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Review Article

Adsorptive separation and influencing factors of bisphenols from environmental matrices using silica-based adsorbents: A review

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Abstract

Bisphenol A (BPA) and its analogues are extensively used in the plastic manufacturing industry and are recognized globally as endocrine-disrupting chemicals. Due to their relatively similar chemical structures, BPA analogues exhibit comparable or even greater toxic effects. These bisphenols have raised significant concerns among the scientific community and environmentalists, and their potential impacts require further investigation and evaluation. Effective techniques for the removal of these emerging contaminants are urgently needed. This review presents a comprehensive and critical summary of recent advancements in silica-based adsorbents for the removal of emerging bisphenols, covering developments from 2010 to the most recently published studies. Silica has played an important role as a sorbent in adsorption technologies due to its unique physicochemical properties, including high surface area, tuneable pore structure, and thermal stability. In this review, the surface modification and functionalization strategies aimed at enhancing adsorption capacity and selectivity are elaborated. Key variables, comparisons between different silica adsorbents, and adsorption mechanisms are discussed in detail. This review serves as a valuable reference for the development of efficient, low-cost, and selective adsorbents for the removal of bisphenols and other emerging contaminants.

Keywords: adsorption, bisphenols, silica, water pollutant removal, safe water access

Introduction

Bisphenol A (BPA) has been classified as an emerging contaminant [1] and ranks among the chemicals with the highest production and consumption volumes [2]. In response to increasing global restrictions on BPA in food-contact materials, the plastic and canning industries are actively searching for alternative substitutes [3]. Bisphenol A and its analogues—such as bisphenol F (BPF), bisphenol B (BPB), bisphenol Z (BPZ), bisphenol AP (BPAP), bisphenol AF (BPAF), bisphenol S (BPS), as well as bisphenol P (BPP)—are a class of similarly structured chemicals that consist of two phenolic rings joined through a bridging carbon or other chemical structures [4].

Numerous studies have reported that these analogues are widely detected in various environmental and biological samples [5]. Unfortunately, recent toxicological studies have revealed that some BPA analogues exhibit similar or even greater toxicity than BPA itself [6]. Among the bisphenols, BPS, BPF, and BPAF are the main substitutes used in the production of epoxy resins and polycarbonate plastics [7]. BPS (Bis(4-hydroxyphenyl) sulfone) is more resistant to heat and sunlight when compared to BPA, and the biodegradability of bisphenol analogues in seawater was ranked in the following order: BPF, BPA, and BPS [8]. BPF (2,2-bis(4-hydroxyphenyl)methane) on the other hand is increasingly used due to its low viscosity and better resistance to solvents than BPA epoxy resins [9] while BPAF (2,2-bis(4-hydroxyphenyl)-hexafluoro propane) is a fluorinated derivative of BPA in which the two methyl groups are replaced with trifluoromethyl groups and it is used as a monomer to produce fluoropolymers and fluoroelastomers, specialty polymers such as polyesters, polyamides, and polyimides [10].

Multiple studies have confirmed that bisphenols are present in various environmental media, including soil, water, biomass, sediment, and air [11]. Water bodies can become contaminated with bisphenols and their degradation products through atmospheric deposition, industrial wastewater, sewage, and runoff, all of which occur during the production, use, aging, and disposal of bisphenol-containing products [12]. It has been well-documented that bisphenols can disrupt endocrine function and cause adverse health effects in humans through both dietary and non-dietary exposure [7]. The chemical structures and physicochemical properties of BPA and its analogues are similar, as shown in **Table 1** [10]. BPA exhibits high hydrophobicity and lipid permeability at neutral pH, with a pKa value of approximately 9.6, which contributes to its bioaccumulation in animals and microorganisms [13].

There is a global estimation for the intake of BPA, which is 30.76 ng kg^{-1} of BPA per day, according to the assessment done on humans for exposure to BPA through ingestion, inhalation, and dermal contact [14]. Due to the significant concerns about the health hazards of bisphenols, both the EU Commission and the U.S. Environmental Protection Agency (US EPA) have enacted stringent regulations, including the tolerable daily intake (TDI) of 0.05 mg BPA/kg body weight/day [15]. Other than that, Canada has amended the Environmental Protection Act in the Official Journal of Canada and added BPA to the list of toxic substances, as well as officially declared BPA as a harmful compound, with its exposure being highly toxic to humans [16].

The analysis of BPA and its analogues in different environmental matrices is crucial and therefore, there is an urgent need to establish new standards for bisphenols, as well as to find the best method to trace and remove them [10]. Reverse osmosis, nanofiltration, and ozone treatment are some of the methods used for BPA removal. However, the adsorption process has been found to be the most cost-effective solution for environmental remediation [17].

Nowadays, the use of silica-based adsorbents for bisphenols removal has become increasingly popular and a focus of considerable interest. Most of the recent reviews have demonstrated a strong interest in silica-based sorbents for use as adsorbents in environmental applications, with a particular focus on the adsorption and extraction of pollutants, as shown in **Table 2**. Researchers have actively explored modifications and functionalization strategies to maximize the effectiveness and applicability of these materials, particularly for the adsorption of bisphenols, especially BPA. Additionally, some reviews have highlighted the importance of understanding adsorption kinetics, thermodynamics, and environmental factors that influence the removal process of BPA.

However, reviews on BPA analogues are limited. Thus, the main objective of this review is to provide the latest developments in the selective adsorption of BPA and its analogues from environmental sources using silica-based adsorbents. For this purpose, recent studies were discussed in terms of adsorption capacity, fitted isotherm and kinetic models, and thermodynamic aspects. The adsorption and separation mechanisms for bisphenols using silica-based adsorbents are also discussed in this review. This review is expected to provide researchers with valuable insights for tailoring new functional groups and tuning the properties of silica-based adsorbents to select, optimize, and apply suitable techniques for the extraction of bisphenols and other emerging contaminants.

Types of silica-based adsorbents

Silica-based adsorbents could be divided into two main categories: those found in nature and those that are man-made. Natural sources like diatomaceous earth and clay are usually easy to get and relatively inexpensive. In contrast, synthetic silicas can be engineered to have specific traits, making them better at removing BPA.

