



REVIEW

Graphene's Role in the Clean-Up Act: Progress in Industrial Wastewater Treatment

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Water scarcity and environmental pollution are the two pressing global challenges that demand innovative solutions. In recent years, graphene-based nanomaterials have emerged as promising candidates for revolutionizing the field of water purification, particularly in the treatment of industrial wastewater. This review article, the first part of a two-part series, comprehensively explores the applications, synthesis methods and mechanisms of graphene-based nanomaterials for industrial wastewater treatment. Industrial activities generate vast quantities of wastewater contaminated with a myriad of organic and inorganic pollutants, heavy metals and dyes. Traditional water treatment methods often fall short in efficiently removing these contaminants, leading to significant environmental repercussions. In contrast, graphene-based nanomaterials exhibit exceptional properties, including a high specific surface area, excellent electrical conductivity and superior mechanical strength, making them highly attractive for wastewater treatment applications. This article begins by providing an overview of the properties of graphene and its derivatives, such as graphene oxide (GO) and reduced graphene oxide (rGO), which form the foundation of graphene-based nanomaterials. Our discussion focuses on the many approaches to the synthesis of these nanomaterials, with particular attention paid to their scalability, cost-effectiveness, and influence on the environment. This review focuses on the various applications of graphene-based nanomaterials in the removal of organic pollutants and dyes through adsorption, coagulation and photocatalysis. The unique structural and chemical properties of graphene-based nanomaterials enable them to selectively adsorb pollutants, even in the presence of competing ions, making them highly efficient in complex wastewater matrices. Furthermore, we explore the mechanistic aspects of pollutant removal by graphene-based nanomaterials, elucidating the role of surface functionalization, intermolecular interactions and photocatalytic processes. In addition to their pollutant removal capabilities, graphene-based nanomaterials also exhibit remarkable regeneration potential, making them economically sustainable for long-term industrial wastewater treatment applications. The challenges associated with large-scale implementation, including cost considerations, environmental impact assessments and regulatory compliance are also discussed. This review aims to provide a comprehensive understanding of the current state of research in graphene-based nanomaterials for industrial wastewater treatment. By critically evaluating the existing literature, this review serves as a valuable resource for researchers and practitioners seeking to advance sustainable and effective strategies for the industrial wastewater treatment through the integration of graphene-based nanomaterials.

Keywords: Graphene-based nanomaterials, Industrial wastewater treatment, Water purification, Pollutant removal.

INTRODUCTION

With the increasing population, as the demand for chemical and pharmaceutical industries increases, a re-evaluation of the issue of environmental pollution was undertaken worldwide at the end of the last century. The inevitable release of waste products, such as medicines, disinfectants, laundry detergents, surfactants, pesticides, dyes, paints, preservatives, food additives and cosmetic products, due to rapid industrialization and

aggressive urbanization, is the main cause of environmental pollution. Water is the most plentiful of all natural resources. Water bubbles, splashes and the flowing of water persist as symbols of existence [1]. The escalating global population is causing a rapid expansion of the chemical industry and industrial waste is the main culprit behind water pollution. These pollutants are discharged into natural water supplies. Therefore, despite the pronounced prevalence of science and technology worldwide, uncontaminated water is becoming a scarce resource.

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This issue has become a major concern for the environment and it is critical to eliminate the harmful and hazardous effluents from streams and increase the quantity and quality of portable water [2]. Modern and advanced technologies are required for wastewater treatment to secure the safety of aquatic life and eradicate dreadful health hazards, ensuring the welfare of society and future generations. Water pollution may be defined as the contamination of water by foreign materials that render it harmful to the health of human beings, animals, plants or aquatic life and make it unfit for domestic, industrial or agricultural use. Pollution of water originates from human activities that, through various pathways, contaminate groundwater or surface water. The major water pollutants and their sources include microorganism pollutants from domestic sewage, organic waste from domestic sewage, animal manures, food processing waste, decaying plants and paper discards, as well as rags and other bio-degradable wastes. Plant nutrients come from chemical fertilizers, toxic heavy metals originate from chemical factories, pesticides stem from chemicals used for killing insects, fungi and weeds and radioactive waste results from mining uranium containing minerals and accidental releases from nuclear plants. Heat also pollutes water and harms aquatic life, primarily due to the hot water discharged by industrial plants used for cooling purposes [3-5].

Wastes can be broadly classified into two categories: sewage and non-sewage. Domestic wastewater is commonly referred to as sewage, originating from various sources such as houses, kitchens, schools, hospitals, restaurants, hotels and public restrooms. In daily life, numerous instances of domestic sewage containing human waste, such as urine and feces, emerge from these buildings. Non-sewage wastewater encompasses various other forms of wastewater that do not contain human waste, including rainwater, stormwater resulting from flooding and water generated by commercial activities and industrial facilities, such as laundrettes, commercial car washes and washing machines. Non-sewage wastewater can be recovered and managed at a low cost, given its absence of human waste [6]. However, wastewater can be classified into distinct types and subtypes for efficient handling and treatment. Stormwater runoff wastewater is one of the rapidly growing sources of pollution. When rainwater strikes lawns, rooftops, driveways, streets, parking lots, construction and industrial sites, a copious number of noxious pollutants such as oils, pesticides, plastics, chemicals, heavy metals and pathogens get rinsed away. It tends to flow into the channeled drains that connect with natural waterways, posing a threat to all life forms dependent on these natural streamways for their survival. Domestic wastewater originates from human household activities, including bathing, laundry, dishwashing, garbage disposal, toilet flushing and food preparation. This wastewater should be effectively managed and treated to ensure its safe and secure discharge into the atmosphere [7].

Domestic wastewater is classified into three categories *viz.* black-water, greywater and yellow water. Blackwater primarily originates from the restrooms and contains fecal matter, urine, soaps, toilet paper and cleaning liquids. It can also come from kitchens and household activities. When water becomes contaminated with pathogens and grease, it can be categorized as

blackwater. Greywater is typically discharged from bathtubs, washing machines, sinks and showers. It is less contaminated as it does not contain fecal matter or urine, making it non-pathogenic and therefore, treatment and recycling of greywater are easier. Yellow water consists mainly of pure urine and lacks the contaminants found in greywater and blackwater, such as chemicals, toilet paper, feces and food particles [8].

Yellow water, which contains nutrients that can transform urine into a viable fertilizer suitable for agricultural use and readily absorbed by plants, should be separated from other wastewater streams. Urine serves as a more environmentally friendly option compared to chemical fertilizers since it contains fewer heavy metals and pathogens. Agriculture wastewater originates from various farm activities, including animal and poultry watering systems, animal feeding operations, barnyard and feedlot runoff, egg washing, processing and slaughterhouse wastewater. Additionally, pollutants such as fertilizers, pesticides, insecticides, fish farm effluents and waste from animal and dairy farming enter nearby waterways, leading to the severe water pollution. The excessive nutrients in animal waste pose a threat to overall water quality, contributing to nutrient pollution, wherein excess nitrogen and phosphorous infiltrate natural waterways, fostering algae growth that can cover the surface water and release toxins. The accumulation of nutrients in water and the subsequent loss of biodiversity are key components of eutrophication [9-11].

Industrial wastewater is generated as a result of the release of major water pollutants in chemical industries. The primary water pollutants in dye manufacturing industries include sulfur, nitrates, chlorine compounds, arsenic, mercury, nickel, cobalt and chromium (known to be carcinogenic). Lead can lead to neurological damage in children and cardiovascular diseases in adults. Meanwhile, the pharmaceutical industries produce cadmium, nickel and phenolic compounds as major pollutants. When inadequately treated water is discharged into streams, it can have adverse impacts on aquatic ecosystems and public health. In case of paper and pulp industries, inorganic pollutants such as ferrous, copper, zinc, nickel and magnesium are toxic to juvenile channel catfish and can accumulate in the liver, ovary, kidney and muscles. Additionally, major organic and inorganic pollutants like methanol, phenol, hexadecanoic acid, β -sitosterol, trimethyl silyl ether, 1-tetradecane, 2-methoxyphenol, trichloroguaiacols, terpenes, benzoic acid and abietic acid have been detected and reported as endocrine-disrupting chemicals (EDCs), which can harm reproductive systems [12].

