Chemical Engineering Science 296 (2024) 120274

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Interplay of adsorptive properties and dielectric reactivity of graphenes upon edge functionalization

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ARTICLE INFO

Keywords:
Adsorption
Edge functionalization
Framing
Graphene
Graphene oxide

ABSTRACT

Graphenes have unique physicochemical properties that can be engineered for pollutant adsorption and their dielectric properties can facilitate subsequent microwave regeneration. First, graphenes have the potential for high-level control of oxygen content at the edges of the material without compromising the conjugated electronic structure of the basal plane. Because the basal plane contains lightly functionalized sp^2-hybridized carbon atoms while sp^3-hybridized ones are on the periphery of the sheets. Edges of graphenes can be oxidized while the basal plane can still be electronically stable with conjugated π-electrons perpendicular to the graphene lattice. For this, graphenes can be oxidized to attain controlled dispersion in water without disrupting the conjugated electron network on the basal plane, which is critical for pollutant adsorption. Graphenes have sheet-like surfaces that form dynamic, porous aggregates in water and can facilitate synthetic organic compound adsorption by the complex interplay of 'pore' accessibility and favorable intermolecular interactions. Thus, studying the role of oxidation of graphenes can help unravel the interplay between inter-sheet distance and the adsorption of synthetic organic compounds. Second, the sp^2 hybridized basal planes of graphenes have mobile π-electrons that are expedient for rapid dielectric heating, which can be harvested for rapid and efficient microwave regeneration. Fundamental research on graphene chemistry can lead to a paradigm shift in the water treatment industry towards the safe and sustainable deployment of regenerable nano-scale adsorbents. This article presents a perspective on how to approach edge functionalization of graphene with an aspiration to advance their safe and sustainable use in water treatment.

1. Introduction

Anthropogenic activities are disrupting the natural aquatic ecosystem at a rate that has never been seen in the history of mankind (Saulnier-Talbot and Lavoie, 2018). This has cascading consequences on human civilization where more than one billion people with no access to safe drinking water and, millions of people dying from diseases related to unsafe drinking water (Rockström et al., 2014). Thousands of human-made, toxic pollutants are entering natural water resources at an increasing rate (Arenas-Sánchez et al., 2016). These contemporary challenges are beyond the capabilities of materials we use for water treatment that date back to the Victorian era. Advancing the fundamental knowledge of material science can help accomplish water treatment applications of modern, groundbreaking materials such as graphenes (Mauter et al., 2018).

Graphene, a Nobel Prize decorated material, had our community's attention because of its unique physicochemical and electronic properties. Today, graphene, graphene-based nanomaterials, and composites have been studied as materials for biomedical applications, energy storage, different classes of sensors, preparation of stable oil–water emulsions, electronics, and environmental applications (Contreras Ortiz et al., 2019; Kuziel et al., 2020; Yu et al., 2017). Especially, for environmental applications, the exceptional properties of graphene can be transformative, particularly for water treatment, where it can be used as an adsorbent (Ersan et al., 2017). To date, several graphene-based adsorbents have been developed through nanomodification of graphene such as covalent and non-covalent functionalization, defect formation, element doping and alterations of morphology by wrinkles, folds and nanoholes formation (*e.g.*, holey graphene) (Bai et al., 2010; Farhan et al., 2023; Rout et al., 2023; Wang et al., 2024; 2016; Yu et al., 2020). After about one decade of research, we have a much better understanding of the material; however, the translation of graphene-based

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https://doi.org/10.1016/j.ces.2024.120274
Received 2 February 2024; Received in revised form 17 May 2024; Accepted 20 May 2024

technologies into water treatment is still in its early stages, and more work is needed. By effective and regenerable use of graphene-based adsorbents, we could minimize energy and material demand, prevent extra loads on landfills or incinerators, lessen the associated health and ecological risks and therefore contribute to the sustainable practice of graphene-enabled water treatment.

