Graphene-based Adsorbents for the Removal of Water Contaminants: A Review

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Abstract

Water (Jal) is one of the most important utility for the entire mankind. Arguably, our mother earth is covered with close to 70% of water spreading across different channels: lakes, rivers, ocean, etc. But how much of the 70% water can be consumed for drinking purpose? A great number of people in poor or developing nations have still no access to unadulterated drinking water, they continue consuming water which may prove harmful to health containing bacteria, germs and other micro-organisms. 'Adsorption' process is considered in this paper to eliminate undesired contaminants from water. Graphene and its derived products have been used to remove the pollutants. The motivation of this study is to help in any capacity conceivable and contribute our bit to the people and nature around us. This paper discusses various graphene-based adsorbents and their equilibrium and maximum adsorption efficiencies for different water pollutants, overviewed with various affecting parameters.

Key words: Graphene; materials; adsorption; water purification; wastewater.

1. Introduction

The populace blast, urbanization, and atmosphere changes have escalated the interest for clean water. It is expected that by the year 2025, several countries will face severe water crises that may represent a significant danger to worldwide wellbeing, financial development, maintainability, and social advancement [14]. Along these lines, accessible water and a practical water cleaning innovation are desperately required for the close future. Water cleaning is the way toward expelling disagreeable synthetic compounds, natural contaminants, suspended solids, and gases from water. The primary use of cleansed water is human utilization (drinking water), yet numerous others are also designed for the requirements of medical, pharmacological, chemical, agricultural, and industrial applications. Among different water refinement methods, some significant techniques have been created in late decades, such as adsorption, physical and chemical treatments, membrane-based separation, and biological treatment. Although these methods have played vital roles in sustaining human society in the past, the ever-expanding interest for protected and clean water has raised the need of novel technologies for water purification [18].

The utilization of nanotechnology in natural remediation has pulled in incredible consideration in recent years. Nano-structured materials speak to a few favourable circumstances for water cleaning because of their remarkable physicochemical and surface properties, such as large specific surface area [24]. Graphene is a 2-D layer of atoms of carbon that are firmly associated in a hexagonal honeycomb matrix at atomic scale. Graphene and graphene-based materials have been viewed as one of the sultriest and most quickly rising research areas because of their distinctive physical and chemical properties. As of late, graphene-based nanocomposites have demonstrated guarantee for application in various water decontamination frameworks. The current part sums up ongoing improvements in the utilization of graphene and graphene-based materials in water purification forms through adsorption [29].

1.1. Graphene

Graphene is the name for an iota thick honeycomb sheet of carbon molecules (Figure 1). It is the building block for other graphitic materials (since a typical carbon atom has a diameter of about 0.33 nanometres, there are about 3 million layers of graphene in 1 mm of graphite). It showcases versatile nature, as it is more elastic than rubber and harder than diamond [30]. To put this in perspective: if a sheet of cling film (like kitchen wrap film) had the same strength as a pristine monolayer of graphene, it would require power applied by a mass of 2000 kg, or an enormous vehicle, to cut it with a pencil.

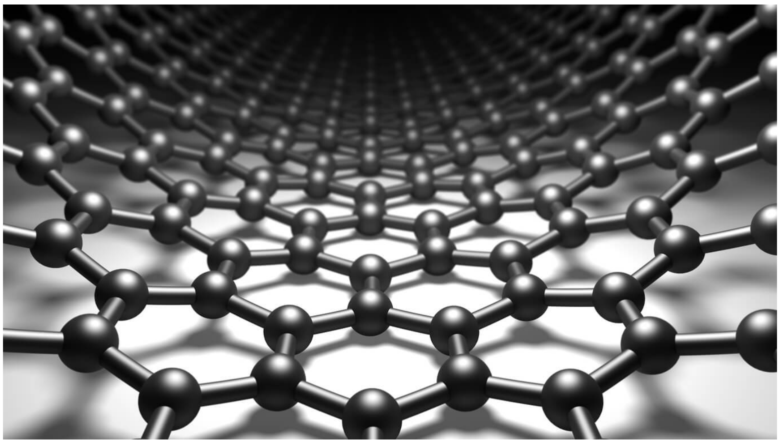

Figure 1: Molecular structure of graphene

Graphene has other stunning qualities (Figure 2): Its high electron versatility is 100x quicker than silicon; it conducts heat 2x superior to precious stone; its electrical conductivity is 13x superior to copper; it ingests just 2.3% of reflecting light; it is impenetrable so that even the littlest particle (helium) can't go through a defect-free monolayer graphene sheet; and its high surface area of 2630 square meters per gram means that with less than 3 grams you could cover an entire soccer field (well, practically speaking you would require 6 grams, since 2630 m2/g is the surface zone for the two sides of a graphene sheet).

