DOI: 10.24996/ijs.2023.64.10.6

ISSN: 0067-2904

Graphene Oxide : A Key Solution for Future: Recent Achievements as A New Adsorbent for Water Treatment Applications: Review

Watheq K. Salih*, Sawsan A. M. Mohammed, Basma A. Abdulmajeed
Chemical Engineering Department, College of Engineering, University of Baghdad, Baghdad, Iraq

Received: 18/8/2022 Accepted: 16/3/2023 Published: 30/10/2023

Abstract
Polluted water has been considered a critical issue nowadays, threatening the environment and lives of living creatures. Because of technological and industrial advancements, as well as increased social activities of humans in various countries, pollution sources have multiplied. To reduce the impact of this problem, many techniques have been developed in order to reach zero discharge pollution. In the last decade, graphene oxide (GO) - a member of the graphene nanomaterials family, has been the focus of many research efforts in the water treatment sector because of its extraordinary properties. This review highlights the research efforts conducted to investigate GO as a novel adsorbent for water treatment applications and recent fulfilments in the last 3 years. The synthesis techniques, properties, and efficiency of this material in water treatment will be explained. All results confirm the future role of GO as an efficient absorbent to solve wastewater purification challenges, but the big challenge is to reduce time and simplify the complicated extent of synthesis stages, besides reducing the high cost of production methods. According to the review, Iraqi researchers' efforts to use this nanomaterial in water purification are still in their early stages.

Keywords: Graphene oxide, Water treatment, Adsorption, Nanomaterials, Hummer method

أوكسيد الجرافين الحل المستقبلي: اخر الانجازات كممتز جديد في تطبيقات معالجة المياه

واثق كريم صالح *, سوسن عبد مسلم محمد ، بسمة عباس عبد المجيد
قسم الهندسة الكيمياوية ، كلية الهندسة ، جامعه بغداد ، بغداد ، العراق

الخلاصة
تعتبر المياه الملوثة قضية خطيرة في الوقت الراهن تهديدها البيئة و حياة المخلوقات الحية. لقد تنوعت مصادر نلوث المياة نتيجة للتطور التقني والصناعي وكنتيجة لزياده الفعاليات الاجتماعية للبشر في مختلف البلدان. و لتقليل اثر هذه المشكلة تم تطوير العديد من التقنيات للوصول الى ماء خالي من الملوثات. في العقد الاخير اصبح اوكسيد الجرافين الذي هو احد افراد عائلة الجرافين النانومترية محورا للعديد من الجهود البحثية في قطاع معالجة المياه لخواصه الاستثنائية. ان مقال المراجعة هذا يسلط الضوء على الجهود المبذولة لاختبار اوكسيد الجرافين كماده ممتزة جديده في تطبيقات معالجة المياه والانجازات المتحققة خلال السنوات الثلاث الاخيرة. إذ يتم شرح طرق التصنيع والخواص وكذلك كفاءة هذه المادة لهذا في معالجة المياه. ان نتائج

*Email: w.saleh1007@coeng.uobaghdad.edu.iq

البحوث المنشورة تؤكد على الدور المستقبلي لأوكسيد الجرافين كمادة ممتزة كفوءة لحل التحديات في مجال تنقية مياه الصرف لكن التحدي الاكبر هو تقليل وتبسيط مراحل التصنيع المعقدة اضافة لتقبيل كلف الانتاج . وطبقا لهذه المقالة فان جهود الباحثين العراقيين في استخدام هذه المادة النانومترية في معالجة المياه الملوثة ما زالت في مراحلها المبكرة .

1. Introduction

1.1. Historical background

The first nanomaterial was a carbon nanotube material that was discovered by chance when Japanese scientist Sumio Iijima saw a thin layer precipitated on the electric discharge electrodes of an electric arc discharge in 1991 [1]. This discovery triggered the nanotechnology revolution, which has continued up to this time. As a result, the properties of carbon nanotubes have widely opened the door for scientists to synthesize other types of carbon nanomaterials in particular and to produce other nanomaterials in general. Since that discovery, nano-carbonous materials have observed a very rapid expansion. The new types are still appearing. The main motivation for this huge development in the synthesis and manufacturing of carbon nanomaterials is their extraordinary characteristics, which allow for their use in different applications with high performance. Figure 1 shows the main types of carbon nanomaterials. In return, each main type is divided into several secondary types. Graphene is one of these types, which has rapidly expanded to include a variety of types with common basic properties [2].

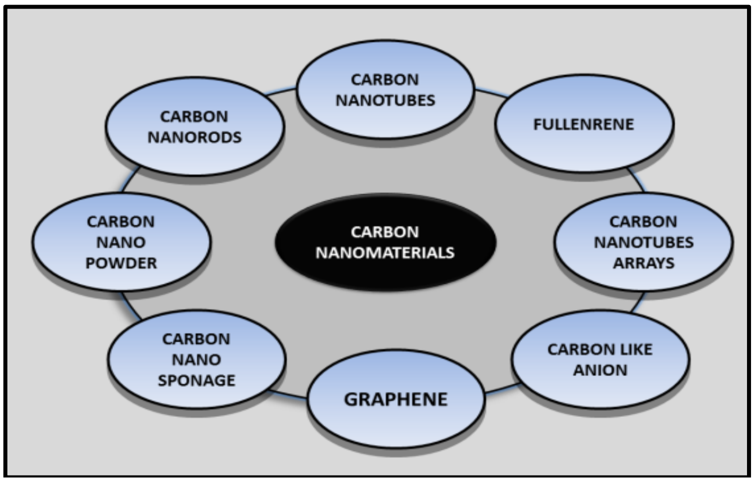

Figure 1: Carbon nanomaterial types

1.2. Graphene, structure classifications and properties

Graphene can be defined as a dual-dimensional hexagonal honeycomb structure in which carbon atoms are arranged and bonded with each other by covalent bonds at the sp^2 orbital. Each carbon atom is bonded with three neighboring atoms, but in general it tends to connect with a fourth to form the benzoic hexagonal structure. The main difference between graphene and graphite is that the latter consists of 3D stacks arranged together [3]. In 2004, Geim and co-workers could isolate a single layer of carbon atom film from graphite using adhesive tape for the first time, and then this material attracted serious attention as a new material [4]. This note was the real start of graphene synthesis and characterization [5]. Figures 2 and 3 show the structures of graphene and graphite.