Major water pollutants in the form of heavy metals include cadmium, lead and mercury. Heavy metals pose a significant global threat to water quality. Cadmium and mercury are known to cause kidney injuries, while lead is responsible for damage to the kidneys, liver, intestines, brain and central nervous system. Mercury poisoning is also linked to minamata disease in humans, leading to damage to the nervous system, particularly the central nervous system, primarily due to methylmercury exposure. These metals are considered cumulative poisons because they accumulate in the body and cannot be naturally expelled. In addition to heavy metals, petrochemical industries contribute to water pollution with toxic petroleum hydrocarbons such as

natural gas (methane), phenolic compounds, alkanes, chloroalkanes and nitrobenzene. Refineries are a significant potential source of water contamination. Incidents like oil tanker breakages at sea result in oil spills in the ocean, creating a broad film of oil on the sea surface known as an oil slick. This poses a severe threat to ecosystems and marine life due to its precarious and toxic nature [13]. Water pollutants from the plastic industry include perfluoro octanoic acid (PFOA), cadmium, mercury, lead and diethyl hexyl phthalate. Plastics degrade into microplastics, infiltrating the marine food chain and causing distress to aquatic life and ecological equilibrium. Textile industries, in particular, discharge significant amounts of organic dye

chemicals, leading to water pollution and altering the physical and chemical properties of freshwater. Coloured dye wastewater is a direct byproduct of dye production and its extensive industrial use. Dyes are highly visible substances, so even a low concentration (< 1 ppm) imparts colour to water. Unfortunately, removing the persistent organic pollutants is a highly complex process [14,15]. The impact that have different types of industrial waste on the surrounding environment can be represented in Fig. 1.

Organic dyes and its impact on health and environment:

Dyes can be classified into two types viz. natural and synthetic dyes. Natural dyes, such as indigo and madder, are sustainable,

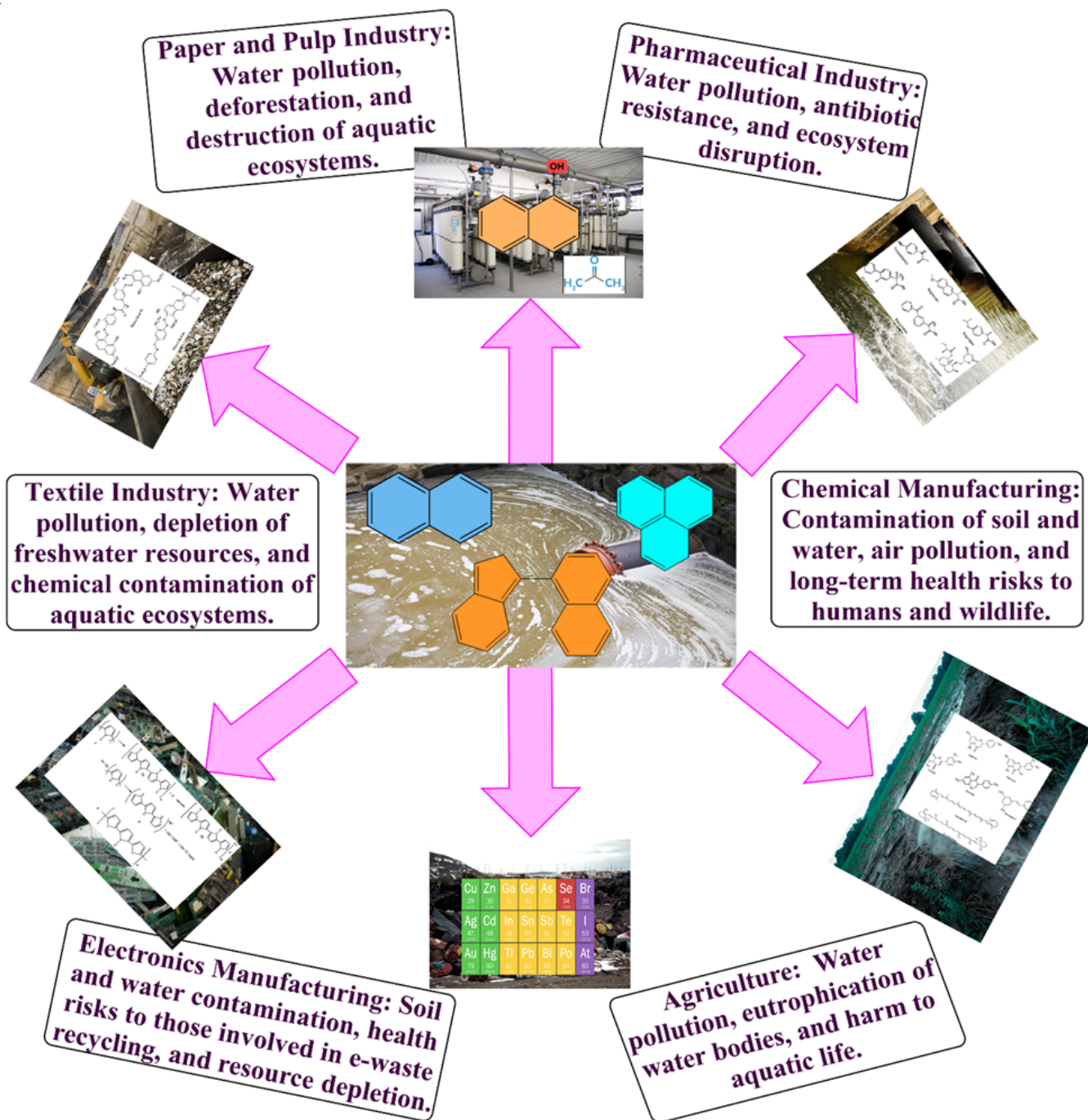


Fig. 1. Sources of various industrial waste and its effect on environment

biodegradable and do not contain noxious substances or chemical additives, as they consist of natural and organic dyes. On the other hand, synthetic organic dyes are simpler, less expensive to synthesize and available in more colors than their natural equivalents, however, they contain harmful chemical ingredients. Synthetic organic dyes play crucial roles in modern-day life, serving various industries, including paper printing, textiles, leather, pulp, cosmetics, food and pharmaceuticals, as well as in the scientific laboratories. These dyes consist of compounds with multiple aromatic rings fused or linked by various C-C, C-O and C-N linkages and they feature functional groups such as -OH, -NH₂ and -SO₃⁻ that enable them to absorb light at various wavelengths and frequencies of the chromophore. These functional groups, known as auxochromes, are responsible for producing colour, making the dyes more hydrophilic in nature and enhancing their binding capacity to textile fabrics [16]. There are around 10,000 different dyes commercially accessible worldwide, each exhibiting an extensive variety of colors and possessing varied chemical structures [17-19].

However, synthetic dyes have a significant adverse impact on the human body and the environment (Fig. 2). The residual dyes obtained from various textile industries discharged into water are carcinogenic to both humans and aquatic life due to the presence of naphthalene, benzamine, anthraquinone and other aromatic moieties. The untreated effluents from the chemical industries cause prevalent health problems such as respiratory sensitization, disorders of the central nervous system, allergic conjunctivitis, skin irritations and various tissue changes

due to the decomposition of dyes into carcinogenic pollutants [20,21]. The effects of azo dyes on human health and the environment are a cause for concern. In general, the production of azo dyes, particularly the highly water-soluble ones with amino groups, high boiling points, low Henry's law constants and high mobility compared to hydrocarbons, in the textile industry poses acute health hazards. This is primarily due to the inhalation of dye particles, which are breakdown products of azo dyes and can lead to various health issues, including permanent blindness, acute tubular necrosis, facial edema and respiratory diseases, as they are absorbed through the lungs, skin and gastrointestinal tract [22-24].

Despite the indisputable contributions of the textile industry, it is accountable for enormous ecological contamination in current advancements. The discharge of effluent containing dyes becomes a significant environmental concern due to its poor biodegradability and toxicity. In textile industry, during the dyeing process, achieving the fixation of 100% dye on textile fibers through physical absorption, which involves van der Waals forces, hydrogen bonds and hydrophobic interactions, is not possible [25]. There is always a fixed portion of dyes that is lost in textile effluents and is washed away by water. The unfixed portion of dyes, which do not bind to fibers, causes significant environmental pollution. Fixation degrees for different dye classes vary. For basic dyes, acid dyes, disperse dyes, direct dyes, reactive dyes, sulphur dyes and metal complex dyes, the percentages of loss in effluents are 0-5%, 5-20%, 0-10%, 5-30%, 10-50%, 10-40% and 2-10%, respectively [26].