Natural silica-based adsorbent

Diatomaceous earth, or DE, is a naturally occurring material composed of fossilized diatoms, which are tiny algae. Its high silica content gives it beneficial properties like chemical inertness, stability at high

Table 1. Summary of recent review articles related to application of silica-based sorbent for the adsorption of bisphenols in environmental matrices

Topic	Year	Remarks	Ref.
4-Nonylphenol adsorption, environmental impact and remediation: A review	2025	Discuss on 4-nonylphenol as well as bisphenol contamination with focus on remediation by adsorption.	[18]
Recent trends in the use of silica-based materials and their composites in extraction techniques: A review	2024	Provide comprehensive overview of the recent trends in the use of silica-based materials and their composites in extraction techniques as well as highlights the advantages of using these materials in extraction processes.	[19]
Agro-waste-based adsorbents as sustainable materials for effective adsorption of Bisphenol A from the environment: A review	2023	Provide an overview of the use of agro-waste based adsorbents as an effective adsorption of BPA. A few examples of silica-based adsorbent were discussed briefly.	[20]
Separation of toxic contaminants from water by silica aerogel-based adsorbents: A comprehensive review	2023	Various types of silica aerogel-based adsorbents as well as its functionalized forms are discussed.	[21]
Magnetic-silica nanocomposites and the functionalized forms for environment and medical applications: A review	2022	This review paper explores the synthesis, properties, and applications of magnetic-silica nanocomposites. They also explore the use of these functionalized nanocomposites in environmental remediation, including the adsorption process.	[22]
Endocrine disrupting effects of bisphenol A exposure and recent advances on its removal by water treatment systems. A review	2019	Discuss a little bit on the use of silica-based sorbent in the adsorption of bisphenol in water treatment system.	[14]
Grafting of Amine functional group on silicate-based material as adsorbent for water purification: A short review	2018	Provides reviews and evidence on the potential use of amine grafted silicate-based materials to remove contaminants through adsorption method.	[23]
Surface functionalization and manipulation of mesoporous silica adsorbents for improved removal of pollutants: A review	2018	This review provides an overview of surface functionalization techniques employed to enhance the performance of mesoporous silica adsorbents for pollutant removal. It serves as a valuable resource for researchers working in the field of adsorption and environmental remediation.	[24]
Adsorptive removal of bisphenol A (BPA) from aqueous solution: A review	2017	Examines the effectiveness of various adsorbents, including modified materials and composites as well as highlighting the importance of understanding adsorption kinetics, thermodynamics, and environmental factors influencing this process.	[2]
Newest applications of molecularly imprinted polymers for extraction of contaminants from environmental and food matrices: A review	2017	The paper emphasizes the unique ability of some silica MIPs to selectively bind target molecules.	[25]

temperatures, and environmental safety [26,27]. The inherent qualities of diatomaceous earth have made it useful across a range of industries and environmental cleanup efforts. To improve how well DE can adsorb particular contaminants, researchers have looked into different modification techniques.

Clay minerals like montmorillonite and kaolinite also have distinct structural and chemical features that enable adsorption. Montmorillonite, with its 2:1 dioctahedral structure, is notable for its high ability to exchange positive ions and its swelling behaviour, both of which enhance the adsorption of different pollutants, such as BPA [28,29]. Kaolinite, another type of clay mineral with a 1:1 structure that is less complex than montmorillonite, has a more limited ability to adsorb substances. This is because it has a smaller surface area and a lower capacity to exchange positive ions compared to montmorillonite [30].

In addition, zeolites are crystalline, tetrahedral aluminosilicate minerals with a highly porous structure, making them effective adsorbents for environmental applications, including the removal of organic pollutants such as bisphenols. Their negatively charged framework is counterbalanced by cations and water molecules within their pores, enhancing their ion exchange capacity [31].

Synthetic silica adsorbents

The use of synthetic silica adsorbents for the removal of BPA from environmental sources has been extensively studied, with various innovative approaches being developed to enhance their efficiency and selectivity. These adsorbents leverage the unique properties of silica, such as its high surface area and tuneable porosity, to effectively capture and remove BPA from water. Amorphous silica, with its irregular structure and high surface area, provides a versatile platform for BPA adsorption.

Mesoporous silica exists in numerous series, such as Mobil Composition of Matter (MCM), Santa Barbara Amorphous (SBA), Hexagonal Mesoporous Silica (HMS), Michigan State University Mesoporous Silica (MSU), and Fudan University Mesoporous Silica (FDU), each distinguished by variations in aperture, particle size, surface area, and synthesis method [32]. The MCM series stands out as the most recognized one among the various mesoporous silicas. MCM-41, MCM-48, and SBA-15 are characterized by their highly ordered pore structures, which play a crucial role in their adsorption abilities. MCM-41, with its hexagonal arrangement of uniform mesopores and a high surface area (ranging from 1000 to 1200 m^2/g), is particularly good at adsorbing BPA [33,34]. In addition, SBA-15 is characterized by its larger pore size and thicker walls compared to MCM-41, offering a high surface area (up to 745 m^2/g) and pore volumes (0.99–1.44 cm^3/g) [35]. MCM-48, which is a cubic mesoporous silica material, has proven to be more effective at extracting BPA than C18, with recovery rates ranging from 87.4% to 95.6% [36]. Magnetic silica composites incorporate magnetic nanoparticles (e.g., iron oxide) into the silica matrix, enabling easy and efficient separation using a simple magnet.

The integration of silica with the targeted binding sites of a molecularly imprinted polymer (MIP) allows silica-based MIPs to achieve greater selectivity and capacity for BPA adsorption. Silica-based MIPs are synthesized by grafting molecularly imprinted copolymer layers onto silica nanoparticles, enabling selective recognition of BPA molecules [37]. Several key advancements have improved the performance of MIPs for BPA removal. Surface imprinting creates binding sites on the MIP surface, enhancing accessibility and adsorption speed [38]. Dummy molecular imprinting simplifies synthesis and reduces template leakage by using BPA analogues as templates [39].

Finally, incorporating magnetic nanoparticles into MIPs allows for easy separation and recovery of the adsorbent [40]. Functionalized silica adsorbents demonstrate high stability and can be reused multiple times without significant loss of adsorption capacity [41]. The introduction of specific functional groups, such as amino, thiol, carboxylic acid, imidazole, and phenyl groups, enhances the selectivity and capacity for BPA adsorption through interactions like hydrogen bonding and π-π interactions [41,42]. Functionalized silica adsorbents, such as those co-immobilized with imidazole, phenyl, and long-chain alkyl groups, have shown enhanced adsorption capacities due to synergistic effects between hydrophobic and hydrogen-bonding interactions. These materials maintain high adsorption capacities even in high-salt solutions [43].

Researchers have also utilized agro-waste like sugarcane ash to synthesize mesoporous silica nanoparticles (MSN) that can adsorb BPA. Modifying these MSNs with substances such as hexadecyltrimethylammonium (CTAB) boosts their adsorption capacity, achieving up to 155.78 mg/g, with rapid adsorption rates [44] . In rice husks, nano silica can be extracted through acid and thermal treatment as an efficient BPA adsorbent in an aqueous solution [45]. Ostensibly, agro-waste-based adsorbents offer a greener and cheaper alternative to conventional adsorbents.

Synthesis and functionalization of silica-based adsorbents

Sol-gel, hydrothermal, and template methods (including soft and hard templating) represent the primary preparation techniques for silica. Through adaptation and modification, these methods yield a wide variety of silica nanoparticles, encompassing solid, porous, and functionalized forms with diverse sizes, shapes, and morphologies, making them suitablle as nanoadsorbents for pollutants [46].

In the sol-gel process, a sol forms via hydrolysis and condensation of highly reactive precursors at specific pH levels, subsequently gelling into a 3D structure through siloxane bond formation between colloidal particles. Conversely, the hydrothermal technique typically involves reacting materials in a high-temperature, high-pressure aqueous solution within a closed system [47]. On the other hand, the microemulsion templating method offers a straightforward and adaptable approach that utilizes a colloidal mixture. This mixture comprises a silicone alkoxide precursor, a suitable surfactant, and a polymer, which then undergoes hydrolysis by solvents and condensation to create a siloxane bond network [24].

