

# Progress and expansions of chitosan-graphene oxide hybrid networks utilizing as adsorbents and their organic dye removal performances: A short review

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Abstract: Increasing concentration of pollutants has significantly affected the quality of water, especially for organic dyes-contained water/wastewater. This is probably reduced effectively through adsorbing the pollutants onto potential materials-based adsorbents. Chitosan/graphene oxide (CTS/GO) hybrid networks utilizing as adsorbents are attracting considerable interest owing to the available functional groups, which can remove plenty of the organic dyes from water/wastewater. In this review, the progress and expansions of CTS/GO hybrid networks-based adsorbents are summarized and discussed in detail. Following the general introduction and properties of each material system (i.e., CTS, GO and CTS/GO hybrid networks), the CTS/GO hybrid networks-based emerging adsorbents with coupling numerous other support materials (i.e., reduced GO, magnetic iron oxide, polymeric materials, metal-organic frameworks, etc.) are compared and discussed in short, at same time that some common organic dyes used in adsorption studies are also introduced. Besides, adsorption technique and mechanisms (i.e., adsorption kinetics and isotherms) focus on the CTS/GO hybrid networks' role are discussed as well. Although there are still some challenges from the reviewed researches, the CTS/GO hybrid networks-based emerging materials can be considered promising and potential adsorbents. Moreover, this review can provide a clear overview of CTS/GO hybrid networks-based adsorbents in organic dye removal applications to readers.

**Keywords:** Chitosan, graphene oxide, adsorbent, organic dyes, adsorption.

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#### INTRODUCTION

So far, organic dyes have been employed plenty in the dyeing, textiles, leather, paper, plastics, and food industries, etc.; thus, these colored matters significantly affect currently environmental problems. Various organic dye types (i.e., reactive, anionic, and cationic dyes, etc.) employed in these industries are toxic and disadvantage to the water source (1-6) (Table 1). Specifically, anionic dyes contain negative charges instead of cationic dyes with positive charges in an aqueous solution that is due to the presence of carboxyl and/or sulfonate groups (2, 5, 6). Among them, the anionic dyes are intensely colored, water-soluble, and manifest acidic features inducing to the most challenging (7, 8).

Additionally, the organic molecular structures of these complex aromatic dyes display more stability inducing to being difficult to biodegrade, which leads to removing the organic dyes from water/wastewater with an economical way is considered to be an important challenge for scientists. In fact, primary methods regarding dye-contaminated water/wastewater treatment (i.e., flotation and sedimentation) cannot be adequate to remove the colored features without general chemical manners. The treatment methods of coagulation, separation, and ion exchange are utilized for removing the color from the dye-contaminated water/wastewater as well; however, the cost is one of the main disadvantages of these methods. Whereas, adsorption is a well-known and effective technique in dye-contaminated water/wastewater treatment applications to remove the color from the dye-containing wastewater (4, 8, 9) that is mainly owing to low cost, flexibility, simple design, and easy operation (4).

So far, material system was extensively applied in different research fields and practical applications (4, 6, 10-12), at same time that remarkable factors of a material-based adsorbent almost regard to possible physical, chemical and interface properties. So, it is necessary to conduct considerably synthetic methods that can obtain better desired results corresponding to multiple researches. Concomitantly, the emerging materials-based adsorbents are found and utilized increasingly more to investigate the color dye-contaminated removal from water/wastewater (13-17), which can be novel, readily available, economical and highly effective adsorbents. Especially, chitosan (CTS) - a biopolymer is attracting interest as a matrix for the development of emerging material-based adsorbent that can be mainly owing to potentially active adsorption sites in a high density of available primary amine (-NH<sub>2</sub>) and hydroxyl (-OH) groups; thus, the CTS is considered an efficient adsorbent (4, 6, 18). Actually, the CTS molecule is not available that is produced through N-deacetylation of chitin (19-22) basing on the replacement of acetamide group at position 2 to distinguish the structure and solubility of CTS and chitin molecules (Figure 1) (i.e., CTS is soluble in weak acids, chitin is inert and insoluble) (19-22). At the same time, graphene oxide (GO) contains plenty of functional surface groups [i.e., carbonyl (>C=O), epoxide (C-O-C), hydroxyl (-OH) and carboxylic (-COOH) groups] (Figure 2a) that can facilitate to couple with positively charged molecules through electrostatic interactions, and