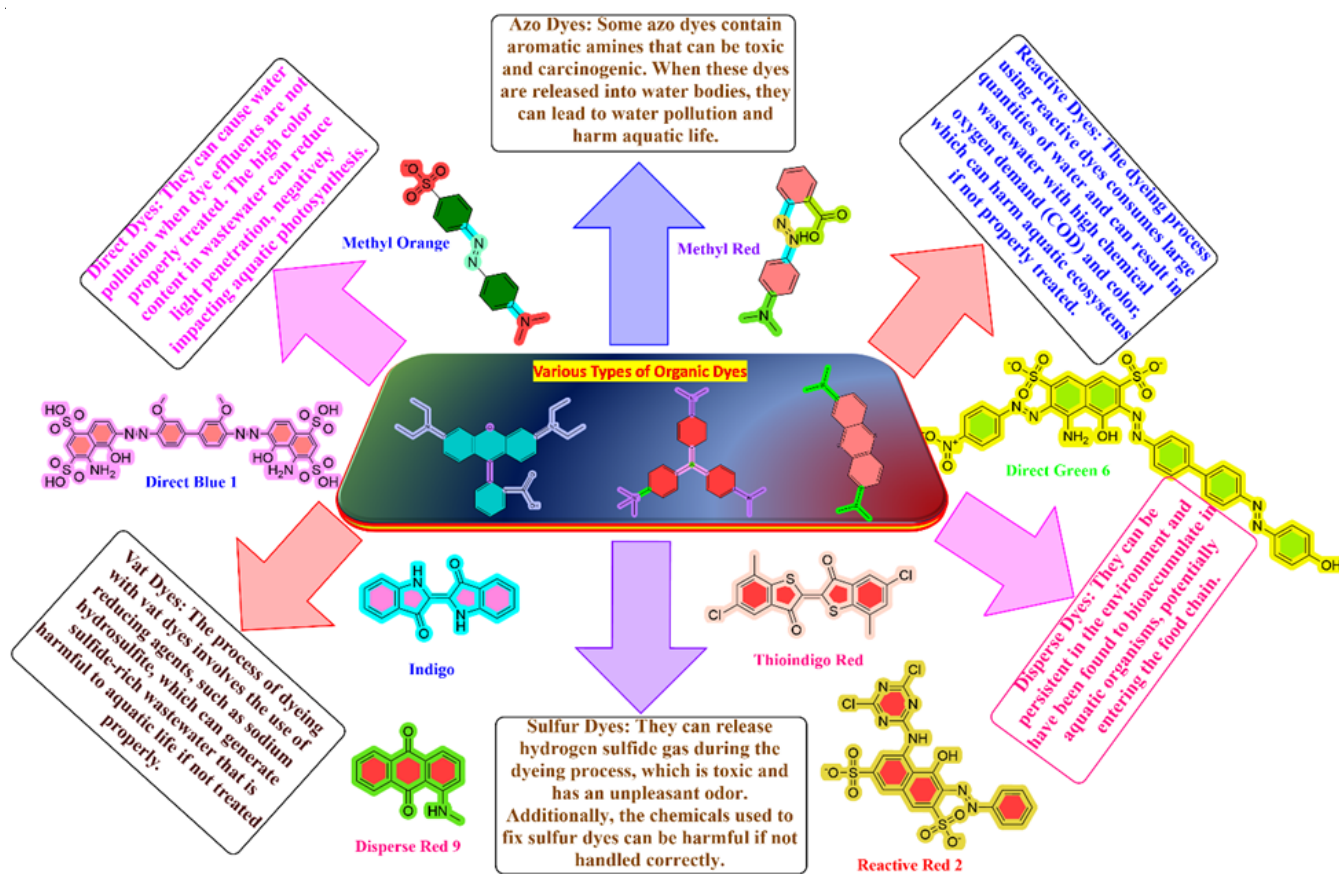


Fig. 2. Various types of organic dyes and their effects on environment and health

These dyes hinder the oxygenation capacity of receiving water in aquatic bodies, as photosynthetic power eventually decreases due to the depletion of sunlight penetration by dyes through the stream. Thus, synthetic organic dyes harm the biological activity of aquatic plants and also contribute to the accumulation of toxic effluents in the bodies of several aquatic life forms, leading to cancer and distress in the biological cycle and ecosystem. It has become a serious challenge for environmental scientists to protect future generations and the environmental ecosystem, especially to save aquatic life in streams and to remove the danger of water supply contamination. In this review, we discuss the purification of wastewater from synthetic organic dyes using graphene and graphene-based nanomaterials [27,28].

Graphene: Graphene, a rising and exceptional material is a one atom thick planar 2D nanosheet packed in honeycomb crystal lattice with a carbon-to-carbon bond length of 0.142 nm, discovered in 2004 by Geim *et al.* [29]. Its unique intrinsic properties make graphene extraordinary. Graphene is incredibly flexible and a single defect-free graphene layer possesses the highest intrinsic tensile strength, capable of withstanding

up to 42 Nm^{-1} , while also having stiffness similar to graphite. Graphene boasts high intrinsic mobility ($200,000 \text{ cm}^2 \text{ v}^{-1} \text{ s}^{-1}$), exceptional crystal quality and chemical tunability. Additionally, it exhibits excellent electrical conductivity, akin to copper, as well as remarkable thermal conductivity (with in-plane conductivity of $5000 \text{ Wm}^{-1} \text{ K}^{-1}$ at room temperature, five times that of copper; though when attached to another substrate, it can drop to as low as $600 \text{ Wm}^{-1} \text{ K}^{-1}$, with cross-plane conductivity at $6 \text{ Wm}^{-1} \text{ K}^{-1}$). Furthermore, graphene has a density four times lower than copper, a high Young's modulus of 1000 Gpa, effective moisture barrier properties and a spacious range of light absorption abilities. Its high optical transparency, due to being only one atom thick, is noteworthy, as are its remarkable electronic properties, as graphene exhibits no bond gap thanks to overlapped π -electrons across its surface, facilitating the easy and fluid movement of electrons. Lastly, its high planar surface area, at $2630 \text{ m}^2 \text{ g}^{-1}$, makes it well-suited for a broad range of applications [30-33] (Fig. 3).

Synthesis of graphene: Graphene, the first lab-made 2D atomic crystal, was explored in 2004 by Geim & Novoselov [29]. For their innovative work, they received the Nobel Prize

