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Review

Removal of contaminants present in water and wastewater by cyclodextrin-based adsorbents: A bibliometric review from 1993 to 2022☆

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ABSTRACT

Cyclodextrin (CD), a cyclic oligosaccharide from enzymatic starch breakdown, plays a crucial role in pharmaceuticals, food, agriculture, textiles, biotechnology, chemicals, and environmental applications, including water and wastewater treatment. In this study, a statistical analysis was performed using VOSviewer and Citespace to scrutinize 2038 articles published from 1993 to 2022. The investigation unveiled a notable upsurge in pertinent articles and citation counts, with China and USA contributing the highest publication volumes. The prevailing research focus predominantly revolves around the application of CD-based materials used as adsorbents to remove conventional contaminants such as dyes and metals. The CD chemistry allows the construction of materials with various architectures, including cross-linked, grafted, hybrid or supported systems. The main adsorbents are cross-linked CD polymers, including nanosponges, fibres and hybrid composites. Additionally, research efforts are actually concentrated on the synthesis of CD-based membranes, CD@graphene oxide, and CD@TiO_2. These materials are proposed as adsorbents to remove emerging pollutants. By employing bibliometric analysis, this study delivers a comprehensive retrospective review and synthesis of research concerning CD-based adsorbents for the removal of contaminants from wastewater, thereby offering valuable insights for future large-scale application of CD-based adsorption materials.

1. Introduction

The rapid advancement of the industry has resulted in the concurrent growth of various sectors, including the textile, paper, printing, battery manufacturing, metal plating, mining operations, metallurgy, and leather manufacturing industries (Dutta et al., 2021; Morin-Crini et al., 2022). The development of these industries has led and still leads inevitably to the discharge of substantial volumes of wastewater into the natural environment, even if these discharges respect the regulations in force in each country (Chen et al., 2020a). Population growth, increased demands, and deteriorating water quality have heightened the demand for clean water supply (Akhtar et al., 2021). Nevertheless, ensuring the provision of clean and safe water, especially in developing countries, has become a formidable challenge and an urgent imperative. The environmental repercussions of wastewater pollutants have been widely acknowledged as constituting a significant health and environmental threat to humanity, natural ecosystems, and other organisms (Garg et al., 2022; Qu et al., 2019).

Conventional wastewater treatment techniques such as physicochemistry, oxidation, adsorption, biological treatment, ion-exchange, and membrane filtration are well-known and used by the industrial sector (Crini and Lichtfouse, 2019). However, due to the context of water scarcity and pollution, industry is asked to make further efforts to reduce their pollution flows to aquatic environments. In the past few decades, there has been a growing emphasis on the development of new efficient wastewater treatment technologies (Rout et al., 2021). These technologies include solvent extraction, evaporation, advanced oxidation, electrochemistry, and biosorption. (Giwa et al., 2021; Adam et al., 2022; Samsami et al., 2020; Yashni et al., 2021; Crini et al., 2019). Among these techniques, adsorption is widely recognized as a

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cost-effective wastewater treatment method and arguably the most used technique with biological treatment (Chai et al., 2021; Rashid et al., 2021). Adsorption is a fundamental surface phenomenon that involves selectively removing pollutants from aqueous solutions by adsorbing solutes (typically referred to as adsorbates) onto solid surfaces (commonly referred to as adsorbents) (Adegoke et al., 2022; Tony, 2022). Adsorption technology can efficiently eliminate pollutants from water at a relatively low cost while avoiding the potential for secondary contamination (Rathi and Kumar, 2021). Compared to alternative processes, adsorption offers lower costs, a more straightforward design, and higher efficiency in removing trace hazardous pollutants (Rathi and Kumar, 2021; Rasheed, 2022).

The most used adsorbent material is activated carbons (Dąbrowski et al., 2005; Qu, 2008). Other commercial materials are also used, such as clays, zeolites, alumina, porous organic resins, or modified silicas. Other materials of natural origin, called biosorbents, have also been proposed (Fakhri et al., 2023). Examples include polysaccharides, aquatic plants, and biomass (Cabral et al., 2022; Panja et al., 2023). Despite extensive research on conventional and unconventional adsorbents, the challenge of finding materials that are both chemically effective, cheap, of natural origin, easy to use and ecologically benign has mobilized many researchers on this theme (Bharadwaz and Jayasuriya, 2020). Thus, new synthetic and modified natural materials such as nanosponges, metal-organic frameworks (MOFs), and molecularly imprinted polymers (MIPs) have been recently proposed (Zhao et al., 2023; Rajkumar et al., 2019; Fernández et al., 2019; Tian et al., 2021; Okasha et al., 2023; Jia et al., 2022; Utzeri et al., 2022; Belenguer-Sapiña et al., 2020; Cova et al., 2021; Zhao et al., 2021).

Biosorbents such as cyclodextrin polymers and cyclodextrin-based composites have also found extensive application in environmental remediation processes (Tian et al., 2021; Okasha et al., 2023; Saravanan et al., 2023; Safapour et al., 2022; Ozelcaglayan and Parker, 2023). Cyclodextrins (CDs) have gained significant attention due to their environmentally friendly, easily available (from renewable and green resources), non-toxicity, biocompatibility and biodegradable properties, coupled with their capacity to adsorb contaminants from wastewater (Qi et al., 2021; Morin-Crini et al., 2021). These macromolecules are naturally occurring cyclic oligosaccharides, and the most widely employed include α-CD, β-CD, and γ-CD, each composed of 6, 7, and 8 D-glucopyranose units, respectively (Gonzalez Pereira et al., 2021; Chodankar et al., 2022). They possess a unique hydrophobic inner cavity and a hydrophilic outer structure, enabling them to encapsulate contaminants and form host-guest complexes, making them suitable candidates for use as adsorbents or biosorbents (Morin-Crini et al., 2018).

Towards the end of the last century, the enormous potential of CDs in the realm of environmental remediation was realized (Fig. 1). Highly cited articles often revolve around oligomers and polymers derived from renewable sources. These sustainable alternatives are not only biocompatible and biodegradable but also reduce our reliance on fossil fuels. A notable trend in recent years is the increasing citation of published articles on CD polymers and CD-based MOF materials not only for water and wastewater applications but also for pharmacy, medicine, and food.

When searching the "SCOPUS" database with "cyclodextrin" and "water treatment" as subjects, a total of 60 articles were classified as review articles from 2021 to 2023 (as of October 7, 2023), with only 13 of these reviews systematically addressing the use of CD-based adsorbents for the removal of wastewater contaminants. Table S1 provides a summary of these thirteen reviews. Despite the existence of several recent review articles in related fields, there has been no comprehensive overview of the research landscape and trends, which limits the ability to offer new insights from a developmental perspective. Bibliometric analysis is a powerful tool for exploring and analyzing scientific data within published articles, unveiling the progress in specific domains and emerging areas (Mejia et al., 2021; Siddiqui et al., 2023). In recent years, this approach has been employed to investigate research trends and prospects in various fields, such as the application of biochar in electrochemical processes (Jiang et al., 2023), the fate of antibiotic resistance genes in solid waste composting (Sun et al., 2023), and phosphorus recovery from livestock and poultry manure (Ran et al., 2023), among others.

This study represents the first systematic bibliometric analysis of publications related to the adsorption of contaminants from aqueous media using CD. The objective is to investigate the progress and trends within this research domain comprehensively. The approach taken was as follows.