Leveraging the high stability of silicon dioxide, the incorporation of organic or inorganic groups onto its surface bridges organic chemistry and inorganic materials. This dual modification grants silicon dioxide both enhanced functionality and maintained thermal stability. As a result, the adsorbents produced through this approach excel at addressing the pollution posed by emerging contaminants and promoting their environmental remediation [32].

The Stöber method, sol-gel process, and aerosol pyrolysis currently represent the most common strategies for applying silica coatings to iron oxide nanoparticles (NPs) [24]. Applying such a coating generally leads to an increase in particle dimensions and a modification of the magnetic characteristics of the resulting composite nanoparticles. It is significant that the silica shell thickness, within a range of 5 to 200 nm, can be controlled through adjustments in ammonia concentration and the tetraethoxysilane (TEOS) to water ratio [24]. Furthermore, the presence of hydroxyl groups on the iron oxide NPs' surface facilitates silica coating in aqueous media, particularly through the Stöber and sol-gel techniques. This results in improved dispersion and a reduction in the aggregation of the magnetic particles. The sol-gel method allows the formation of uniform and tuneable structure of silica through hydrolysis and condensation of alkoxysilanes under controlled pH conditions [48]. Coupling agents such as (3-amino-propyl) triethoxysilane (APTES) or (3-merhydryl propyl) trimethoxysilane (MPTMS) could be used for surface functionalization and their role has been proven to enhance the adsorption capacity and selectivity for removing bisphenols from environmental matrices [48].

Commonly grafted onto the surface and within the pores of silica are amine, halo, and thiol groups, which can be selected to tailor the material's acidity or basicity. Beyond this tuning, these initial functional groups serve as a foundation for further chemical modifications, transforming silica into a solid support for organic reactions. Grafted functional groups, thro ugh their chemical derivatives, can stabilize metal ce nters via the chelation effect [49]. Surface modification of mesoporous silica primarily relies on silane coupling agents such as γ-methylacrylloxy propyl trimethoxysilane (MPS), (3-amino-propyl) triethoxysilane (APTES), vinyl triethoxysilane (VTES), and (3-merhydryl propyl) trimethoxysilane (MPTMS). Upon hydrolysis, these organosilanes react with the active hydroxyl groups on the silica core, forming a bond. The amino, vinyl, and mercaptan groups present at the ends of these silane molecules enhance the surface reactivity of the silica, facilitating subsequent attachment of other organic functional groups [32].

By modifying the surface properties of nanoparticles (NPs), it becomes possible to achieve faster adsorption rates, higher adsorption capacities, and improved selectivity. Functionalized silica nanoparticles also offer the capability in targeting specific molecules for removal. Researchers leverage surface engineering of these NPs to develop materials with a high affinity for particular molecules, leading to more efficient and selective contaminant removal from water [50]. The reactive silanol groups on silica nanoparticle surfaces enable the relatively simple grafting of diverse organic functionalities via alkoxysilane chemistry, a widely reported method for imparting desired properties. The increasing availability of various functional alkoxysilanes has further streamlined this grafting process [46]. This functionalization enhances the adsorption capacity of the silica nanoparticles, and the subsequent grafting of organic groups or polymers creates core-shell structures that improve their colloidal stability.

Removal of bisphenols using silica-based adsorbents

Researchers have explored several methods for BPA removal from water, and adsorption is considered a particularly promising and well-investigated option [13]. Adsorption is a popular method for pollutant removal due to its effectiveness, simplicity, and reliable outcomes. The cost-effectiveness of this method largely depends on how well the adsorbent material works. Silicates, zeolites, and activated carbon are commonly used to adsorb pollutants because of their physical characteristics, like surface area and pore structure [51].

The unique ability to tailor surface functional groups, along with distinctive structural and surface chemistry, has been extensively researched for water pollutant adsorption in recent years. There is also been notable progress in the field of surface modification of these materials, such as the impregnation of Fe_3O_4, $AlCl_3$, $MgSiO_3$ and $ZnCl_2$, which enhances the adsorption capacity [51]. Mesoporous silica can be used as ideal adsorbent to endocrine-disrupting molecules, showing approximately 95% removal, which can be justified by the presence of electrostatic interactions and hydrophobic effect [18]. Furthermore, high adsorption selectivity for some endocrine-disrupting chemicals (EDCs) was achieved by simply grafting hydrophobic functional groups onto mesoporous silica. In particular, organic-inorganic hybrid mesoporous silicas containing phenyl groups outperformed activated carbon in water solutions for BPA adsorption capacity and selectivity. This enhanced selectivity is probably due to the specific interactions between the adsorbent's dual hydrophobic and hydrophilic surface characteristics and the two benzene rings and two hydroxyl groups of BPA [52].

The creation of magnetic nanocomposites for water purification through adsorption frequently utilizes silica, as it offers chemical stability, a large surface area, and the capability for functional modification [53]. In the Stöber method, tetraethylorthosilicate (TEOS) or other organosilanes are mixed in ethanol, ammonia, and water while stirring to form particles with a size that depends on the concentration of the solvents and silicates [13]. Generally, the obtained nanomaterial exhibits a uniform core-shell structure, $Fe_3O_4@SiO_2$, which is being applied for the adsorption of inorganic elements such as bisphenols in aqueous systems. Modification of silanol groups is frequently necessary to prevent basic analytes from being adsorbed, as this can negatively impact the adsorbent recovery. Numerous nanomaterials, such as iron oxide nanocrystals synthesized by a co-precipitation method, were decorated with an aminopropyl-functionalized silica layer [54]. The addition of SiO_2 not only reduced superparamagnetic iron oxide stacking and nanocrystal agglomeration but also yielded a highly tuneable surface area, leading to improved BPA binding and adsorption capabilities [2].

Dong et al. synthesized zeolite from coal fly ash (ZFA), which was then modified with hexadecyltrimethylammonium (HDTMA) and was examined for the adsorption of BPA from water [55]. Based on the results, ZFA had no affinity for BPA, however, surfactant-modified ZFA (SMZFA) showed a high adsorption capacity. Zeolite modification for BPA removal was explored using the cationic surfactants cetyltrimethylammonium bromide (CTAB) and cetylpyridinium bromide (CPB) [56]. The optimal results were achieved by treating pre-treated zeolite with 10 % CTAB for 36 hours at 25°C [2]. The mesoporous silica SBA-15 exhibits a hydrophilic character and a negative surface charge. Pore size control is achieved through manipulation of the reactant ratio. Furthermore, the hydrolysis rate and the interaction level between the assembled template and the SiO_2 polymer matrix are identified as key parameters governing the morphology and size of SBA-15. Consistent with previous reports, SBA-15, akin to MCM-41, is characterized by substantial pore volumes and surface areas. The hexagonal channels within SBA-15 possess diameters spanning 5 to 30 nm, while the pore walls exhibit thicknesses within the range of 3 to 6 nm [19].