which attracts significantly in combing with polymer matrices for enhancement of organic dye removal, mainly based on its high surface area and water solubility (4, 6, 23). Indeed, plenty of studies have shown the GO's excellent ability to adsorb various organic dyes through electrostatic interaction,  $\pi$ -stacking and hydrogen bonding (24-28). However, the organic dye adsorption performance of GO alone is lower than that of modified GO-based adsorbents (24, 25, 28).

Furthermore, the stability of GO nanosheets in colloidal solutions can lead to coupling easily with the CTS molecules (i.e., the CTS is often dissolved in acetic acid) to reach CTS/GO hybrid networks with a homogeneous mixture (Figure 2b). The CTS is known as a positively charged biopolymer that is mainly owing to protonation of the amino (-NH<sub>2</sub>) groups leading to attracting the negatively charged GO nanosheets (Figure 2b). These electrostatic interactions coupling with both hydrogen bonding and  $\pi$ -stacking have facilitated the formation of the CTS/GO hybrid networks (Figure 2b) to reach the more stable networks-based materials hybrid with outstandingly mechanical and thermal features (6, 29). Indeed, the measurement of Fouriertransform infrared spectroscopy (FTIR) (30) has been successfully employed to demonstrate the possible reaction between the amino  $(-NH_2)$ groups of the CTS chains and the carboxylic (-COOH) groups of GO basing on the formation of amide (-NHCO-) linkages (Figure 2b). So far, the CTS/GO hybrid networks can be easily attained through various methods (i.e., the supplement of NaOH, violent shaking, sonication, freeze-drying, etc.) (6, 31-33), and which can also be well combined with other support material sources to develop and expand these CTS/GO hybrid networks that are applied in organic dye removal applications, especially for becoming costeffective, eco-friendly and recyclable adsorbents. In this review article, the adsorption technique regarding the general adsorption theory and the adsorption kinetics/isotherms are introduced in detail. Significantly, the recent progress and expansions of emerging CTS/GO hybrid networks-based adsorbents for various organic dye removal applications are summarized and compared to favor the reader with a clear overview.

| Organic<br>dyes    | Table 1: Information           Structures  | Dye<br>class | λ <sub>max</sub><br>(nm) | M.W.<br>(g⋅mol <sup>-1</sup> ) | Contamination sources   |
|--------------------|--|--------------|--------------------------|--------------------------------|---|
| Methylene<br>Blue  | $\begin{array}{c} H_{3}C_{N} \\ H_{3}C_{N} \\ CH_{3} \\ \end{array} \begin{array}{c} N \\ S \\ H_{3} \\ CH_{3} \\ \end{array} \begin{array}{c} CI' \\ CH_{3} \\ CH_{3} \\ CH_{3} \\ \end{array} $  | +            | 665                      | 319.85                         | Textile, Leather,<br>Dyestuff, Paper and<br>Plastic industries;<br>Pharmaceutical uses;<br>Laundry activity |
| Rhodamine<br>B     |  | +            | 558                      | 479.02                         | Natural sources;<br>Textile, Dyestuff,<br>Food, Leather and<br>Plastic industries;<br>Laundry activity      |
| Malachite<br>green | CH <sub>3</sub><br>H <sub>3</sub> C <sup>-N⊕⊙CI</sup> CH <sub>3</sub><br>CH <sub>3</sub> C <sup>-N⊕</sup> CH <sub>3</sub>  | +            | 617                      | 364.91                         | Textile, Leather and<br>Dyestuff industries;<br>Lab activities;<br>Laundry activity                         |
| Methyl<br>Orange   | $H_{3C}$ $N$   | -            | 464                      | 327.33                         | Textile, Foodstuffs,<br>Paper and Leather<br>industries; Lab<br>activities; Laundry<br>activity             |
| Congo Red          | $\begin{array}{c} H_2 N \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ $  | -            | 498                      | 696.67                         | Textile, Leather and Dyestuff industries  |
| Brilliant<br>green | $\begin{array}{c} CH_{3} \\ \oplus \\ N \\ CH_{3} \\ HSO_{4} \\ H_{3}C \\ N \\ CH_{3} \end{array}$   | +            | 625                      | 475.60                         | Pigments, Dyestuff<br>and Chemical<br>industries;<br>Pharmaceutical uses                                    |
| Crystal<br>violet  | $\begin{array}{c} \begin{array}{c} \begin{array}{c} \begin{array}{c} \begin{array}{c} \begin{array}{c} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \end{array} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \begin{array}{c} \\ \\ \\ \end{array} \\ \\ \\ \end{array} \\ \\ \end{array} \\ \\ \end{array} \\ \\ \\ \\ \end{array} \\$ | +            | 590                      | 407.98                         | Textile, Leather and<br>Dyestuff industries;<br>Laundry activity  |