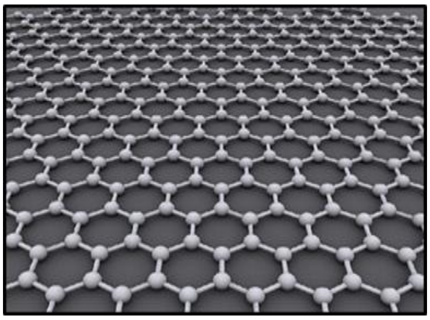

Figure 2 : Graphene's victual structure [6]

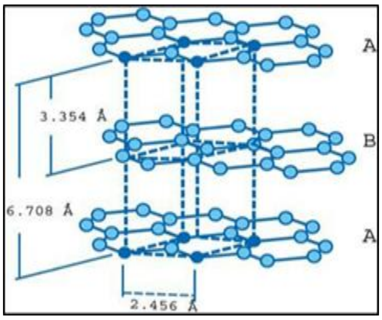

Figure 3 : Graphite structure [7]

The unique properties of graphene have contributed to making it an effective alternative for a variety of substances in a wide range of applications and industries. The first of these benefits is that it is a super electrical and thermal conductor. Besides, it possesses high mechanical properties, such as high elasticity; its E-modulus is about 1.1 terapascal. The strength resistance of graphene is approximately 200 times that of steel. So, it resembles a diamond but is lighter in weight. It also has antibacterial properties and, due to its high density, acts as a good barrier to water passing through. As well, it is characterized by a high surface area of about 2630 m^2/g [8-10]. Moreover, graphene has extraordinary optical and electronic properties, so it is suitable for the solar and electronic industries [11].The graphene family consists of several types because of its rapid expansion in recent years. Figure 4 clarifies the graphene derivatives. Graphene oxide (GO) is considered the most prominent example in the graphene group, which has been a main topic for a wide range of applications.

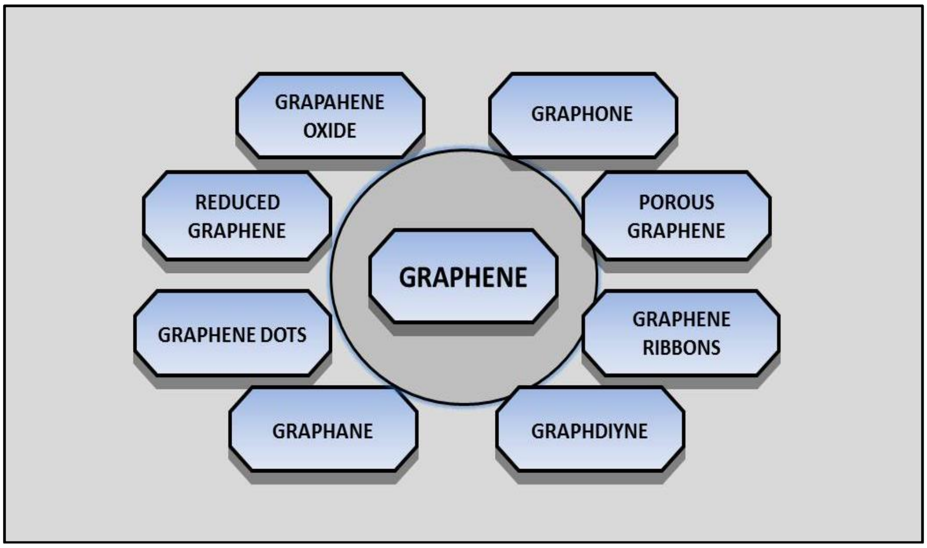

Figure 4: Graphene group members

2. Graphene oxide (GO)

2.1. History background

The history of graphene oxide dates back to 163 years ago. The first GO synthesis was done by Brody in 1859 [12]. At that date, no one had investigated its unique properties, But it acquired a new importance after the nanotechnology revolution because the properties of GO were reassessed according to new concepts and implications. As a result, the tests have approved that GO is classified as a 2D nanomaterial, in which its thickness is at the nanoscale while other dimensions are in the microscale range with outstanding characteristics [10,13].