Besides that, in a study by Li et al. [57], dual-responsive magnetic and thermal nanomaterials ($Fe@SiO_2@$PNIPAM and $Fe@SiO_2$) were synthesized and used to adsorb four phenolic compounds: bisphenol A, tetrabromobisphenol A, 4-tert-octylphenol, and 4-nonylphenol. Under optimized conditions, 4-nonylphenol showed over 90% adsorption efficiency within two hours, with maximum adsorption capacities of 2.4, 6.8, 24.7, and 49.3 mg/g for bisphenol A, tetrabromobisphenol A, 4-tert-octylphenol, and 4-nonylphenol, respectively [57].

Liu et al. employed a surfactant structure-directing agent to synthesize a hydrophobic mesoporous material (MCM-41-d) at room temperature and subsequently evaluated its effectiveness in removing BPA [11]. While the maximum adsorption capacity of MCM-41-d has been documented, catalytic activity in such silica MCM-41 molecular sieves necessitates modification through framework substitution to introduce active sites. Separately, MCM-48 has been employed for the removal of Congo red dye, phosphate, and nitrate from water samples, as well as for the extraction of EDCs [58]. **Figure 1** shows the schematic illustration of the experimental procedure for MI-SBA-15 preparation [49].

Organic-inorganic hybrid mesoporous silica materials, synthesized by covalently bonding organic moieties to the internal surface of mesoporous silica, have attracted considerable attention in the adsorption of a specific compound. Two common functionalization strategies include grafting organic species onto the pore walls and co-synthesis, which involves the co-condensation of alkoxysilane and organosilane precursors within a structured template. For instance, alkyl-grafted mesoporous silica materials for the selective adsorption of alkylphenols were prepared by Inumaru et al. [59].

Other than that, Yamaguchi et al. [60] concluded that organic-inorganic hybrid mesoporous silicas that had been synthesized through the co-condensation method showed a high adsorption capacity for BPA compared to those synthesized by the grafting method [61]. The co-condensation method yields superior BPA adsorption capacity over grafting because of fundamental differences in material structure. For example, co-condensation (a one-pot synthesis) incorporates the beta-cyclodextrin (BCD) functional groups directly into the growing silica framework, leading to a much higher density and more uniform distribution of the active BCD cavities throughout the entire material volume. In contrast, the grafting (post-synthesis) method only attaches BCD primarily to the surface and near the pore mouths of the pre-formed silica, often causing pore clogging and restricting the diffusion of BPA to internal sites, thus drastically limiting the number of effective host-guest inclusion sites available for BPA capture.

Besides, Bhattarai et al. created silica-coated cyclodextrin polymer adsorbents to remove emerging pollutants like steroid hormones (17-β-estradiol), bisphenol A, and perfluorooctanoic acid, employing three distinct functionalization methods to attach cyclodextrin onto silica [62]. The first method involved polymerizing cyclodextrin directly onto silica using coupling agents, next is supporting cyclodextrin on silica with glycidoxypropyl trimethoxysilane and 3-aminopropyl triethoxysilane copolymers, and lastly, coating silica with cyclodextrin using both cross-linking agents and copolymers. Across fourteen synthesized adsorbents, created under varying conditions like solvent, temperature, and reactant concentration, adsorption tests revealed that the material prepared with hexamethylene diisocyanate as a cross-linking agent and dimethyl sulfoxide as a solvent exhibited the best performance, achieving over 90% removal for 17-β-estradiol, perfluorooctanoic acid, and bisphenol A [53]. Overall, the Bhattarai et al. study, while commendably performing a systematic optimization across fourteen adsorbents to achieve high, broad removal (>90% for diverse contaminants), is fundamentally limited by its incomplete practical validation. The core critique is the absence of crucial data on long-term stability and cost-effective reusability which is the metrics essential for real-world application, despite good initial regeneration results.

Bucur et al. also has synthesized silica particles modified with two types of beta-cyclodextrin (BCD), confirmed by Fourier transform-infrared spectroscopy (FT-IR) and thermogravimetric analysis (TGA), and proposed a formation mechanism using contact angle, Brunauer-Emmett-Teller (BET), and scanning electron microscopy (SEM). These modified silicas effectively removed bisphenol A (BPA) from wastewater, reaching adsorption equilibrium in 180 minutes with capacities of 107 mg/g (BCD-OH) and 112 mg/g (BCD-NH_2), following a pseudo-second-order model and fitting the Freundlich isotherm, thus highlighting BCD's impact on adsorption [63]. The Langmuir model typically describes adsorption dominated by chemical interactions between the adsorbate and adsorbent, and can also apply to some physical adsorption scenarios, provided the adsorbent surface has a single type of active site. The results, however, suggest a heterogeneity of active sites on the materials, likely due to the higher activity of the BCD compared to the Si-OH sites on the silica during adsorption [63]. This study suffers from several practical and mechanistic limitations. The slow kinetics, taking 180 minutes to reach equilibrium, is a significant drawback for efficient flow-through or industrial wastewater treatment compared to faster alternatives. Mechanistically, while the Freundlich isotherm and pseudo-second-order model confirm a complex, heterogeneous adsorption driven by BCD inclusion, the study is critically limited by its omission of essential reusability data, leaving the practical sustainability and cost-effectiveness of the BCD-modified material completely unevaluated for long-term real-world application.

Other silica-based adsorbent, which is a highly selective material, MI-SBA-15, was successfully synthesized using a semi covalent method for recognizing and separating BPA from water [58]. This material possessed an ordered mesoporous structure with high surface area and pore volume, demonstrating rapid rebinding, high capacity, and excellent selectivity for BPA in rebinding experiments. The adsorption behaviour followed pseudo-second-order and Langmuir-Freundlich models. MI-SBA-15 exhibited significant selectivity for BPA over hydroquinone (HDQ), 4,4'-biphenol (BP) and diethylstilbestrol (DES) with relative selectivity coefficients of 132.13, 193.53, and 172.45 respectively. They also showed good stability and reusability for at least eight cycles and was effectively used for the selective separation of BPA from water samples with high recovery. Other examples of applications of silica-based adsorbents for the adsorption of BPA and its analogues from environmental sources including their details on adsorption conditions, the isotherm, kinetic and thermodynamics model results are shown in **Table 3**.

Silica-based adsorbents also have demonstrated high efficiency in BPA removal, with renewable mesoporous silica nanoparticles (MSN-CTAB, Q_{max} = 155.78 mg/g) and Fe_3O_4@SiO_2@PANI-GO (Q_{max} = 125.3 mg/g) showing the best adsorption capacities [64]. For BPA adsorption studies, the Langmuir and Freundlich isotherms are the most widely used models to describe adsorption behaviour on different materials. The Langmuir model typically excels at describing monolayer adsorption on uniform surfaces, while the Freundlich model is better suited for multilayer adsorption and heterogeneous surfaces with multiple active sites, particularly in porous adsorbents [65]. The Langmuir model was the most common, showing that BPA formed a single layer on the adsorbent surface. Some adsorbents manage to fit two different isotherm models like Langmuir and Freundlich models because the real-world adsorption process is often a complex combination of the idealized mechanisms described by each model, rather than a perfect fit to just one.