(1) List the institutions and authors of the most relevant publications;

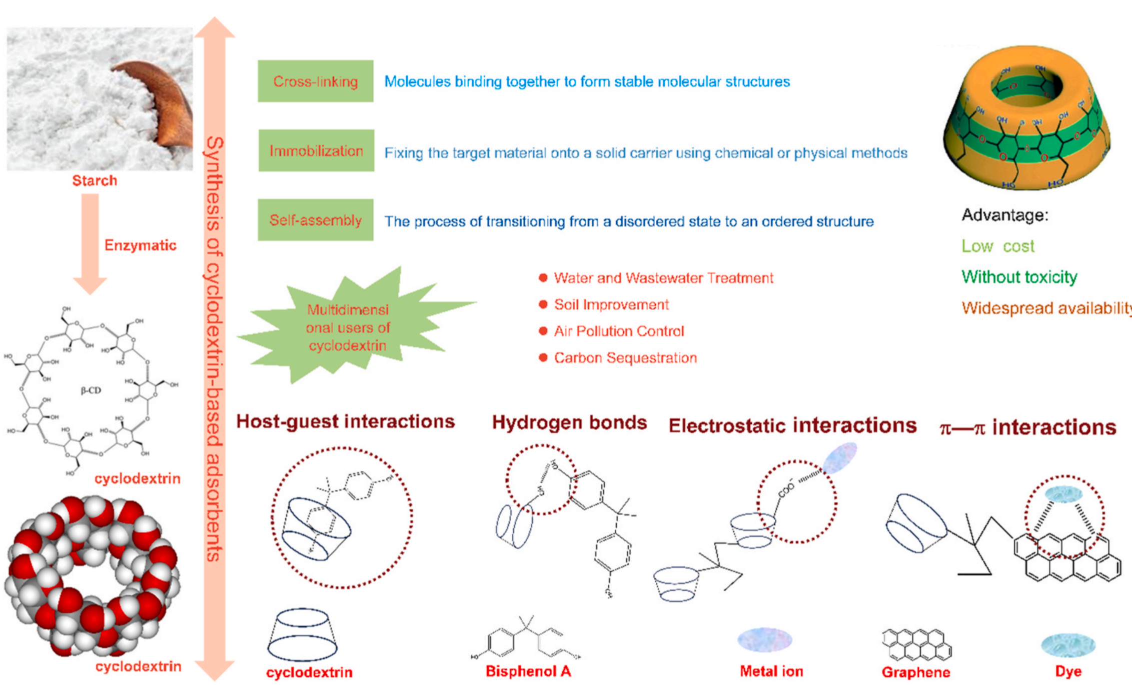

Fig. 1. The application of CD-based adsorbents.

(2) Summarize current research hotspots and their advancements based on co-citation analysis of literature, keywords, and co-cited topics;
(3) Identify current challenges and outline future research directions.

This research endeavors to provide a comprehensive understanding of the current state of research and future research directions within this field. Furthermore, it will serve as a reference and inspiration for subsequent related studies while laying a theoretical foundation for the environmental applications of CD.

2. Methodology

2.1. Data sources

The literature metrics analysis of this study adhered to the technical workflow depicted in Fig. 2. This research collected literature from the Web of Science (WOS) Core Collection, focusing on evaluating the research progress and developmental trends in using CD-based adsorbents for wastewater pollutant removal. The literature search adhered to specific criteria:

(1) The search strategy was TS = ((sorption or removal) NOT (soil or gas or CO_2 OR H_2S)) AND TS = (cyclodextrin);
(2) Publications were limited to the period from January 1, 1993 to December 31, 2022;
(3) Only English-language publications were considered;
(4) The subject categories encompassed fields related to the environment and chemistry.

Following these procedures, all duplicate and irrelevant literature was manually excluded. Ultimately, 2038 articles were included in the final analysis.

2.2. Bibliometric analysis

CiteSpace is currently the most commonly used software for bibliometric analysis, suitable for dynamic, complex, and staged analyses (Shi and Liu, 2019). It can present visual results in three modes: "cluster view," "timeline," and "time zone" (Wang and Lu, 2020). VOSviewer (Visualization of Similarities Viewer) allows for the analysis of literature keywords, authors, publication years, institutions, citation information, and more (McAllister et al., 2022). Based on data characteristics, VOSviewer performs network visualization, overlapping cross-visualization,

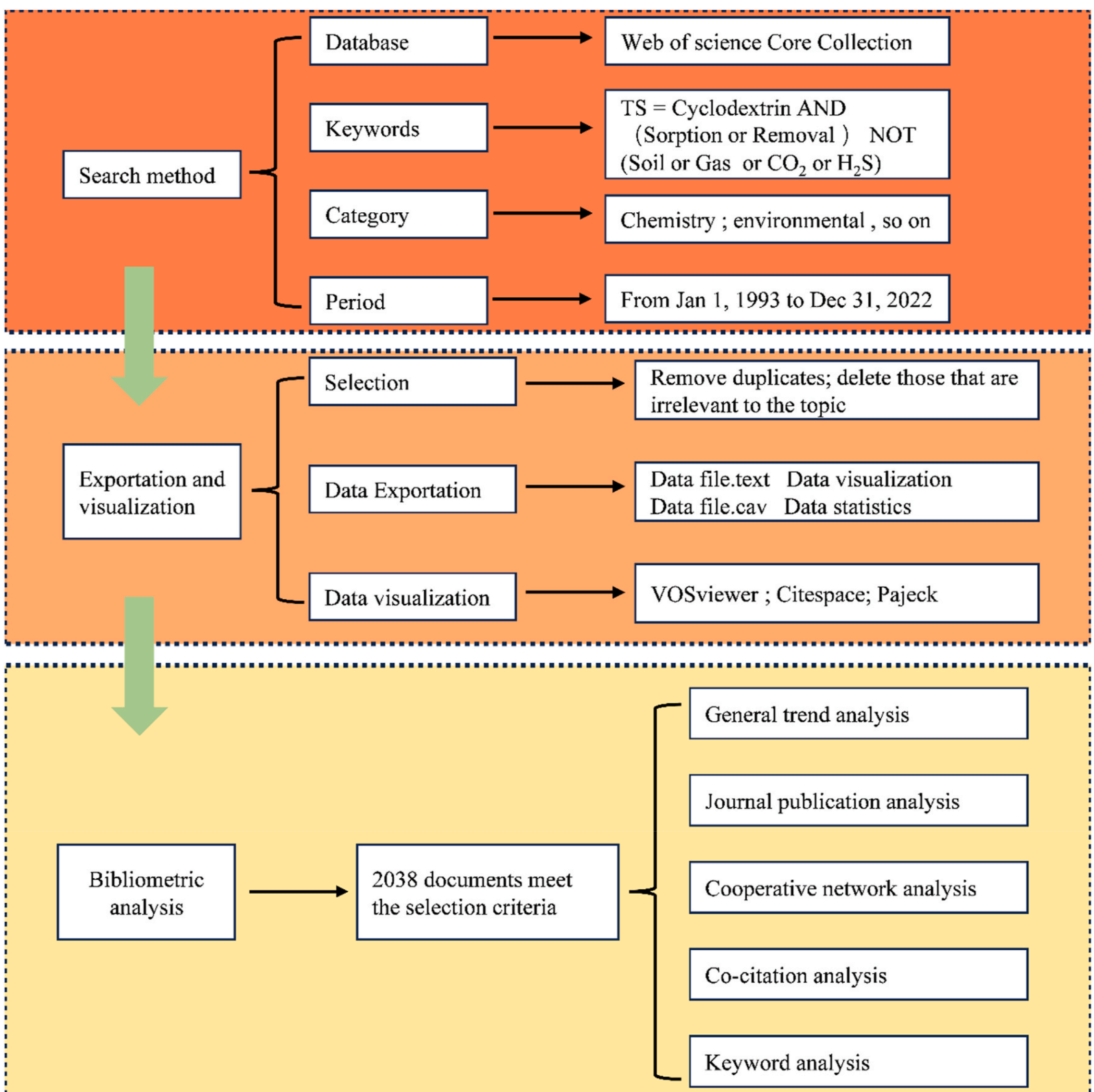

Fig. 2. The workflow for literature metrics analysis in this study.

and density visualization, offering robust graphic representation capabilities for visualizing various knowledge units within the literature, particularly for analyzing large sample datasets (Sood et al., 2021).