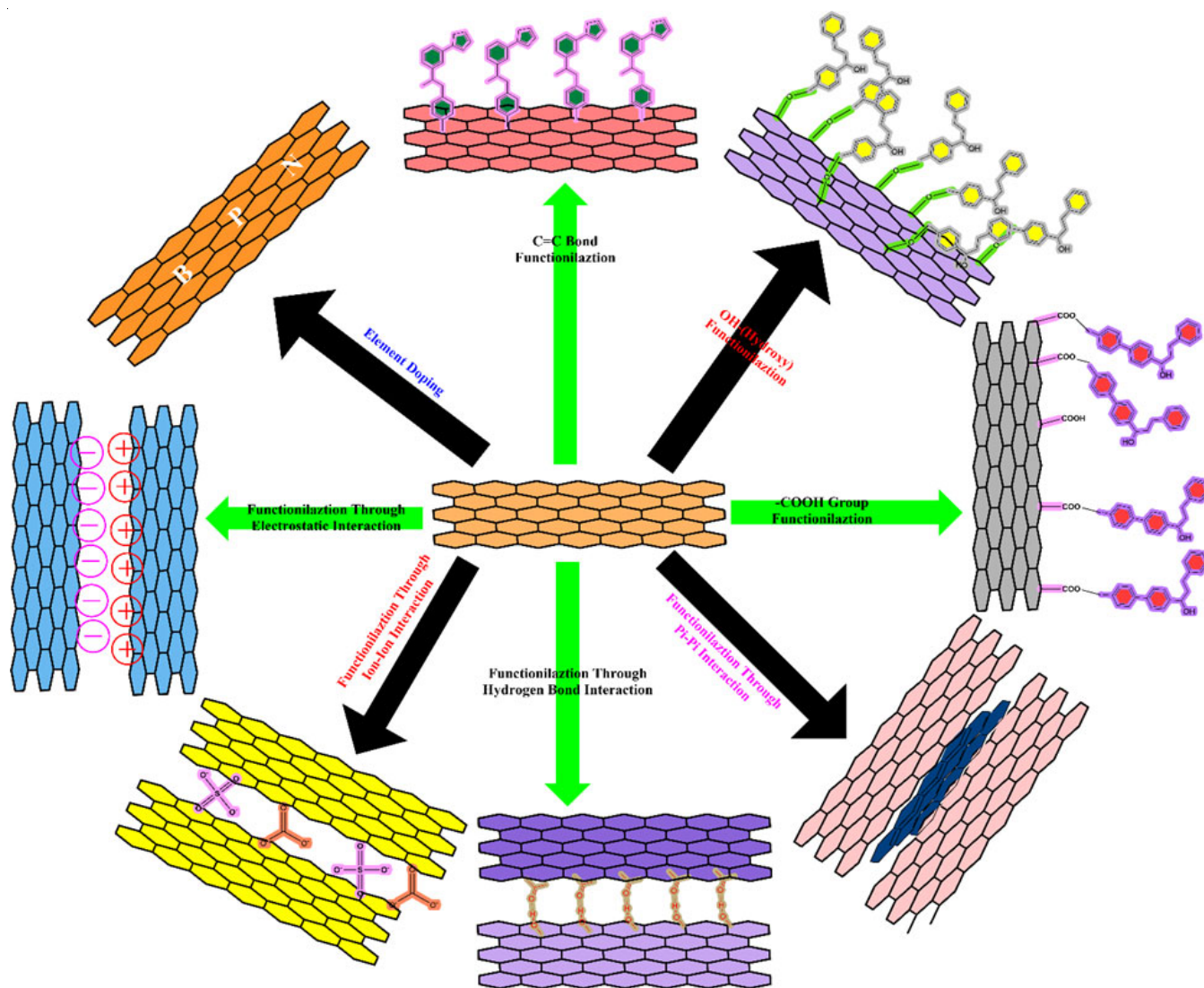


Fig. 3. Surface modification of graphene oxide for various applications

in 2010 in physics. Graphene can be wrapped up, shaping zero-dimensional fullerene. Rolling up graphene sheets produces 1D carbon nanotubes. Single-walled CNTs are produced from a single layer of carbon atoms, whereas multi-walled CNTs are produced when comprising a few layers of graphene sheets held together by weak van der Waals forces, responsible for the formation of 3D graphene. Hence, the thinnest material in the universe, graphene is the fundamental building block of CNTs, fullerene and graphite [34].

Top-down approach

Mechanical exfoliation: This is a top-down technique of nanotechnology. Micromechanical exfoliation of graphite is used to extract one-atom-thick 2D carbon nanofiller, graphene flakes, which was first acknowledged as a method for graphene discovery by Geim & Novoselov [29]. The interlayer distance and interlayer bond energy in a graphite sheet between two monoatomic layers of graphene are 3.34 Å and 2 eV/nm², respectively. In graphite, graphene layers stack together due to the overlap of filled π -orbitals perpendicular to the graphene sheet, associating van der Waals forces. Weak forces and a large lattice gap in the perpendicular direction of graphene flakes are attributed to the exfoliation of graphite. Exfoliation includes the peeling of highly ordered pyrolytic graphite (HOPG) using adhesive tape. Few-layer graphene (FLG) can also be produced by this method. Geim & Novoselov [29] developed a graphene film stabilized on silicon support (Si/SiO₂), with the largest film size being 10 μm^2 . Despite this simplest and most economical method being recurrently used in laboratory experiments for producing high-quality graphene, the yield of graphene monolayer is low. Therefore, this method is not appropriate for the scalable production of high-purity graphene. The production of the largest size unmodified graphene is the only advantage of this method [35].

Liquid phase exfoliation: It is a new top-down approach to graphene synthesis charged with the wide-reaching manufacture of graphene. It involves the dispersion of graphite flakes into a solvent using intercalating agents, reducing the interaction between layers in graphite and leading to exfoliation by sonication or high-shear mixing to obtain a stable dispersion of monolayer or few-layers defect-free graphene. The surfactants act as walls, preventing the assemblage of graphene to form graphite by surrounding the single atomic layer of graphene as stacking up of graphene flakes expedites graphite formation. The ultrasonication time is typically 60 min with 250-500 W power. The most widely used solvent for this method is N-methyl-2-pyrrolidone (NMP) as recommended by Coleman *et al.* [36]. Owing to the fact that single and multiple-layer graphene produced by this method, as a consequence, difficulty in separation and low yield are the main pitfalls of this method. But the fabrication of unmodified graphene by this method is economical [37].

(a) Sonication: Sonication is one of the liquid-phase exfoliation (LPE) methods with colossal potential for producing mono-layer or few-layers graphene at relatively high concentrations. Ultrasonic waves proliferate during compression and rarefaction through a medium, exerting high and low pressure

areas that are accountable for the production of cavitation bubbles, as these high- and low-pressure areas push and pull at the molecules. The broadening and subsidence of cavitation bubbles hence make the power of sonication influential in persuading physical and chemical changes. Sonication-assisted LPE includes the following steps: dispersion of graphite in a solvent, followed by exfoliation of the dispersion, especially *via* cavitation bubbles in sonication and repeated filtering of the solution for separating graphene based on their size and the number of layers to obtain purified graphene. There are two types of sonication tips, A and B and both types of sonication can be employed simultaneously or individually [38].

(b) High-shear mixing: It involves the exfoliation of graphite in a liquid under shear force through the use of high shear mixers. A significant breakthrough in graphene synthesis using a new exfoliation technique emerged from Coleman *et al.* [36] for extensive film growth. They revealed that a highly concentrated dispersion of graphene nanosheets can be achieved through high shear mixing of graphite in solvents. The shear exfoliation method is more effective than sonication. One of the emerging applications of high shear mixing is its implementation in synthesizing graphene oxide (GO) using a modified Hummer's method.

(c) Micro-fluidization: It is a high-pressure homogenization technique and involves the exertion of high pressure on fluids, which impels the fluid to traverse a microchannel with a diameter less than 100 μm . This process is superior to sonication high shear mixing because of the high shear rate (greater than 10^6 s^{-1}) that is widespread throughout the entire fluid region. This exfoliation technique is prompted by cavitation, shear force and collision. Karagiannidis *et al.* [39] performed bespoke exfoliation of graphite in an SDS aqueous solution using a microfluidic processor. The yield was 100% and no additional washing for distillation or centrifugation occurred during exfoliation under a shear rate condition of 10^8 s^{-1} .

Electrochemical exfoliation: This technique presents a promising approach for the effective production of graphene from graphite on a large scale. An applied voltage causes the ionic species to propel and intercalate into the graphite electrode. Then, these ionic species pass over the interstitial region of the graphite and the local formation of gases (oxygen, sulfur dioxide) swells the interlayer distance in graphene. As a result, the van der Waals force is broken, accelerating the exfoliation process and producing graphene sheets. The electrochemically exfoliated graphene nanosheets (GNs) are completely dispersed by sonication. For example, Salverda *et al.* [40] reported that the exfoliation of graphite rod used as anode, when applying a 10 V direct current, produced the bilayer graphene sheets with a C/O ratio of 11.7 and an average lateral size of approximately 0.5 to 1 μm . Single-step functionalization and high electrical conductivity are advantages of this method. The cost of ionic liquids and the erosion of the graphite electrode during the process hinder the feasibility of this route.

Chemical reduction of graphene oxide (GO): Graphene oxide (GO) is synthesized by the oxidation of graphite using conc. H₂SO₄, HNO₃ and KMnO₄ as oxidants. This process involves Brodie's method, Staudenmaier's method and the most compre-

hensively used Hummer's method. Previous studies have demonstrated that chemically reduced graphene possesses hydroxyl groups on the layer plane and edges [41].

Bottom-up approach

Chemical vapour deposition (CVD) method: CVD is the most significantly used propitious technique of the bottom-up approach due to its easy accessibility monetarily, optimum production of yield and facile laboratory arrangement, forming single-layer and few-layer graphene films. CVD encompasses the deposition of a thin film of high-quality graphene on the surface of transition substrates like Ni, Cu, Pd, Pt and Ir. CVD growth of graphene onto Ni and Cu is suitable for productivity [42-44].

Thermal CVD method (hot wall CVD): The copper substrate is placed inside a quartz-made furnace tube, heated to around 1000 °C. Low or atmospheric pressure is maintained by a pump in the reaction chamber. The first step involved introducing a flow of hydrogen gas for annealing to remove the oxide layer from the metal surface. During the annealing step, hydrogen molecules or atomic hydrogen chemically adsorbed onto the copper surface. During the growth stage, hydrocarbon gas, mainly methane blended with hydrogen, was introduced into the reactor at a fixed ratio through the gas delivery system. The ratio of hydrocarbon to hydrogen gas played a crucial role in the CVD growth of graphene. Insufficient H₂ flow could be responsible for the presence of an oxidized metal layer, while excess H₂ flow caused graphene to etch away. Subsequently, dehydrogenation of methane and the formation of carbon radicals occurred on the Cu surface due to the emergence of active sites caused by atomic hydrogen, facilitating the deposition of graphene on the surface of the Cu substrate. As a result, the kinetics of chemisorption of methane on the copper surface diminished, attributed to the obstruction of active sites on the Cu surface by atomic hydrogen. Since H₂ gas had higher diffusivity and dissolvability in Cu, a high hydrogen circulation in the reactor reduced the number of surface sites and consequently decreased the kinetic growth of graphene. After completing the chemical reaction, carbon was deposited on copper and it was necessary to cool the reaction chamber to room temperature to prevent the accumulation of deposited carbon, which could lead to the formation of multilayer graphene. Therefore, single-layer graphene was synthesized using the thermal CVD method. Similarly, in case of nickel, due to the low dispersibility of hydrogen, it recombined on the nickel surface at a higher hydrogen circulation rate. Consequently, additional active sites on the nickel surface for the dissociative chemisorption of hydrocarbons were left unused [32,45-47].