In anticipation of employing graphenes, there must be a long and windy journey of technology maturation if we can envision its pragmatic use today. Graphene nanosheets can be integrated into municipal water treatment systems as suspended adsorbents, similar to powdered activated carbon. The mixing can facilitate mass transfer and minimize intraparticle diffusion times. It is important to note, though, that subsequent separation would be required to ensure their recovery from treated water. Graphenes can also be integrated to municipal or domestic systems in fixed beds in the form of graphene-based macrostructures such as aerogels, sponges, pellets, composites, beads, or granules (Hirani et al., 2022; Khalil et al., 2022; Low et al., 2024; Masud et al., 2021; Schmidt et al., 2023; Zhao et al., 2012). Fixed-bed reactors are generally restrained by reduced surface areas, lower mass transfer rates, and large hydraulic head demand. On the other hand, the fixed-bed configuration eliminates the need for future separation and introduces the ability of adsorbent regeneration. Regardless of the envisioned application, understanding fundamental mechanisms that govern the adsorption and regeneration will empower the pursuit of graphene-enabled water treatment. This perspective article aims to discuss possible advances in modern knowledge of graphene edge chemistry that are expedient for water treatment. Specifically, we will review (i) the role of graphene edge functionalization in improving key properties that are relevant to water treatment applications: dispersion in water and selective adsorption in the presence of natural organic matter (NOM) and (ii) discuss some research gaps that could enhance our understanding of graphene edge and surface chemistry to develop microwave regenerable carbon nanoadsorbents.

2. Edge-tailored graphenes for adsorption of synthetic organic compounds

Graphenes are two-dimensional quantum materials with negligible masses when compared to their electron densities (Keimer and Moore, 2017). Their electron density is localized above and below the hexagonal plane as a result of sp^2 hybridization and the carbon atoms display different bonding energies depending on their location within their structures. Graphenes have edge atoms, corners, and defects unlike other common carbon allotropes; their functionalization presents an opportunity to increase the oxygen content without completely deteriorating their π-electron network on the basal plane, which is critical for pollutant adsorption. There are possibly countless opportunities to harvest these properties for water treatment, but this section will discuss three of them. First, there is the possibility of high-level control of oxygen content at the edges of the material without compromising the conjugated electronic structure of the basal plane. The oxygen content of graphene is important for controlled dispersion in water, whereas the integrity of the basal plane is important for pollutant adsorption. According to the well-celebrated Lerf-Klinowski model, graphenes are archetypal bipartite systems with lightly functionalized sp^2-hybridized carbons such as hydroxyl and epoxy groups whereas the edges of the graphenes can host sp^3-hybridized carbon atoms. The reactivity of the edge carbon atoms of graphene differs from the relatively stable basal plane. Edges of graphenes in the absence of a reactant have metastable σ and π bonds with unsaturated sp^2 orbitals, whereas the basal plane is electronically stable with conjugated π electrons perpendicular to the graphene lattice (Bellunato et al., 2016; He et al., 1998). The split of σ bonds between adjacent carbon atoms in the *p*-conjugation network results in the formation of edges in graphene. As represented in Fig. 1, mainly, two types of edge configurations, named zigzag and armchair edges, are possible based on the orientation of the edge along the basal plane structure. However, a perfect zigzag or armchair is difficult to obtain, and the edges, in reality, have more complex geometries, a combination of both zigzag and armchair segments known as a “chiral edge”. The conjugation system and bond energies are different on a zig-zag edge compared to an armchair one, resulting significant differences in reactivity of these sites as shown in Fig. 1 (Radovic and Bockrath, 2005; Suenaga and Koshino, 2010; Zhang et al., 2013). In addition to that, local defects, such as imperfections and vacancies, which can break the conjugated network, can also be considered as edges. The edge reactivity and electronic properties of graphene are highly dependent on the edge chemistry and geometrical shape. Armchair edges can form triple bonds (carbyne) to reduce electron energy, whereas zigzag edges have p_z electrons on each outer carbon atom, resulting in a chemically reactive radical singlet configuration (carbene) that contributes to the energetic “edge state”. Furthermore, the edge functionalization with various atoms or groups and the formation of defects can alter the chemical and electronic properties of graphene (Acik and Chabal, 2011; Girit et al., 2009; Hyun et al., 2015; Jia et al., 2011; Wagner et al., 2013). For example, the binding of hydrogen atoms to graphene edges can stabilize the dangling bonds and influence the electronic properties by inducing localized states at the Fermi level, which can modify the electronic properties by altering band structures and magnetic and thermal properties, making graphene suitable for electronic and spintronic applications (He et al., 2014; Li

