Review Article: Photocatalytic Degradation of Malachite Green Dye by Metal Oxide Nanoparticles - Mini Review

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ABSTRACT

A wide range of hard adulterants in waste water are generated from different industries as a by-product of the organic compound. In this review, the cationic dye Malachite Green removal from wastewater by photocatalytic route by using various metal oxide nanoparticles is analysed. The effect of some specific parameters like the method of preparing the catalyst, the initial concentration of dye, the amount of nanocatalyst required for degradation, the initial pH of dye solution, type of light source used, and time of light exposure required for removal of dye were specially incorporated. Some general conclusions were made based on the pivotal analysis of literature available. With a view to reach higher removal efficiency, in an economical manner, some optimal testing conditions on degradation of these hard dyes should be carried out.

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Content

1. Introduction

Water is a one of the basic source for a human to survive on the earth. Among the total availability of water resources, only 1% of water is utilized for human consumption [1]. The adulterants from various industries like dye manufacturing industries, apparel industries, paper pulp mill industries, reed mat industries, tanneries, and printing industries increase a great threat to our natural resources [2]. Textile industry is the one which releases a huge amount of waste i.e. the organic dyes, chemicals, heavy metals, and oil in water bodies create a mass disaster to the environment because of its venomous nature and it acts as an agent for cancer disease [3]. Textile dyes are highly soluble in water, due to this high release of dark dye the entry of sunlight is blocked causing a severe damage to the aquatic organisms and humans in and around the areas of these industries [4]. These dyes are approximately ten thousand in number. On viewing the large scale of usage, the azo dye is the one which comprises of the immense and critical damage [5].

Malachite Green (*N*-methylated diaminotriphenylmethane) is an organic compound containing a green colour crystal with a metallic lustre used in various dye stuff industries. It is one among the most effective chemicals all over the world [6]. It is highly toxic and should be treated before discharging it into the natural resources. It has a noxious effect on liver, gills, kidney, intestine, and various damages to mammalian cells. It may lead to cancer when inhaled; causes the skin irritation with pain when come into contact [7]. The direct discharge of MG dye in hydrosphere causes an imbalance to environment. This untreated water used for the irrigation purpose might result in reducing the quality of crop production [8]. These dyes are banned in many countries, but are still in use due to its low cost, accessibility, and potency [9].

Treating these dye wastes before discharging into water bodies is the most crucial need to be performed. One in eight people on the earth lack access to clean drinking water. Nearly one billion people have no access to safe drinking water. In developing countries, 3.5 million people die each year due to inadequate sanitation and hygiene. Around 1.5 million children die due to water-borne diseases. By 2025, 1.8 billion people will be living in countries or regions with absolute water scarcity. This problem in water scarcity is going to make a worldwide systematized hazard [10]. To overcome this, research in appropriate material and perfect treatment method/ devices should be implemented in an urgent scale.

This mini review contains the process in literatures from 2018 to 2022 for the degradation of malachite green dye by photocatalytic technique by using various metal oxide nanoparticles.

2. Dye Treatment Methods

For the betterment of human society and to resolve inadequate drinking water availability, both the portable water and waste water should be treated. Various physical, chemical, and biological ways have been employed to remove the contaminants in water (**Figure 1**). The electrocoagulation process [11], adsorption, enzyme degradation [12], ion exchange, membrane filtration, chemical precipitation, flocculation, chemical oxidation [13], bacterial decomposition, electrochemical decolourisation, ozonisation [14] nano photocatalysis, ceramic nano filtration membrane, biofilms, [15], organic resin, electrolysis, reverse osmosis, hybrid materials, oxidation, and electrolysis, biofoulents [16]. The properties of these toxic dyes make the traditional methods less effective in decolourisation. Yet, the light based adsorbent (photocatalysis) method has come out with an excellent capability of degrading these dyes.