 Table 1: Information of some organic dves.



**Figure 1:** Preparation of chitin and CTS molecules from the waste supply sources.

## **ADSORPTION TECHNIQUE**

Adsorption is known as a surface phenomenon, which is occurred at the surface or interface positions. In another word, it mentions a procedure in which a material is gathered at a surface by its gaseous or liquid solid surroundings. In 1881, the term "adsorption" was firstly introduced by Heinrich Kayser – a German physicist (1853–1940) in order to distinguish surface accumulation from intermolecular penetration, as well as which assumes surface accumulation of a material to be a fundamental feature of an adsorption process (4). Theoretically, two adsorption types count physisorption and chemisorption. If the attractive forces between the adsorbed molecules and the solid surface relate to van der Waals forces, which are usually weak forces inducing reversible adsorption to be named physisorption. In comparison, chemisorption involves chemical bonds between the adsorbed molecules and the solid surface, which is higher strength, leading to hard removal of the chemisorbed molecules from the solid surface (4).

Besides, adsorption techniques are extensively employed to remove plenty of pollutants from water/wastewater, especially for organic dyes that are not readily biodegradable. The general mechanism of dye adsorption consists of three steps during the process of color removal following as (34): (i) diffusion of organic dye molecules - adsorbates presented availably in the bulk solution can be onto the material-based adsorbent surface; (ii) the organic dye adsorption on the material-based adsorbent surface can base on molecular interactions (i.e., "film diffusion"); and (iii) diffusion of organic dye molecules can be moved from the surface to the inside of the materials-based adsorbents (i.e., "surface diffusion" or "pore diffusion"). The processes of diffusion and surface reaction occurring on the materials-based adsorbents are described in Figure 3. Overall, agitation and concentration of the organic dye molecules in the solution probably impact to the first step. The nature of the organic dye molecules directly influences to the second step, for example, cationic and anionic features. At the same time, the third step regards to the rate-confirming stage in this process that surely impacts to the adsorption of organic dye molecules on the

substrates. In particular, external diffusion occurring in the adsorption processes is the most efficacy, while intraparticle diffusion will attain a longer contact time in non-flow systems. Moreover, these are influenced by physicochemical factors, examples for interactions of organic dyes and adsorbents, particle sizes, surface areas of adsorbents, pH values, temperatures, and contact times.



Figure 2: Schematic of chemical structure of GO (a) and possible interactions in CTS/GO hybrid networks (b).



Figure 3: The processes of diffusion and surface reaction occurring on the materials-based adsorbents.