2.2. Structure and properties of GO

GO is classified as a non-stoichiometric chemical material in which the ratios of carbon, oxygen, and hydrogen are variable depending on the synthesis methods and manufacturing conditions. Using various theories and explanations, the scientists created a punctual description model for GO structure. The early explanations, which had been fulfilled by Hofmann, Rudolf, Ruess, Scholz, and Boehm [14] were not successful because the first two researchers supposed that GO consists of epoxy groups distributed randomly on the graphite basal layer. In response, others attempted to modify this model by adding different groups to the basal carbon, such as hydroxyl and carboxyl, in addition to epoxy groups, and they proposed a less ordered structure for GO containing C-C with C-C bonds [15]. The novel and most convincing models were formulated by Lerf-Klinowski [16] and Denkany [17]. According to two models, GO, as a member of the graphene family, has a similar structure to graphene. Both of these compounds have a hexagonal carbon lattice, but the GO sheet is usually distorted where it is bonded to the oxygen groups. GO has a two-dimensional carbon sheet with various oxygenated functional groups on its basal planes and peripheries. As usual, the thickness between its layers is about 1 nm, while the lateral dimensions fall in a range between a few nanometers and several microns. The main groups on the basal plane are hydroxyl and epoxide groups. In the outermost layer, groups such as aldehydes, carboxylic acids, and ketones exist. Furthermore, reduced graphene (rGO) can be synthesized from GO *via* different methods with the same structure except that the number of oxygen atoms will be reduced, leading to differences in the characteristics of rGO [17-20]. Figure 5 shows the structures of graphite or graphene, GO, and rGO.

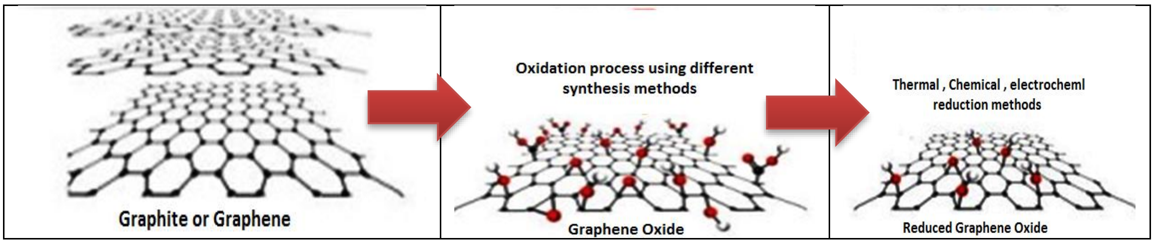

Figure 5: Graphene, graphene oxide, and reduced graphene oxide structures [21]

2.3. Graphene oxide characteristics

Because of the oxygen present in its lattice interlayers, GO has distinct properties that differ from those of graphene. These differences have contributed to its use in new applications where graphene cannot be used. The important properties of GO can be outlined in the following [5,12,22, and 23].

1. Hydrophilic material.
2. Low electrical conductor.
3. The functionality: It is easy to modify GO with different function groups.
4. It can easily come back to graphene at an effective cost compared with CVD and mechanical exfoliation.
5. High mechanical properties, but not as good as graphene.
6. Low thermal conductivity.
7. Unusual electronic properties.

2.4. Synthesis methods

The main idea of GO synthesis methods is the strong oxidation process of graphite. Following the synthesis of Broody GO, manufacturing efforts have continued to seek the most effective method with the lowest cost and the least negative impact on the environment, as well as the shortest possible synthesis time [15]. So, many methods have appeared using different techniques. However, the chemical oxidation method is still the most effective method for now.

Despite this, all synthesis methods have resulted in the production of GO, albeit with significant differences in the properties of each product. Table 1 gives details about the chemical oxidation methods [16, 24-30].

Table 1 : Chemical oxidation methods

Method name	Oxidants	Toxicity	Advantages	Disadvantages
Broodie 1859	$KClO_3$, HNO_3	Yes	First synthesis method	Weak acidity Imperfect structure Small crystallite
Staudenmaier 1898	$KClO_3$ ($NaClO_3$), HNO_3, H_2SO_4	Yes	All reaction steps were one in one vessel, so it was a modification of the Broodie method	Time of experiment is very long dangerous one May be explosive because of The liberating chlorine dioxide
Hummers and Hofmann 1937	$KMnO_4$, H_2SO_4, $NaNO_3$,	No	The degree of oxidation is higher than the two previous methods	Long period of time spent Complex purification process The oxidation is still not complete
Modified Hummers 1958	$KMnO_4$, H_2SO_4, $NaNO_3$,	No	Sufficient degree of oxidation	Long period of time spent Complex purification process
Tour (improved) Hummers 2010	$KMnO_4$,H_2SO_4, H_3PO_4	No	Reduced defects in the basal plane, high oxidation degree, green method, organized structure	Long period of time spent Complex purification process

The modified and improved methods are the traditional and popular techniques that are used to produce GO in the present time. Furthermore, there are novel attempts to synthesize GO using novel technologies such as, electrochemical [31], microwave [32], ultrasonic [33], and laser ablation [34].

2.5. GO characterization methods

Different characterization methods have been used to specify all the properties of GO. Table 2 shows the details of these tests and their results.

Table 2 : Characterization methods

Test	Function	Typical	Ref.
XRD	Crystallinity, phase, and structure in addition to confirm the GO formation	2theat = 9-11°	16 35 36 37 38 39 40 41
XPS	Specify the surface oxidation ratio	Two peaks at Cis and OIs 284 and 530 eV	
Raman	To analyse the number of layers, edge chirality, and doping levels	D, G, 2D, bands at 1350, 1580 and 2700 nm, respectively	
AFM	Specify the thickness of the layers	thickness of layer is about 1 nm	
SEM	Morphology and structural examination	Folded and curling	
EDS	Elemental calculation	C/O is about 1.02	
TEM	Thickness and defects of the surface	The thickness must be in the nanometre range	
NMR	The structure of GO	Resonance: 59 nm (C-O-C), 68 nm (hydroxyl	