Plasma enhanced chemical vapour deposition method (PECVD): PECVD method is based on a number of plasma sources, such as microwave, radiofrequency and direct current arc discharges. The approach can be used for large-scale graphene synthesis without a catalyst, but it is expensive and requires gas phase precursor substrates. The PECVD growth of graphene can be achieved at low temperatures in less than 5 min, although the resulting graphene is of lower quality compared to that obtained using thermal CVD methodology. Singh *et al.* [48]

fabricated the PECVD growth of graphene at substrate temperatures ranging from 600 to 900 °C using 5-100% methane gas in hydrogen at a pressure of 2 pascals in the reactor. A 900-watt radiofrequency power plasma was employed and a thin film of plasma was deposited on the substrate within 5-10 min [48-50].

Pyrolysis: The solvothermal method is one of the chemical synthetic procedures for graphene material. The pyrolysis of sodium ethoxide, using sonication, constructs graphene sheets to unfurl smoothly, with dimensions of up to 10 μm. An inexpensive method and high-purity, functionalized graphene at low temperatures are two benefits of the process. However, the method involves a large number of flaws, so low-quality graphene is acquired through this procedure [51].

Epitaxial growth on silicon carbide substrates: Thermal decomposition of the surface layer of SiC occurs at nearly 1000 °C (high temperature), assuming the epitaxial growth of a single and multiple-layer, high-quality graphene on a single-crystalline SiC surface with a 100 nm size. Deposition of a single-crystalline film arises on a single-crystalline substrate and produces an epitaxial film. The method, on that account, is named as epitaxial growth. When the deposited film and single-crystalline substrate on which deposition transpires are of the same material, the process is called homoepitaxial growth and the deposited single-crystalline film is called a homoepitaxial layer. When the deposited film and substrate are of different materials, the process is called heteroepitaxial growth and the deposited film on the substrate is called a heteroepitaxial layer. Thermal decomposition of the SiC surface, assuming epitaxial growth of graphene on a single-crystalline SiC substrate, was already developed. In this process, a CO₂ laser was used as a heating source for a single-step, fast-widening process of a uniform graphene film on SiC. Advantages of this method include high-quality graphene and economy and the method controls the layering order of epitaxial graphene. The main stumbling block of this method is obtaining graphene in very small quantities [52,53].

Synthesis of graphene oxide (GO): The process of chemically oxidizing natural graphite using various techniques produces graphene oxide. Graphene oxide was first synthesized by Brodie in 1859. In this method, potassium chlorate was added to slurry of graphite in fuming nitric acid. It was the first methodology of GO synthesis that took 3-4 h to complete at 60 °C. It had been overlooked up until that point. The method thenceforth was upgraded by Staudenmaier. Two-thirds of fuming HNO₃ was reinstated with conc. H₂SO₄ as per Staudenmaier's method of preparing GO. The reaction occurred at room temperature for 96 h, but due to evolution of toxic gases like ClO₂, NO₂ and N₂O₄, it became indispensable to get advanced oxidation method. Extensively used Hummer's method was stumbled on by Hummers & Offeman in 1958 [54-56]. According to Hummer's method, the epitaxial layer of graphite was wrecked to achieve GO by robust stirring of graphite powder with concentrated H₂SO₄. Still generation of NO₂, N₂O₄ like toxic gases and heavy metal pollution made flaws in this oxidation method. And product holded Na⁺ and NO₃⁻ ions were difficult to remove. Hence, hereafter various refinement and

moderation had been taken place over Hummer's method. Marcano *et al.* [57] fabricated the ameliorated method of synthesis GO *in situ* using KMnO_4 as an oxidizing agent in conc. H_2SO_4 and less corrosive H_3PO_4 at 50 °C for 12 h. Despite of higher yield of GO origination with its structural uniformity without evolvment of toxic gases, the method of GO preparation was exorbitant. Peng *et al.* [58] invented a coherent method to draw up GO using K_2FeO_4 /conc. H_2SO_4 as an oxidant system. Shen *et al.* [59] formulated a synthetic method at 110 °C using benzoyl chloride as an oxidant to build up GO. This effectual methodology of constructing GO took 10 min only. Still, it sprang up as an exceedingly perilous method owing to explosive nature of benzoyl chloride at elevated temperature. But high depletion of oxidants and prolonged duration of accomplishing the reaction make it challenging to fabricate high scalable GO in an economical but efficacious way [60-63].

Synthesis of reduced graphene oxide (rGO): The production of rGO can be consummated by reducing graphene oxide (GO) using reductants like sodium borohydride, alcohol, phenol. Su *et al.* [64] observed the formation of highly conductive rGO using ethanol vapour. Alcohol as a reductant does not impair edge cytology of GO making the reduction method safe. Wang *et al.* [65] fabricated synthetic procedure of rGO by peeling of GO followed by reflux in hydroquinone. Regardless of emergence of rGO with structural consistency, proneness of the obtained product to aggregate anew within some hours in water is liable for impracticability of the procedure. Reducing graphene oxide (GO), which has an intact hexagonal lattice structure of carbon atoms, to a high enough temperature in a vacuum with inert gas at 1000 °C produces rGO [66-68].

Modification of graphene

Chemical modification: The accumulation of graphene sheet can be dwindled during reduction process without altering inherent properties of graphene or GO by functionalization of oxygen group present on the surface of GO. Geng *et al.* [69] and Wei *et al.* [70] proposed that the oxidation followed by chemical functionalization leads to good dispersion and a stabilization of graphene to avert agglomeration. Chemical modification emerges appealingly as it amplifies solubility and interaction of graphene with organic polymer according to Worsley *et al.* [71] and Niyogi *et al.* [72]. Hence, the chemical modification facilitates alteration of surface chemistry that leads to change in the physio-chemical properties of graphene. If surface functional groups change chemically during its chemical transformation retaining the scaffold intact to modify chemically, then the process is regarded as primary modification. And when the chemical reaction takes place between surface functional groups and other compounds like small molecules or polymers, the process can be contemplated as secondary modification. Hence, introduction of new functional groups on surface of graphene or GO attributes excellent characteristics in them [73,74].

(a) Reduction of GO in a stabilization medium: According to Park *et al.* [75] addition of KOH strong base into aqueous dispersion of GO leads to confer negative charges with oxygen containing moieties (hydroxyl, epoxy and carboxylic acid

groups) on surface of GO sheet. This attribute coating of negative charges and K^+ around GO sheet. The KOH modified GO, on treatment with hydrazine monohydrate produced stable homogeneous suspension of hKMG.

(b) Covalent modification: The carboxylic acid groups, hydroxy and epoxy groups located at GO edge are utilized in chemical functionalization, such as, addition, isocyanation, diisocyanation, esterification and diazotization [76].

(i) Carbon skeleton functionalization: Zhong *et al.* [77] used solution phase graphene which was dispersed in 0.5% SDS (surfactant) aqueous solution and at 45 °C and stirred with 4-propargyloxydiazobenzenetetrafluoroborate for 8 h long. Further functionalization of graphene took place by click chemistry reaction of obtained product with Azide-dpeg4 acid in presence of sodium ascorbate, CuSO_4 and THPTA. Modified graphene became improved in its water solubility and has potential application as biosensors [78].

(ii) Hydroxy functionalization: The hydroxy functional modification is mainly carried out when profuse of hydroxyl groups are contained by surface of GO sheet's layer. Yang *et al.* [79] fabricated graphene-based polystyrene where surface modifying agents diminished hydrophilic character of GO in polar solvents like THF, DMF and CHCl_3 . Formed azidolated GO (GO-N_3) and alkyne functionalized polystyrene was transplanted on the surface of GO to give graphene-based polystyrene. A novel method for double functionalizing graphene oxide (GO) under mild alkaline conditions has been developed by Guo *et al.* [80]. The process involves a two-step covalent modification of GO: firstly, one functional group is attached through an epoxide ring-opening reaction, followed by the conjugation of a second functional group containing an amine function to benzoquinone linked to GO. The resulting doubly functionalized GO was thoroughly characterized using various techniques, confirming the successful sequential modification of GO with two distinct functional groups.

