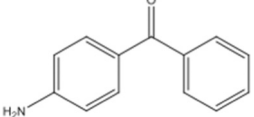
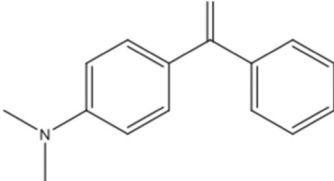
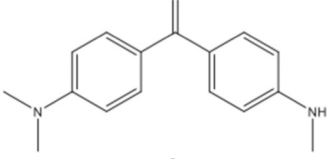
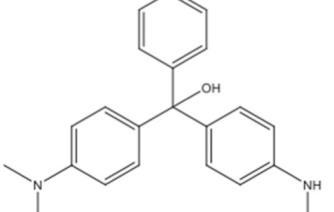
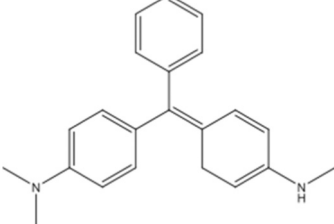
Entry	Chemical formula proposed	Theoretical mass to charge ratio (m/z)	Structural formula proposed
1.	C ₁₃ H ₁₁ NO	197.08	
2.	C ₁₄ H ₁₃ NO	227.12	
3.	C ₁₆ H ₁₈ N ₂ O	256.15	
4.	C ₂₁ H ₂₂ N ₂ O	318.17	
5.	C ₂₂ H ₂₄ N ₂	318.20	

Table 3
Electrochemical oxidation of MG.

Electrochemical method	Conditions	Performance	References
Ultrasonic assisted electrochemical method	Titanium anode; graphite anode; 15.0 g/L Na ₂ SO ₄ ; 300 W ultrasonic power and 20 V.	94.92% degradation of MG	[55]
One-compartment batch reactor electrochemical method	A stainless steel as cathode and boron-doped diamond as anode; pH = 3; Na ₂ SO ₄ as electrolyte; current intensity of 32 mA cm ⁻² .	98% degradation efficiency within 1 h.	[24]
Catalyst-assisted electrochemical method	Pb/β-PbO ₂ , Ti/β-PbO ₂ , SS316/β-PbO ₂ and G/β-PbO ₂ electrodes; I = 4 mA/cm ² , pH = 7.0	100% MG removal efficiency	[6]
Electrochemical method	A current density of 1.44 mA cm ⁻² ; 2 g/L NaCl; 50 mg L ⁻¹ MG concentration; lead oxide anode; graphite cathode; room temperature.	Good removal efficiency but the efficiency was affected by the presence of different ions in the system.	[58]

Table 4
Application of different microorganisms for biodegradation of MG dye.

Microorganism	Type	Results	References
<i>Stenotrophomonas maltophilia</i>	Bacterium	<i>S.maltophilia</i> isolate TPMD-1 derived from composing neem oilcake effectively degenerated both MG and CV dyes. The rate of MG degradation was mainly in neutral pH at 28–30 °C.	[2]
<i>Fusarium oxysporum</i> HUIB02	Fungal laccase	At optimal conditions, malachite green dye degradation effectiveness by laccase attained ≥ 90% using dye concentration of ≤ 100 mgL ⁻¹ . 88.1% dye removal was realized at a concentration of 1000 mgL ⁻¹ of malachite green.	[67]
<i>Pseudomonas sp. strain DY1</i>	Bacterium	90.3–97.2% malachite green dye decolorization efficiency was attained by <i>Pseudomonas sp. strain DY1</i> within 24 h at concentrations of 100–1000 mgL ⁻¹ under shaking mode	[20]
<i>Pithophora sp.</i>	Algae	<i>Pithophora sp.</i> Demonstrated maximum biosorption capacity of 117.647 mgg ⁻¹ at 30 °C while using MG dye concentrations of 20–100 mgL ⁻¹ .	[34]
<i>Trichoderma harzianum strain HZN10</i>	Fungal laccase	HPLC characterization showed that roughly 100% of MG, 90.0% of MB and 60.0% of CR dyes at a concentration of 200 mgL ⁻¹ decolorized in 16.0, 18.0 and 20.0 h, respectively by laccase immobilized on sol-gel matrix in the presence of 1-HBT mediator.	[8]
Crude protease from <i>Bacillus cereus</i> strain KM201428	Bacterial Protease	Crude protease enzyme from <i>Bacillus Cereus</i> Strain KM201428 exhibited the ability to degrade and decolorize MG dye 100%.	[8]
<i>Pseudomonas veronii</i> JW3–6	Bacterium	About 93.5% of 50.0 mgL ⁻¹ of MG was degraded by <i>Pseudomonas veronii</i> JW3–6 within 7 days	[60]
<i>Pseudomonas sp. ESP540</i>	Bacterium	<i>Pseudomonas sp. ESP540</i> manifested higher capability for degrading MG (86.88%) at NaCl concentrations of 1–3%. The strain <i>ESP540</i> exhibited biodegradation of MG of 800 mg L ⁻¹ dye concentration.	[35]
<i>Bacillus sp. AS2</i>	Bacterium	Maximum dye degradation achieved for MB, CR, and MG by <i>Bacillus sp. AS2</i> was 97.01 ± 0.36%, 92.49 ± 0.21%, and 94.23 ± 0.47% respectively.	[57]