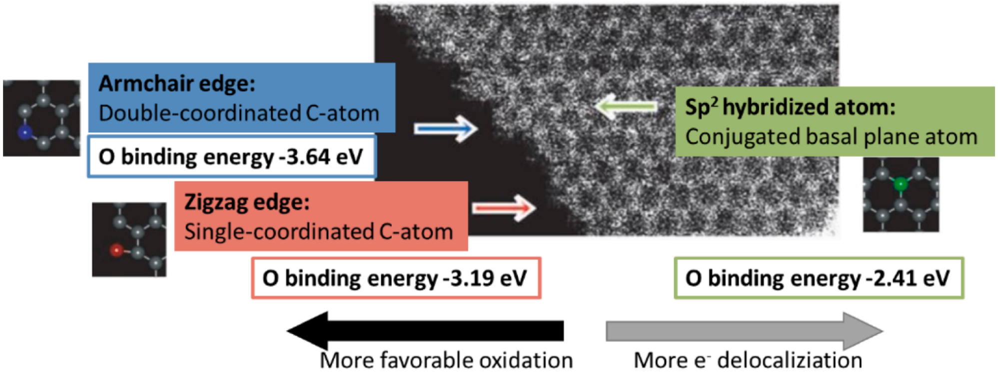

Fig. 1. Comparison of oxygen binding energies at the edge and on the basal plane of graphene surface signifying the affinity of single- and double-coordinated carbon atoms with oxygen (Radovic and Bockrath, 2005; Suenaga and Koshino, 2010). The background of the image shows a scanning transmission electron microscope (STEM) annular dark field (ADF) image of the edge region of a single layer graphene sheet (reproduced with permission from Springer Nature Publishing Group, Copyright 2010) (Suenaga and Koshino, 2010). The right-hand side of the image is the hexagonal network of carbon atoms, and the black blank area on the left-hand side is the vacuum region during imaging.

et al., 2015; Wang et al., 2014). Similarly, oxygen and nitrogen functionalization at the edges has been shown to introduce p-type and n-type doping characteristics tuning the electronic and optical properties of graphenes, respectively, which are critical for fabricating graphene-based electronic devices (Li et al., 2015). Also, holey graphenes have been created by forming nanoholes on the basal plane of graphene. These nanoholes provide active sites like the edges and also act as a sieve, allowing ions and smaller molecules to pass through. These unique properties make them promising materials for water treatment for targeted and selective pollutant removal (Wang et al., 2024). Oxygen-containing functionalities on graphene edges can improve the hydrophilicity of graphene as well as provide active sites for the adsorption of both organic (polar and monopolar) and certain inorganic contaminants. Therefore, based on the oxygen binding energy on different configurations of graphene edges and basal plane (Fig. 1), when the oxygen content of graphene sheets is systematically increased, carboxylic acid groups can first react with the carbon atoms on the periphery and leave a higher degree of conjugation.

Additionally, the hydroxyl and epoxy groups can appear on the basal plane, by decreasing the level of conjugation (Fig. 1). Molecular dynamic simulations confirm the structurally optimized homo-oxidant framing by using the edge-localized, non-bonding molecular orbitals or via carbyne and carbene intermediate formation at armchair or zigzag sites, respectively (Basiuk et al., 2017; Goswami et al., 2019; Radovic and Bockrath, 2005; Sheka and Popova, 2013; Zhang et al., 2013). However, little experimental evidence is available to show the synthesis of edge-oxidized graphenes. Functionalizing the graphenes with edge-localized C-O bonding may sound theoretically achievable, but experimental work is needed to prove the cornerstone of this approach.

Second, graphene nanosheets have large surfaces *i.e.*, theoretically around 2,600 $m^2\ g^{-1}$ for two-sided, single layer sheets. They form dynamic, porous aggregates in water that can be controlled by their flake size, oxygen content, and water chemistry. These properties can facilitate adsorption by the complex interplay of ‘pore’ accessibility and favorable intermolecular interactions such as π-π electron donor–acceptor interactions, Coulombic interactions, H-bonding, and non-specific van der Waals forces (*e.g.*, London dispersion forces) (Ersan et al., 2017). The presence of oxygen-containing functional groups on edges can further provide active adsorption sites for some polar and polarizable organics as well as ionic compounds. For example, the dissociation of carboxylic groups to $-COO^-$ can facilitate the adsorption of cationic contaminants such as heavy metals (*e.g.*, Pb^{+2}, Hg^{+2}) and cationic organic compounds (*e.g.*, cationic dyes). Similarly, the dissociation of $-C-OH$ groups to $-C^+$ can provide adsorption sites for anionic compounds such as arsenate (*e.g.*, AsO_4^{3-}), anionic dyes, and some pesticides (Rout et al., 2023; Tolkou et al., 2020).