In addition, adsorption isotherms and kinetics are suitable models to evaluate the adsorption process (35). Specifically, adsorption isotherms can reach through a change in the initial concentrations of organic dye solutions, and the other corresponding parameters are constants (i.e., contact times, temperatures, pH values, and stirring speeds). Basically, the removal efficiency will attain high values through increases of the organic dye concentrations until no further uptake, which is named saturation or equilibrium state. The common adsorption isotherm models are listed in Table 2, which are on basic of the original and linearized forms. Concomitantly, many other models can also be employed to elucidate the mechanism of organic dye adsorption on a material-based adsorbent, e.g., common adsorption kinetic models (Table 2 and Figure 3). In fact, the adsorption kinetics are usually defined by establishing plots of the adsorbed quantity vs. a time function to manifest the possible nature of the adsorption process and the kinetics. Overall, in order to more easily understand, this review has been summarized on basic of various adsorbent types studied from the progress and expansions of CTS/GO hybrid networks-based adsorbents coupled with numerous other support materials (i.e., reduced GO, magnetic iron oxide, polymeric materials, metal-organic frameworks, etc.), as well as organic dye removal efficiency and adsorption mechanisms onto these emerging adsorbents.

| Adsorption isotherms    | Original form  | Linearized form  | Plot   | Constants<br>attained from<br>the plot            |
|-------------------------|--|--|--|---|
| Langmuir                | $q_e = \frac{q_m K_L C_e}{1 + K_L C_e}$  | $\frac{C_{e}}{q_{e}} = \frac{1}{q_{m}K_{L}} + \frac{C_{e}}{q_{m}}$   | $rac{C_e}{q_e}$ vs $C_e$  | $\boldsymbol{q}_{m}$ and $\boldsymbol{K}_{L}$     |
| Freundlich              | $q_e = K_F \frac{C_e^{1/n}}{n}$  | $\log q_e = \log K_F + \frac{1}{n} \log C_e$   | $\logq_{_e}^{} \ ^{\text{vs}} \logC_{_e}^{}$                                       | $\boldsymbol{K}_{\boldsymbol{F}}^{}$ and n        |
| BET                     | $q_{e} = \frac{K_{B}C_{e}q_{m}}{(C_{s} - C_{e})[1 + (K_{B} - 1)(\frac{C_{e}}{C_{s}})]}$  | $\frac{C_{e}}{q_{e}(C_{s}-C_{e})} = \left(\frac{1}{K_{B}q_{m}}\right) + \left(\frac{K_{B}-1}{K_{B}q_{m}}\right)\left(\frac{C_{e}}{C_{s}}\right)$ | $\frac{C_{e}}{q_{e}(C_{s}\!-\!C_{e})} \ \text{vs} \ \big(\frac{C_{e}}{C_{s}}\big)$ | $K_{_{\rm B}}$ and $q_{_{\rm m}}$                 |
| Temkin                  | $q_e = \frac{R.T.}{b} \ln \left( A_T C_e \right)$  | $q_e = \frac{R.T.}{b} \ln A_T + \frac{R.T.}{b} \ln C_e$  | $\ln C_e q_e^{vs}$   | $\boldsymbol{A}_{_{T}}$ and b                     |
| Dubinin-Radushkevich    | $q_e = q_s e^{-K_{D,R,}\epsilon^2}$  | $\ln q_{e} = \ln q_{s} - K_{D.R.} \varepsilon^{2}$ $\varepsilon = R T \ln \left(1 + \frac{1}{C_{e}}\right)$                                      | $\ln q_e \sim \epsilon^2$  | $K_{\text{D.R.}}\ \text{and}\ \boldsymbol{q}_{s}$ |
| Toth                    | $\mathbf{q}_{e} = \frac{\mathbf{K}_{\mathrm{T}} \mathbf{C}_{e}}{\sqrt[t]{\left(\mathbf{a}_{\mathrm{T}} + \mathbf{C}_{e}^{\mathrm{t}}\right)}}$ | $\log q_e = 2\log K_T - \frac{1}{t}\log a_T + \left(2 - \frac{1}{t}\right)\log C_e$  | $\log q_e^{vs} \log C_e^{vs}$  | $K_{_{\rm T}}$ and $a_{_{\rm T}}$                 |
| Redlich-Peterson        | $q_e = \frac{A \times C_e}{1 + B C_e^{\beta}}$   | $\ln \frac{C_e}{q_e} = \beta \ln C_e - \ln A$  | $\ln \frac{C_e}{q_e}$ vs $\ln C_e$   | A and $\beta$                                     |
| Adsorption kinetics     | Linearized form  | Plot   | Constants a<br>plot  | attained from the                                 |
| Pseudo-first-order      | $\ln(q_e-q_t)=\ln q_t$   | $q_e - k_1 t$ $ln(q_e - q_t) v$  | s <sub>t</sub> k <sub>1</sub>  |   |
| Pseudo-second-order     | $\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$  | $rac{t}{q_t}$ vs $rac{t}{q_e}$   | k <sub>2</sub>   |   |
| Elovich                 | $q_t = \frac{1}{\beta} \ln \alpha \beta + \frac{1}{\beta} l$   | nt q <sub>t</sub> vs ln t  | $\alpha$ and $\beta$   |   |
| Intraparticle diffusion | $q_t = k_t t^{0.5} + 1$  | $q_t$ vs $t^{0.5}$   | ${\boldsymbol{k}}_t$ and I   |   |