results indicated that “sound-assisted” electrochemical oxidation recorded highest MG degradation, as high as 94.92%, with applied voltage, electrolyte concentration and ultrasonic power of 20 V, 15.0 g/L Na₂SO₄ and 300 W, respectively [55]. As a matter of importance is the optimization of the ultrasonic power as many bubbles can be generated when the power is more than a certain value, thereby keeping the bubbles expanding, growing, and bursting near the electrode plate. Ultimately, this leads to a sudden pressure rise close to the electrode, thereby affecting the electrode surface, rate of electrolytic reaction and MG degradation efficiency [19,75]. In terms of pathway of degradation, a slight change in wavelength (blue-shifting) in degraded MG was observed using UV-spectroscopic technique. This unequivocally implies N-demethylation of MG while peaks at 423 and 300 nm recorded are indications of cleavage of conjugated structures of MG by hydroperoxyl ion (HO₂) attacking carbocation and nitrogen ion on MG [29]. Also, a peak at 368 nm represents a compound formed from the attack of center carbocation [76] by hydroperoxyl radical to form diphenyl ketone derivatives [28,29]. When these findings from UV-Vis spectrometric study were combined with other elucidating techniques like high performance liquid chromatography – mass spectrometry, possible intermediate products of degradation are presented in Table 2. Some other investigations that involved the use of electrochemical methods for degrading MG are summarized in Table 3.

3.3. Biological removal methods

The use of biological methods for textile dye decolorization/biodegradation has gained attention due to their thriving versatility, eco-friendliness, cost effectiveness, mild reaction condition and

complete mineralization of toxic organic chromophore into nontoxic end products [72,73]. The use of microorganisms like yeasts, fungus, enzymes, bacteria and algae that have the capability to fragment and absorb dyes are among the widely employed biological methods in dye removal [11]. Effective application of microorganisms and their enzymes greatly depends on dye concentration, contact time, load of microorganisms or enzyme concentration, temperature, pH, dissolved salt concentration and concentration of dissolved oxygen in the system [1]. Table 4 shows different microorganisms employed in biodegradation of MG dye from the textile effluent.

3.3.1. Biocatalytic degradation

Enzyme extracted from organisms and plants have for long been applied in dye degradation. Their adaptability, simplicity, and efficacy even at moderate reacting environment gives them a superiority over conventional physical-chemical treatment techniques. Both intracellular and extracellular enzyme employed with different success as biochemical means of dye decolorization in wastewaters. Application of enzymes in effluent treatment is gaining prominence over the use of whole living microorganisms since effluent may contain contaminants which can inhibit microbial growth [44]. Enzymes are highly specific, extraordinarily effective, ecologically ecofriendly and function under mild reaction conditions of temperature and pH. Since one can manipulate the enzyme concentration as well as enzyme effluent ration, enzymatic treatment can be applied for treating large quantity of effluents in comparison to other biological methods. Both anaerobic and aerobic environment can be modified to increase enzyme-related dye degradation. Biodegradation under shaking conditions could play a consequential function in increasing oxygen accessibility in the liquid media,

thus vitalizing the degradation capability [2].