The adsorption coefficients of synthetic organic compounds (K_D) have already been studied in the literature for low molecular weight synthetic organic compounds (<300 g mol^{-1}) and are reported in clean systems as -1 to 6 L^{-1} g at 0.1 % of adsorbate solubility under equilibrium conditions (Ersan et al., 2017). However, adsorption of synthetic organic compounds by edge-tailored graphenes under natural organic matter (NOM) competition is not well-studied, although NOM is ubiquitous in natural source waters and causes adsorption capacity losses due to pore blockage and active site competition (Ersan et al., 2019). If selective edge oxidation is studied, the inner sheet distance of graphenes can be adjusted to sieve out bulky NOM molecules and minimize the penalty for pollutant adsorption capacity. It should be noted that the natural source waters and treated waters will have differences in size and hydrophobicities ($SUVA_{254}$) thus, representative NOMs would be key to test for this. Similarly, some graphenes show faster adsorption kinetics than activated carbons due to their open, sheet like morphologies, which could have important engineering benefits such as shorter retention times and better utilization of adsorption sites. The lack of inner pore diffusion, though, can be replaced by inner sheet diffusion but may increase the rate, by which adsorbates are adsorbed by graphenes (Bi et al., 2024). However, the effects of edge modification on inner sheet distance are not well reported in the literature. Lastly, the increased ionic strength of solutions could result in “salting out” of compounds from water and change the homoaggregation state of edge-modified graphene sheets (Ersan et al., 2019). In brief, graphenes can be used for pollutant adsorption and their surface chemistries can be tuned to control the sheet stacking distances (homoaggregation) for selective adsorption under NOM competition and high ionic strength solutions (Fig. 2). More research is needed to unravel these mechanistic insights.

Third, the sp^2 hybridized basal plane of graphenes has mobile, conjugated π-electrons that could measure carrier mobility at room temperature as large as $\mu > 10{,}000\ cm^2\ V^{-1}s^{-1}$ that is expedient for dielectric heating (Gosling et al., 2021). The reactivity of graphenes to microwave irradiation can be harvested for rapid and efficient regeneration. However, compared to the first two properties, the fundamental knowledge of how to deploy microwaves for graphene regeneration is even more limited, and research is in its infancy. Research in this area can contribute to the generation of seminal knowledge and advance our understanding of graphene-microwave interactions. In summary, synthetic organic compound adsorptive properties in natural waters, aggregation, and dielectric reactivity of graphenes rely on the sp^2-hybridized electron-rich surface, disposition, and quantity of surficial oxygen-containing functional groups, thus, crucial advances can be made if precise graphene chemistry is studied mechanistically for environmentally relevant synthetic organic compound and pertinent water treatment conditions.

The number of synthetic organic compounds in natural source waters are in the order of 10,000s. In addition, the emergence of new classes of organic pollutants, such as per- and polyfluoroalkyl substances overwhelm the policy makers and water treatment industries. To understand the effects of synthetic organic compound properties on adsorption by edge-tailored graphenes, comprehensive testing of the emerging and environmentally pertinent synthetic organic compounds will be needed. However, performing adsorption experiments for thousands of synthetic organic compounds would be costly, laborious, and time-consuming. Therefore, one possible approach is to utilize the existing database of graphenes and graphene oxides and form a mechanistic link between the disposition of oxygen-containing functional groups and their ability to adsorb pollutants. In other words, forming a mathematical connection between the edge *vs.* bulk oxidized graphene oxides with respect to compound properties may enable the utilization of a decade of adsorption literature to quickly reveal insights regarding the adsorption propensity of synthetic organic compounds by edge-oxidized graphene oxides. However, this requires systematic experimental work to ensure that the graphene properties are changed in a controlled fashion, eliminating the fluctuations caused by graphene’s purity, synthesis, and other variables.

3. Theoretical computational boundaries for edge functionalization

To better understand the theroretical limits on edge-functionalization, initial theoretical computations are presented in Fig. 3 based on some simplifying assumptions. Roughly 0.5–4.0 % of the graphene atoms were computed to be on edges depending on the flake size within 100–1,000 nm flakes. Therefore, if oxygen content is precisely increased then the shifts in synthetic organic compound adsorption under NOM loading can be monitored as a function of graphene characteristics and reveal edge oxidation mechanism. The increasing surface oxygen concurrently changes graphene polarity, flake size, and stacking distance. In addition, the roles of folds, and defects are not fully understood; hence, more research would need to focus on unfolding the imperfections of graphene oxidation. These create a complex model to decode; however, systematic adsorption data that is complemented with material characterization can provide mechanistic insights to help advance our understanding. The comparison between edge *vs.* bulk