In this bibliometric research, VOSviewer (version 1.6.19) and Citespace (version 6.1. R6) were used to analyze and visualize the dataset exported from the WOS (Du et al., 2023). Citation reports were exported from WOS, including statistics on annual publication volume, annual citation count, categories, and source journals. Visualizations were carried out in VOSviewer, including analyses of countries, institutions, subject categories, and source journals' associations (Sun et al., 2023; Pech and Delgado, 2020). In CiteSpace, the study's time frame was set to 1993–01 ~ 2022–12, with a "Year per Slice" parameter of 3, while other settings remained at default values. Centralities analysis, keyword burst analysis, and keyword clustering analysis were performed (Sun et al., 2023). Additionally, this research also utilized software such as *Scimago Graphica* and *Pajek* to optimize and adjust visual outputs generated by VOSviewer (Roldan-Valadez et al., 2019).

3. Results and discussion

This article conducted a bibliometric analysis according to Fig. 2, and the results and discussion are as shown in **Text S1 (**Supporting **information).**

4. Current progress and research trends

In Citespace, the year range is configured to span from 2018 to 2023 with a time interval of 1 year. Keywords and references are selected as the analysis categories, enabling burst analysis and cluster analysis to be conducted on topics, keywords, and cited documents. Upon the analysis, a comprehensive overview is provided, highlighting the latest developments and research focal points in the removal of water pollutants from aquatic systems using CD. Furthermore, the pertinent frameworks and fundamental issues are delineated.

4.1. Application of CD-based adsorbents

4.1.1. Application of CD in the removal of organic micropollutants in water

A recent review has underscored the removal of organic micropollutants, including dyes, phenols, benzene derivatives, polycyclic aromatic hydrocarbons, naphthylamines and pesticides, as a primary challenge in wastewater treatment (Zdarta et al., 2022; Fajal et al., 2023; Obayomi et al., 2022). Among these contaminants, dye molecules were the most studied substances.

The above challenge arises from a substantial body of research demonstrating that conventional wastewater treatment plants cannot wholly eliminate highly toxic and carcinogenic organic micropollutants, leading to their discharge into the environment (Garcia-Becerra and Ortiz, 2018; Luo et al., 2014; Kosek et al., 2020). In response to the concern, activated carbon adsorption has been recently adopted by some European Union countries, and it is likely that more countries will follow suit (Shabtai and Mishael, 2018). However, like other adsorbents, activated carbon rapidly loses its effectiveness in the presence of high organic load and high salinity in wastewater, resulting in a significant decline in performance (Srivastava et al., 2021; Ofiera et al., 2024; Wang et al., 2022). This is due to the fact that in wastewater with a high organic load, a large number of organic molecules will quickly saturate the available adsorption sites on the absorbents (Ofiera et al., 2024). In addition, increasing inorganic salt concentrations mask charged sites on adsorbents, suppressing electrostatic interactions and thus affecting dye adsorption variability (Wang et al., 2022). Besides, carbon reactors are sensitive to the presence of particles in water and clog quickly. Furthermore, the high cost of thermal regeneration for activated carbon may lead to its degradation. Due to its unique nano-scale hydrophobic cavity structure, CD holds promise as the next-generation functional adsorbent (Vunain et al., 2016). This study reviews typical research conducted in recent years on the use of CD to remove organics, as detailed in Table S2.

Current research predominantly focuses on the simultaneous removal of several to dozens of target organic micropollutants (e.g., drugs, cosmetics, phthalates) found in water from urban treatment plants. One study introduced a β-CD polymer that efficiently removes 90 types of organics from water (Ling et al., 2020). However, it is worth noting that multiple studies indicate that the efficiency of CD-based adsorbents in removing organics is contingent upon the affinity of each specific pollutant for the CD polymer (Alzate-Sánchez et al., 2019; Fenyvesi et al., 2020). Therefore, it is imperative to modify CD materials effectively. Encouragingly, there have been pilot cases where CD-based adsorbents have been employed at a pilot scale to eliminate harmful trace pollutants from post-treated wastewater (Fenyvesi et al., 2020). This study demonstrates that even at the pilot scale, CD-based adsorbents can remove selected trace pollutants from wastewater that still contain a variety of other mineral and organic pollutants, some of which are present at higher concentrations. Adsorption results were mainly explained by just two crucial parameters: the presence of a CD cavity and the degree of cross-linking of the CD polymers used in the batch method.

4.1.2. Application of CD in the removal of dye in water

CD-based adsorbents such as cross-linked polymers have a history of several decades in the treatment of dye wastewater, yet this field continues to garner significant attention (Liu et al., 2021a; Yadav et al., 2020). The structure of the dye molecules adapts well to the cavity of the CDs. This is the reason why dyes used by the textile and paper industries, for example, have been extensively studied. The mechanism by which CD-based adsorbents remove dyes depends significantly on the structure of the material used in the batch (Safapour et al., 2022). Previous comprehensive reviews have indicated that the mechanism for CD-based adsorbents in dye removal is complicated, involving hydrophobic complexation, electrostatic interactions, hydrogen bonds, and Yoshida interactions (Liu et al., 2020a; Crini et al., 2022; Chen et al., 2024). In this study (Part 3), the frequency of dye-related terms appearing in keywords was analyzed, reflecting their level of attention. In descending order of frequency, they are methylene blue (125 occurrences), methyl orange (27 occurrences), malachite green (22 occurrences), cationic dye (20 occurrences), azo dye (18 occurrences), anionic dye (16 occurrences), and organic dye (11 occurrences). Methylene blue, in particular, is a representative cationic dye widely used as a chemical indicator, dye, biological stain, and pharmaceutical agent. Its direct discharge without treatment poses a significant threat to water resources, land, and the health of humans, animals, and plants (Qi et al., 2021). Interestingly, even if a dye molecule is too large, it can be complexed by the CD polymers, irrespective of the size of the CD ring. All the results were mainly explained by the network structure of the adsorbents and their swelling properties, closely related to the degree of cross-linking. Many studies conclude that CD materials are particularly suitable for removing dyes.

The impact of several factors (contact time, dose of material to be used, pH of the solution, ionic strength) on the decontamination performance of the materials was widely described, and the results were modelled. Among these factors that influence the adsorption results, the pH of the solutions plays an important role not only in the ionization of the adsorbent but also in the contaminant to complex. For adsorption processes involving electrostatic interactions, when the pH value of the solution changes, the surface properties of the adsorbent and contaminants may change, which in turn affects the electrostatic interaction between the adsorbent and pollutants (Shi et al., 2022). In addition, the character of contaminants and the functional groups of adsorbents may change with changes in solution pH, thereby affecting the complexation process (Liu et al., 2020a; Aigbe et al., 2021). The pH value is a critical variable and an essential factor influencing the adsorption of dyes by CDs (Shi et al., 2022; Aigbe et al., 2021). Therefore, in certain situations, the selectivity of the adsorbent for dyes can be adjusted by altering the pH of the solution (Cai et al., 2020). The point of zero charge (pHpzc) is a fundamental parameter (Zhou et al., 2018; Jia et al., 2020) but is rarely reported in the literature. When the pH is greater than the pHpzc, the adsorbent carries a negative charge, facilitating the adsorption of cationic dyes (Liu et al., 2020a; Moulahcene et al., 2023).

Conversely, when the pH is lower than the pHpzc, the adsorption of anionic dyes becomes viable as the adsorbent surface is positively charged. Table 1 presents the optimal adsorption capacity and pH values for different types of CD-based adsorbents regarding dyes. From this table, it is evident that the optimal pH values vary within a relatively wide range. Additionally, in contrast to electrostatic interactions and cavity inclusion, complexation for dye removal necessitates an optimal pH value below 7.0 (Liu et al., 2020a). It can be explained by the fact that dye removal via the complexation between dye molecules and amino groups that can accept protons under acidic conditions on CD adsorbents is more efficient (Liu et al., 2020a). However, this strongly depends on the kind of material used. Another point must be noted: interestingly, as CD-based polymers do not alter the pH of the effluents, it is not necessary to maintain the initial pH of the waters during tests. From an industrial point of view, this fact is essential.