3. Photocatalytic Treatment of Dyes

The photocatalytic treatment covers a heterogeneous photocatalysis, semiconducting absorbent which degrades numerous impurities upon illumination to the light including the aerosphere organic pollutants. It comes up with several conveniences over the standard methods of treating industrial

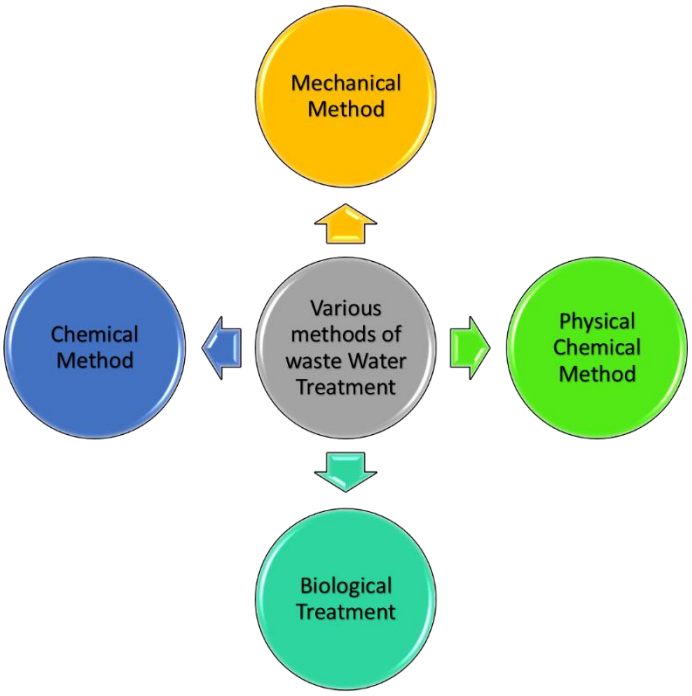

Figure 1. Various methods of treating industrial waste

wastewater. For instance, within a short span of time, the dye material gets completely degraded by photocatalyst even at room temperature. More importantly, there is no formation of toxic residues, it completely breaks down the organic pollutants into unthreatening by-products i.e. water and carbon-dioxide [17]. Some optimal conditions during the photocatalysis reaction also influence the degradation efficiency. They are the light source which is used for irradiation (sunlight is the naturally available light source), the time period which the dye get exposed to the light, pH value of the dye solution, amount of catalyst required for efficient degradation, the distance between light source and dye, the temperature maintained during the reaction, and concentration of the dye solution [18].

In the beginning, the ordinary porous materials and those made under nano size are used as an adsorbent like activated carbon, CNTs, mesoporous silica, and chitosan bead. These materials exposed some constraints such as struggling during operation, less efficient, high cost, need for high amount of energy. An alternate material as photocatalyst is highly recommended which should overcome the previous drawbacks [19].

4. Metal Oxide Nanoparticle as Photocatalyst

Different catalysts have been engineered to enhance the photocatalytic process. Metal oxide nanoparticles are less toxic and can be easily oxidized into hydroxides or oxides. Another main property is its band gap which enables both the oxidation and reduction process by observing energy from the light source. The metal oxides with less band gap make it efficient under the visible light i.e. low of cost and less time consuming because of its absorption shifts to red band [20]. Many metal oxide nanoparticles are employed as an efficient photocatalyst. Some of them are TiO_2, ZnO, Bi_2O_3, Fe_2O_4 [21], WO_3, CuO, Cu_2O [22], SnO_2, CeO_2, $BiVO_4$, Bi_2WO_6, $InTaO_4$, $Zn_{1.7}GeN_{1.80}$, $ZnAl_2O_4$, and ZnGaNO [23]. These metal oxides have much morphology such as nanoparticle, nano spheres, nanofibers, nanotubes, nano ribbons, nano sheets, etc. These morphologies also affect the efficiency of a photocatalyst [24]. The following table (**Table 1**) provides the Malachite green dye degradation by various metal oxide nanoparticles, the experimental condition, and its degradation efficiency.