| Table 2: Common adsorption | n isotherm and kinetic models. |
|----------------------------|--------------------------------|
|----------------------------|--------------------------------|

### VARIOUS CTS/GO HYBRID NETWORKS-BASED ADSORBENTS AND ORGANIC DYE ADSORPTION PERFORMANCES

As known, CTS is often dissolved in acetic acid, and GO is well stable in colloidal solutions, which can easily couple together to attain CTS/GO hybrid networks with a homogeneous mixture through electrostatic interactions, hydrogen bonding, and  $\pi$ -stacking resulting in outstandingly mechanical and thermal features (6, 29). Concomitantly, the CTS/GO hybrid networks-based materials can be prepared from various methods such as supplement of NaOH, violent shaking, sonication, freeze-drying, etc. (6, 31-33). As a result, the CTS/GO hybrid networks-based materials have been formed with various shapes such as membranes, beads, sponges, and columns (6, 36-39), and which are all utilized successfully as effective adsorbents for organic dye removal from water/wastewater (40).

Notably, other components - support materials have been supplemented to enhance the organic dye adsorption performance of the CTS/GO hybrid networks-based materials in recent years; in particular, two-dimensional GO has also been developed to reduced GO (rGO) or threedimensional GO-based aerogels. Typically, the GO is formed through oxidizing graphite (i.e., Hummers method) (41), while the rGO is synthesized by reducing the GO (i.e., various thermal methods, reducing agents, electrochemical Actually, reduction) (42-45). the oxvaencontaining functional groups on the GO are difficult to reduce, which still contained some on the rGO. Besides, the rGO can combine with various metals or metal oxide particles (46), facilitating their incorporation within the CTS molecule. For instance, CTS/rGO hybrid network has been coupled with iron oxide (Fe<sub>3</sub>O<sub>4</sub>) to attain magnetic CTS/rGO hybrid network-based adsorbents that are effectively applied for the cefixime adsorption (47) (Table 3). Concomitantly, a comparison of dye removal performance of CTS/GO and CTS/rGO hybrid networks utilizing as adsorbents indicated that the CTS/GO hybrid network has adsorbed efficiently more (48) (Table 3), which can involve to the presence of more numerous available functional groups offering a combination of electrostatic interactions, hydrogen bonding and  $\pi$ stacking with the organic dye molecules (4, 6, 28). As a result, the pseudo-second-order model studied in this research was the best model, indicated that the organic dve adsorption could be induced on the chemical rate-limiting step (i.e., electron share or covalent forces).