Among the materials, cross-linked polymers and graphene-based nanocomposites with CD have been extensively employed for dye adsorption (Liu et al., 2014; Tan and Hu, 2017; Li et al., 2018), with their adsorption mechanism depicted in Fig. 3. One study developed a three-dimensional porous structure of β-CD-oxidized graphene aerogel, which exhibited excellent adsorption capabilities for dyes compared to traditional adsorbents. For anionic dyes, it achieved adsorption capacities of 439 mg/g for methylene blue and 388 mg/g for Rhodamine B, and for cationic dyes, it achieved adsorption capacities of 234 mg/g for acid red 87 and 167 mg/g for methyl orange (Nie et al., 2021). Even after five cycles, the adsorption capacity remained above 80% (Nie et al., 2021). Regardless of the structure of the molecule and its ionic character, CD materials have strong adsorption capabilities. Moreover, to facilitate the rapid separation of adsorbents dispersed in water, another study developed magnetic β-CD@oxidized graphene composite materials. These materials achieved removal efficiencies exceeding 95% for 50 mg/L of methylene blue, Congo red, rhodamine B, and malachite green (Li et al., 2021a). Even after eight adsorption-desorption cycles, the removal rates for methylene blue and Congo red remained at 95.0% and 85.0%, respectively. The ability to regenerate and reuse materials is often cited in publications.

4.1.3. *Application of CD in the removal of metal ions in water*

It is well-established that CDs can also effectively remove minerals such as metals from aqueous solutions through electrostatic interactions, complexation, coordination, or ion exchange mechanisms (Morin-Crini et al., 2018). Through the utilization of WOS in conjunction with CiteSpace to analyze keyword frequency, it has been observed that the order of metal cations adsorbed by CDs is as follows: Pb(II) > Cr(VI) > Cu(II) > Cd(II) > U(VI). Recent reviews have also corroborated the effectiveness of CDs as efficient adsorbents for metal cations, boron, and fluorides (Tian et al., 2021; Liu et al., 2020a; Goyal et al., 2023).

Current research emphasizes the simultaneous removal of several pollutants from controlled solutions or complex mixtures, owing to the distinct adsorption mechanisms of various types of pollutants within CD adsorbents. For example, one study synthesized a β-CD/polyethyleneimine bi-functional polymer capable of concurrently removing hydroquinone and Pb(II) from water (Xu et al., 2022). In this case, electrostatic and coordination interactions are responsible for Pb(II) adsorption, while CD's inclusion and hydrogen bonding interactions govern the adsorption of hydroquinone. Another instance involves the environmentally friendly preparation of a novel β-CD/ZrO_2 nanocomposite material, which efficiently removes Pb(II) and BPA from water. Oxygen-containing groups are primarily responsible for binding Pb(II), while the β-CD cavity adsorbs BPA through host-guest

Table 1
The optimal adsorption capacity (Q_{max} in mg/g) and pH values for different types of CD-based adsorbents regarding the complexation of various dye molecules.

Adsorbent	Dye	Q_{max} (mg/g)	pH value	Ref.
PDA/β-CD	methylene blue	106.8	6	Wang et al. (2020a)
PDA/β-CD	neutral red	51.7	6	Wang et al. (2020a)
PDA/β-CD/GO/Fe_3O_4–NH_2	methylene blue	75.0	10	Hu et al. (2023)
PDA/β-CD/GO/Fe_3O_4–NH_2	Congo red	104.0	7	Hu et al. (2023)
CDP-EA	methyl orange	290.0	2.5	Shi et al. (2022)
CDP-EA	Congo red	1250.0	2.5	Shi et al. (2022)
β-CD/chitosan	indigo Carmine	1000.0	3	Kekes and Tzia (2020)
β-CD-BPDA	methylene blue	96.2	3–11	Lagiewka et al. (2023)
PA-β-CD	methylene blue	1095.0	6~10	Li et al. (2022)
PA-β-CD	basic green 4	2005.6	6~10	Li et al. (2022)
PA-β-CD	astrazon pink	1736.3	6~10	Li et al. (2022)
PA-β-CD	crystal violet	1930.2	6~10	Li et al. (2022)
Ce/CCD	reactive blue 4	29.6	3	Sirajudheen et al. (2020)
Ce/CCD	indigo carmine	30.6	3	Sirajudheen et al. (2020)
Ce/CCD	acid blue 158	30.6	3	Sirajudheen et al. (2020)
CD-CA/PDA	methylene blue	583.0	6–12	Chen et al. (2020b)
CD-CA/PDA	malachite green	1174.7	6–12	Chen et al. (2020b)
CD-CA/PDA	crystal violet	473.0	6–12	Chen et al. (2020b)
CD/CA-g-PDMAEMA	methylene blue	165.8	4	Zhou et al. (2018)
CD/CA-g-PDMAEMA	methyl orange	335.5	11	Zhou et al. (2018)
CDNS	basic red 46	101.4	5	Li et al. (2020)
CDNS	rhodamine B	52.3	6~8	Li et al. (2020)
Cl-CD/CA	methylene blue	1523.6	12	Morin-Crini et al. (2018)
Cl-CD/CA	crystal violet	1012.2	6–12	Morin-Crini et al. (2018)
Cl-CD/CA	neutral red	419.2	3–12	Morin-Crini et al. (2018)
TiO_2/Gly/β -CD NPs	methylene blue	82.0	8	Li et al. (2020)
TiO_2/Gly/β -CD NPs	acid blue 113	76.9	8	Li et al. (2020)
TiO_2/Gly/β -CD NPs	methyl orange	384.6	5	Li et al. (2020)
TiO_2/Gly/β -CD NPs	disperse red 1	138.9	9	Li et al. (2020)
TiO_2/γ-CD	methylene blue	134.0	9	Mousavi et al. (2019)
TiO_2/γ-CD	malachite green	244.0	7	Mousavi et al. (2019)
TiO_2/γ-CD	crystal violet	213.0	9	Mousavi et al. (2019)
TiO2/γ-CD	disperse red 1	238.0	9	Mousavi et al. (2019)
TiO_2/γ-CD	acid blue 113	157.0	9	Mousavi et al. (2019)
TiO_2/γ-CD	Congo red	5000.0	4	Mousavi et al. (2019)
RGO-β CD-ECH	malachite green	902.1	8	Rout and Jena (2022)
CM-β-CDP	methylene blue	1030.9	10	Liu et al. (2020b)
CDP-DEA	Congo red	813 0.0	5	Xu et al. (2020)
CDP-DEA	orange G	442.0	3	Xu et al. (2020)
β-CD	methylene blue	47.4	8	Usman et al. (2021)