In short, a dual fit indicates that the complex, heterogeneous nature of the adsorbent's surface and pore structure allows its adsorption data to be statistically well-represented by both models, confirming the presence of different potential active sites and interaction modes. The pseudo-first-order (PFO) and pseudo-second-order (PSO) models are commonly used in BPA removal studies to fit kinetic data and explore the interactions between BPA and different adsorbents. PFO implies that the adsorption rate is controlled by the availability of unoccupied sites on the adsorbent, decreasing as these sites fill. PSO, however, suggests that chemisorption, involving electron exchange between BPA and the adsorbent surface, governs the adsorption process [65]. Most studies found that the adsorption process followed the pseudo-second-order model, meaning it depended on how BPA molecules interacted with the adsorbent.

Thermodynamic analysis confirmed that adsorption was generally spontaneous and exothermic, though some materials performed better at higher temperatures (endothermic behaviour). Many adsorbents maintained high efficiency over multiple cycles, making functionalized mesoporous silica materials promising candidates for BPA removal in water treatment applications. Free energy serves as an indicator of a process's spontaneity; a larger negative value reflects a sorption process that is more energetically favourable [66]. The feasibility and spontaneity of BP adsorption on silica were confirmed by a negative Gibbs free energy (ΔG), whereas the positive enthalpy change (ΔH) showed the process to be endothermic.

In reviewing the existing literature on the removal of bisphenols using silica-based adsorbents (**Table 2**), the most outstanding adsorbent in terms of removal performance and efficiency is the $Fe_3O_4@Si_2\text{-}C_{18}$, which demonstrated by the high adsorption capacity Qmax of 473.224 mg/g for BPA[67]. For the adsorption conditions, the typical optimum initial concentration of bisphenols ranged from 10 to 100 mg/L, with the temperature around 25°C (except for one study conducted at 40°C), and an optimized pH ranging from 3.0 to 9.0, depending on the prepared adsorbent and the type of bisphenol studied. Bisphenols such as BPA, BPF, BPE, and BPS have been used in removal investigations, with BPA being the most commonly studied for adsorption using silica-based adsorbents. The Langmuir isotherm model was the most commonly applied, indicating monolayer adsorption behaviour, followed by the Freundlich model for more heterogeneous systems. The pseudo-second-order (PSO) kinetic model was the primary kinetic model used, suggesting chemisorption as the dominant adsorption mechanism. Thermodynamic studies reported spontaneous and exothermic processes, as well as characteristics of physical adsorption. Silica-based adsorbents demonstrated good reusability, with many able to be reused for 5 to 8 cycles without any significant decrease in removal efficiency.

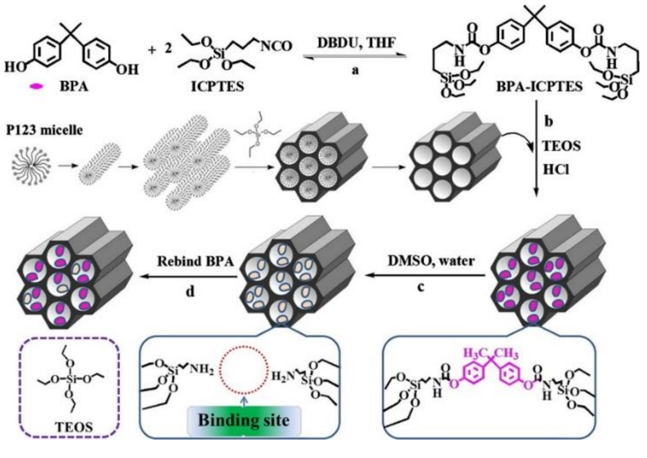

Figure 1. Schematic illustration of experimental procedure for MI-SBA-15 preparation. Reprinted from Ref. [54]. Copyright (2018) with permission from Elsevier.

Table 2. Applications of silica-based adsorbents for the adsorption of BPA and its analogues from environmental sources

No.	Adsorbent	BPs	Porosity	Adsorption Condition	Isotherm model	Kinetic and Thermodynamic model	Qmax/RE	Reusability	Ref.
1	$CoFe_2O_4@SiO_2$-P4VP)	BPA	Spherical structure with magnetic core of ca. 11nm, silica layer 4nm	IC: 100 mg/L T: 25 pH: 7.15 AD: 46.6 mg/g	Freundlich	PSO	46.6 mg/g	8 cycles	[13]
2	PES–SiO_2 composite membranes	BPA	-	IC: 10 mg/L T: 25 pH: 7.0	Langmuir	PSO	53 µg/g	5 cycles with slight decrease in performance	[68]
3	Zeolite imidazole frameworks ZIF-8	BPA	High porosity, large surface area	BPA dosage: 0.4 g T: 25 pH: 6.0	Langmuir	PSO	98.84%	Reusable	[69]
4	GAC with different silicate binders	BPA	Varied based on binder type	IC: 100 mg/L T: 25 pH: 5.0	Langmuir	PSO	Varied depending on binder and conditions (GAC-A: 45.5 mg/g)	Not explicitly stated, likely multiple cycles	[70]
5	Zeolite modified with CTAB	BPA	High surface area, improved hydrophobicity due to CTAB modification	IC: 10-100 mg/L T: 25 pH: 7.0	Langmuir	PSO	23.1 mg/g	Multiple cycles possible	[55]
6	Renewable (MSN) from sugarcane waste ash, functionalized with CTAB	BPA	High surface area, large pore volume	IC: varied T: 25 pH: 4.0-9.0	Langmuir	Fast adsorption kinetics	155.78 mg/g	Multiple cycles possible	[44]

7	D-MIMS	BPA	High surface area, large pore volume	IC: varied T: 25 pH: 5.0-7.0	Langmuir, Freundlich	PSO	High Qmax (expected due to imprinting)	Multiple cycles possible	[71]
8	m-DMI-MSNP	BPA, BPF, BPE	Mesoporous	IC: varied T: 25 pH: 7.0	Langmuir	PSO	38.75 mg/g, 93.1-97.8%	Multiple cycles with slight decrease in performance	[72]
9	PP-g-CaAlg@SiO_2	BPA	Mesoporous	IC: 0.1 g/L T: 25 pH: 6.0	Langmuir	PSO	High Qmax (expected due to synergistic effects)	Multiple cycles possible, good stability	[73]
10	Magnetic dodecyl chitosan/silica composite (MDCS)	BPA, BPS, BPF	High surface area, improved hydrophobicity due to dodecyl modification	IC: varied T: 25 pH: 6.0	Langmuir	PSO	BPA (17.6 mg/g), BPS (15.2 mg/g), BPF (13.8 mg/g)	5 cycles with minimal loss of efficiency	[6]
11	Ionic liquid functionalized polymer (ILFP), grafting1-butyl-3-vinylimidazolium bromide onto the silica surface	BPA	Rough surface with a lower specific surface area	T: 25 pH: 3.0-10.0	Langmuir	PSO, ΔH° = 36.30 kJ/mol ΔG° = 21.53 kJ/mol Spontaneous, exothermic and physical adsorption process	-	-	[74]
12	Mesoporous silica from batik sludge impregnated with aluminum hydroxide (MS-Al)	BPA	Mesoporous	IC: varied T: 25 pH: 3.0-9.0	Langmuir, Freundlich	PFO, PSO	83.53 mg/g	5 cycles with minimal loss of efficiency	[51]
13	SiO_2 modified biobased hydrolyzed hollow	BPA	High surface area, large pore volume	IC: varied T: 25 pH: 6.0	Langmuir	PSO	60.24 mg/g	5 cycles with minimal loss of efficiency	[75]