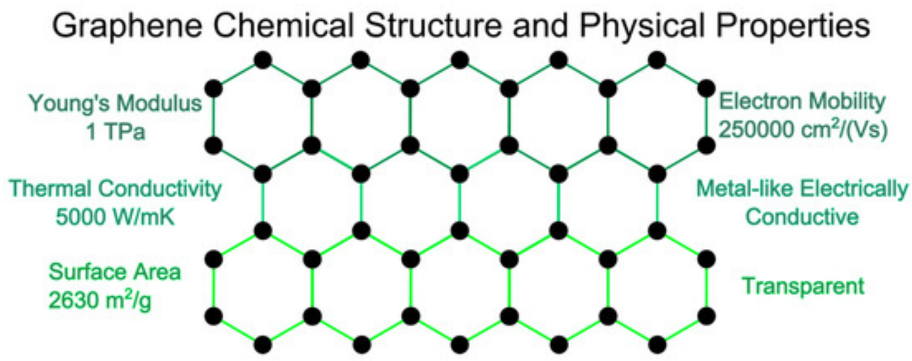

Figure 2: Graphene hexagonal honeycomb chemical structure and its exceptional physical properties. The black dots denote carbon atoms. [21]

Graphene is the essential structure obstruct for other graphitic materials; it also represents a conceptually new class of materials that are only one atom thick, supposed two-dimensional (2D) materials (they are called 2D on the grounds that they reaches out in just two measurements: length and width; as the material is just a single iota thick, the third measurement, height, is viewed as zero). Graphene is also very attractive for the fabrication of mixed-dimensional van der Waals heterostructures that could be helped through hybridizing graphene with 0D quantum dabs or nanoparticles, 1D nanostructures such as nanowires or carbon nanotubes, or 3D bulk materials [13,16].

1.2. Water-Purification Methods Using Graphene-Based Materials

Since Geim and Novoselov achieved the Nobel Prize in physics in 2010 [1], graphene has attracted growing interest in different research areas due to its great surface area, high thermal and electrical conductivity, crack and breaking quality, great mechanical properties, prevalent portability as a charge carrier, proper optical transmittance, specific stability [16]. The length of molecule bonds in the carbon layer of graphene is 0.142nm. The plan of graphene sheets on each other makes graphite a three-dimensional material with interplanar gaps of 0.335nm [10,11].

Graphene functionalization with oxygen-containing functional groups, for instance, carboxyl, hydroxyl, ether, and epoxy make graphene oxide (GO), a broadly utilized carbon-based material. Various composites of graphene have additionally been set up through joining of different materials, for example, inorganic particles and polymers. Graphene and graphene-based materials have as of late picked up the consideration of researchers as perfect substitutes for carbon nanotubes (CNTs) in environmental remediation processes. Contrasted with CNTs, the creation forms for GO and reduced graphene oxide (RGO) are very simple with no need for complex apparatus or specific catalysts. Graphene material can be coordinated by graphite's chemical exfoliation, achieving an end product free of catalyst residues, and no further purification is required. The accompanying areas centre around late advances in the use of graphene-based materials in diverse water purification strategies, with a significant accentuation on the adsorption.

2. Literature Review: Adsorption

Adsorption is a customary, compelling process for wastewater treatment as a result of its convenience, ease of operation, and simplicity of design. This procedure can be used for the exclusion of dissolved contaminants staying after chemical and biological oxidation treatments [26]. Carbon-based materials, for example, activated carbon are the most utilized adsorbent in this process Recently, graphene-based composites have been successfully used for the adsorption of different pollutants including heavy metals, organic dyes, halomethanes, and other pollutants because of their high specific surface area and electron-rich environment. Graphene-based adsorbents have a few points of interest over different materials, for example, CNTs. The single layer structure of graphene materials provides two basal planes and huge surface region accessible for contaminant adsorption, while the inner walls of CNTs are not open to contaminations. In addition, as the most generally utilized compound of graphene, GO has numerous oxygen-containing functional groups that impart a hydrophilic nature and high negative charge density to GO. These functional groups effectively improve the adsorption of pollutants by GO sheets [4].

