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Visible Light Photocatalysis: The Role of Metal Oxide/Graphene Oxide Nanocomposites in Mitigating Aging-Associated Pollutants

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ARTICLE DETAILS

ABSTRACT

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Nanocomposites of graphene oxides (GOs) and metal oxides (MOs) have garnered tremendous attention in photocatalysis due to their remarkable properties and versatile applications. The combination of GOs and MOs leverage unique properties of both graphene and metal oxides like mechanical strength, excellent electrical conductivity, high catalytic activity, stability and large surface area. MO/GO have wide range of potential applications, particularly in visible photocatalysis for environmental remediation. These MO/GO nanocomposites act as photocatalysts in the presence of visible light, breaking down the harmful organic pollutants and chemicals from air, water and soil which cause plethora of harmful effects on health and decrease the quality of life. Various synthesis methodologies have been developed to fabricate these nanocomposites, each offering specific advantages based on the desired properties and applications. This review highlights the various synthesis procedures for MO/GO nanocomposites, focusing on their visible light harnessing for environmental pollutants degradation. Further this review seeks to provide insight into the efficiency, selectivity and optimization of various MO/GO nanocomposites for environmental applications which may lead to healthy lifespan or health span.

1. Introduction

Photocatalysis is one of the most efficient strategies for wastewater remediation using semiconductor-based materials. TiO₂ and ZnO are the widely used commercially available photocatalysts offering multifarious advantages like low cost, stability and environment friendliness¹. Though metal oxides have proved to be quite effective but still there is a lot of improvement scope for them due to their large reactive oxygen species (ROS) recombination rate, ineffective to work in the desired wavelength range, low surface area etc.,^{2,3}. Performance of metal oxide photocatalysts can be enhanced using metal loading^{4,5} and ion doping⁶, composite formation using 2 D Carbon-based materials like graphene, graphitic carbon nitride etc^{7,8}. Graphene, a 2-D carbon material has unique electronic and mechanic properties offers phenomenal properties for preparation of nanocomposites in enhancement of photocatalytic applications⁹. As most of the sunlight constitutes visible region, visible photocatalysis is highly demanding¹⁰. Fabrication of nanocomposites (NCs) which demonstrate the characteristics of both the constituent material i.e. graphene and metal oxides have overcome the drawbacks of both the counterparts¹¹. Graphene oxide (GO) has similar properties to graphene but has gathered immense attention due to the presence of introduced hydroxyl and carboxyl groups¹².

Metal oxide / Graphene oxide (MO/GO) nanocomposites can efficiently utilise the combinative merits of both to cover the three excellent attributes: increased absorptivity of pollutants, promotion of visible catalysis and efficient charge transportation and separation for ideal photocatalysis¹³. There have been various methods of synthesis of metal

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oxides/graphene oxides nanocomposites and metal oxides/reduced graphene oxides for visible photocatalysis such as hydrothermal method¹⁴, spread coating method¹⁵, covalent grafting¹⁶, solution phase approach¹⁷. Visible photocatalysis using metal oxide/graphene oxide and reduced graphene oxide nanocomposites has been verified on various organic pollutants like Methylene blue (MB), Methyl orange (MO), Rhodamine-B (Rh-B) dyes¹⁸, benzoic acid, PAHs, pharmaceutical drugs, heavy metals and pathogenic microbes¹⁹ which are directly or indirectly related to degrading the life span of humans. Photocatalytic efficiency depends majorly on reactive oxygen species (ROS) generation e.g. hydroxy (OH[•]) radicals and suppressing recombination of electron-hole pairs²⁰. On the other hand, anti-aging properties depends on the antioxidant nature of the materials, thus dual functional materials are best fit to solve the both the problems. Under certain conditions (such as light exposure or Fenton reactions), metal oxides can act as ROS generators, which makes them valuable for uses like antibacterial treatments or pollutant degradation. On the other hand, particularly in biomedical settings, they can also function as antioxidants, scavenging ROS and preventing oxidative stress. Metal oxides are useful in both environmental and medicinal applications because of their dual functionality. As graphene is an excellent acceptor of electrons, the presence of graphene in the composites enhances the catalytic performance of the semiconductor metal oxides²⁰. On the other hand, it enhances the both the photocatalytic activity in visible light as well the antioxidant nature of graphene maintains the ROS release hence reduces the recombination rate of ROS.