(iii) Carboxyl functionalization: Xu *et al.* [81] used compound containing porphyrin ring for the preparation of modified GO using covalent attachment. Porphyrin ring on surface of GO sheet modified GO in presence of SOCl_2 to activate carboxyl group on GO surface to make reaction with porphyrin ring. Moderated one is conductive and used as conductive membrane Salavagione *et al.* [82] prepared modified GO, after dispersing GO functionalized with PVA, functionalized product becomes soluble in H_2O and DMF. Modification of GO by adding isocyanate to GO dispersion in anhydrous DMF was observed by Stankovich *et al.* [83]. The obtained derivative was insoluble in water but, dispersible in polar aprotic solvents (DMF, DMSO, N-methyl pyrrolidone, hexamethyl-triamido phosphate. Zhang *et al.* [84] reported the diisocyanate-modified GO by adding organic diisocyanate to dispersed GO (DMF is dispersing medium). Cui *et al.* [85] reported the chemoselective aerobic oxidation of primary alcohols to either aldehydes or carboxylic acids in the presence of nitric acid. After refluxing and purifying, the obtained crude product of ODA-modified GO prevailed its solubility in THF, CCl_4 and 1,2-dichloroethane. According to report of Bonanni *et al.* [86] chemically reduced (CrGO) produced from reductive treatment of GO by

reducing agent hydrazine in aqueous medium undergoes functionalization by isobutyronitrile group to give CN-modified CrGO (CrGO-CN). Refluxing CrGO-CN with CH₃OH and NaOH was embodied on the GO surface by this method based on free-radical addition reaction [76,87,88].

(c) Nucleophilic substitution: Bourlinos *et al.* [89] fabricated amino acid (in alkaline medium)-modified graphene. Here, the epoxide groups on GO undergoes nucleophilic attack by -NH₂ end group. The amino acid-functionalized GO dispersed in CHCl₃/THF/toluene/DCM. The nucleophilic attack of amine to aqueous dispersion of GO is one of the magnificent design to dispersion of GO [89,90].

Non-covalent modification: The *sp*² structure of graphene crystal lattice is compromised due to chemical bond formation between organic molecule and graphene in covalent functionalization. But in case of non-covalent functionalization, the functional groups adsorbed on surface of graphene by π - π bond interaction, H-bond interaction, ion-ion interaction and electrostatic interaction. Owing to the fact that no covalent bond formulation between the functional group and graphene transpires, in consequence, wreckage of carbon skeleton does not arise and no flaws in chemical and electronic properties come about. Introduction of organic molecules on surface expands modified graphene to be used as supercapacitor electrode materials [91,92].

Pi-Pi interaction: Liu *et al.* [93] outlined poly(N-isopropylacrylamide) [PNIPAAm] modified graphene sustained aqueous dispersion of graphene by strong π - π interaction between π -orbitals of graphene and PNIPAAm. Pyrene terminated PNIPAAm was first melted in water subsequently, aqueous dispersion of graphene was added. Lee *et al.* [94] cobbled together tetradecane derivative with a dendritic polyether branch as modifier. Polyether chain refined dispersibility and stabilization of graphene surface layer. The interaction was synergistic interface interaction of π - π interaction. The carbon framework mainly accomplished using C=C in aromatic ring of graphene or GO could be functionalized using tetrapylene derivative [95].

(b) H-Bond interaction: Patil *et al.* [96] delineated DNA modified graphene where H-bonding interaction was prevailed between graphene surface and DNA boosted dispersibility, stability and lipophilicity of graphene. This method is defended and dependable due to no embracement of grimes. DNA-functionalized graphene has a prospective application as biomedicine. Some research groups probed GO-doxorubicin hydrochloride nanohybrid (GO-DXR) ameliorated dispersibility, stability and solubility [96].

(c) Ion-ion Interaction: Li *et al.* [97] fabricated more emulsifiable and soluble sodium dodecyl benzene sulphonated (SDBS) modified rGO. Ion-ion interaction between -COOH on rGO and SDBS arose stable dispersion of rGO. Ge *et al.* [98] looked into amine terminated polymers functionalized rGO stemmed, proliferated dispersibility and boosted lipophilicity.

(d) Electrostatic interaction: Bhunia *et al.* [99] outlined the generation of well dispersed graphene by charge repulsion. Controlled reduction of GO using hydrazine modifying agent perpetuated -COOH group on GO surface, whereas, hydroxyl groups and epoxy bonds were abolished which made carboxylic acid groups on GO surface to diffuse by repulsion of charges.

Accordingly, the origination of stable colloidal solution was supervised [99].

Electrochemical modification: In accordance with the idea of quantitative structure and property relationship, the functional groups are incorporated into molecules by the utilization of a functionalized target group design. However, the method of synthesizing various forms of graphene and graphene oxide is a complicated one. This procedure involves the introduction of a large number of other special groups, which makes it difficult to regulate the reaction and acquire the products that are desired [100].

Polymer/graphene composite: There are various methodologies to form graphene filled different polymer composites. Polymers inapposite for *in situ* polymerization approach, could be employed shaping graphene-based polymer nanocomposite broadly using melt intercalation polymerization methodology have been already reported [101-103].

Transition metal oxide doped graphene or GO nanocomposite: The binary or ternary nanocomposites based on GO/rGO and metal oxides possess a high level of photocatalytic activity. Individual metal oxides like TiO₂, ZnO₂, *etc.* in UV region manifest photocatalytic activities with sizeable band space (energy difference between valence band and conduction band) of 3 eV for TiO₂ and 3.37 eV for ZnO. When photon of UV range hits on surface of transition metal oxides and consequently photoexcitation occurs from valence band to conduction band, then generation of electrons and consecutive holes transpire. The efficiency of photodegradation in transition metal oxides is reduced due to electron and hole recombination. Transition metal oxides disperse on GO surface due to compact binding of it with oxygen functionalities (carbonyl, epoxy, hydroxyl and carboxyl functional groups) on the surface of graphene oxide (GO). Transition metal oxide doped GO/rGO composites result in a reduction in the energy level of the conduction band, leading to a decrease in the band energy gap [104-106]. Actually, during striking of photon of UV region on the transition metal oxide (TMO)/GO or rGO surface, the electron from conduction band of TMO transports to GO or rGO surface and thus electron-hole recombination gets impeded which is escalated the photodegradation capacity of TMO doped GO or rGO composite. The photo-induced electron transfer to GO or rGO surface from CB of TMO is ascribed to good electron accepting ability of GO [107-111].

Degradation of organic dyes: The lipophilic nature of aromatic organic effluents is responsible for the more toxic nature of aromatic hydrocarbons compared to aliphatic hydrocarbons. Graphene and its nanocomposites have great potential for use in wastewater absorption due to its adsorption, oxidation and catalytic capabilities [112-115]. Hence, graphene and its oxide-based composites have been traversed extensively for eliminating the organic pollutants from stream. The following subsection focuses on the adsorption of various organic pollutants that encompass dyes utilizing graphene and its related nanomaterials-based platform.

Removal of cationic dyes

Methylene blue: A lot of inspections have been taken on for assessment of the adsorption efficiency of carbon nanotube

and its composites, graphene, GO, rGO and other graphene nanocomposites against methylene blue dye emerging from the chemical industry accountable for deleterious effects on health and environment. Hydrophobic nature, large surface area and functionalization, such characteristics of graphene make it a superior carbon build adsorbent in comparison to activated carbons (AC) and carbon nanotubes (CNT). Graphene has an appreciable removal efficiency against methylene blue dye from its aqueous solution [116-118]. The pseudo second order model was predominant in removal of methylene blue dye using graphene. Adsorption of methylene blue dye onto graphene is a spontaneous and endothermic process and its adsorption equilibrium is well adapted to Langmuir adsorption isotherm. Hydrothermal treatment of GO sheets with thiourea produced a porous graphene sponge with high surface area and good operability that adsorbs impurities well [116-118]. Oxidative treatment of graphite powder by several methods (Hummer's methods, Brodie's methods, Staudenmaier's method) produce GO having large surface area and intensified the electrostatic interaction with adsorbate dye. The GO has been modified to introduce various oxygen functional groups, which include carboxyl, ketone, hydroxyl and epoxy group at basal and edge planes and make possible adsorption of cationic dyes onto negatively charged surface of graphene through electrostatic interactions [119,120]. Yang *et al.* [120] reported almost complete removal efficiency of GO for methylene blue at pH 6 and 298 K temperature obeying the Freundlich adsorption isotherm. The mechanism of removal of toxic organic dyes by GO is the π - π interaction and cation-anion interaction [121, 122].