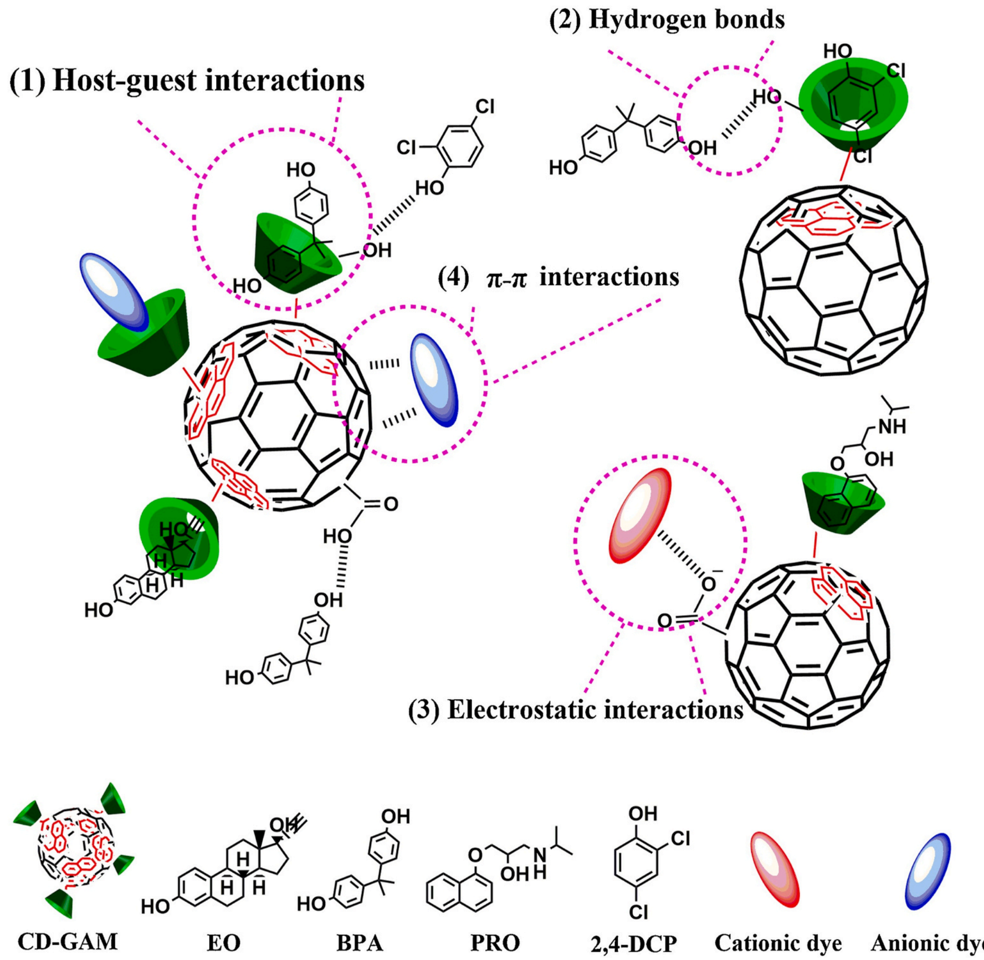

Fig. 3. Schematic illustrating the adsorption mechanisms of CD-graphene oxide aerogel microspheres for dyes and organic micropollutants (Nie et al., 2021).

interactions (Usman et al., 2021). A similar ability to simultaneously remove Pb(II) and BPA from water using modified materials was exemplified in Liu et al.'s work utilizing magnetic cellulose fibers (Liu et al., 2022). Research on competitive effects during the adsorption process is also a focus of current studies on CD's adsorption of metal ions (Morin-Crini et al., 2018). A typical study utilized β-CD functionalized diatomite (D) as an environmental adsorbent to adsorb Cs^+ and Ba^{2+} ions from water (El-Sherbeeny et al., 2021). The study reported that the efficiency of β-CD/D during Cs^+ and Ba^{2+} removal was only slightly affected by the presence of anions (HCO_3^-, SO_4^{2-}, NO_3^-, and PO_4^{3-}) but was significantly impacted by the presence of other metals (Cd^{2+}, Pb^{2+}, Zn^{2+}, and Co^{2+}). Additionally, multiple studies suggest CD adsorbents are capable of effectively removing various pollutants (metals, bisphenol A, cationic dyes, atrazine) simultaneously (Chen et al., 2020b; Usman et al., 2021; Liu et al., 2022; Qu et al., 2020; Qu et al., 2021). This result was attributed to different adsorption mechanisms acting simultaneously (Usman et al., 2021). To conclude, all these works highlight the polyfunctionality of CD materials capable of interacting with both mineral, metallic, and organic contaminants.

4.2. Development of new CD-based materials

With a hydrophobic internal cavity combined with hydrophilic external properties, cyclodextrin molecules can also combine with a variety of other materials, such as polysaccharides (cellulose, chitosan), nanoparticles, fibres, graphene, carbon nanotubes, covalent organic frameworks (COFs), metal-organic frameworks (MOFs) and molecularly imprinted polymers (MIPs), to produce new intelligent macromolecular platforms for applications not only in water and wastewater treatment but also in pharmacy, medicine, textiles, foods and sensors (Namgung et al., 2014; Costoya et al., 2017; Nafee et al., 2015; Feng et al., 2021). So, several research groups have opened the way for future progress through the application of CD-based materials to physical and organic chemistry, material design, and water engineering. Among proposed materials, graphene oxide materials containing cyclodextrins deserve particular attention due to their excellent adsorption properties because of their bounteous structure and rich oxygen functional groups combined with the presence of the CD cavity.

4.2.1. Development of CD/GO composite materials

Over the past decade, the utilization of carbon-based materials such as graphene oxide (GO) in wastewater treatment has garnered extensive attention due to their large specific surface area and abundant surface functional groups, rendering them excellent adsorbents (Soffian et al., 2022; Nasrollahzadeh et al., 2021). However, the adsorption capacity of pristine GO is limited, underscoring the crucial significance of its modification to broaden its practical applications (Li et al., 2021b). Therefore, the modification of GO is of great significance for expanding its practical application. The cyclic structure and hydroxyl-rich nature of CDs endow them with the capability to encapsulate pollutants (Nie et al., 2021). Consequently, CDs emerge as an ideal choice for enhancing the adsorption capacity of GO through modification (Nie et al., 2021; Wu et al., 2021). The surface of GO contains a large and reactive number of hydroxyl, carboxyl and epoxide functional groups on its skeleton, facilitating its modification by CD molecules. A previous review has indicated that CD-grafted oxidized graphene can be synthesized using various techniques, including chemical precipitation, sonochemical methods, and layer-by-layer assembly (Kumari et al., 2020). However, changing the structure of GO is a challenge because any modification can cause a dramatic chemical change in its base properties. GO particles exhibit small particle sizes, making their separation challenging, and their environmental exposure poses potential health and environmental concerns (Jiang et al., 2020; Hu et al., 2023). Consequently, magnetic β-CD/GO composite materials are currently being widely developed.

The preparation of β-CD/GO composites via chemical precipitation is considered an economical, effective, and environmentally friendly method. As shown in Fig. 4a, Ma et al. introduced β-CD into an alkaline solution (solution A) and dispersed graphene oxide (GO) in a solution containing Fe(II) and Fe(III) under ultrasonic conditions (solution B) (Ma et al., 2018). Subsequently, solution B was added dropwise to solution A to facilitate the reaction, resulting in the collection of the precipitate (β-CD/MGO) (Ma et al., 2018). It can be seen that β-CD/MGO nanohybrids exhibited maximum adsorption capacities of 279.21 mg/g for Pb(II), 51.29 mg/g for Cu(II), and 93.97 mg/g for methylene blue, underscoring the economic effectiveness of β-CD/MGO in practical applications. This result can be explained by the presence of electrostatic interaction between oxygen functional groups and adsorbents (partly from van der Waals force), by the interaction between aromatic rings and GO structures of dye molecules (Π-Π interactions), and by the formation of inclusion complex owing to the interaction between organic dyes and CD molecules. Some research has also proposed a physical sieving mechanism controlled by the interlayer spacing of CD-GO material, mainly when membranes are used.

The sonochemical method is a technique that accelerates chemical reactions through the application of mechanical waves, thereby enhancing chemical yields, occasionally accompanied by chemical precipitation (Hachem et al., 2022). In a study, a mixture of GO, β-CD, and $Cu(NO_3)_2{\cdot}3H_2O$ was dispersed in water through ultrasonication (Yakout et al., 2021). Subsequently, $NaBH_4$ was added dropwise to the mixture described earlier, and the black solid was separated via centrifugation. This process chemically reduced GO to obtain β-CD/rGO nanocomposite material. It is noteworthy that β-CD/rGO exhibited maximum adsorption capacities for tetracycline, oxytetracycline, and streptomycin of 403.2 mg/g, 476.2 mg/g, and 434.8 mg/g, respectively. Even after five cycles of adsorption-desorption, the removal efficiency only decreased by 3.7%, indicating the potential practical application of β-CD/rGO in actual wastewater remediation.