This review will examine different synthesis techniques for visible light-active metal oxides, graphene oxide (GO), and reduced graphene oxide (rGO). This is an overview of how several researchers have described them. After then, the emphasis will change to their use in visible light-driven photocatalysis, going into detail into the fundamental mechanics. The review will also look at several water pollutants that have a detrimental effect on human health and lifespan, showing how these toxins lead to problems associated with aging. Nanocomposites based on metal oxides and graphene offer a shared solution to the issues of water pollution and declining human health outcomes. To shed light on these materials' potential for commercial uses and overall viability, their capacity to breakdown pollutants under visible light will be examined and contrasted in tabular form.

2. Synthesis strategies for fabrication of MO/GO Nanocomposites

Common methods for graphene preparation include in situ growth, solution mixing, and hydrothermal or solvothermal techniques. Innovative approaches such as epitaxial growth via chemical vapor deposition, chemical reduction of graphite oxide, mechanical and liquid-phase exfoliation, in situ electron beam irradiation, laser reduction of polymer sheets, and carbon nanotube unzipping are also used^{21, 22}. Graphene synthesis has been categorized into two approaches: top-down, which reduces van der Waals forces in graphite, and bottom-up, where molecular building blocks form graphene layers through chemical, catalytic, or thermal methods²³. Metal oxide nanoparticles are commonly synthesized through techniques like sol-gel processes, hydrothermal and solvothermal methods, chemical co-precipitation, and thermal decomposition. Advanced methods include flame spray pyrolysis, microwave-assisted synthesis, and electrochemical techniques^{24,25,26}. These approaches enable precise control over nanoparticle size, shape, and composition. Synthesis methods are broadly divided into bottom-up approaches, where nanoparticles are assembled from molecular precursors, and top-down approaches, which involve breaking down larger materials into nanoscale particles through mechanical or chemical processes²⁷. Various synthesis procedures have been utilized for the synthesis of MO/GO nanocomposites are synthesized by techniques that are generally based on two approaches "top-down and bottom-up. Some of the techniques are thermal hydrolysis, spread coating, covalent grafting, hydrothermal synthesis, solvothermal, sol-gel^{28,29,30}, etc.

2.1 Thermal Hydrolysis:

In this synthesis at elevated temperatures chemical breakdown of substances takes place in water. In this method, the deposition of metal oxides on graphene sheets takes place in a controlled manner. Synthesis of Fe₃O₄ deposited graphene using thermal hydrolysis technique proved to be an excellent material for the removal of Arsenic from water²⁸.

2.2 Spread Coating:

In this method, very thin layers of metal oxides are coated onto the graphene oxide sheets to create uniform films that enhance the surface interaction between the graphene and metal oxide components. Using this technique researchers fabricated magnetic nanocomposites in which three different nanoparticles (TiO₂, Fe₂O₃, Ag) were loaded onto reduced graphene oxide for the successful degradation of Crystal violet dye from wastewater²⁹.

2.3 Covalent Grafting:

In this synthesis route metal oxides are covalently grafted onto the graphene sheets leading to better stability and interfacial interaction in the nanocomposites. Using this technique TiO₂-graphene nanocomposites have been fabricated for enhanced photocatalytic applications³⁰. Though this method is efficient but also time-consuming.

2.4 Hydrothermal:

In this method high pressure and high-temperature conditions are maintained using superheated solvents inside autoclaves to create nanoparticles. Highly crystalline materials are formed using this technique as high pressure and temperature enhances the solubility and reactivity of the nanoparticles. Using hydrothermal technique visible light active TiO₂/graphene have been fabricated for methylene blue dye degradation³¹. The schematic diagram shows the basic fundamentals for the synthesis of nanocomposite.

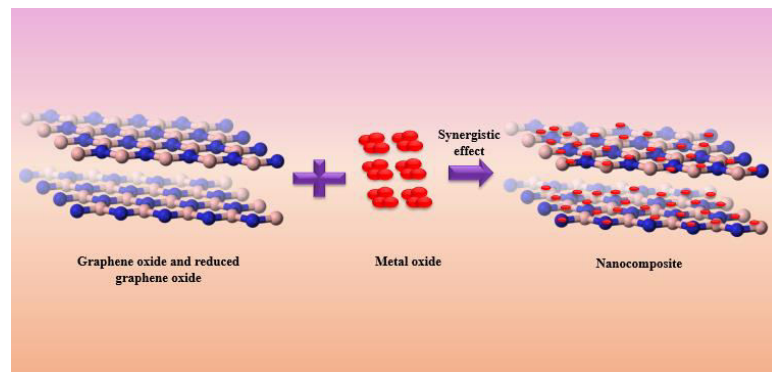


Fig. 1. General representation of fabrication of GO/MO nanocomposite. *Source:* Adapted from Nano energy³²