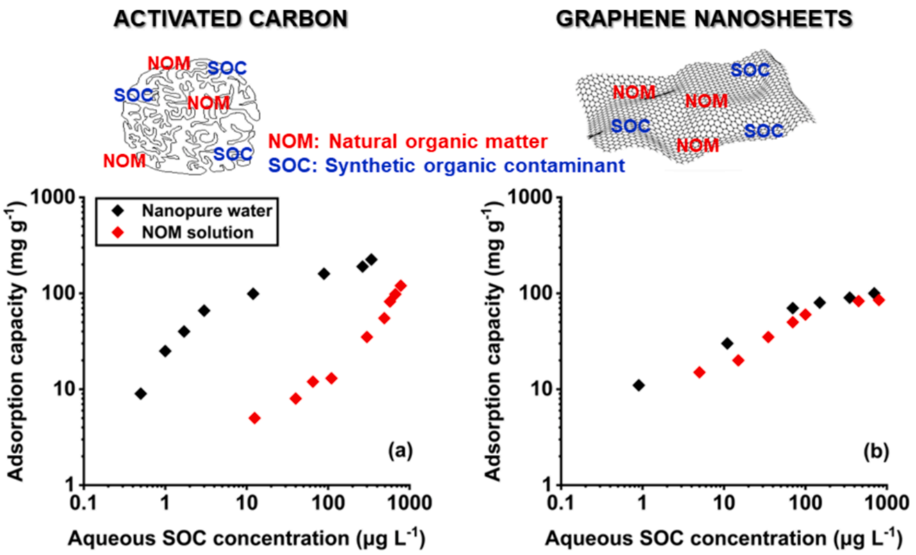

Fig.2. Schematic depiction of adsorption of synthetic organic compound under NOM competition (top). Adsorption isotherms of phenanthrene in nanopure water and 3 mg L^{-1} NOM by activated carbon (a) and graphene (b), respectively (Ersan et al., 2016).

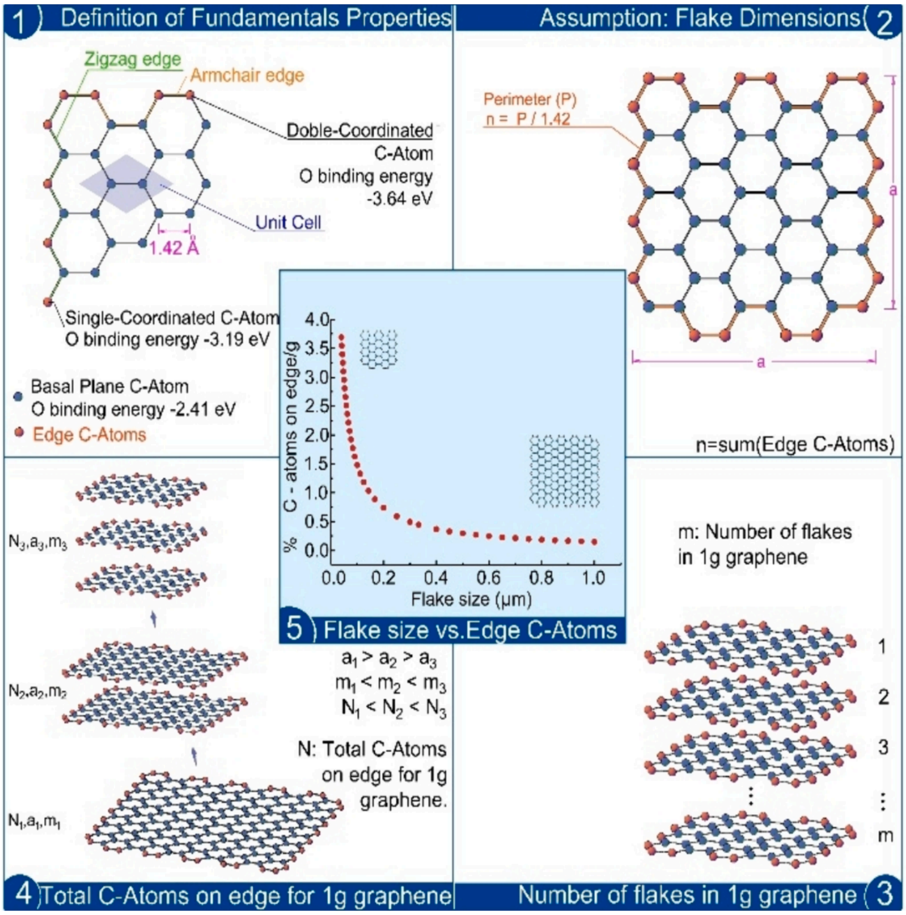

Fig.3. Assumptions, definitions, and theoretical computations for the percentage of C-atoms on the edge of graphene as a function of flake edge length.

graphene oxides can be accomplished by systematically oxidizing the surface with small increments of oxidizing and reducing conditions while testing the total oxygen content and inner sheet distance.