The application of layer-by-layer assembly for pollutant removal is primarily exemplified by molecular imprinting (Liu et al., 2020a). It has been reported that the adsorption process of 2D GO is hampered by its aggregation tendencies and challenges associated with separation and recovery, thus constraining its utility (Liu et al., 2018; Geim, 2009). A recent study has introduced a 3D graphene-based aerogel, denoted as γ-GO/CD aerogel, which is meticulously assembled layer by layer from

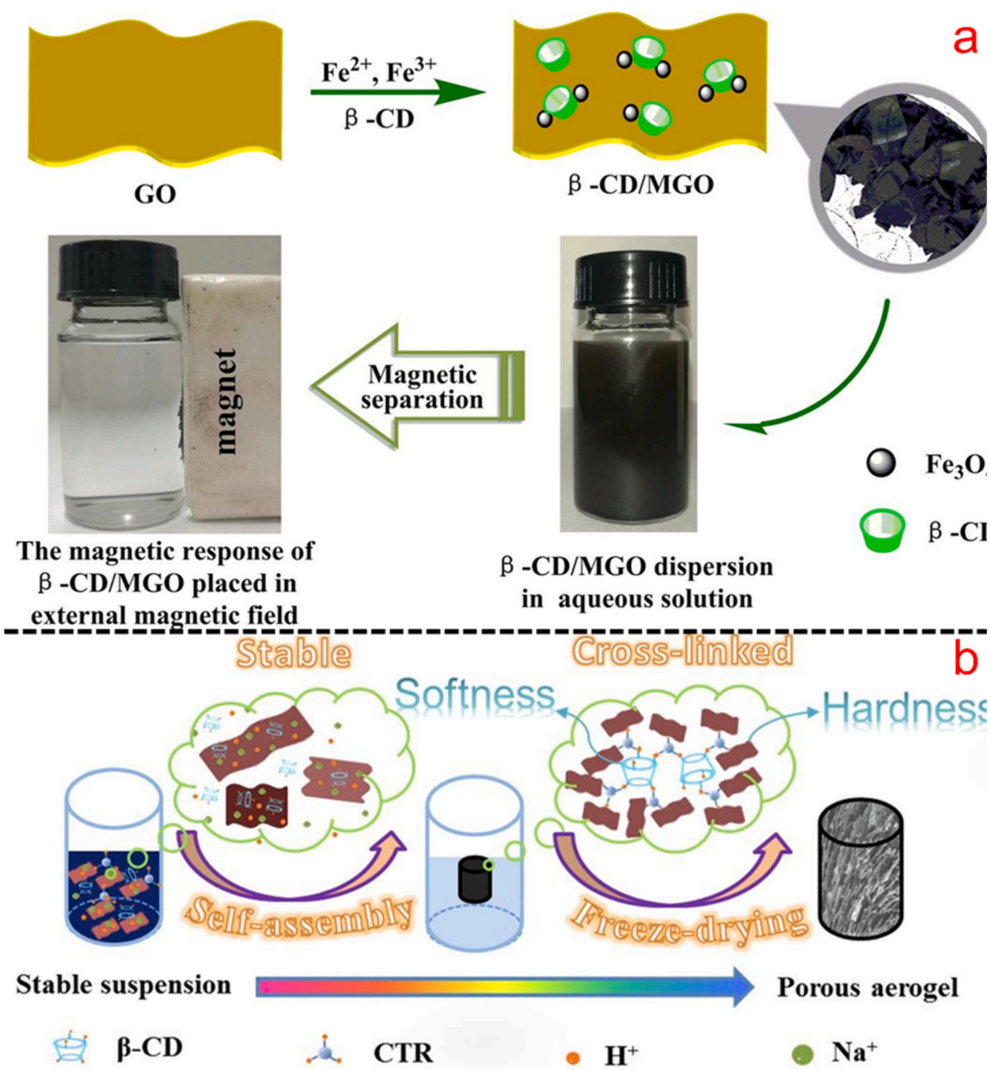

Fig. 4. Schematic of CD/GO synthesis methods (a: chemical precipitation; b: layer-by-layer assembly) (Ma et al., 2018; Sun et al., 2019).

two-dimensional nanosheets (Fig. 4b) (Sun et al., 2019). The incorporation of soft β-CD within the hard GO nanosheets, concurrently with the self-assembly process of the aerogel, has significantly contributed to the formation of a structurally superior and stable r-GO-CD aerogel (Sun et al., 2019). The adsorption efficiency of this material for bisphenol A is approximately 30 times greater than that of γ-GO.

4.2.2. Development of CD cross-linked polymers

The literature data does show that CDs can be incorporated into graphitic structures. However, CD polymers remain the way to go due to better control during syntheses, resulting in systems with higher specific surfaces, controlled porosity, and the possibility of grafting ligands that increase the selectivity of materials. Indeed, commercial CD-epichlorohydrin polymers have been known for half a century as adsorbents to eliminate various contaminants (Morin-Crini et al., 2018; Morin-Crini and Crini, 2013; Gidwani and Vyas, 2014; Crini, 2021; Landy et al., 2012; Liu et al., 2021b; Liu et al., 2020c). These cross-linked polymers are easily obtained by copolymerization of CD molecules and epichlorohydrin as a coupling agent in an alkaline medium. There is no doubt that among the various CD materials, CD cross-linked polymers are the most widely studied adsorbents, and an extremely large number of articles have been published. All the studies showed that cross-linked polymers were capable of simultaneously removing both organic and inorganic contaminants from water and wastewater to desirable levels. Even at low concentrations and in the presence of high salinity, the CD materials were efficient in contaminant removal because of their high adsorption properties and selectivity. Numerous researchers also pointed out other advantages, such as the low cost of the materials, easy recyclability, regeneration, or disposal (incineration) (Fenyvesi et al., 2020; Alsbaiee et al., 2016; Fenyvesi and Sohajda, 2022). In addition, the action mechanisms are now well-known (Morin-Crini et al., 2018). However, CD polymers are still basically at the laboratory-scale work stage, mainly due to their higher cost compared to conventional activated carbon. In addition, they will probably never replace carbons, but these materials could help improve the capture of target molecules (e.g., the beta-blocker propranolol or the carcinogen herbicide 2,4-dichlorophenol). Another advantage of CD polymers is the ease of their regeneration compared to that of carbons, which requires a high energy input (up to 1000 °C of heating in an inert atmosphere) if the path of regeneration is privileged. In practice, once saturated, the carbons are changed and disposed of by incineration.

4.2.3. Development of CD-based membranes

Powdered CD-based adsorbents suffer from slow pollutant removal rates due to limited intraparticle diffusion. Furthermore, the recovery of used CD adsorbents necessitates additional filtration steps, incurring energy consumption (Xiao et al., 2014). Additionally, adsorption is commonly conducted in a batch mode, hindering continuous operations (Xiao et al., 2014; Liu et al., 2020d). Moreover, fixed-bed adsorption encounters the drawback of compaction, requiring higher pressure for increased permeability (Zhao et al., 2022; Lu et al., 2022). To minimize energy and time consumption, incorporating nano-scale adsorbents into porous membranes is imperative, thus enabling the development of a continuous and energy-efficient water purification process (Wang et al., 2019a; Wang et al., 2020b).

Benefiting from the intrinsic structure of CD molecules and the complexation characteristic of their inclusion complexes, CDs are used in chromatography, for example, to recognize isomers. In general, the chiral separation membrane prepared with CDs shows higher permselectivity and stability compared with other chiral selectors. So, these CD-based membranes have been proposed in the field of water treatment membranes. They can be used for drinking water and removal of emerging contaminants, and the results published confirm that these new systems display improved performances in terms of selectivity, rejection, permeation, and flux with reduced fouling propensities.