2.5 List of water Contaminants Accelerating the Aging Process:

Recalcitrant water pollutants contribute to aging directly or indirectly by accelerating oxidative stress, skin damage, and other biological processes. Some of the key pollutants³³⁻³⁹ leading to the aging process are given below

2.5.1 Heavy Metals:

Lead, Arsenic, Mercury are the most prominent heavy metals leading to both water contamination and aging. These metals generate free radicals, leading to oxidative stress in the cells which in turns breaks the collagen and elastin of skin leading to reduced skin firmness and elasticity. Also, long term exposure to the heavy metals leads to damaged internal organs causing overall premature aging.

2.5.2 Chlorine and Chlorinated Compounds:

Chlorine and chlorinated agents are worldwide used as a useful technique for water purification. The byproducts formed during the chlorination process strip the natural oils from skin making it more vulnerable to damage.

2.5.3. Pesticides and Herbicides

Pesticides and herbicides are being used tremendously in the agriculture sector. It can leach from the soil and is released into the water bodies causing both soil and water pollution. It contains chemicals which are endocrine disruptors interfering with the hormone balance and also promote oxidative stress in the cells.

2.5.3 Polycyclic Aromatic Hydrocarbons (PAHs)

PAHs are often present in water bodies near the industrial areas can easily penetrate the skin and induce DNA damage which can accelerate skin aging and lead to hyperpigmentation, rough skin texture and loss of elasticity.

2.5.4 Microplastics and Nanoplastics

Nanoplastics and microplastics have become a significant threat to the environment and human health. Microplastics have been found in drinking water, agricultural products, seafood making an easy route into the human body. Many toxic chemicals like persistent organic pollutants (POPs), Bisphenol (BPA) and phthalates can get attached to the microplastics disrupting human health.

2.5.5 Pharmaceutical Residues

Pharmaceuticals like drugs, antibiotics, hormones enter water systems can accumulate inside the body disrupting the endocrine system and metabolic processes. These residues also increase the risks of drug resistance leading to multiple disorders and weak immune system in humans.

2.5.6 Volatile Organic Compounds (VOCs)

VOCs, found in plastics, fuel, industrial solvents can induce oxidative stress and DNA damage accelerating skin aging as well as certain cancers.

2.5.7 Nitrates and Nitrites

These chemicals generally found in agricultural runoff and can convert into nitrosamines which are carcinogenic and cause oxidative stress.

Thus, most of the water contaminants contribute negatively on human health leading to reducing the life span of humans thus causing aging by inducing oxidative stress.

2.6 Role of ROS in elevating the aging process

Due to the detrimental effects of reactive oxygen species (ROS), which are byproducts of regular cellular metabolism, oxidative stress is a major factor in aging. ROS are very reactive chemicals that have the ability to harm essential biological constituents such as proteins, lipids, and DNA. ROS-induced accumulated damage causes cellular malfunction over time, which accelerates aging. ROS-induced DNA damage builds up with age, leading to mutations and compromising cells'

capacity to proliferate and repair them. Similarly, oxidized proteins lose their structure and function, which leads to age-related diseases such neurodegenerative disorders and cellular inefficiencies. Lipid peroxidation, in which ROS assault cell membranes, impairs cellular integrity and communication, hence exacerbating tissue damage⁴⁰. Due to the detrimental effects of reactive oxygen species (ROS), which are byproducts of regular cellular metabolism, oxidative stress is a major factor in aging. ROS are very reactive chemicals that can harm essential biological constituents such as proteins, lipids, and DNA. ROS-induced accumulated damage causes cellular malfunction over time, which accelerates aging (Figure 2).

ROS-induced DNA damage builds up with age, leading to mutations and compromising cells' capacity to proliferate and repair themselves. Similarly, oxidized proteins lose their structure and function, which leads to age-related diseases such neurodegenerative disorders and cellular inefficiencies. Lipid peroxidation, in which ROS assault cell membranes, impairs cellular integrity and communication, hence exacerbating tissue damage. Cellular components are harmed by this damage, which results in organelles like mitochondria malfunctioning⁴⁰. Oxidative stress is exacerbated by the concurrent reduction of the body's antioxidant defense system, which includes enzymes such as glutathione peroxidase (GSH-Px), catalase (CAT), and superoxide dismutase (SOD). Age-related illnesses like dementia, neurological diseases, weakened immune systems, and cardiovascular issues are all influenced by these processes. Oxidative stress is shown in the Figure 2 as a primary cause of aging and associated diseases.