Table 1. Malachite green dye degradation by various metal oxide nanoparticles, experimental condition, and its degradation efficiency

Metal oxide NPs	Method of synthesis	Morphology	Photocatalytic experiment condition	Degradation efficiency (%)	Reference
rGO/CuS	co-precipitation	Irregular hexagonal	100 mg photocatalyst, 10 ppm dye, under sunlight at room temperature	97.60	25
Hematite	Combustion	Spherical and irregular	Presence of H_2O_2, 20 ppm dye, UV source of 250 W, 0.1 g catalyst, 70 min	100	26
Cobalt Oxide	Sol-gel	Spherical	50 mg nanocatalysts, 100 mL of 1 × 10-5 M dye, Xe lamp of 80 mW/cm2, 100 min	91.20	27
Cu2O	Sonochemical	Uniform icosahedron	10 ppm dye, 10 mg catalyst, visible lamp, 45 min	91.89	28
ZnO	Sol-gel	Spherical	10 ppm dye concentration, 20 mg catalyst, UV lamp, 40 min	99	29
CuO-$Gd_2Ti_2O_7$	Two phase fabrication	-	Under visible lamp in 90 min	86.60	30
Ag-CdSe/GO	Solvothermal	Hexagonal	5 ppm dye, visible lamp, 25 min	97	31
Co- doped TiO_2	Hydrothermal	Agglomerated and non-uniform	Sunlight, UV lamp, visible light, 5mg Co^{2+}-TiO_2, 180 min	82, 31,and 74	32
ZnO	coprecipitation	Hexagonal	10 ppm dye, UV lamp, 180 min	89	33
TiO_2, ZnO	Synthetic	Spherical rod shape	9W UV lamp, pH 3, temp-25 deg, 30 ppm dye, 16 min	79.4 and 97.5	34
WO_3 Decorated 2D Graphene Sheet	Microwave irradiation	Highly porous 2D rGO sheets	UV lamp	97	35
rGO-Fe_3O_4/TiO_2	Hydrothermal	Spherical	100 ppm dye, 15 mg catalyst, 55 min, visible lamp	99	36
Copper Ferrite-Graphene Oxide	Hydrothermal	Hexagonal	0.01 g GO/$CuFe_2O_4$ was spread into 10 mL dye solution, 240 min	90.70	37
Fe(III)-Cross-linked alginate-carboxymethyl cellulose	-	Bead shape	pH=4, 10 ppm dye, 0.1 g catalyst, UV lamp, 30 min	98.80	38
ZnO	Green synthesis	Irregular hexagon	100 ppm dye, 10 mM catalyst, 150 min	Complete	39
CuO/ZnO	Sol-gel	-	180 min	79.70	40