Polyvinylidene fluoride (PVDF) is a commonly used membrane material due to its processability, chemical resistance, wear resistance, high thermal stability resulting from the highly electronegative fluorine atoms and carbon-fluorine solid bonds, excellent thermal-mechanical properties, and chemical compatibility compared to other polymeric materials (Lu et al., 2022; Chen et al., 2022). Chen et al. developed a novel hydrophilic PVDF composite membrane (β-CD@ZIF-8/PVDF) via a deep infiltration method, using β-CD and zeolitic imidazolate framework-8 (ZIF-8) nanoparticles, as depicted in Fig. 5a (Chen et al., 2022). The saturation adsorption capacities of β-CD@ZIF-8/PVDF micro-membrane adsorbents for Pb^{2+} and Cu^{2+} were found to be 708.130 mg/g and 651.379 mg/g, respectively, demonstrating significant industrial applicability. It is attributed to enhanced contact within enclosed spaces and the provision of more active sites within the membrane pores. In another study, a membrane was prepared through simple solution mixing of β-CD and PVDF, followed by membrane casting, as illustrated in Fig. 5b (Wang et al., 2019a). It not only effectively removes individual organic micro-pollutants but is also capable of complete removal of mixtures, along with good adsorption regeneration capacity.

Biomass-based electrospun nanofiber membranes, such as cellulose-based, chitosan-based, and lignin-based membranes, are considered ideal materials for water pollution control due to their low cost, environmental friendliness and ease of modification (Chabalala et al., 2021; Teng et al., 2011; Fan et al., 2019; Zhao et al., 2015). Numerous studies have shown that introducing functional materials into biomass-based materials is an effective way to enhance adsorption capabilities (Chabalala et al., 2021; Fan et al., 2019). Lv et al. prepared a porous β-CD-modified cellulose nanofiber membrane (CA-P-CDP) for the treatment of trace bisphenol contaminants in water. The resulting CA-P-CDP exhibited a porous structure, stable crystalline structure, good thermal stability, and abundant functional groups, significantly enhancing the adsorption and recovery of bisphenol pollutants (Lv et al., 2021). Another study involved the synthesis of a novel electrospun β-CD/CS/PVA nanofiber membrane for the rapid removal of organic micro-pollutants and heavy metal ions from water simultaneously (Fan et al., 2019). Overall, the β-CD/CS/PVA nanofiber membrane can efficiently remove organic micro-pollutants and heavy metal micro-pollutants from river water, offering substantial potential in treating large volumes of drinking water or wastewater.

4.2.4. Development of CD@TiO_2 composite materials

Another area that attracts the attention of researchers is photocatalytic technology. Photocatalysis, regarded as an emerging green approach, has found widespread application in the remediation of environmental pollutants (Deng et al., 2021; Gopinath et al., 2020). Nevertheless, its efficiency is significantly impacted by the recombination of electrons and holes (Mousavi and Mohammadi, 2018; García-Díaz et al., 2020; Zhu et al., 2018). Prior research has illuminated that bolstering the interaction between pollutants and the catalyst, coupled with the enhancement of the transference of charge carriers at the interface, contributes to the augmentation of photocatalytic efficiency (Chen et al., 2018; Zhang et al., 2018). Furthermore, the diminished adsorption capacity of TiO_2 for hydrophobic pollutants substantially restricts its utility in the control of such contaminants (Yadav et al., 2022). CDs function as reservoirs for retaining pollutants within their cavities, effectively serving as a conduit between organic pollutants and photocatalysts.

The exorbitant cost impedes the application of noble metal-modified photocatalytic nanomaterials. The proportion of CDs within composite materials significantly influences adsorption efficiency (Wang et al., 2019b). Research has seen the synthesis of carboxymethyl-β-CD-modified titanium dioxide as a noble metal-free catalyst for the efficient removal of organic pollutants (Zhou et al., 2020). This study compared the photoelectrochemical properties and photocatalytic activity of CM-β-CD-P25 and Au–P25. Notably, CM-β-CD-P25 (2:1) hybrid nanoparticles exhibited exceptional photoelectrochemical

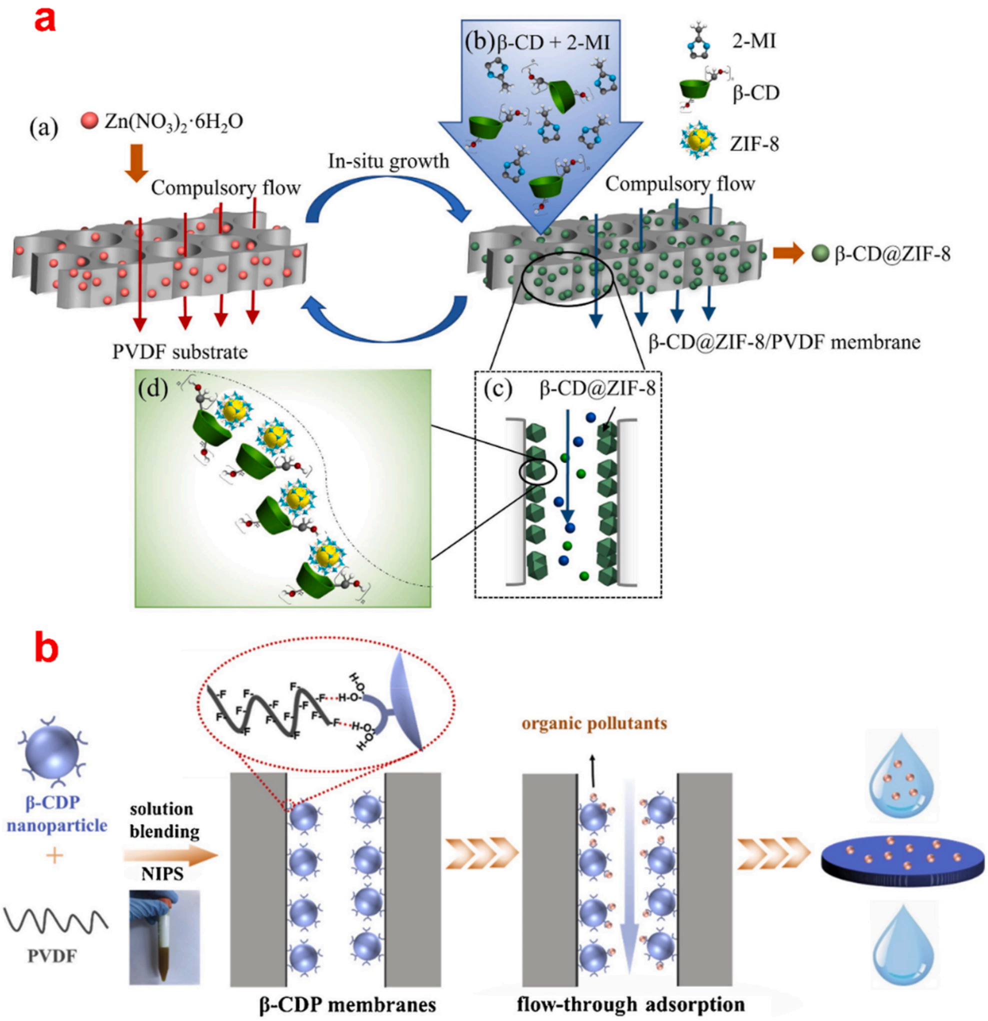

Fig. 5. Schematic of CD/PVDF synthesis membrane material (a: β-CD@ZIF-8/PVDF; b: β-CD/PVDF) (Wang et al., 2019a; Chen et al., 2022).

properties and outperformed gold-modified TiO_2 (Au–P25) in terms of the removal of BPA, phenol, and sulfonamides.