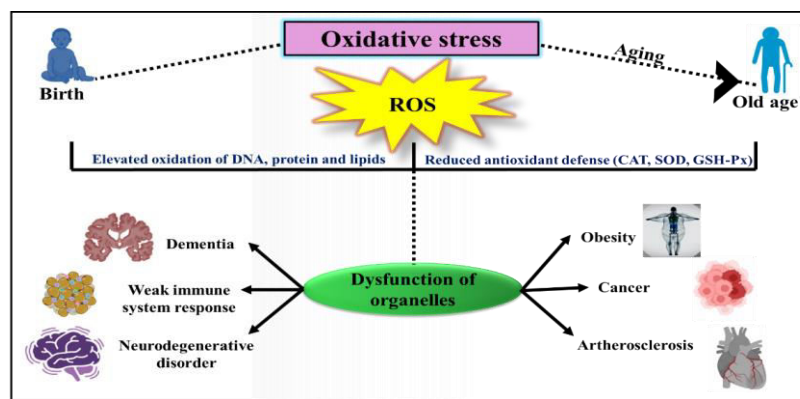


Fig. 2. Role of oxidative stress in aging process

3 Characterisation

Graphene oxides/metal oxides or reduced metal oxides nanocomposites can be characterised using X-ray diffraction (XRD), Scanning electron microscope (SEM) with energy dispersive X-ray diffraction (EDX), Transmission electron microscope (TEM), Fourier transform infrared spectroscopy (FTIR), X-ray photoelectron spectroscopy (XPS), Thermogravimetry analysis (TGA). SEM reveals the exfoliated sheet or leaf like morphology of Graphene oxide sheets onto which the deposition of metal oxides can be clearly seen. Also, the size of nanocomposites can be confirmed through the TEM results⁴¹⁻⁴³. XRD patterns of metal oxide, graphite, and the nanocomposite shows the characteristic peak for the exact identification of the components. 2θ value is at 26.40° for graphite which reduces to 11.2° for graphene oxide (GO) showing the oxidation process. This peak in GO is due to various oxygen containing functional groups on the surface of GO⁴¹. For investigating the functional groups in the nanocomposites FTIR studies are performed which gives the GO peaks at 1730 cm and 1630 cm corresponding to C=O and C=C stretching and peaks reported in literature⁴⁴. XPS tells the binding energies of the nanocomposites and also the phenomena of adsorption of dyes and their degradation can also be analysed using XPS⁴⁵. For studying the thermal stability of MO/GO nanocomposites TGA can be performed and weight loss of different components of nanocomposites is checked⁴⁶.

4 Visible Photocatalysis

Visible Photocatalysis refers to the process in which visible light is used to activate a substrate (photocatalyst) which generally facilitates the rate of reaction but itself remains unconsumed⁴⁷. The efficiency of visible photocatalysis of metal oxide increases to many folds after forming composite with GO or rGO⁴⁸. After absorption of the light source excitation of an electron from the valence band to the conduction band occurs, and recombination should be suppressed for the overall efficiency of the photoinduced process^{49,50}. Photocatalysis in visible light is cost-effective, more efficient, and reusable. Two very important methods are widely used to evaluate visible photocatalysis performance and efficiency: Colour decolourisation and absorbance reduction from UV-Vis spectrophotometer to calculate the % efficiency of degradation. Graphene oxide/Metal oxide nanocomposites, due to enhancement in their properties, have better visible photocatalytic efficiency than other materials. The mechanism of visible photocatalysis utilizing metal oxide/graphene nanocomposites involves several key steps. Initially, a visible light source excites electrons in the metal oxide, such as titanium dioxide (TiO_2) or zinc oxide (ZnO), leading to the generation of electron-hole pairs. The excited electrons transition to the conduction band, while holes are created in the valence band. This process facilitates the efficient separation of charges, with electrons transferring to the graphene layer. Subsequently, these electrons engage in reduction reactions with electron acceptors like oxygen, generating reactive oxygen species (ROS), while the holes react with water or organic

substances to produce hydroxyl radicals ($\bullet\text{OH}$). Overall, this photocatalytic process effectively leads to the degradation of pollutants in the environment⁵⁰.