ZnO	Green synthesis	Uniform spherical	fluorescent lamp (λ>400, 90 W) 10 ppm dye solution, 0.05 g catalyst, natural pH, 90 min	92	41
$CuWO_4$-RGO	Hydrothermal	Agglomerated with polycrystalline nature	2 ppm dye, 50 mg catalyst, 370 W mercury halide visible light, 60 min	93	42
$CuWO_4$-GO	Ball-milling	Micro-structured	0.05 g catalyst, 10 ppm dye, visible lamp, 80 min	95	43
RGO/TiO_2 /PANCMA	Electron spun	Hair like	3 mg catalyst, 100 ppm dye solution, pH=1, UV lamp, 60 min	91.40	44
CuO-modified silicon nanowires	Electroless deposition technique	Nanowires	Room temperature, UV (λ = 350–400 nm) or visible light (λ = 400–700 nm), presence of PMS, 100 min	98	45
La_2CuO_4 - decorated ZnO	Green synthesis	Non-uniform Spherical shape	2.5 mg photocatalyst, 25 mL dye, visible lamp (125 W) for 120 min	91	46
AgO, CuO, ZnO, and NiO	Electro biosynthesis	-	10 ppm dye, 0.5 g/L of catalyst, pH of 2, visible light irradiation, 60 min	91.70, 94, 98.50, and 97.30	47
GO	Hummer's method	Aggregates of irregular particles	500 W Halogen lamp, 2 mL dye, 20 mg of catalyst and 30 min	74	48
CuO	Green synthesis	-	Visible light	Complete	49
CuO	Biosynthesis	Rod like	pH 7	58	50
CuO	Co-precipitation	Spherical	0.03 g catalyst, 50 mL dye, room temperature	94.26	51
$La_{1-y}GdyNi_{1-x}Fe_xO_3$	Micro-emulsion	Spherical	10 mg catalyst, 10 ppm dye, Visible light, 50 min	96.40	52
TiO_2	Hydrothermal	Sphere	0.8 g catalyst, 5 x 10^{-6} dye, pH-8, UV Light, 120 min	95	53
Mn doped $ZnAl_2O_4$	Precipitation	Nano-sized grains	Visible light, room temperature,120 min	57	54
NiO	Combustion	Flower like	60 ppm dye, 0.2 g catalyst, 308 K temperature, 120 min	89	55
$ZnVFeO_4$	Precipitation	Hexagonal	1 g catalyst, dye solution at pH=3, Visible lamp, 180 min	90.1	56
TiO_2–NiO	Sonochemical	-	20 wt % catalyst, UV light	High catalytic performance	57
Bi_2MoO_6	Hard-template process	Irregular	10.4 ppm dye, 4 mg catalyst, 100 W, 6500 K LED floodlight, 480 min	91	58

TiO_2	Wet diffusional impregnation	Tetragonal	3 g catalyst, 80 ppm concentration, visible light, 180 min	Faster degradation	59
ZnO	Co-precipitation	Hexagonal	Visible light, 120 min, minimum dosage of 0.1 g catalyst	95	60
ZnO/CuO	Hydrothermal	-	0.2 g catalyst, 15 ppm dye, pH-10, UV lamp, 240 min	82	61
Chitosan–zinc oxide	Chemical Precipitation	Hexagonal	2.3 mg/L, dosage of 0.6 g and pH 8 within 180 min	98.5	62
$CuBi_2O_4$	Hydrothermal	Cubic like	10 ppm dye, catalyst of 0.1 g/ml w/v%, T=298K, pH=3	65	63
GO	Hummers Method	-	UV lamp , 90 min	85	64
Mn_3O_4	Ultra-sound assisted method	-	101- 162 mg catalyst, visible lamp	Maximum degradation	65
CuO/ZnO	Green synthesis	Spherical and cubic	1.8 X 10^{-5} M catalyst, 100 ppm, Microwave irradiation, 90 min	80	66
Ag_2O	Biosynthesis	Spherical	100 ppm dye, sunlight (600–800 lE m^{-2} s^{-1}), 240 min	94.7	67
TiO_2-inulin-Fe_3O_4	Simple method	-	Presence of light	Better removal	68
ZrO	Biosynthesis	-	50 ppm dye, 0.01% catalyst, room temp, visible light, 45 min	Complete degradation	69
CuO	Precipitation	Rod	10 ppm dye and 10 mg of catalyst	92.40	70
CuO nanosheets	Low temperature synthesis	Leaf like	Day light, 60 min. UV light, 60 min	94.80, 90.10	71
$CuMn_2O_4$	Co-precipitation	Flake like	1.370 × 10^{-5} M dye,0.5 mg catalyst, Sunlight, 60 min	92	72
$Na_{12}[CuNiW_{18}O_{62}]$	-	Uniform size	pH=7.5, 5.00 x 10^{-5} M, 0.10 g catalyst and light intensity 50.0 $mWcm^{-2}$, 160 min	Degradation at good rate	73
ZnO	Green synthesis	Spherical	70 ppm, 90 min.	90.70	74
CA/TiO_2	Green synthesis	Spherical globules	Visible light, 10 ppm dye, 240 min, 100 mg of catalyst	91.94	75
ZnO	Green synthesis	-	UV irradiation	69	76
GO with TiO_2	One step synthesis	Spherical	50 mg catalyst, 100 ppm dye, UV irradiation , 30 min	90	77