	polymer particles								
14	Graphene Oxide Assimilated Magnetic Silica Polyaniline Composite ($Fe_3O_4@SiO_2@PANI$-GO)	BPA	High surface area, improved adsorption capacity due to synergistic effect of components	IC: varied T: 25 pH: 6.0	Langmuir	PSO	125.3 mg/g	5 cycles with minimal loss of efficiency	[64]
15	Three functionalized silica adsorbents (with imidazole, phenyl, and long-chain alkyl groups)	BPA	High surface area, varied pore properties based on functionalization	IC: varied T: varied pH: 7.0	Freundlich	PSO, $\Delta H^\circ = -17.83, -13.53, -5.16$ kJ/mol $\Delta G^\circ = -18.06, -15.90, -13.14$ Exothermic process, physisorption interaction	120 mg/g (imidazole), 95 mg/g (phenyl), 80 mg/g (alkyl groups)	multiple cycles	[76]
16	$Fe_3O_4@SiO_2$-C18	BPA	Large surface area and pore volume	IC: 10-100 mg/L T: 40 pH: 6.0	Langmuir	PSO	473.224 mg/g	Reused several times without a considerable loss in removal efficiency	[67]

Abbreviations:

PSO – pseudo-second-order; PFO – pseudo-first-order; RE – removal efficiency; $CoFe_2O_4@SiO_2$-P4VP) - cobalt ferrite core-shell nanoparticles coated with silica and decorated with poly(4-vinylpyridine); m-DMI-MSNPs - magnetic dummy molecularly imprinted mesoporous silica nanoparticles; PP-g-CaAlg@SiO_2 - Polypropylene fiber grafted calcium alginate with mesoporous silica; MSN-CTAB - mesoporous silica nanoparticles functionalized with hexadecyltrimethylammonium; CTAB - Cetyltrimethylammonium bromide. IC – initial concentration, T – Temperature

Optimization strategies for the adsorption of bisphenols

The adsorption of bisphenols onto silica-based adsorbents is affected by several operational parameters that determine the efficiency of the removal process. Operational parameters such as contact time, temperature, adsorbent dosage, ionic strength, and pH play critical roles in optimizing adsorption conditions. Understanding the impact of these parameters is essential for enhancing adsorption capacity, improving selectivity, and ensuring cost-effective pollutant removal from aqueous solutions. This section explores the significance of these parameters, their influence on adsorption performance, and insights from various studies to determine optimal conditions for bisphenols removal. **Table 3** summarizes the optimization parameters for silica-based adsorbents in the adsorption of bisphenols.

Effect of contact time

For bisphenol adsorption, the duration of contact between the adsorbent material and the bisphenol being targeted is a critical factor in assessing when equilibrium is reached. Examining this helps determine the ideal contact time, which directly impacts the effectiveness and cost-efficiency of the adsorption process [77]. Additionally, optimizing the adsorption process to minimize costs while maintaining high adsorption efficiency requires a well-designed experimental approach that balances adsorption capacity and contact time effectively. Theoretically, optimizing contact time provides valuable insights for determining an appropriate kinetic model, which can be experimentally validated through the significance of linearized expressions.

For instance, Mahmad et al. [78], in their study on the adsorption of BPA using a zeolite imidazolate framework, incorporated three additional parameters namely BPA concentration, adsorbent dosage, and pH, along with contact time in the design of experiments (DoE) for multivariate analysis based on the Box-Behnken Design (BBD). **Figure 2** illustrates the 3-dimension plot of ZIF-8 for BPA adsorption using BBD [65]. An optimal adsorption efficiency was achieved at contact times of 6 minutes and 40 minutes when a higher adsorbent dosage was used. This was attributed to the increased surface area of 1299 m² g⁻¹, which facilitated greater interaction between the adsorbent and adsorbate.

Meanwhile, Gong et al. [79] observed that the adsorption of a developed mesoporous magnetic composite material (Fe_3O_4@SiO_2/CTAB-SiO_2) increased exponentially within the first 30 minutes, achieving a removal rate of over 90% before stabilizing beyond this point. Adsorption kinetics, analysed using the intraparticle diffusion model, revealed a two-step process, with the second step identified as the rate-limiting stage. This indicates that while the initial adsorption phase exhibits a high adsorption rate, the rate gradually declines as contact time increases.

Chen et al. [80] compared the performance of magnetic dummy molecularly imprinted mesoporous silica nanoparticles (m-DMI-MSNPs) with their non-imprinted counterpart (m-NI-MSNPs) and found that the specific recognition sites on m-DMI-MSNPs were tailored for BPA adsorption, resulting in significantly higher adsorption efficiency than m-NI-MSNPs. Additionally, the efficient mass transfer of BPA into the imprinted cavities of m-DMI-MSNPs enabled equilibrium to be reached within just 10 minutes.

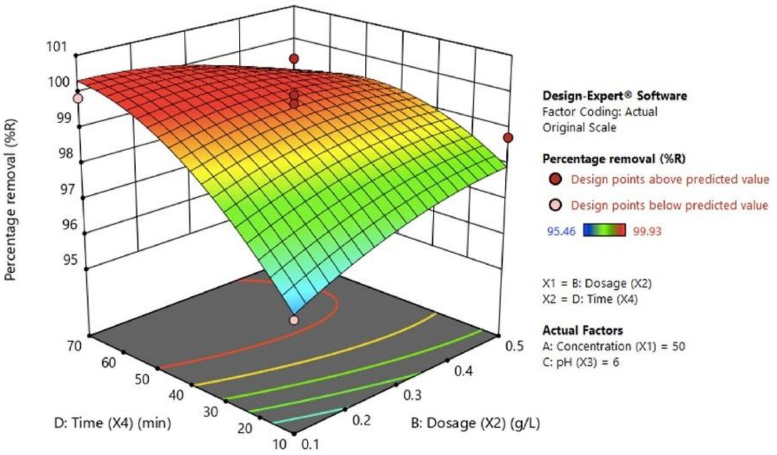

Figure 2. 3–dimension plot of ZIF–8 for BPA adsorption. Reprinted from Ref. [65]. Copyright (2023) with permission from Elsevier

Peng et al. [81] reported that a hierarchical porous metal-organic framework (MOF) coated with large mesoporous SBA-15 (ZIF-8@SBA-15) exhibited twice the adsorption efficiency and nearly 20 times the adsorption rate constant compared to pure ZIF-8. This superior performance was attributed to the ability of ZIF-8@SBA-15 to reach adsorption equilibrium within approximately 2 minutes, whereas ZIF-8 required around 100 minutes to achieve equilibrium. The hierarchical micro-mesoporous structure of ZIF-8@SBA-15 facilitated stronger π–π stacking interactions between the imidazole moieties and BPA, enabling enhanced mass transfer and improved adsorption properties.