Table 1: Some of the recently published studies on graphene-based adsorbents used for water purification

Graphene-Based Adsorbent	Target Pollutant	Adsorption Capacity at Equilibrium (mg/g)	Maximum Adsorption Capacity (mg/g)	Performance	Reference
Magnetic Graphene oxide (mGO)	Arsenate	>30	>50	The adsorption of As(V) on MGO decreased with ascending pH due to the electrostatic interaction. In view of the focus and nature of existing together anions and cations, adsorption of As(V) on MGO was significantly affected	[9]
Ce-Fe/RGO	Congo Red	92	179.5	The main thrust for the adsorption was electrostatic activity between adsorbent materials with positive electricity and Congo Red with negative electricity	[23]
Graphene Oxide-$MnFe_2O_4$	Pb(II), As(III), As(V)	Pb(II): 350 As(III): 65 As(V): 100	Pb(II): 673 As(III): 146 As(V): 207	The exceptional adsorption property was due to the combination of the extraordinary layered nature (permitting extreme surface zone) of the hybrid system and the good adsorption capabilities of both GO and $MnFe_2O_4$ nanoparticles	[27]
Thiol-functionalized magnetite/GO	Hg^{2+}	30.94	289.9	The synthesized adsorbent exhibited a higher adsorption	[12]

				capacity compared to magnetite/GO and the bare GO because of the integrated adsorption of thiol groups and magnetite nanocrystals	
GO-$ZrO(OH)_2$	As(III), As(V)	As(III): 89.53 As(V): 79.05	As(III): 95.15 As(V): 84.89	The GO-$ZrO(OH)_2$ nanocomposite showed high adsorption capacity in a broad pH scale, and the monolayer adsorption amounts were 95.15 mg/g for As(III) and 84.89 mg/g for As(V), which are 3.54 and 4.64 times that of $ZrO(OH)_2$	[34]
Magnetite/Graphene Oxide (M/GO)	Cobalt(II)	>10	12.98	The ionic strength-independent and pH-dependent Co(II) sorption on M/GO indicated that the sorption mechanism of Co(II) was inner sphere surface complexation at low pH levels, while the expulsion of Co(II) was finished by synchronous precipitation and inner-sphere surface complexation at high pH values	[17]
Graphene supported Fe-Mg oxide composite	Arsenic	25.5	103.9	The prepared composite exhibited significant fast adsorption of arsenic over a wide range of solution pHs, with exceptional durability and recyclability	[6]
RGO-Fe(0)-Fe_3O_4	Cr(VI), Pb(II), Cd(II), Hg(II)	As(III): >40 Cr(VI): >25 Pb(II): >20 Cd(II): >5 Hg(II): >20	As(III): >40 Cr(VI): >30 Pb(II): >20 Cd(II): <5 Hg(II): >20	The porous RGO-Fe(0)-Fe_3O_4 material was highly efficient in adsorbing heavy metal ions and employed for catalytic oxidation	[22]

				reactions	
Smart Magnetic graphene (SMG)	Cr(VI), As(V), Pb(II)	Cr(VI): <6 As(V): <5 Pb(II): <4	Cr(VI): 4.86 As(V): 3.26 Pb(II): 6	The SMG possesses increased the adsorption sites with tuneable superparamagnetic properties, encouraging the adsorption and attractive division of aqueous Cr(VI), As(V), and Pb(II) with close to 99% removal efficiencies	[7]
Magnetic GO/poly (vinyl alcohol) (mGO/PVA)	Methyle-ne Blue and Methyl Violet	>150 by mGO/PVA-50%	Methylene Blue: 270.94 Methyl Violet: 221.23	The mGO/PVA not only exhibited strong paramagnetic property, but also showed obvious GO content-dependent enhancement in adsorption capacity for organic dyes	[35]
Fe_3O_4/RGO	As(V), Ni(II), Pb(II)	As(V): 15.97 Ni(II): 18.88 Pb(II): 15.63	As(V): 58.48 Ni(II): 76.34 Pb(II): 65.79	Fe_3O_4/RGO showed superparamagnetic properties at room temperature and saturation magnetization. On Fe3O4/RGO, the adsorption models of As(V), Ni(II) and Pb(II) indicated surface heterogeneity and monolayer adsorption of the adsorbents	[19]