		130 nm sp^2 (C=C), 167 nm carbonyl carbon (C=O)
TGA	Weight stability with temperature	Depending on purity, defects, and volatile content, substances like humidity
FT-IR	Bonding structure and types	1620 cm^{-1} for water 3300-3400 cm^{-1} for C-OH 3600 wide shoulder for OH
Zeta potential	Suspension stability of GO	The value of stable suspension is greater than 30 mV
Surface area	Porosity of material and number of active sites	From 10 m^2g^{-1} to 2000 m^2g^{-1}
UV-Visible	Wavelength of GO	Wide peak 190-900 nm but the maximum at 230 nm
Thermal and electrical conductivity	Showing the thermal and electrical conductivity	Thermal conductivity is no more than 2000 W/mK 4.07 10^{-7} S/cm

3. GO applications in water treatment

Water treatment is an essential concern nowadays because of the population increase and the lack of surface water on the whole earth, besides the diverse chemical pollutants discharged as a result of industrial, medical, agricultural, and domestic activities [4]. The purification of wastewater to obtain high-quality water is done with different techniques, such as mechanical, chemical, biological, and physical methods. Nowadays, one of the effective topics occupying considerable ranking in international scientific research is the adsorption technique, which can be defined as the surface process that leads to the movement of polluted molecules from liquid media to the surfaces of solid particles [24,44]. This method has gained more attention because it is inexpensive and simple. Additionally, a wide range of materials, either nanomaterials or micromaterials, can be used as adsorbents. Lately, many papers have focused on investigating **GO** as a new absorbent to purify wastewater in general and to treat medical wastewater in particular, due to its structural characteristics and its special surface properties. Table 3 lists some scientific advancements achieved during the previous three years involving the use of GO as a novel water treatment adsorbent.

Table 3 : Science contributions for using GO as an adsorbent

Years and Author	GO Synthesis method	Wastewater type and technique	Pollutant removal type	Important results	Ref.
Zhang *et al.* (2020)	Modified Hummers	wastewater by adsorption using GO/Sodium alginate	Pb, Cd, Mn, Mg	Pb removal 92.8% Cd removal 91.1% Mn removal 89% Mg removal 85.7%	45
Huang *et al.* (2020)	Modified Hummers	Industrial wastewater eliminating by adsorption using modified amino-GO	Cr, Cu, Pb, Cd	Adsorption capacity of Cd, Cu Pb, Cr is, 71.89 mg/g , 26.25 mg/g, 10.04 mg/g, 280.11 mg/g	46
Bhattacharya *et al.* (2020)	Modified Hummers	Medical wastewater removal by adsorption	Carbamazepine	Removal efficiency is 99% with adsorption capacity is 6.1g.L^{-1}. the results showed that GO can be used for 8 times without change in its capacity	47

Qurat-Ul-Ain *et al.* (2020)	Modified Hummers	Heavy metals removal by magnetic GO	Lead, zinc, copper and nickel ions	Results showed that MGO had high efficiency to remove heavy metals ions Langmuir model with pseudo second order were fitted with adsorption process	48
Salihi *et al.* (2021)	Modified Hummers	Medical wastewater utilizing adsorption	Antibiotic TMP and INH	The adsorption capacity was 204.08 mg/g for TMP and 13.89 mg/g for INH The acidity of solution have strong effect on adsorption	43
Liu *et al.* (2021)	Modified Hummers	GO- TiO_2 composite using adsorption	Sewage by adsorption and photocatalysis	The new composite proved its efficiency to treat sewage wastewater with adsorption capacity 167.92 mg/g	49
Senguptal *et al.* (2021)	Modified Hummer	Arsenic remove groundwater using GO/Fe_3O_4 composite	As	As removal by GO =71.64% As removal by GO/Fe_3O_4 = 81.6%	50
Politaeva *et al.* (2022)	Electrochemical	Industrial by GO-chitosan granules	Cu cation	The adsorption efficiency was 96% with adsorption capacity is 20 g/L	51
Januário *et al.* (2022)	Commercial GO	Medical residue removal by activated carbon-GO combination	chloroquine dipyrone,	The new combination absorbent showed potential to remove these drugs. The capacity was 37.65 and 63.43 for CQN and DIP. Lagnumir and pseudo - 2nd order are applied to both drugs	52
Le *et al.* (2022)	Modified Hummers	Industrial wastewater by GO-Fe_3O_4-Ag adsorbent	Nitrogen compounds and phosphate (PO_4^{-3})	This complex showed its high efficiency in the total removal of nitrogen and phosphate	53
Ajeel *et al.* (2022)	Modified Hummers	Dye removal by sodium alginate/GO composite	Methylene blue	The results were fitted to the Freundlich model. The pH value was 7.78, the time was 12 hours, the temperature was 15 °C, and the removal ratio was 91.26%	54
Kadhim and Saleh (2022)	Modified Hummers	Cooper from industrial wastewater using chitosan/GO	Copper	The Freundlich model was applied, and pseudo-second order was fitted with a 99.4% removal ratio	55
Yang *et al.* (2022)	Modified Hummers	Drug residue and dye from wastewater	Rhodamine B Enrofloxacin	Maximum capacity was 107.23 mg/g for rhodamine B 45.035mg/g for enrofloxacin	56
Mizhir *et al.* (2022)	Modified Hummers	Dye remove by Go and grafted GO	Bismarck brown dye	The maximum adsorption capacities of GO and Grafted GO are 833.33 and 1000 mg/g, respectively.	57

3.1. Mechanism of GO adsorbent

The adsorption mechanism of GO is unclear until now, so it needs more research before a clear interpretation can indeed be made [58]. At the moment, the proposed mechanism of GO adsorption is based on hydrogen hydrophilic bonding and interactions between the adsorbate and the adsorbent. This interaction is responsible for the efficiency of the adsorption process [24]. Furthermore, the pH role in the adsorption process is critical in determining the efficiency of using GO as an adsorbent to remove various pollutants from water [59, 60].