Catalyst stability and reusability are also integral considerations (Zhang et al., 2021). Research efforts have resulted in the preparation of Fe–N co-doped TiO_2/β-CD materials, which can efficiently eliminate organic dyes through photocatalysis and maintain high stability after five successive usages (Mottola et al., 2022). Another study introduced TiO_2@ACD@RGO, demonstrating significantly higher photocatalytic BPA degradation activity under ultraviolet irradiation compared to pure TiO_2 and TiO_2@RGO (1.87 and 1.34 times, respectively) (Mottola et al., 2022). Impressively, even after five repeated cycles, it exhibited outstanding stability and reusability. Wang et al. synthesized CD-Ni/-TiO_2 via a one-step method, showcasing its superior degradation performance for hydrophobic molecules and sustaining stability over five consecutive cycles (Wang et al., 2020c). The main disadvantages of these materials are the difficulties of synthesis and their price.

5. Current challenges and future development trends

5.1. Current challenges

Despite numerous previous works, some limitations persist in building upon the overview mentioned above of the hotspots in CD-based adsorption of pollutants in water and wastewater:

(1) Industrial production: There are no industrial sectors for the production of CD-based materials, with the exception of cross-linked polymers, of which a few industries are capable of supplying large quantities. Start-ups have recently been created on the use of CD polymers for complex emerging pollutants. These polymers are indeed able to eliminate drugs, cosmetics, and fluorinated pollutants, which suggests opportunities for development.

(2) Cost issue: One key limiting factor for the industrial application of CD-based adsorbents is their cost in comparison to commercially available activated carbon. These materials are more expensive than commercial activated carbon and organic resins. In addition, the quantities to be used in the water treatment processes are also higher than those of conventional, which does not encourage industry to turn to CD materials. However, they have an advantage in terms of water engineering. Indeed, most CD-based adsorbents are easily regenerable under mild conditions, making them more cost-effective and profitable in the long run. At the same time, activated carbon is typically used as a one-time consumable that must be replaced after saturation because its regeneration is costly. The state of the bibliographic art shows that recent efforts have been made to produce low-cost CD-based hydrogels with significantly lower synthesis costs than traditional materials by implementing green chemistry processes. However, all studies are still in the laboratory stage.

(3) Selectivity: With the ease of regeneration, selectivity is another advantage often cited in the literature. The selectivity of CD-based adsorbents refers to their adsorption capacity for specific pollutants relative to their affinity or adsorption preference for other substances. Even in the presence of high water salinity, materials are able to complex trace substances in complex aqueous matrices. Indeed, the advantage of this selectivity is the targeted removal of specific pollutants in polycontaminated effluents, but it also poses a drawback in terms of efficiently removing multiple pollutants simultaneously. Therefore, specific modifications are necessary to enhance their adsorption efficiency when targeting the efficient removal of dozens or even hundreds of pollutants simultaneously.
(4) Engineering application: Recent and increasing results show that CD polymers are capable of treating actual water. However, as already mentioned, most of the studies published so far have been small-scale laboratory experiments designed to demonstrate the applicability of CD adsorbents. In reality, there is a significant gap between experimental testing and engineering application, and results obtained in experiments may not necessarily translate correctly to real-world applications. Therefore, it is essential to continue to demonstrate the applicability of CD adsorbents for actual water and wastewater on a larger scale and to couple the chemical efficiency obtained with a potential decrease in the impact of water in terms of toxicity. Moreover, more and more works are published, pooling the skills of chemists and biologists.

5.2. Future research directions

Considering the current research landscape and the challenges mentioned above, future endeavors in the realm of CD-based adsorbents should concentrate on the following aspects:

(1) Price of the materials: The replacement of a mature technology by another innovative solution implies that the latter ultimately brings a performance gain for an equal or even lower cost. For most applications, unfortunately, paying more for increased performance is a luxury. CD-based materials remain expensive for now.
(2) Development of low-cost composite materials with high efficiency and regeneration: This involves altering the structure or introducing functional groups to enhance the adsorption performance of CD-based materials. Exploring cost-effective CD-based composite materials and methods is essential. Additionally, efforts should be directed toward the development of efficient recovery and regeneration techniques to minimize the loss of CD-based adsorbents.
(3) Application of artificial intelligence (AI) in CD-based adsorbents: Machine learning models can learn complex relationships from extensive data, enabling rapid prediction and analysis. Machine learning predictions for CD-based adsorbents can reduce experimental costs and enhance research and production efficiency. Furthermore, AI models can be employed to select CD-based materials for adsorbing specific pollutants. Lastly, various aspects of CD-based adsorbent production processes can be enhanced through AI optimization.
(4) Practical engineering applications of CD-Based materials: Currently, most research on CD-based adsorption materials are primarily conducted in laboratory settings and has yielded promising results. However, real-life and industrial wastewater situations tend to be more complex, making the performance more challenging. Hence, future research should involve large-scale testing under laboratory conditions that replicate actual wastewater systems.
(5) Post-Treatment considerations: After CD-based adsorbents have adsorbed organic pollutants (dyes, pesticides), metals, or emerging pollutants from water or wastewater, it is imperative to assess the cost and environmental implications of post-treatment thoroughly. This evaluation is vital in terms of economic feasibility and environmental impact. The life cycle of materials is a concept that must also be taken into account in future research and development. Few data are accessible on this point.

6. Conclusion

Although the evolution of scientific production leaves little doubt about this, as this bibliometric study shows, this is the first work providing a thorough evaluation and classification of CD-based material for contaminant removal. Indeed, this study represents the first application of bibliometrics to analyze the research progress on this theme. The analysis encompasses 2038 papers published on the WOS between 1993 and 2022. The findings reveal a steady annual increase in the number of publications and citations in this field, with an estimated annual citation count reaching 36,710 by 2030. Unsurprisingly, CD-based materials are of growing interest, stimulated not only by the intrinsic properties of CD molecules but also by the exceptional performance of their derivatives used as adsorbents.

Notably, China, France, Canada, and USA are the leading contributors in terms of publications. In terms of institutions, some of the most relevant contributors are the Chinese Academy of Sciences (the most prolific institution), the University of Franche-Comté, the University of Johannesburg, the University of Saskatchewan, the Sichuan University, and the University of Malaya, to name a few. Among the researchers, Crini G from the University of Franche-Comté, Wilson LD from the University of Saskatchewan, and Wang Y from the Dalian University of Technology have the highest number of publications with the highest citation. The primary publications have appeared in journals such as "Carbohydrate Polymers" and the "Journal of Hazardous Materials," which gives an idea of the impact of the research. This also reflects an interdisciplinary trend spanning the fields of Physics, Chemistry, Polymer Science, Materials Science, Chemical Engineering, and Environmental Science.

The analysis of the keywords indicates the main actual areas of research being conducted: (1) fabrication and characterization of new CD-based architectures, (2) control and analytical monitoring of water before and after use of these materials to highlight the interest of the process, (3) application of CD adsorbents to treat actual water in pilot-scale tests.

The current research focus predominantly revolves around the application of CD-based adsorption materials for the removal of organics and metals. Most works focus on the study of environmental concerns concerning water parameters of a qualitative and quantitative nature. Additionally, research efforts are concentrated on the synthesis of composites containing CD molecules (MOFs, MIPs), CD@GO, CD-based membranes, and CD@TiO_2 materials. Despite significant advancements in CD-based adsorbents, this field faces notable challenges that necessitate resolution, including cost issues, adsorption selectivity, and engineering applications. The results of this study provide valuable insights for CD product manufacturers, researchers, environmental management companies, and policymakers.

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CRediT authorship contribution statement

Chong Liu: Writing – review & editing, Writing – original draft, Methodology, Conceptualization. **Grégorio Crini:** Writing – review & editing, Writing – original draft, Supervision. **Lee D. Wilson:** Writing –

review & editing. **Paramasivan Balasubramanian:** Writing – review & editing. **Fayong Li:** Writing – review & editing, Writing – original draft.

Declaration of competing interest

The authors 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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2024.123815.

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