SnO_2 nanorods	-	Rod	15 min, visible light	24.70	78
FeO	Biosynthesis	Spherical	60 min, catalyst (0.5×10^{-8} - 2.0×10^{-8} mol/dm^3), dye (1.0×10^{-5} - 5.0×10^{-5} mol/dm3), pH (5), and high temperature (25-35 °C)	88	79
ZnO	Wet chemical	Nanoplates	Sunlight, 15 min	80	80
Al/RGO/Ag	Layer by layer assembly technique	Large, wrinkled and thin layered structure	20 mL of 1×10^{-5} dye, 1.02 % catalyst, 3 min	80.9	81
$MnFe_2O_4$	Microwave assisted combustion	Irregular shape agglomerates	30 mg catalyst, 50 ppm dye, 60 min under natural pH condition	Complete	82
Fe-Cu binary oxides.	Co-precipitation	Roughly spherical	50 ppm dye, 15 mg photocatalyst and placed under UV lamp (6 W, 254 nm with pH=7	95	83
Cu_2O	Electro-deposition	Cubic and regular shape	Catalyst dosage of 0.07 g, dye concentration of 4 ppm, light power of 250 W with natural pH, 120 min	78.185	84
MAl_2O_4	Chemical precipitation	Spherical	5 mg photocatalyst, $1x10^{-4}$M dye, 250 W tungsten halogen lamp, 120 min	80.4	85
Ti-BALDH	Simple method	Irregular	Varying dye concentration from 5 ppm to 20 ppm, visible light irradiation	88.5, 90.80, 91.7, and 94.2	86
Xanthan gum /SiO_2	Ultra sonication with polymerization	Lobule	10 mg catalyst, 450 ppm of MG dye, pH=7, temperature of 30 °C, 480 min	99.5	87
CuO	Thermal evaporation and annealing	Change in morphology is observed	Sunlight , 160 min	88.4	88
ZnO-SnO_2	Photopolymerization	Homogenous structure without pores	45 min of irradiation, pH-5 under Xe type, visible light with 1 g of photocatalyst	100	89
NiO/zeolite	Impregnation	-	LED irradiation, 240 min 10-50 ppm of water, 1.5 g of catalyst, pH 4.6	92 - 46	90

FeO	eco-cordial method	Non-uniform aggregates	20 mg catalyst, 50 mL of dye exposed for 5-h sunlight irradiation	97	91
$Mn_{0.5}Cu_{0.5}Cr_2O_4$	Hydrothermal	Uniformly distributed and closely packed particles	10 ppm dye, under visible light with 40 mg of catalyst, H_2O_2, 40 min	96	92
$ZnFe_2O_4$	Probe sonication	Spongy like	Under sunlight, UV lamp, it took about 180 min	98, 88	93
CuO	Thermal evaporation and annealing	Change in morphology is observed	Sunlight , 160 min	88.4	88
ZnO-SnO_2	Photopolymerization	Homogenous structure without pores	45 min of irradiation, pH-5 under Xe type, visible light with 1 g of photocatalyst	100	89
ZnO	Co-precipitation	Flower like	10 ppm dye, 0.5 mg catalyst, under UV irradiation (1000 Ultraviolet cross linker, energy x100 μ j/CM^2), 180 min	89	94

5. Conclusion

Though there are various ways to treat the dye contaminated water, photocatalysis plays a major role in waste water treatment. The industrial dye waste water which makes a great threat to the society can be resolved by the photocatalytic degradation method which has been widely employed. The work presented in this review showed that metal oxide nanoparticles can act as an efficient photocatalyst in treating industrial dye waste. These photocatalytic metal oxides can be reused after the proper cleaning treatment after separation by using centrifuge. By using the abundant naturally available source solar light, these industrial dyes should be treated before discharging into water bodies. The scarcity for the proper and safe drinking water can be resolved by photocatalytic treatment and the society can be benefitted.

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