4. Challenges and future solutions

Many difficulties and limitations remain in the way of using GO as an effective adsorbent. The first is that the production of GO is still at the laboratory scale due to the complex and long stages that must be reduced to reach bulk production. Another important limitation is related to defects in the GO structure because of various manufacturing methods, which are still in use and have impressive effects on the properties of GO. These need to be improved in order to easily use GO with polymers or other nanomaterials to produce composites, especially in the membrane industry. Subsequently, bulk production of GO is still very limited because of these reasons, which contribute to making the synthesis process expensive. To address these issues, efforts should be focused on developing synthesis methods that yield high purity products as well as a cost-effective technology that yields an uncomplicated bulk production method that yields a cost-effective product.

4.1. Future developments and projects

The main limitations of using GO as an adsorbent are its high price and the complicated successive steps of the manufacturing process, as shown previously. However, several ambitious scientific projects have been launched to use GO as an adsorbent in water treatment plants while also identifying simple and cost-effective methods to reduce the complexity and high cost of production. One of these projects is a Monash University project (the CRC project) in cooperation with two Australian private companies in 2017. The project intends to use GO as a filtration medium, either as GO membrane composites or as traditional adsorbents coated with GO, on a commercial scale to purify wastewater while reducing electricity consumption. Up until now, initial results showed the potential merits of using GO in the water treatment sector [61]. Khalifa University in the UAE recently announced the launch of the RIC-2D project in 2022. This project is aimed at funding and supporting global scientific research on graphene and its derivatives, as well as 2D carbon nanomaterials, for water and energy applications [62]. In the UK, the University of Manchester could secure sufficient funding in 2022 for its project, which is aiming to produce graphene-based membranes in order to recover lithium and other materials from brines and wastewater produced from battery manufacturing for reuse in the same industry again [63]. In addition, in 2022, GO commercial scale membranes were synthesized by NematiQ, an Australian developer company, where GO flat sheets of 1000 m in length and 1 m in width were achieved as a result of five years of hard scientific research and development, a milestone success in the GO membrane industry [64]. These projects assure the importance of GO in future applications, especially in the water treatment sector.

5. GO ranking in Iraqi scientific research

The importance of GO was explained in detail in previous sections showing the international efforts of scientists throughout the entire world. In Iraq, the scientific efforts to utilize GO as an adsorbent are still in the beginning for many reasons, but the most effective reason is the time consumption of preparation. Nonetheless, the activity of Iraqi researchers has begun to increase in the recent five years. For example, the published papers that were published in academic journals (IAJS) inside Iraq are about 31 papers from 2015 to July 12, 2022 according to the Iraqi academic journals database [65]. However, only 5 papers dealt with GO as an adsorbent. In other words, 16.161% of GO papers published in national Iraqi journals dealt with the topic of water treatment. Iraqi researchers, on the other hand, have published 52 papers in Science Direct International journals for various applications since 2015, as shown in Figure 6. Nonetheless, the number of papers using GO as an adsorbent is still limited, accounting for only 5 (9.62%) of the total number of published papers.

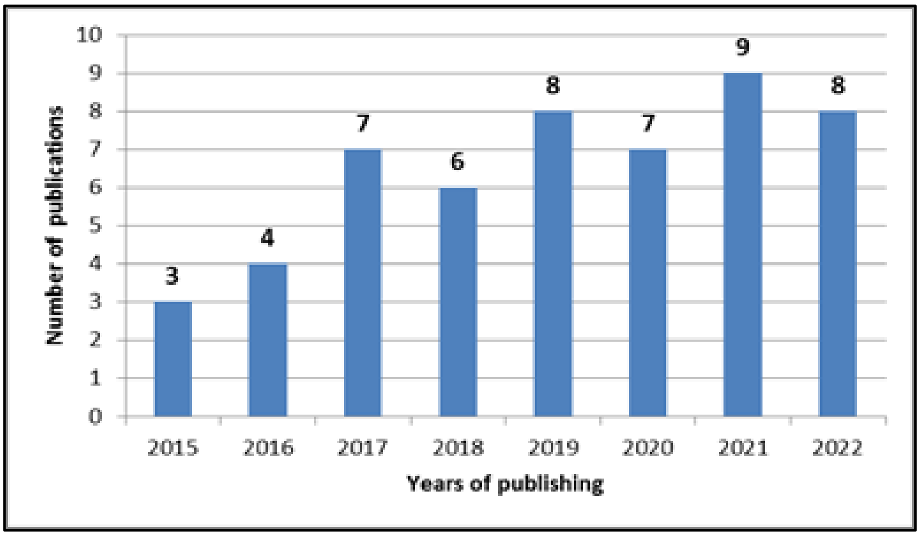

Figure 6: Iraqi researchers published papers in peer-reviewed journals

6. Conclusions

Novel materials for water treatment, particularly adsorbents, are currently regarded as a critical topic in scientific research. So, the properties and characteristics of **GO** may make it a promising future water treatment material. Different synthesis methods have been used for 165 years. The results, which are recorded in international papers, indicate its effective ability to treat different water contaminants with high adsorbent capacity. GO has begun to be used in the industrial-scale production of water purification materials and tools containing GO as a novel nanomaterial absorbent that can play an important role in water treatment in the near future, particularly in large projects. The main obstacle to using GO extensively is the complex successive oxidation stages of synthesis methods that keep production of GO in the laboratory or small-scale industrial range, which contributes to making the price of GO expensive compared with other types of adsorbent. In Iraq, scientific efforts to apply GO adsorbent in water treatment are still limited, as evidenced by publications in national and international journals.