Majority of the biocatalyst that have been explored for dye biodegradation prospects belongs to oxidoreductases class of enzyme. During reactions, these enzymes participates in electron transfer to effect dye biodegradation [44]. Oxidative enzyme like peroxidases needs hydrogen peroxide or alkyl peroxide perform as the electron acceptor while laccases use molecular oxygen for the same purpose. Peroxidases secreted by roots of the plant have been used in degradation of dyes including brilliant blue, MG and methyl orange [32]. Lignin peroxidase and manganese peroxidase are also employed in dye degradation. Oxidative enzymes have expansive scope of substrate peculiarly about phenols and amines that may not have definite substrate binding locations. Despite these advantages of biocatalytic degradation and decolorization, the major hindrance in the use of enzymes is their exorbitant cost. Presently, wastewater treatment by use of biocatalyst on a large scale is not economically prudent due to high cost.

3.3.2. Biodegradation

Biodegradation refers to biologically interceded fragmentation of chemical compounds; it is an energy-dependent approach and entails disintegration of dye structure/chromophore into different byproducts by the act of diverse enzymes [3]. Biological degradation of MG dye results in a moderately nontoxic compound owing to breakdown of bond in dye structure, which assist in color elimination from effluent. Dye biodegradation can be categorized as either aerobically or anaerobic or combination or both. In an aerobic technique, microorganisms are used in treatment of pollutants in the presence of adequate dissolved oxygen while anaerobic process involve the use of microorganisms in the absence of oxygen [1]. The effectiveness of biological degradation technique is greatly determined by the selection of microorganisms and enzymes activity. Aerobic biodegradation of malachite green by crude protease from *Bacillus cereus* strain KM201428 resulted in production of several intermediate metabolites product before complete mineralization and decolorization. Some of the intermediate product based on GC-MS and LC-QTOF-MS examination included Leuco malachite green, 4-[4-(dimethylamino) benzyl]benzoic acid, (4-aminophenyl) [4-(dimethylamino)phenyl]methanol, [4-(dimethylamino)phenyl](phenyl) methanol, 4-(hydroxymethyl) phenol, bis(4-aminophenyl)methanol, (E)- 2-Hydroxy-4'-dimethylamino-stilbene, 6-amino-6-oxohexanoic acid, Benzenemethanamine, N-phenyl, Cyclohexanamine, 6-amino-6-oxohexanoic acid [71]. Five intermediate metabolites were identified in the biodegradation of MG by *Pseudomonas veronii*: leucomalachite green, 4-(dimethylamino) benzophenone, 4-dimethylaminophenol, benzaldehyde, and hydroquinone [61]. These results showed that the dyes underwent reductive splitting relatively easily under anaerobic conditions.

Moreover, the integration of physical treatment methods such as adsorption with biological methods can greatly increase the cost-effectiveness of treatment process in terms of time, infrastructure cost, effluent volume and quantity of enzyme or microorganisms deployed. Dye pre-concentrated helps in controlling the efficacy of treatment process, lower the cost related to water and energy consumption and final recycling or waste disposal. It is worth noting that a higher degree of biodegradation and biodecoloration may be achieved when the biodegradation treatment process incorporates a microbial community that complement each other as opposed to the use of pure cultures. Studies have shown that microbial consortia (comprising both fungi and bacteria) are more potent than pure cultures due to synergistic effect [3]. This is because fungus can initiate the biodegradation of the dye (which is not responsive to biodegradation by bacteria). However, for complete dye removal, both bacteria and fungus can utilize the resultant metabolite as nutrients source for growth. MG dye was utilized as a single source of carbon in the biodecolorization media of two fungal species, *Aspergillus flavus* and *Alternaria solani* which decolorized 99.78% and 91.72% of 30- μ M dye solution within six days respectively [4]. The findings suggest that both fungal species synergistically use the dye for their thriving.