Rovani et al. [44], on the other hand found that rapid adsorption capacity was observed for the first 20 minutes contact time due to the reason of great affinity interaction between BPA and develop mesoporous silica nanoparticles functionalized with hexadecyltrimethylammonium (MSN-CTAB). This adsorption continues to reach maximum 93.3% adsorption beyond 180 minutes but in slow steps which due to possibility of monolayer formation of BPA on the MSN-CTAB surface.

Effect of temperature

In the adsorption process, temperature is a critical factor influencing both the adsorption rate and capacity. Additionally, it enables researchers to evaluate thermodynamic parameters such as Gibbs free energy (ΔG), enthalpy (ΔH), and entropy (ΔS), providing valuable insights into the feasibility and mechanism of adsorption. Optimizing the temperature can enhance adsorbate uptake while preventing degradation or loss of integrity of both the adsorbent and adsorbate, ultimately preserving adsorption performance [82].

In this regards, Gong et al. [79] investigated the effect of temperature on the adsorption of BPA using Fe_3O_4@SiO_2/CTAB-SiO_2 composite and found that 35ºC was the optimal temperature, providing a conducive environment for the efficient diffusion of BPA into the adsorbent.

Meanwhile, Xu et al. [83] obtained a slightly lower BPA adsorption was observed at 25ºC when using PP-g-CaAlg@SiO_2. This is attributed to the increase in solute solubility at higher temperatures, which weakens hydrogen bonding and the hydrophobic effect, ultimately hindering the adsorption process.

Effect of adsorbent dosage

The adsorbent amount was one of the aspects that affected the adsorption capacity of target phenols, depending on the initial concentration, pH, and the desired removal efficiency. Increasing the adsorbent amount resulted in more active sites available for interaction, with a minimum reported amount ranging from 10 mg to 100 mg due to the adsorbent mass affecting binding capacity and efficiency [89]. Plotting the amount of adsorbent versus adsorption efficiency showed that they were directly proportional (**Figure 3**) [90].

The mesoporous silica spheres doped with CuO (CuO/MSS) were employed by Liang et al. to remove BPA by activating peroxydisulfate (PDS) through an electron transfer mechanism [91]. The high surface area and mesoporous structure enhanced adsorption efficiency, while stripping the BPA off resulted in high removal efficiency (80%) after multiple cycles. Silica-based adsorbents significantly enhanced the sorption capacity for heavy metals, indicating that various modifications could be beneficial for organic pollutants like BPA.

Mourao et al. highlighted that the use of commercial silica binders in granulated activated carbon resulted in higher adsorption competencies for BPA, ranging from 37.0 to 54.8 mg/g compared to unmodified carbon [92]. These granulated adsorbents were suitable for practical applications in wastewater treatment due to their structural stability and enhanced mechanical properties.

Another study utilized magnetic dummy molecularly imprinted mesoporous silica nanoparticles (m-DMI-MSNPs), which showed high selectivity and rapid adsorption kinetics for BPA (38.75 mg/g) [93]. This type of adsorbent exhibited removal efficiencies of 93.1% to 97.8% across various water samples for different types of bisphenols. The removal efficiency increased with higher concentrations of nanotubes (MWCNTs/SiO_2/CS), from 69% to 79.5%, as the dosage increased from 0.33 to 0.78 g/L with a maximum BPA concentration of 6.5 mg/L [90].

Recently, Dura et al. used a natural zeolite with high silica content, known as clinoptilolite, which demonstrated a promising result with an adsorption capacity of 50 mg/g for BPA [94]. Characterization of clinoptilolite, with its pore size ranging from 30 nm to 10 μm, showed that it was suitable for adsorbing BPA. Unfortunately, a further increase in the adsorbent amount led to saturation, where particle aggregation occurred, resulting in no significant impact on the final adsorbate concentration [95,96].

Tokuyama et al. synthesized a poly (NIPA)/silica gel composite as an adsorbent and found that, depending on the ideal contact time, pH, and temperature, higher BPA concentrations required a significant amount of adsorbent to achieve successful removal [97,98].

Table 3. Summary of the optimization strategies for silica-based adsorbents in the adsorption of bisphenols

Adsorbent	Analyte	Matrix	Optimization Type	Instrument	Adsorbent Dosage (mg)	pH	Contact Time (min)	Analyte Concentration (mg/L)	Temperature (ºC)	Ionic Strength	Removal Efficiency (%)	Ref.
Zeolite imidazole frameworks (ZIF-8)	BPA	Water	BBD	UV-Vis spectrophotometer	0.3	6	40	50	25		99.93	[78]
Fe_3O_4@SiO_2/CTAB-SiO2	BPA	Water	OVAT	HPLC-FLD	10	6.5	30	-	35		93.2	[79]
m-DMI-MSNPs	BPA, BPF, BPE	Water	OVAT	HPLC-DAD	10	-	10	0.25	25		95	[80]
ZIF-8@SBA-15	BPA	Water	OVAT	HPLC-DAD	-	10	~2	-	25		-	[81]
PP-g-CaAlg@SiO_2	BPA	Water	OVAT	UV-Vis spectrophotometer	-	6	-	-	25		-	[83]
CuO/MSS	BPA	Water	OVAT	HPLC-UV	-	9	45	50	-		>80	[84]
MSN-CTAB	BPA	Water	OVAT	UV-Vis spectrophotometer	100	11	65	100	25		>93	[44]
$CoFe_2O_4$@SiO_2-P4VP	BPA	Water	RSM	UV-Vis spectrophotometer	-	7.15	-	100	25		-	[85]
Sil-(HDTMS-MPS)-VIm, Sil-(HDTMS-MPS)-S and	BPA	Water	OVAT	UV-Vis spectrophotometer	-	4-8	-	-	-	0.2 mol/L Na_2SO_4 and NaCl	-	[86]

Adsorbent	**Analyte**	**Matrix**	**Optimization Type**	**Instrument**	**Adsorbent Dosage (mg)**	**pH**	**Contact Time (min)**	**Analyte Concentration (mg/L)**	**Temperature (ºC)**	**Ionic Strength**	**Removal Efficiency (%)**	**Ref.**
Sil-MPS-(S-VIm)												
MSN-CTAB	BPA	Water	OVAT	UV-Vis spectrophotometer	-	11	60	100	-	-	>90	[44]
Fe_3O_4@SiO_2@PANI-GO	BPA	Water	OVAT	UV-Vis spectrophotometer	100	6	210	-	25	-	96.2	[64]
m-DMI-MSNPs	BPA, BPE, BPF	Water	OVAT	HPLC-DAD	-	-	<1	-	-	-	93-97	[87]
Polyvinylpyridine/SiO_2/APTMS and polyvinylpyridine/SiO_2	BPA	Water	OVAT	HPLC-DAD UV-Vis spectrophotometer	-	9	-	-	-	-	99.5	[88]
SiO_2-BCD-OH and SiO_2-BCD-NH_2	BPA	Water	OVAT	UV-Vis spectrophotometer	-	-	180	-	-	-	-	[63]

Abbreviations:
BBD – Box-Behnken Design; OVAT - One-Variable-at-a-Time; HPLC-FLD - high-performance liquid chromatography-fluorescence detection; m-DMI-MSNPs - magnetic dummy molecularly imprinted mesoporous silica nanoparticles; HPLC-DAD - high-performance liquid chromatography-diode array detector; PP-g-CaAlg@SiO_2; MSN-CTAB - mesoporous silica nanoparticles functionalized with hexadecyltrimethylammonium ; MSNCTAB - mesoporous silica nanoparticles functionalized with hexadecyltrimethylammonium; Fe_3O_4@SiO_2@PANI-GO - magnetic graphene oxide assimilated magnetic silica polyaniline; m-DMI-MSNPs - magnetic dummy molecularly imprinted mesoporous silica nanoparticles; MWCNTs/ SiO_2/CS - magnetic multi-wall carbon nanotubes modified with chitosan biopolymer.