The rGO produced by reduction of GO through chemical, thermal or hybrid treatment has sturdy properties and hopeful applications due to high mouldability, surface area, electron motility, conductivity, chemical firmness and good adherence. Tewari *et al.* [123] studied the reduction of GO with ascorbic acid generates rGO hydrogel with 3D porous materials and good mechanical stability and uniform pore size. Only 0.6 g/L of adsorbent dose can adsorb approximately 100% of methylene blue within 2 h at room temperature. Ramesha *et al.* [124] stipulated removal efficiency of rGO hydrogel is 7.85 mg/g at pH 6 for methylene blue which could be achieved by 2 h of contact. But it is difficult to abolish and isolate graphene and GO from rehabilitated water, so to prevent recontamination in water, application of binary composite of GO and rGOs in wastewater purification have been inspected sweepingly. Along with this, tendency of graphene sheets forming aggregates and thereby bringing down obtainable surface area make functionalized graphene, the most recommendable adsorbent for synthetic organic dye removal.

Nanocomposite of graphene enhance adsorption activity by detaining assemblage of graphene and hence enlarge accessible surface area of graphene sheet. Wang *et al.* [125] blended the graphene-based magnetite (G/Fe₃O₄) nanocomposite and employed it successfully for removal of methylene blue dye with adsorption capacity 43.82 mg/g and 45.27 mg/g at 298 K and (298 ± 0.5) K, respectively following Langmuir isotherm and pseudo second order kinetic model. Similarly, graphene-

CNT hybrid composite also shows the good adsorption performance with adsorption capacity 81.97 mg/g for methylene blue obeying Freundlich isotherm and pseudo-second order kinetic model [126-131].

Graphene based nanocomposites also act as photocatalyst and amplify the photodegradation activities. Functionalized graphene, graphene composite with photocatalytic material deteriorate the organic dyes. Zinc oxide nanorods show 66% photodegradation capacity for methylene blue dye but Nipane *et al.* [132] observed the 99% degradation activity against methylene blue using rGO-ZnONR (zinc oxide nanorod). The ZnO-nanoparticles/rGO (ZnO-NPs/rGO) also shows similar 99.5% degradation capacity as reported by Azarang *et al.* [133]. Another modified ZnFe₂O₄/G photocatalyst fabricated by Lu *et al.* [134] degrade methylene blue dye almost 100%. Furthermore, rGO/Fe₃O₄ and G-SnO₂ nanocomposites also photodegrade methylene blue completely as reported by Vinodkumar *et al.* [135] and Seema *et al.* [136], respectively. Another kind of graphene material, GO-BiOBr is also found to be a promising nanocomposite, which photodegrades 98% as reported by Vadivel *et al.* [137]. Liu *et al.* [138] reported 96.2% and 96.0% degradation activity by using GO-Ni₂Fe₂O₄ (act as photo-Fenton catalyst in the presence of UV-Vis light) and Bi₂O₃-rGO nanocomposites, respectively. Chandra *et al.* [139] reported approximately 84% photodegradation of methylene blue dye using graphene nanosheets doped Mn₂O₃ (Mn₂O₃-G) composite.

Rhodamine blue: Graphene sponge exhibited removal capacity of 72.5 mg/g within 4 h at 298 K towards rhodamine blue dye as reported by Zhao *et al.* [140]. Rani *et al.* [141] successfully removed the rhodamine B from water by zirconium oxide/graphene at higher pH within 80 min. Tiwari *et al.* [142] showed 97% removal efficiency of rhodamine blue B dye using rGO based hydrogel. The several different types of graphene based materials *e.g.* graphene/c-MWCNT, polystyrene-Fe₃O₄-GO, Magnetite rGO (MRGO), rGO-ZnO nanocomposites, carboxy-GO-polyethyleneimine, CuI-rGO, β -SnWO₄-rGO, CdSe-G-TiO₂ composite have been reported by various researchers for the photodegradation of rhodamine blue B dye at different capacities [114,143-149].

Other cationic dyes

Methyl green: Methyl Green (MG) is a synthetic dye used in the biological staining. While specific information about its environmental impact is limited, synthetic dyes in general can pose risks to ecosystems due to their production process and potential release into water bodies. Several researchers had carried out the elimination of methyl green dye in different water bodies. For example, Bradder *et al.* [150] reported the adsorption capacity of graphene oxide (GO) towards methyl green dye to be 351 mg/g. The endothermic adsorption process followed the Langmuir isotherm and the adsorption kinetics conformed to pseudo-second-order kinetics. Murcia *et al.* [151] modified membrane surfaces with reduced graphene oxide (rGO) for the effective removal of methyl green dye. Later on, Li *et al.* [152] employed SP-PVA (sulphonated graphene enhanced polyvinyl alcohol) for adsorbing methyl green dye, and GO-doped NiFe₂O₄ was utilized for catalyzing the photodegra-

dation of methyl green dye under visible light and in the presence of oxalic acid. Sharma *et al.* [153] evaluated the adsorption parameters of methyl green onto the reduced graphene oxide (rGO) nanosheets.

Methyl violet: Methyl violet dye is another kind of a synthetic dye with potential ecological concerns. Its manufacturing process and discharge into water systems can contribute to water pollution, affecting aquatic life. Diagboya *et al.* [154] observed that the GO-tripolyphosphate hybrid (GPM) exhibited a removal efficiency of greater than 2540 mg/g against methyl violet dye, making it a more preferable adsorbent for cationic dyes than anionic dyes. Ramesha *et al.* [124] reported an adsorption capacity of 2.47 mg/g for the graphene sponge towards methyl violet dye.

In short, several authors had also carried out the research work on different cationic dyes using modified graphene based nanocomposites for their elimination with adsorption efficiencies and the details are summarized in Table-1.

Removal of anionic dyes

Most of the research that has been conducted on the acute toxic effects of anionic surfactants on aquatic animals has focused on the juvenile and adult life stages. There has been a lack of attention paid to the implications that this phenomena has on the early stages of psychological development. The hydrophilic surfaces are oriented towards the aqueous phase, while the lipophilic terminals of the molecules are located at the core of the micelle. These micelles have the ability to dissolve compounds that are otherwise insoluble in water. This mechanism is highly significant in various physiological processes, such as the dissolution of cholesterol in bile. Studies on the removal of various kinds of anionic dyes using graphene oxide have been carried out by researchers from all over the world. The discussion below will be focused on the elimination of the anionic dyes that have been the subject of the most extensive studies.

Methyl orange: An anionic azo dye commonly used, methyl orange (MO), is toxic to both humans and wildlife unless it undergoes harmless treatment prior to discharge. Since azo dyes have greater stability against temperature, chemicals,

and detergents as compared to natural dyes, it is necessary to eliminate the methyl orange from the environment. Nipen *et al.* [168] traversed the photolytic degradation of methyl orange dye upto 78% by using rGO-ZnO nanorod nanocomposite. Lu *et al.* [169] observed that ZnFe₂O₄/G nanocomposite in presence of H₂O₂ could remove MO dye with excellent degradation capacity of 96% under visible light. Chen *et al.* [170] succeeded to degrade MO by 71% by employing rate of CdSe-G-TiO₂ nanocomposite. Whereas Han *et al.* [171] used Ag₃PO₄ and flocculent like TiO₂/G nanocomposite to degrade MO at 86.7%. A new graphene material, β-SnWO₄-rGO prepared by Buledi *et al.* [172] possess the photocatalytic degradation efficiency of 90% towards the MO dye. Wang *et al.* [173,174] prepared Cu₂O/GO/chitin composite film and Cu₂O/PA/rGO (PA = *n*-propylamine) nanocomposite and found that these materials degrade MO dye with outstanding removal efficiency of 92% and 95%, respectively. Li *et al.* [175] explored the adsorption of MO dye by G/CoFe₂O₄ composite and provided the new insights into the green reduction of GO by bagasse and the formation of rGO/bagasse material presented a great potential in the disposal of dye wastewater. Shen *et al.* [176] reported the graphene-based cement composites for the photocatalytic degradation of rGO-ZnCd against MO dye, which could degrade 10⁻⁵ mol/L amount of synthetic MO dye. Filice *et al.* [177] explored the use of Nafion® (DuPont) membranes containing a graphene-based materials in the photolytic degradation of MO with 71% degradation efficiency.

The ability of graphene oxide and its related materials to degrade other anionic dyes is also evaluated by several researchers as summarized in Table-2, which also provides a summary of the degradation efficacy of various anionic dyes.