Increasing the oxygen content of graphenes by up to 0.5–4.0 wt% can be hypothesized to keep the inner sheet distance relatively unchanged because most edges can be assumed as oxidized. The increase in C-O bonding will increase the overall polarity (the conceptualization can be seen in Fig. 4); therefore, the inner sheet sieving mechanism may maintain the adsorption of small molecular weight synthetic organic compounds under NOM competition. This is speculated because large NOM molecules will not compete with synthetic organic compounds for the same adsorption sites at the “inner sheet” locations. Increasing the

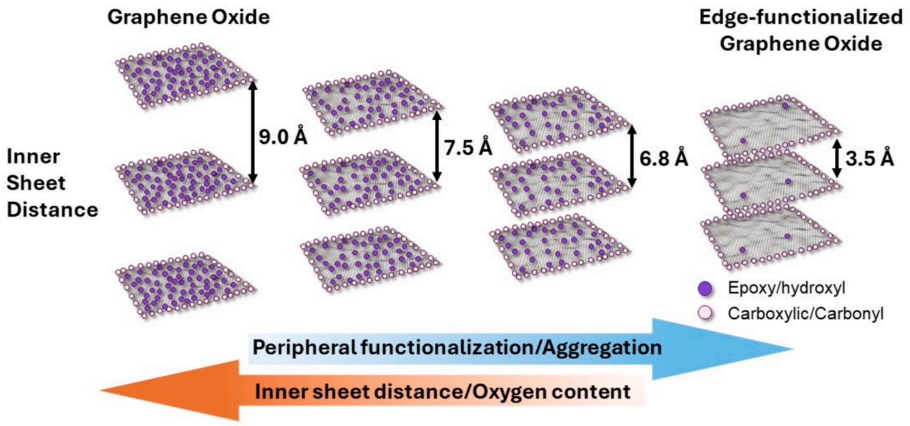

Fig.4. Conceptualization of edge *vs.* bulk oxidation with examples of quantitatively comparing inner sheet distances and oxygen content. Inner sheet distances acquired from Sekimoto et al., 2017.

oxygen content to greater than 4.0 wt% can increase their dispersion in water due to polar interactions. Although this can reveal some adsorption sites, this may not increase the overall adsorption of synthetic organic compounds under NOM competition because the oxidized sites would be expected to host water clusters and repel hydrophobic synthetic organic compounds, unlike the slightly oxidized (or edge-functionalized) graphenes.

4. Dielectric regeneration of graphene nanosheets by microwave irradiation

The electronic structure of graphene nanosheets allows the conversion of microwave energy into heat efficiently more than typical lossy materials that are used in industrial applications such as silicon carbide and activated carbon (Fig. 5) (Apul et al., 2016; Bozkurt and Apul, 2020; Lafaille et al., 2021; Menéndez et al., 2010). Due to the hyperreactivity of graphenes during microwave irradiation, the fundamental aspects of microwave-graphene interactions can be rewarding for energy-efficient heating applications. For example, microwave heating can become an alternative to conventional rotary kilns and multiple hearth furnaces at thermal regeneration facilities because microwaves can selectively heat spent graphenes and achieve desorption and thermolysis of synthetic organic compounds. Therefore, it is construed that microwave-induced graphene regeneration could minimize the energy demand and contribute to the sustainable practice of graphene-enabled water treatment. There are ongoing synthesis efforts of graphene beads, sponges, and pellets in literature, and hence this research can be complementary to recovering spent graphenes (Allgayer et al., 2020; Yousefi et al., 2019). It should be noted that full-scale microwave reactors at 2.45 GHz have been safely implemented in industrial applications for four decades and they provide rapid, efficient, and targeted heating with no start-up delays (Falciglia et al., 2018; Hong et al., 2004; Krouzek et al., 2018). For this, an array of controlled edge-tailored graphenes can be used to systematically reveal key microwave interactions. An ambient atmosphere could thermally anneal graphene oxides by decomposing oxygen-containing functional groups and can produce H_2O, CO, and CO_2 (Hu et al., 2012; McAllister et al., 2007; Pei and Cheng, 2012).