Conflict of Interest:

The authors declare that they have no conflicts of interest.

References

[1] M. Kolahdouz, B. Xu, A. F. Nasiri, M. Fathollahzadeh, M. Manian, H. Aghababa, Y. Wu, and H. H. Radamson, "Carbon-Related Materials: Graphene and Carbon Nanotubes in Semiconductor Applications and Design", *Micromachines*, vol. 2022, no.13, Article ID 1257, 2022.

[2] A. Tiwari, "Graphene bioelectronics", *Elsevier publisher*, Cambridge, UK, 2018.

[3] S. M. Rangappa, J. Parameswaranpillai, S. Siengchin, and L. Kroll, "Lightweight polymer composite structures: Design and manufacturing techniques", *CRC publisher*, FL, USA, 2020.

[4] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva and A. A. Firsov, "Electric field effect in automatically thin carbon films", SCIENCE , vol. 306, no. 3696, pp. 666-669 , 2004

[5] T. J. E. Proctor, D. A. M. Armada, and A. Vijayaraghavan, "An introduction to graphene and carbon nanotubes", CRC press, FL. USA, 2017.

[6] A. A. Berlin, V. F. Kablov, A. A. Pimerzin, and S. S. Zlotsky, "Key elements in polymers for engineers and chemists: form data to applications", *Apple Academic*, Oakville, Canada, 2014

[7] V. Skakalova and A. B. Kaiser, "Graphene: properties, preparation, characterization and applications", 2nd Ed., *Woodhead Puplisher*, MA, USA, 2020.

[8] A. Alessandro, A. L. Materazzi, and F. Ubertini, "Nanotechnology in cement-based construction", *Jenny Stanford Publisher*, Temasek Avenue, Singapore, 2020.

[9] S. Mishra and D. Hansora, "Graphene nanomaterials: fabrication, properties and applications" Pan *Stanford Publishing*, Boulevard, Singapore, 2018.

[10] P. Avouris, T. F. Heinz, and T. Low, "2D Materials", *Cambridge University Press*, Cambridge, UK, 2017.

[11] P. Samorì and V. Palermo, "Flexible carbon-based electronics", *Wiley VCH*, Weinheim, Germany, 2018.

[12] P. P. Brisebois and M. Siaj, "Harvesting graphene oxide - years 1859 to 2019: A review of its ructure, synthesis, properties and exfoliation", *Journal of Materials Chemistry C*, vol. 2020, no. 8, pp. 1517-1547, 2020.

[13] C. E. Banks and D. A. C. Brownson, "2D materials: characterization, production and applications", *CRC press*, FL, USA, 2018.

[14] H. Luo, "A theortical study of graphene oxide chemical structure", Master thesis , The school of mechanical and mining engineering , University of Queensland , Australia, 2017

[15] L. Sun, "Structure and synthesis of graphene oxide", *Chinese Journal of Chemical Engineering* , vol. 27, no. 10, pp. 2251-2260, 2019.

[16] A. Siklitskaya, E. Gacka, D. Larowska, M. Mazurkiewicz‑Pawlicka, A. Malolepszy, L. Stobiński, B. Marciniak, A. Lewandowska‑Andrałojć and A. Kubas, "Lerf- Klinowski type models of graphene oxide and reduced graphene oxide are robust in analyzing non-covalent functionalization with porphyrins", Scientific reports, vol. 2021, no. 11, Article ID 7977, 14 pages, 2021.

[17] Q. Qiao, C. Liu, W. Gao, and L. Huanga, "Graphene oxide model with desirable structural and chemical properties", *Carbon*, vol. 143, no. 2019, pp. 566-577, 2019.

[18] A. M. Dimiev, and S. Eigler, "Graphene oxide: fundamentals and applications", *Wiley and Sons*, West Sussex, UK, 2017.

[19] F. Pendolino, and N. Armata, "Graphene oxide in environmental remediation process", *Springer*, Cham, Switzerland, 2017.

[20] W. Gao, "Graphene oxide: Reduction recipes, spectroscopy, and applications", *Springer*, Cham, Switzerland, 2015.

[21] A. N. Banerjee, "Graphene and its derivatives as biomedical materials: future prospects and challenges". *Interface Focus*, vol. 8, no. 3, Article ID 20170056, 22 pages, 2018.

[22] A. T. Dideikin and A. Y. Vul, "Graphene oxide and derivatives: the place in graphene family", *Frontiers in Physics.* vol. 6, no. 2018, Article ID 149, 13 pages, 2019.

[23] A. T. Smith, A. M. Chance, S. Zeng, B. Liu, and L. Sun, "Synthesis, properties, and applications of graphene oxide/reduced graphene oxide and their nanocomposites", *Nano Materials Science*, vol. 1, no. 1, pp. 31-47, 2019.

[24] A. Ahamad, S. E. Siddiqui, and P. Singh, "Contamination of water: health risk assessment and treatment strategies", *Elsevier*, London, UK, 2021.

[25] R. Ikrama, B. M. Jana, and W. Ahmad, "An overview of industrial scalable production of graphene oxide and analytical approaches for synthesis and characterization", *The Journal of Materials Research and Technology*, vol. 9, no. 5, pp. 11587-11610, 2020.

[26] S. Sali, H. R. Mackey, and A. A. Abdala, "Effect of graphene oxide synthesis method on properties and performance of polysulfone-graphene oxide mixed matrix membranes", *Nanomaterials*, vol. 9, no. 5, Article ID 769, 16 pages, 2019.