So far, magnetic CTS-based adsorbent has emerged as a potential material for removing pollutants from water/wastewater. There has recently been a considerable interest in the magnetic CTS/GO hybrid network-based material as well (49, 50) (Table 3); notably, these magnetic materials have shown a fast removal performance. Obviously, the magnetism approach can facilitate

well for separating the adsorbents from water/wastewater based on a simple magnetic procedure (51). In fact, separation of CTS/GO hybrid network-based materials using conventional sedimentation and filtration techniques is an important challenge due to blocked and lost filters inducing secondary contaminations. Hence, the choice of  $Fe_3O_4$  is the most efficacy, owing to its low toxicity, good compatibility and high magnetic features (52). For preparation methods of the magnetic CTS/GO hybrid network-based materials, it can be easily created via in- or ex-situ approach, or a combination of both these approaches (49, 53), or alterations of these two methods (50, 54) (i.e., CTS/ Fe<sub>3</sub>O<sub>4</sub> hybrid network can be first created before being coupled with GO, or opposite). Nonetheless, the shape and size of the Fe<sub>3</sub>O<sub>4</sub> nanoparticles are also important characteristics to couple with the CTS/GO hybrid network-based materials for organic dye removal performances. Although the  $Fe_3O_4$  nanoparticles can be easily clustered that is mainly due to their magnetic nature, several studies have also be conducted to control the suitable shape and size with the research aims (54-56). For example, Fe<sub>3</sub>O<sub>4</sub> particles could be attained with the spherical shapes and ~45 nm of size by Shafaati et al. (55), but their particle size has increased with incorporating the CTS, suggesting that the CTS chains could probably induce linking together for the neighboring Fe<sub>3</sub>O<sub>4</sub> particles. Silica is employed to coat the Fe<sub>3</sub>O<sub>4</sub> particles resulting from the significantly reduced agglomeration (56). Thereby, the use of silica is a promising approach to coat and protect magnetic iron-containing particles in the CTS/GO hybrid networks, as well as stability of these materials need to be investigated more to not leach the GO flakes or the magnetic iron oxide particles.

In addition, the CTS/GO (or CTS/rGO) hybrid networks-based materials with three-dimensional porous structures acting as scaffolds (i.e., aerogel, foam, sponge, column) can be fabricated through the available wrinkling and blending of GO nanosheets. These scaffolds are effectively applied in the organic dye removal applications owing to high surface areas, low mass densities (porous structure) and good mechanical strength (57, 58); besides, these scaffolds-based adsorbents are also easily regained from the aqueous solution after completing the adsorption procedure. Indeed, the GO or rGO alone with three-dimensional structures reach relatively poor stability in water, but this stability can be improved significantly through a combination of the three-dimensional GO or rGO network and the CTS molecules. For instance, the three-dimensional CTS/GO hybrid network-based columns have been successfully applied in five adsorption-regeneration cycles reaching >90%adsorption performances (6, 32), and the layered CTS/GO hybrid network-based sponges have also been attained good stability and effective resulting recvclability in>80%-adsorption performances over five adsorption-regeneration cycles (59). Besides, plenty of studies on CTS/GO

hybrid network-based aerogel have been successfully fabricated to remove effectively azo dyes, anionic and cationic dyes (60, 61) from water.

Furthermore, other additives - support materials have been coupled with the CTS/GO hybrid network to become potential and outstanding materials-based adsorbents in environmental applications. For example,  $\beta\mbox{-cyclodextrin}$  (Figure 4) has been combined with the CTS/GO hybrid network to enhance the adsorption performance of methylene blue following GO < CTS/GO < CTS/GO/B-cyclodextrin networks (62), similarly for a study on the adsorption performance of hydroquinone and dye molecules (63) (Table 3). Besides, polypyrrole (Figure 4) has also been used to couple with the CTS/GO hybrid network basing on polymerization of the pyrrole monomer in the above-mentioned hybrid network, resulting in the ternary CTS/GO/polypyrrole hybrid network-based material that could become efficient adsorbents in the organic dye removal applications (64, 65) (Table 3). Interestedly, these hybrid networksbased adsorbents could incorporate magnetic nanoparticles to improve the separation of these adsorbents from water/wastewater after completing the organic dye adsorption process (66) (Table 3). Concomitantly, other polymers with high molecular weights (i.e., polydopamine, polyacrylamide, and polyacrylate are shown in Figure 4) have also been employed to combine with the CTS/GO hybrid network (67-69) that could significantly enhance the swelling and adsorption performances of these hybrid networksbased adsorbents (Table 3). In another word, these support materials have offered different binding sites to enhance the adsorption performances, and which are considered promising materials-based adsorbents.