Removal of phenolic effluents using graphene oxide-related materials

Removal of phenols: Oxygen-containing groups on the surface of exfoliated GO, is liable for electrostatic adsorption of phenolic sewage as it results evolution of large negative charge density on graphene's surface. Hence the electrostatic interaction between phenolic adsorbate cationic groups and

TABLE-1
ADSORPTION CHARACTERISTICS OF VARIOUS GRAPHENE-BASED MATERIALS FOR CATIONIC DYES REMOVAL

Adsorbent	Dye	Adsorption capacity	Ref.
Chitin/GO nanocomposite	Neutral red	57 × 10 ⁻² mmol/g	[155]
Graphene-Fe ₃ O ₄ nanocomposite	Pararosaniline	198.2 mg/g	[156]
Magnetic Fe ₃ O ₄ @graphene composite	Methylene blue	45.27 mg/g	[157]
Magnetic graphene oxide	Methylene blue	64.23 mg/g	[158]
GO-NH ₂ (amino functionalized graphene oxide)	Rhodamine B	3333 mg/L	[159]
Magnetic-sulfonic graphene nanocomposite	Safranin T	199.3 mg/g	[160]
	Neutral red	216.8 mg/g	
	Victoria blue	200.6 mg/g	
GO-Fe ₃ O ₄ hybrid composite	Methylene blue	167.3 mg/g	[161]
	Neutral red	171.3 mg/g	
Reduced graphene oxide-Fe ₃ O ₄ nanoparticles	Rhodamine B	40.01 mg/g	[162]
Magnetic graphene-Fe ₃ O ₄ @carbon hybrid	Methylene blue	73.26 mg/g	[163]
Ferromagnetic hematite@graphene nanocomposites	Rhodamine B	90.8%	[164]
Magnetite/reduced graphene oxide nanocomposites	Rhodamine B	91%	[165]
	Malachite green	94%	
Modified nanographite/Fe ₃ O ₄ composite	Methyl violet	144.7-151.5 mg/g	[166]
Acetone reduced graphene oxide (ARGO)/Fe ₃ O ₄ composite	Rhodamine 6G	93.37 mg/g	[167]

TABLE-2
 ADSORPTION CHARACTERISTICS OF VARIOUS GRAPHENE-BASED MATERIALS FOR ANIONIC DYES REMOVAL

Adsorbent	Dye	Adsorption capacity	Ref.
Mn ₂ O ₃ -G composite	Eosin	80%	[139]
GO/chitosan composite	Fuchsin acid	175.5 mg/g	[179]
CS, GO-coated biopolymer sponge	Congo red	145.6 mg/g	[180]
3D GO-Polyethyleneimine	AM dye	800 mg/g	[181]
Pt-TiO ₂ /G	Orange 7	99%	[182]
Mn ₃ O ₄ -RGO nanohybrid	Orange II	100%	[183]
Co ₃ O ₄ /GO nanocomposite	Orange II	100%	[184]
NiO/GNS nanocomposite	Congo red	99.56%	[185]
Poly(diallyl dimethyl ammonium chloride) PDDA/GO hydrogel	Ponceau S	188.679 mg/g	[186]
PDDA/GO	Trypan blue	50.025 mg/g	[186]
Zerovalent iron nanoparticle (3DG-Fe) – decorated 3D graphene	Orange IV	94.5%	[187]

oxygen-containing groups comes about which quickens adsorption of positively charged heavy metals. Along with, pi electron rich graphene fabric also conveys firm pi-pi interaction with negative electron cloud of aromatic organic pollutants like phenols, bisphenol-A, *etc.* [188,189]. Several researchers [190-192] established that π - π interaction is most doable and consummate root for adsorbing phenols because of electrostatic repulsion between aromatic ring of adsorbate (aromatic organic effluents) and great negative charge density on the exfoliated GO resulting pre-eminent adsorption potentiality of reduced graphene oxide which has insufficiency of oxygen-containing groups on GO and positively charged polymer polypyrrole functionalized graphene. Table-3 summarizes the removal

efficiency data of phenols and other phenols using graphene oxide-related materials.

Conclusion, challenges and future perspective

In recent years, the exploration of graphene and graphene-based materials for industrial wastewater treatment has explored their exceptional potential as advanced materials in addressing the pressing challenges of water pollution and scarcity. This concise review summarized the significant advancements in utilizing graphene and its derivatives for various treatment processes, highlighting their outstanding physico-chemical properties that enable efficient adsorption, catalysis and membrane based separation. The versatility of graphene based materials

TABLE-3
 REMOVAL OF PHENOLIC EFFLUENTS USING GRAPHENE OXIDE-RELATED MATERIALS

Adsorbent	Dye	Adsorption capacity	Ref.
Phenols			
GO/PPy	Phenols	201.4 mg/g	[193]
NiO@GNCC composite	Phenols	159 mg/g	[194]
GAs-MS nanocomposite	Phenols	90 mg/g	[195]
H-RGO and T-RGO	Phenols	31.1 mg/g and 20 mg/g	[196]
TiO ₂ -TGO	Phenols	99.3% and 96% for 8 h and 3 h, respectively	[197]
GO and GO-PNIPAM	Phenols	10.2 mg/g and 10 mg/g	[198-200]
Bisphenol-A			
3D-GO	Bisphenol-A	421 mg/g	[201]
Ag ₃ PO ₄ /RGH	Bisphenol-A	15 mg/g	[202]
RGH-AgBr@rGO.	Bisphenol-A	80%	[203]
N-RGO (at 6.6 pH), RGO-CD (at pH 7) and GO-CD at pH 9	Bisphenol-A	356 mg/g and 346 mg/g and 373 mg/g, respectively at 298K	[204-206]
Cu-BDC@GO, GO/HDTMA and RGO-MNPs nanocomposites	Bisphenol-A	182 mg/g, 141 mg/g and 123 mg/g	[207-209]
Cu-BDC@GrO	Bisphenol-A	87%	[196]
NG/NH ₂ -MCM-41 and H-RGO composites	Bisphenol-A	71.5 mg/g and 70.4 mg/g	[210,211]
Other phenols			
3D GO (at 298 K and pH 5)	2,4,6-Trichlorophenol	586 mg/g	[204]
	2,4-Dichlorophenol	399 mg/g	
	4-Chlorophenol	238 mg/g	
	2-Chlorophenol	191 mg/g	
GO	Hydroquinone	55 mg/g	[212]
	<i>p</i> -Nitrophenol	1021 mg/g	[213]
N-RGO and GAs-MS nanocomposite	Bisphenol F	286 mg/g	[204]
	Hydroquinone	67 mg/g	
Graphene	4-Chloro-2-nitrophenol	25 mg/g	[214]
Graphene	Catechol	66 mg/g	[215]
	Resorcinol	22 mg/g	
NG/NH ₂ -MCM-41 composite	<i>p</i> -Cresol	68.9 mg/g	[216]

has been demonstrated across a spectrum of contaminants, including heavy metals, organic pollutants and emerging contaminants, indicating their applicability in diverse industrial settings. The reviewed literature underscores the remarkable adsorption capacities of graphene-based materials, attributed to their high surface area, unique porous structure and functionalization capabilities. Additionally, these materials have exhibited exceptional catalytic properties, promoting the degradation of recalcitrant pollutants through various advanced oxidation processes. The integration of graphene based membranes into filtration and separation technologies offers a promising avenue to achieve selective pollutant removal with enhanced efficiency and durability.

Despite the promising strides made in the field of graphene based materials for industrial wastewater treatment, several challenges merit consideration. One of the primary challenges is the scalable synthesis of high-quality graphene based materials with consistent properties. The reproducibility and cost-effectiveness of synthesis methods remain significant hurdles that must be overcome to facilitate large-scale applications. The impact of co-existing contaminants, pH variations and competing ions on adsorption and catalytic processes requires comprehensive investigation to ensure the materials' reliability under practical conditions. Furthermore, the stability and long-term performance of graphene based materials in industrial settings necessitate further exploration. Their susceptibility to fouling, aggregation and mechanical stresses could potentially hinder their sustained effectiveness and operational lifespan.

Looking ahead, the field of graphene and graphene-based materials for industrial wastewater treatment holds immense promise. Researchers and stakeholders should collaboratively address the aforementioned challenges to unlock the full potential of these materials. To achieve this, continued efforts in developing novel synthesis techniques that yield consistent and cost-effective graphene-based materials are essential. Advanced characterization techniques and computational modeling will contribute to a deeper understanding of the materials' interactions with complex pollutants, helping the design of tailored solutions. The integration of graphene-based materials into hybrid systems, combining their strengths with complementary technologies such as nanotechnology, bioremediation and other advanced materials, could lead to synergistic effects and enhanced overall performance. Furthermore, interdisciplinary collaborations between material scientists, chemists, engineers and environmental experts will foster innovative solutions that cater to the specific needs of different industries. Long-term studies assessing the stability, recyclability and environmental impact of graphene-based materials are imperative for establishing their credibility and sustainability as a viable treatment option. Regulatory frameworks must also evolve to ensure the safe and responsible deployment of these advanced materials in industrial wastewater treatment. In conclusion, the journey of graphene and its derivatives from laboratory marvels to industrially applicable materials for wastewater treatment is underway. With concerted efforts to overcome challenges and a forward looking approach, graphene-based materials are poised to revolutionize industrial wastewater treatment, contributing significantly to a more sustainable and water-secure future.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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