3.4. Coupling of individual technologies for MG removal

Another effective method of eliminating MG from wastewater is the coupling of individual techniques. Adsorption has been combined with other removal techniques to remove malachite green from water and wastewater. In an investigation, adsorption was combined with electrocoagulation and peanut shell was used as the adsorbent. The use of the combined techniques was found to be better than the use of ordinary electrocoagulation. In 5 mins, there was a 98% MG removal which is 23% higher than what was obtained when ordinary electrocoagulation was adopted for the removal under similar optimum conditions[70]. Similarly, plant biochar and menthe plants were reported as adsorbent for MG removal in an investigation that coupled both adsorption and electrochemical techniques for MG removal. The results obtained from the cyclic voltammetry revealed that the coupled redox reaction was reversible at the interface of the adsorbent and the dye molecules. The maximum adsorption capacity of 322.58 mg g⁻¹ was obtained [54]. Removal of MG has also been achieved by coupling adsorption with photocatalysis. Kant et al.,[33] used both adsorptive and photocatalytic properties of Fe_{0.01}Ni_{0.01}Zn_{0.98}O/polyacrylamide nanocomposite to degrade 96.13% of MG in the water samples. Another combined technique that has been reported is the combination of ultrasound and electrochemical degradation. The maximum degradation of 94.92% was obtained with the combination of the ultrasonic power of 300 W and 20 V. The anode used was ruthenium-iridium plated titanium while the cathode chosen was graphite [55]. All these point to the fact that combining different removal techniques is also effective for the decontamination of wastewater from MG."

4. Outlook and practical-knowledge gaps

Summarily, findings from the electrochemical advanced oxidation processes and ozonation indicate that mineralization of the toxic MG is possible, but it comes with challenges of chemical consumption. The use of different potentially toxic or less toxic chemicals like ozone and other reagents, are harmful themselves. The question is: the reactive oxygen species, are they completely removed from the treated water after mineralization? For town water supply water treatment, would these chemical removal approaches be safer, cost-friendly, and easier to operate? These questions need to be investigated in improving the various methods on chemical removal approach. In addition, as much as chemical technique is not green, research need to be focused on quantification of degradation products (metabolites) from the various advanced oxidation processes and this is scarcely studied or reported, while priority research should also be placed on the use of natural materials as photocatalyst, such as clay minerals, to make the photochemical method green.

Furthermore, it is evident that biological degradation of organic dyes has attracted considerable recognition, as a cost-effective and eco-friendly methods of treating wastewaters contaminated with dye though not fully exploited by industries. Efforts need to be directed in screening for more effective, affordable, and safe microorganisms /community microorganisms what can work under diverse environmental conditions in biodegradation of dyes. It is worth noting that disappearance of color during biodegradation process does not guarantee elimination of dye toxicity. The prospects of biodegradation techniques should include the creation of a real time dynamic analytical tools that researchers and industries could use to virtually analyze the resultant byproducts/metabolites, ascertain their toxicity before final discharge in the environment. Importantly, it is vital to analyze the nature of the degraded compounds by toxicity analysis before implementation of the bioremediation technique on a large scale. Such a strategy will assist in developing a scientifically sound environmental policies and laws on regulation of undoubtedly harmful dyes and management of toxic industrial effluent.

5. Conclusions

This review has discussed the successful applications of ozonation, electrooxidation, and biological treatment methods in removing MG from wastewater streams. Complete degradation of MG within a short period of time by ozonation, electrooxidation, and biological removal techniques have been achieved by incorporating Fenton reagent, and biocatalyst/microbial community, respectively. Most importantly, the application of sound waves has proved to be effective at enhancing the degradation of MG using ozonation and electrochemical oxidation processes, owing to the formation of free radicals. Unfortunately, all these treatment methods require relatively high energy/operation costs. However, a significant enhancement in the effectiveness of MG removal along with reduction of costs can be achieved by using combined treatment techniques. Overall, this review provides cutting-edge information on the above-mentioned techniques that would be valuable for further developments in the field.

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.

Data Availability

No data was used for the research described in the article.

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