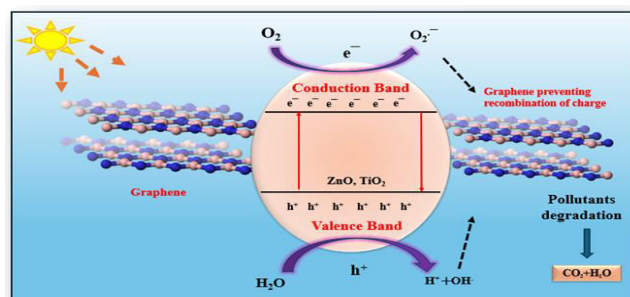


Fig. 3. Mechanism of visible light photodegradation of MO/GO nanocomposite. Several nanocomposites have been utilized for the degradation of pollutants from water which are tabulated in Table 1 showing the comparison, efficiency rate and target pollutants in presence of visible light⁵¹⁻⁵⁹.

5 Conclusion

One of the most prevalent issues affecting human health span and shortening human lifespan is water contamination, which may be resolved via graphene and metal oxide-based nanocomposites. The works related to synthesis, characterization and visible photocatalytic application of GO or rGO/ Metal oxide nanocomposites using various methodologies have been discussed and compared on various parameters in which simple impregnation methodology of synthesis is easy, efficient, and cost-effective. Also, in comparison to only metal oxide nanoparticles or nanocomposites the incorporation of rGO or GO drastically enhances the photocatalytic efficiency for the pollutants in the visible range along with high reproducible rates. Metal oxides and graphene nanocomposites have a promising future in wastewater treatment because of their capacity to improve sustainability, efficiency, and selectivity which are not possible through conventional routes. These materials should be crucial in solving the world's water pollution problems as research progresses since they provide scalable, affordable, and ecologically friendly solutions.

Table 1. Some recently published studies focused on Metal oxide-GO nanocomposites and their effectiveness in degrading major pollutants for environmental remediation under visible light conditions.

Nanocomposite	Method of synthesis	Target pollutant	Graphene incorporation method	Photocatalytic efficiency under visible light
TiO ₂ /Graphene ⁵¹	Hydrothermal	Methylene blue dye	In situ reduction of graphene oxide (GO)	~98% degradation in 60-90 min
ZnO/Graphene ⁵²	Sol-gel	Phenol	Solution mixing with reduced graphene oxide	~90% degradation in 120 min
Fe ₂ O ₃ /Graphene ⁵³	Hydrothermal	Rhodamine B dye	GO mixed in the precursor	~85% degradation in 180 min
CuO/Graphene ⁵⁴	Chemical co-precipitation	Methyl orange dye	GO added during metal oxide precipitation	~88% degradation in 120 min
SnO ₂ /Graphene ⁵⁵	Solvothermal	Methyl orange dye	GO dispersed and reduced simultaneously	~95% degradation in 60 min
CeO ₂ /Graphene ⁵⁶	Thermal decomposition	Rhodamine B dye	GO reduced by high-temperature process	~92% degradation in 90 min
ZnO-GO/Nanocellulose ⁵⁷	Hydrothermal	Ciprofloxacin organic pollutant	ZnO mixed with GO and nanocellulose	~98% degradation in 120 min
GO/Goethite ⁵⁸	Hydrothermal	Tylosin organic pollutant	GO mixed with goethite nanoparticles	~84% degradation in 120 min
CuO-CeO ₂ /GO ⁵⁹	Hydrothermal	Tetracycline and Methyl orange dye	CuO mixed with CeO ₂ and GO	~90% and ~95% in 120 min respectively

Metal oxide nanoparticles exhibit unique antioxidant properties, enabling them to neutralize reactive oxygen species (ROS) such as superoxide anions, hydroxyl radicals, and hydrogen peroxide. By mimicking the activity of antioxidant enzymes, these nanoparticles play a vital role in combating oxidative stress, which is both a cause and consequence of aging and age-related diseases. Graphene and graphene oxide itself have antioxidant properties thus proving the composites capability of removing anti-aging pollutants from water without itself doing much harm addressing both health and ecological concerns.

6. Author Contributions

Aishwarya Singh wrote the original draft and did editing. Meenakshi Sharma conceptualized the idea, reviewed the original draft and did final editing.

7. Conflict of Interest

Authors declare no Conflict of interest.

8. Funding Information

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