High oxidation levels of graphenes can be detrimental to microwave reactivity despite increasing polar C-O bonds, which would indicate the importance of the π-electron system for microwave reactivity. Deoxygenation of graphene under microwave irradiation was reported at temperatures ~1,000 °C in a nitrogen environment, whereas graphene did not show any notable reaction or changes in C: O ratios at lower temperatures (Chen et al., 2016). Another possible reaction pathway under aggressive heating is incidental oxidation of the surface due to localized heating under microwave irradiation in the ambient atmosphere due to hot spots (Chen et al., 2016). Therefore, to preclude thermally induced redox reactions, deoxygenation, oxidation, and

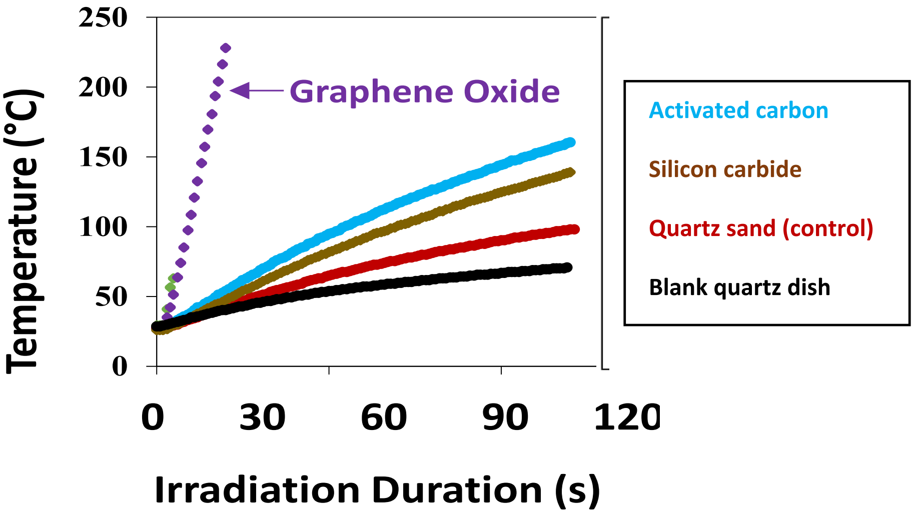

Fig.5. Heating profiles of graphene oxide (new data) *vs.* typical microwave absorbing materials tested using the same sample mass in quartz dishes mixed in quartz sand under 2.45 GHz microwave irradiation for up to 2 min from our previous study (Lafaille et al., 2021).

subsequent gas build-up between sheets: (i) graphenes with less oxygen-containing functional groups on the basal plane need to be used as the starting material and (ii) moderate heating temperatures below annealing point (*i.e.*, ~400 °C) must be reached at (iii) slower rates (<10 °C) during microwave irradiation. Additionally, optimum conditions for temperature limits, as well as microwave power, frequency, and exposure time, need to be considered to preserve the graphenic structure of graphene-based adsorbents during microwave-assisted regeneration. Previous studies have reported that maintaining the temperature below the sublimation point, *i.e.,* ~3,600 °C in a vacuum, is critical during thermal exposure of graphenes (Jing et al., 2021). However, for practical applications, it is important to control the temperature up to ~600 °C to prevent oxidative damage and preserve the graphenic structure, especially in atmospheric conditions (Liu et al., 2019). Previously, a typical microwave power range of 300–800 W, and a standard frequency of 2.45 GHz have been effectively used for microwave-assisted synthesis of carbon-based nanomaterials without causing any damage to structural properties (Adeola et al., 2023). Microwave irradiation exposure typically varies based on the organic contaminant type, load, and microwave settings (Peyravi et al., 2022; Shoushtarian et al., 2020).

Moreover, the electronic structure of graphenes can catalyze the thermolysis of adsorbed organic compounds and cause partial transformation at temperatures significantly lower than the analyte's expected thermolysis temperatures as a function of synthetic organic compound reactivity, volatility, and microwave energy input. Microwave regeneration experiments can demonstrate the catalytic impact of graphene's electronic structure on decomposition *vs.* desorption of adsorbates for a suite of environmentally pertinent and structurally related compounds. This can enable harvesting the microwave energy very efficiently to degrade adsorbates, so repetitive use of the material can be possible while minimizing the risks associated with released synthetic organic compounds or harmful transformation byproducts during regeneration if complete thermal mineralization can be accomplished. This research can particularly be rewarding if a rise in temperatures can thermolyze compounds that are considered persistent (*e.g.*, per- and polyfluoroalkyl substances) while regenerating the spent graphene nanosheets.