[27] W. Huang, "Nano-papers: from nano-chemistry and nano-manufacturing to advanced applications", *Elsevier*, Cham, Switzerland, 2018.

[28] D. R. Dreyer, S. Park, C. W. Bielawski, and R. S. Ruoff, "The chemistry of graphene oxide", *Chemical Society Reviews*, vol. 39, no. 1, pp. 228-240, 2010.

[29] A. Mohammeda, J. G. Sanjayan, A. Nazari, and N. T. K. Al-Saad, "The role of graphene oxide in limited long-term carbonation of cement-based matrix", *Construction and Building Materials*, vol. 168, no. 2018, 858-866, 2018.

[30] S. A. Yaseen, G. A. Yiseen, and Z. Li," Synthesis of calcium carbonate in alkali solution based on graphene oxide and reduced graphene oxide", *Journal of Solid State Chemistry*, vol. 262, no. 2018, pp. 127-134,2018.

[31] A. M. Díez-Pascual, C. Sainz-Urruela, C. Vallés, S. Vera-López, and M. P. S. Andrés, "Tailorable synthesis of highly oxidized graphene oxides via an environmentally-friendly electrochemical process", *Nanomaterials* , vol. 10, no. 2, Article ID 239, 16 pages, 2020.

[32] A. Ibarra-Hernández, A. Vega-Rios, and V. Osuna, "Synthesis of graphite oxide with different surface oxygen contents assisted microwave radiation", *Nanomaterials*, vol. 8, no. 2, Article ID 106, 17 pages, 2018.

[33] M. Sabbaghan, H. Charkhan, M. Ghalkhani, and J. Beheshtian, "Ultrasonic route synthesis, characterization and electrochemical study of graphene oxide and reduced graphene oxide", *Research on Chemical Intermediates,* vol. 45, pp. 487-505, 2019.

[34] A. K. Jeryo and Q. A. Abbas, "Synthesis of graphene oxide nanoparticles by laser ablation system", *Iraqi Journal of Science*, vol. 61, no. 9, pp. 2241-2250, 2020.

[35] A. Al-Ahmed and Inamuddin, "Graphene from natural sources: synthesis characterization and applications", *CRC*, FL, USA, 2023.

[36] A. Tampieri, M. Szabó, F. Medina, and H. Gulyás, "A brief introduction to the basics of NMR spectroscopy and selected examples of its applications to materials characterization", *Physical Sciences Reviews*, vol. 6 , no. 1, Article ID 20190086, 41 pages, 2021.

[37] E. Aliyev, V. Filiz, M. M. Khan, Y. J. Lee, C. Abetz, and V. Abetz, "Structural characterization of graphene oxide: surface functional groups and fractionated oxidative debris", *Nanomaterials*, vol. 9, no. 8, Article ID1180, 15 pages, 2019.

[38] A. W. Rokmana, A Asriani, H Suhendar, K Triyana, A Kusumaatmaja, and I Santoso, "The optical properties of thin film reduced graphene oxide/poly (3,4-ethylenedioxtriophene): poly (styrene sulfonate) (PEDOT:PSS) fabricated by Spin coating", *Journal of Physics: Conference Series*, 1011012007, 2018.

[39] Y. Yang, J. Cao, N. Wei, D. Meng, L. Wang, G. Ren, R. Yan, and N. Zhang, "Thermal conductivity of defective graphene oxide: A molecular dynamic study", *Molecules*, vol. 24, no. 6, Article ID 1103, 10 pages, 2019.

[40] T. R. B. Ramakrishna, D. P. Killeen, T. D. Nalder, S. N. Marshall, W. Yang, and C. J. Barrow, "Quantifying graphene oxide reduction using spectroscopic techniques: A chemometric analysis", *Applied Spectroscopy*, vol. 72, no. 12, pp. 1764-1773, 2018.

[41] E. Jaafar, M. Kashif, S. K. Sahari, and Z. Ngaini, "Study on morphological, optical and electrical properties of graphene oxide (GO) and reduced graphene oxide (rGO)", *Materials Science Forum*, vol. 917, pp. 112-116, 2018.

[42] F. T. Johra, J. Lee, and W. Jung, "Facile and safe graphene preparation on solution based platform", *Journal of Industrial and Engineering Chemistry*, vol. 20, pp. 2883-2887, 2014

[43] E. Ç. Salihi, J. Wang, G. Kabacaoğlu, S. Kırkulak, and L. Šiller, "Graphene oxide as a new generation adsorbent for the removal of antibiotics from waters", *Separation Science and Technology*, vol. 56, no.3, pp. 453-461, 2021.

[44] S. E. Jorgensen, and B. Fath, "Encyclopedia of ecology", *Elsevier B.V.* Amsterdam, Netherlands, 2008.

[45] Y. Zhanga, F. Lianga, P. Dianb, and Z. Chenb, "Novel graphene oxide based adsorbents for removing water pollutants Pb^{+2}, Cd^{+2}, Mn^{+2}, and Mg^{+2} ions", *Desalination and Water Treatment*, vol. 206,no. 2020, pp. 263-272, 2020.

[46] H. Huang, Y. Wang, Y. Zhang, Z. Niu, and X. Li, "Amino-functionalized graphene oxide for Cr(VI), Cu(II), Pb(II) and Cd(II) removal from industrial wastewater", *Open Chemitry*, vol. 18, no. 1, pp. 97-107, 2020.