More notably, metal-organic frameworks (MOFs) have recently attracted much interest in

environmental applications that is mainly due to their high surface areas and porosities and controlled pore structures. Thus, they have been used to add into the CTS/GO hybrid network that enhances adsorption performance (70, 71). Unfortunately, the MOFs are powdered materials that are hard to separate from water, which is its main limitation in environmental applications. Thereby, the CTS/GO hybrid network can be contributed like a matrix to encapsulate these powdered materials that can couple with magnetic iron to result in a necessary magnetic separation. Besides, plenty of other support materials has also been combined with the CTS/GO hybrid network; the use of hydroxyapatite for examples,  $[Ca_{10}(PO_4)_6(OH)_2]$  has increased strength and organic pollutant adsorption performance (72), the use of silica (numerous silanol groups, Si-OH) has favored a good dispersion of GO nanosheets within CTS to reach effective adsorbents (73, 74), the use of triethylenetetramine and lignosulfonate (Figure 4) have supplied amine groups and different binding sites to enhance adsorption performance, respectively (75-77) (Table 3). In general, these studies manifested that these materials could become cost-effective, eco-friendly, and recyclable adsorbents for organic dye removal applications. Additionally, other biopolymers (i.e., gelatin, alginate, heparin, and cellulose are shown in Figure 4) have been blended with the CTS to create polymeric mixtures, which are then coupled with the GO nanosheets to attain outstanding performing adsorbents (78-81). Overall, the pseudo-second-order model studied in these researches were the best model; nonetheless, the kinetic studies need to investigate more, and the chemical/physical characterizations of the CTS can be focused more on the degree of deacetylation levels, molecular weight, etc., similarly, for the GO. Also, it is needed to develop further suitable recovery systems, as well as preparation process of the adsorbents should be employed green solvents and material sources.



| Figure 4: Chemical structures of various additive    | s.  |
|--|-----|
| Adsorption data of various CTS/GO hybrid networks-ba | haa |

| Table 3. Adsorption data of various CTS/GO hybrid networks-based adsorbents. |  |  |                                  |                              |
|--|--|--|----------------------------------|------------------------------|
| Adsorbents   | Organic dyes   | Adsorption<br>performance<br>(%, or mg·g <sup>-1</sup> ) | Adsorption<br>isotherm<br>models | Adsorption kinetic<br>models |
| CTS/GO_1/15 (6)  | Methylene blue,<br>Rhodamine B,<br>Congo Red,<br>Methyl orange | ~95.0%,<br>>90.0%,<br>~70.0%,<br>~80.0%                  |                                  |                              |
| CTS/GO (48)  | Reactive Red   | 32.2 mg⋅g <sup>-1</sup>                                  | Langmuir                         | Pseudo-second order          |
| Magnetic CTS/rGO<br>(47)   | Cefixime   | 30.6 mg·g⁻¹  | Freundlich                       |                              |
| Magnetic CTS/GO<br>(49)  | Methylene blue   | 74.93 mg⋅g <sup>-1</sup>                                 | Freundlich                       | Pseudo-second order          |
| CTS/GO/β-<br>cyclodextrin (62)   | Methylene blue   | 1134.0 mg⋅g <sup>-1</sup>                                | Freundlich                       | Pseudo-second order          |
| Magnetic<br>CTS/GO/β-<br>cyclodextrin (63)                                   | Methylene blue   | 84.3 mg·g <sup>-1</sup>                                  | Langmuir                         | Pseudo-second order          |