During microwave regeneration, intermolecular attractive forces between the graphene surface and the adsorbates would be overcome, and the molecules would be liberated from the graphene surface without degradation first (Fig. 6). This is because the intermolecular interactions are typically much weaker than the covalent bonds that form organic molecules. This can lead to the speculation that thermolysis takes place in the gas phase and graphenes only play the role of converting microwaves to heat. Therefore, if efficient thermal diffusion and good gas phase control are accomplished, then engineering practices could be more desirable. On the other hand, extremely hot spots on the graphene surface can generate •OH radicals and contribute to the destruction of the compounds via oxidation (Fig. 6). The oxidation pathways would be much more complicated than pyrolysis, and a systematic evaluation of radical quenchers and proper controls would be needed to reveal these pathways. Compounds can be carefully selected for microwave regeneration at temperatures based on their boiling points and oxidation potential. All in all, more research in simplified conditions can instigate new discoveries but the work must bridge the long-term engineering vision.

5. Environmental and engineering implications

Global water scarcity is an increasing contemporary challenge of providing access to safe, sustainable, and equitable drinking water and it is becoming relevant to more people day by day. We need a paradigm shift to overcome it in the coming decades. This perspective discussed one possible research pathway to advance cutting-edge knowledge for graphene-based water treatment systems that aim at utilizing the unique properties of graphene nanosheets. This route is a steep learning curve for material scientists and environmental/chemical engineers, but it may be rewarding. Advancing the knowledge in this discipline will have direct implications on our understanding of other graphitic carbon materials and will help advance the edge chemistry literature for similar 2-D materials. In addition, microwave-induced graphene regeneration for nanomaterials needs to be at the forefront of research, which can facilitate discussion about implementing microwaves for granular activated carbon regeneration. Microwave irradiation is a mature, field-proven, and safe technology and can contribute to the global regeneration industry. Currently, the regeneration industry aims to recover roughly 80–85 % of the substrate because of physical losses, accidental carbon burning, and losses to reactor walls. The ability to integrate microwaves into the existing regeneration infrastructure either by ex-situ regeneration plants or accomplishing it in situ via implanted antennas in packed columns can minimize the loss of adsorbents during transport, downtime during carbon switch and regeneration, and the overall cost of the regeneration process.

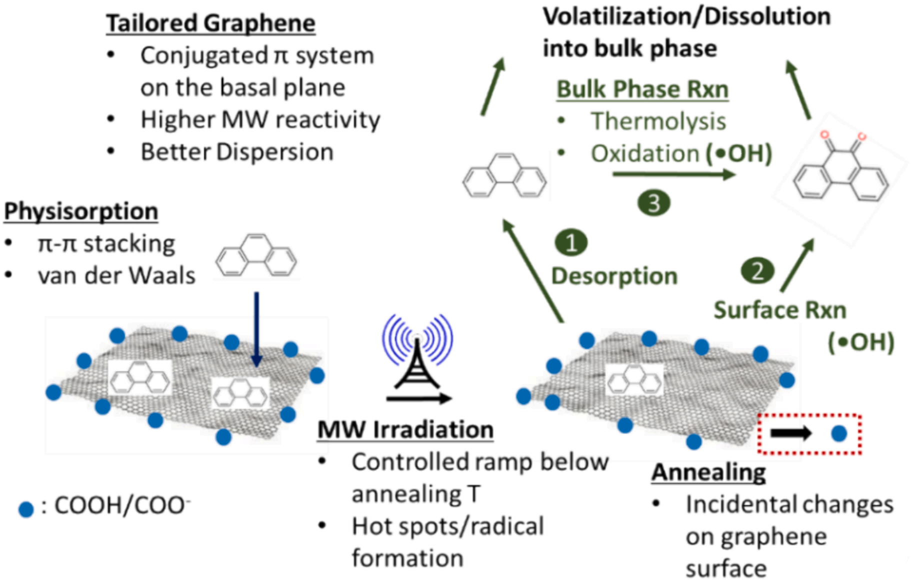

Fig.6. Conceptual model for regeneration mechanism of edge-tailored graphenes during microwave irradiation of phenanthrene-laden graphene.

CRediT authorship contribution statement

Onur G. Apul: Conceptualization, Funding acquisition, Project administration, Resources, Writing – original draft, Writing – review & editing. **Manisha Choudhary:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Onur Apul reports financial support was provided by National Science Foundation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

This work was supported by National Science Foundation through 2219832 and L.L. Bean PFAS Research Program. The manuscript has not been subjected to the peer and policy review of the agencies and therefore does not necessarily reflect their views.

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