[47] S. Bhattacharya, P. Banerjee, P. Das, A. Bhowal, S. K. Majumder, and P. Ghosh, "Removal of aqueous carbamazepine using graphene oxide nanoplatelets: process modelling and optimization", *Sustainable Environment Research.*, vol. 30, Article ID 17, 12 pages, 2020.
[48] Qurat-Ul- Ain, M. U. Farooq, and M. I. Jalees, "Application of magnetic graphene oxide for water purification: heavy metals removal and disinfection", *Journal of Water Process Engineering*, vol. 33, no. 2020, Article ID 101044, 12 pages, 2020.
[49] S. Liu, Z. Su, Yi Liu, L. Yi, Z. Chen, and Z. Liu, "Mechanism and purification effect of photocatalytic wastewater treatment using graphene oxide-doped titanium dioxide composite nanomaterials", *Water*, vol. 13, no. 14, Article ID 1915, 12 pages, 2021.
[50] S. Sengupta, A. Pari, L. Biswas, P. Shit, K. Bhattacharyya, and A. P. Chattopadhyay, "Adsorption of arsenic on graphene oxide, reduced graphene oxide, and their Fe_3O_4 doped nanocomposites", *Biointerface Research in Applied Chemistry*, vol. 12, no. 5, pp. 6196- 6210, 2021.
[51] N. Politaeva, A. Yakovlev, E. Yakovleva, V. Chelysheva, K. Tarantseva, S. Efremova, L. Mukhametova, and S. Ilyashenko, "Graphene oxide-chitosan composites for water treatment from copper cations", *Water,* vol. 14, no. 9, Article ID 1430, 2022.
[52] E. F. D. Januário, Y. J. Fachina, G. Wernke, G. M. M. Demiti, L.B. Beltran, R. Bergamasco, and A. M. S. Vieira, "Application of activated carbon functionalized with graphene oxide for efficient removal of COVID-19 treatment-related pharmaceuticals from water", *Chemosphere,* vol. 289, no. 2022, Article ID 133213, 2022.
[53] T. T. H. Le, T. T. Ngo, T. H. H. Nguyen, T. D. Pham, T. X. H. Vu and Q. V. Tran. "Green nanoarchitectonics using cleistocalyx operculatus leaf extract in the preparation of multifunctional graphene oxide/Fe_3O_4/Ag nanomaterials for water decontamination and disinfection". *Journal of Inorganic and Organometallic Polymers and Materials*, vol. 32, pp. 547-559, 2022.
[54] S. J. Ajeel, A. A. Beddai, and A. M. N. Almohaisen, "Preparation of alginate/graphene oxide composite for methylene blue removal", *Materials Today: Proceedings*, vol. 51, no. 1, pp.289-297, 2022.
[55] H. H. Kadhim and K. A. Saleh, "Removing of copper ions from industrial wastewater using graphene oxide/chitosan nanocomposite", *Iraqi Journal of Science*, vol. 63, no. 5, pp. 1894-1908, 2022.
[56] J. Yang, S. Shojaei, and S. Shojaei, "Removal of drug and dye from aqueous solutions by graphene oxide: adsorption studies and chemometrics methods", *npj Clean water*, vol. 5, no. 5, Article ID 5, 10 pages, 2022.
[57] A. A. Mizhir, H. S. Al-Lami, and A. A. Abdulwahid, "Kinetic, isotherm, and thermodynamic study of bismarck brown dye adsorption onto graphene oxide and graphene oxide-grafted-poly (*n*-butyl methacrylate-co-methacrylic acid)", *Baghdad Science Journal*, vol. 19, no. 1, pp. 132-140.
[58] Y. Ai, Y. Liu, Y. Huo, C. Zhao, L. Sun, B. Han, X. Caoc, and X. Wang, "Insights into the adsorption mechanism and dynamic behavior of tetracycline antibiotics on reduced graphene oxide (RGO) and graphene oxide (GO) materials", *Environmental Science: Nano*, vol. 6, Article ID 3336, 2019.
[59] D. Robati, M. Rajabi, O. Moradi, F. Najafi, I. Tyagi, S. Agarwal, and V. K. Gupta, "Kinetics and thermodynamics of malachite green dye adsorption from aqueous solutions on graphene oxide and reduced graphene oxide", *Journal of Molecular Liquids*, vol. 214, no. 2016, pp.259-263, 2016.
[60] E. E. Ghadim, F. Manouchehri, G. Soleimani, H. Hossein, S. Kimiagar, and S. Nafisi, "Adsorption properties of tetracycline onto graphene oxide: equilibrium, kinetic and thermodynamic studies", *PLoS One*, vol. 8, Article ID e79254, 2013.
[61] CRC project, power efficient wastewater treatment using graphene oxide technology, https://business.gov.au/grants-and-programs/cooperative-research-centres-projects-crcp-grants/customer-stories/ionic-industries, Access at 15- June- 2022.
[62] RIC-20 project, 2022 https://www.graphene-info.com/uae-launches-global-graphene-funding-initiative, last access at 22 - June - 2022.
[63] DLE project, 2022, https://www.myscience.org/news/wire /geic_partner_watercycle_ technologies _ secures_funding_for_lithium_extraction_process-2022-manchester, last access at 25 – June – 2022.

[64] NematiQ's graphene membranes, https://www.proactiveinvestors.com/companies/news/976181 /clean-teq-subsidiary-produces-commercial-scale-graphene-membrane-in-milestone-achievement-shares-up-976181.html, last access at 25 – June – 2022.
[65] Iraqi academic Scientific journals: https://www.iasj.net/iasj/search? addFilter=year: %222022%22&fromYear=&query=Graphene%20oxide&currentYear=2022&toYear