| Polypyrrole/CTS/GO<br>(64)                 | Ponceau 4R                                  | 5.4 mg⋅g <sup>-1</sup>  | Langmuir   | Pseudo-second order                        |
|--|---|---|------------|--|
| Magnetic<br>Polypyrrole/CTS/GO<br>(66)     | Ponceau 4R                                  | 5.1 mg⋅g <sup>-1</sup>  | Langmuir   | Pseudo-second order                        |
| CTS/<br>Polyacrylamide/GO<br>(67)          | Methylene blue                              | 510.2 mg·g <sup>-1</sup>  | Langmuir   | Pseudo-second order                        |
| CTS/polyacrylate/<br>GO (68)               | Methylene blue,<br>Food yellow 3            | 296.5 mg·g <sup>-1</sup> ,<br>280.3 mg·g <sup>-1</sup>                            |            | Pseudo-first order,<br>Pseudo-second order |
| CTS/GO/<br>Hydroxyapatite (72)             | Congo Red,<br>Acid Red 1,<br>Reactive Red 2 | 43.1 mg·g <sup>-1</sup> ,<br>41.3 mg·g <sup>-1</sup> ,<br>40.0 mg·g <sup>-1</sup> | Freundlich | Pseudo-second order                        |
| CTS/<br>lignosulfonate/GO<br>(75)          | Methylene blue                              | 1023.9 mg·g <sup>-1</sup>   | Langmuir   | Pseudo-second order                        |
| Magnetic<br>CTS/lignosulfonate/<br>GO (76) | Methylene blue                              | 253.5 mg·g <sup>-1</sup>  | Langmuir   | Pseudo-second order                        |

## CONCLUSIONS AND FUTURE RESEARCH OUTLOOKS

With a large number of publications, the CTS/GO hybrid networks-based materials are employed as potential and interesting adsorbents for various organic dye removal applications. In summary, the CTS/GO hybrid networks easily couple with numerous other components or additives - support materials (i.e., rGO, magnetic iron oxide, polymeric materials, MOFs, etc.) that is attracting considerable attention, as well as the CTS/GO hybrid networks-based materials have attained not only magnificent adsorption capacity with organic dyes but also heavy metal ions. Nonetheless, several challenges still exist for removing a variety of pollutants directly implicating in terms of costs, which regards the regeneration of the CTS/GO hybrid networks-based adsorbents. These adsorbents should be effectively regenerated to can be employed multiple times in the adsorption process, which relates to washing-solvents and natures of polysaccharide/carbon-based materials (CTS/GO); thus, the adsorption capacity can reduce with each cycle of adsorption-regeneration (i.e., the polysaccharide hydrolysis on the CTS). These require an offer of more longer-lasting and cost-effective CTS/GO hybrid networks-based adsorbents.

In addition to the effective performances of these adsorbents, they also need to be removed from the aqueous solution after completing the adsorption purposes, especially for the magnetic CTS/GO hybrid networks. Concomitantly, there are also concerns about the environmental influences CTS/GO hybrid of other networks-based adsorbents, which can adversely impact to the water environment if these materials-based adsorbents are leached. Consequently, the CTS/GO hybrid emerging networks-based adsorbents have to be stable and not leach regarding other support materials (i.e., magnetic iron oxide particles, GO flakes, and MOFs, etc.), which is considered as one of the importantly

environmental perspectives. The other studies on intraparticle diffusion and adsorption kinetics also need to be investigated further, although most studied kinetic models regarded to pseudo-secondorder models. These emerging materials-based adsorbents are attractive and potential materials that is probably due to the abundance supply source from chitin (seafood waste sources), and expansion of these materials in the various preparation methods and high-performance studies. Besides, the recent development has attended to new two-dimensional materials (i.e., MoS<sub>2</sub>, MXenes, and MoSe<sub>2</sub>), which are potential support materials utilized as emerging adsorbents and can easily couple with the CTS or CTS/GO hybrid networks to generate novel materials-based adsorbents with outstanding performances.

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