3. Other Methods

A brisk study of the literature shows that there is an increasing pattern toward developing graphene-based bactericidal materials for different ecological and biomedical applications. The antibacterial action of graphene and its derivatives, including GO and RGO, was accounted for by a few researchers [36, 33, 5, 15, 25, 31, 3]. It is believed that the cell layer of the microscopic organisms is harmed by direct contact of the microorganisms with the extremely sharp edges of the graphene nanosheets that cause the bacterial inactivation. Furthermore, it was discovered that the RGO nanosheets are more poisonous to the microorganisms than unreduced GO nanosheets. The better antibacterial action of the reduced nanosheets was attributed to the better charge movement between the microscopic organisms and the more sharpened edges of the reduced nanosheets, during the contact interaction [20, 32]. Deng et al. [2] prepared a graphene sponge enriched with Cu nanoparticles as a novel bactericidal filter for water disinfection. The incorporated composite could inactivate over 99.9% of feasible E. coli cells with a 5-min hydraulic retention time, affirming its effective antibacterial activity. Pourbeyram and others [28] revealed that water dispersive CuO/RGO nanocomposite has a lot higher antibacterial activity contrasted with Cu^{+2}/GO and CuO nanoparticles. Vatanpour et al. additionally improved the antifouling and antibacterial ability of the polyethersulfone membranes by consolidating Ag-GO nanocomposite [31]. A graphene-based composite was used in photocatalysis and ozonation combined framework for mineralization of Bisphenol A [8]. Liao et al. synthesized the TiO_2-RGO hybrid by the liquid-phase deposition (LPD) technique and saw that the recombination of photogenerated charges is essentially decreased by RGO commitment. The total organic carbon (TOC) exclusion effectiveness was 98.4% utilizing TiO_2-RGO/O_3/UV framework in 45min, which was 1.17 times higher than that of TiO_2/O_3/UV framework. Additionally, the TiO_2-RGO hybrid presented high stability after recycling in five continuous runs.

4. Findings and Analysis

Despite enormous growth in the application of graphene-based materials for water and wastewater purification, there are still some challenges that restrict its application at a large scale and industrial cases. Some of the key problems is the concentration of graphene in aqueous solutions, which limits the mobility of the impurity and its interaction and therefore reduces the material's varying surface region. This test was conquered by numerous specialists by altering graphene materials with surface treatment and functional groups. It is well-proven that the presence of functional hydrophilic groups (groups containing oxygen) on the graphene surface can improve their dispersion and decrease their aggregation. The detachment of graphene-based materials utilized in a water purification procedure is another challenge which often needs membrane filters or high-speed centrifugation. As another alternative, and increasingly functional arrangement, magnetic graphene-based composites were suggested using an external magnetic field with promising performance and easy separation.

Like various nanomaterials, graphene-based materials' useful uses are legally subordinated to their quality, processability, and environmental aspects. Graphene, GO, and RGO 's easy, renewable, and financially clever planning is yet a challenging operation. In addition, transforming events into increasingly stable, fast, and professional graphene-based mixing techniques is of great importance for generating huge quantities at economically viable rates. With respect to the environmental implications, the release of graphene-based materials during a water treatment process, and the impact of their harm on living things, human exposure, and the environment, to minimize the associated health and environmental impact, should be tackled seriously.

5. Conclusion and Future Scope

Owing to its unusual physicochemical properties, Graphene, an individual 2-D sheet of carbon particles reinforced in a hexagonal honeycomb cross section, has drawn growing interest in recent years. One of the intriguing and exciting application fields of the materials built on graphene is wastewater treatment. In this paper, a review of recent advancements in the application of graphene and graphene-based materials in the water refinement strategies taking into consideration adsorption was presented with an emphasis on the most recent scientific reports. The graphene-based adsorbents present high contaminants expulsion effectiveness due to the large surface area available for adsorption. Moreover, the presence of a large number of oxygenated functional groups on the GO sheets bestows a hydrophilic nature and high negative charge density to the GO surface and improves the adsorption of contaminations. More scientific and technical efforts and exhaustive studies are still needed to overcome the difficulties of graphene-based materials thinking about their preparation, physical and structural properties, reasonability, and reproducibility, as well as improvements in their dispersion, compatibility, and stability in different water purification systems. Likewise, further examination is required to assess the cost viability of enormous scope utilization of graphene-based materials and checking their abiding permanence under practical conditions in wastewater purification.

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