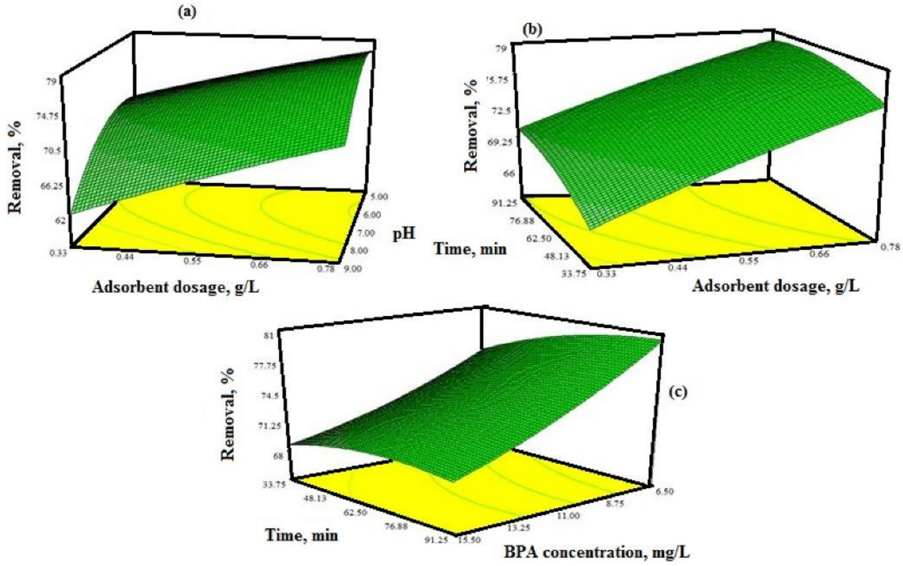

Figure 3. The response surface of the BPA adsorption on the MWCNTs/SiO_2/CS. The effect of (a, b) initial BPA concentration and time, (c, d) MWCNTs/SiO2/CS dose and time. Reprinted from Ref. [87]. Copyright (2020) with permission from Elsevier

Another study on silica-based adsorbents reported that the maximum adsorption capacity of the MIL-53(Al)@SiO_2 composite was 134.68 mg/g, as determined by Chatterjee et al. [98]. Since adsorption uptake decreased as the number of available adsorbent sites increased, some active sites remained vacant. The optimum amount of adsorbent selected was 1 g/L, achieving a removal percentage of 85%.

Recently, Hadoudi et al. demonstrated that bentonite-CTAB achieved a maximum adsorption capacity of 256.41 mg/g [99]. While silica-based adsorbents, such as silica gel, were commonly used in adsorption processes, bentonite-CTAB operated on different principles due to its clay mineral composition. The interactions involved in the adsorption process included electrostatic attraction and hydrogen bonding, which were influenced by the unique properties of bentonite clay. It followed pseudo-second-order kinetics, indicating chemisorption, with equilibrium reached within half an hour under optimal conditions. Adsorbents with a larger surface area required smaller quantities to achieve the same level of removal compared to those with a smaller surface area [100–102]. Furthermore, large-scale applications benefited from lower adsorbent doses since they were more cost-effective and required a minimal amount.

Effect of ionic strength

Further exploration of the factors affecting the removal of BPA included the effect of ionic strength, which was systematically examined. It was found that increasing salt concentration had different impacts on the adsorption affinities for the target analyte. Ionic strength had the least impact on the adsorption process, as stated by Fu et al., indicating that sodium chloride (NaCl) opposing effects of electrostatic screening and salting-out were insufficiently convincing [100]. Adding salt caused the adsorbent to undergo “squeezing-out”, which was harsh for target analyte adsorption and also competed with BPA for active sites [96,103,104].

Yilmaz demonstrated that additional increases in NaCl content increased the solution’s viscosity and reduced the removal rate and mass transfer due to competing adsorption. Enhancement in adsorption was only observed with lower concentrations of salt (<0.1M) due to the salting-out effect, which led to lower solubility of BPA and increased its hydrophobicity [105–107]. The effect of ionic strength on BPA removal was found to be insignificant, as indicated by studies using Palygorskite-montmorillonite as an adsorbent. The ionic strength was verified in the range of 0–0.1M of calcium chloride ($CaCl_2$), which did not extensively affect the adsorption capacity for BPA under the tested conditions [108]. The research indicated that as the ionic strength increased, using potassium chloride (KCl) at 0.01 M and 0.1 M, adsorption capacity remained poor. This suggested that higher ionic strength may have competed with BPA for adsorption sites on the Clinoptilolite [94].

However, several studies reported a positive and significant effect on the adsorption rate and increased mass transfer of BPA by adding salt. It appeared that an increase in ionic strength did not substantially degrade the adsorption ability for BPA since the composite hydrogel-derived iron/nitrogen co-doped carbon (FeN@CP800) hydrogel maintained its adsorption efficiency under a range of coexisting ion species and ionic strengths. The stability and efficacy of the adsorption process may have improved, as demonstrated by Tie et al., who achieved an adsorption capacity of 84% after five cycles [109].

Higher BPA adsorption was attributed to the high concentration of NaCl, which Hlekelele et al. demonstrated influenced the surface charge of aromatic polymeric adsorbents and favoured π–π dispersion interactions [100,107]. Overall, the results indicated that, depending on the applied NaCl concentration, ionic strength could either improve or reduce the effectiveness of BPA removal.

Effect of pH

The adsorption rate of pollutants like BPA onto adsorbents was highly affected by the solution pH, as pH altered the surface charge of the adsorbent and the ionization of the pollutant, which in turn affected their interactions [95]. The adsorption rate influenced by pH was often related to the point of zero charge (pH_pzc) [96,110]. This referred to the pH value at which the adsorbent surface had a net neutral charge. The adsorbent surface became positively charged when the solution pH was lower than the pH_pzc [94]. This condition enhanced the adsorption of anionic pollutants, as the positively charged surface attracted negatively charged molecules, and vice versa. Generally, adsorption tended to be higher at acidic to neutral pH levels, where favourable interactions occurred between the adsorbent and BPA.

Several studies reported that the optimum pH range for the highest adsorption capacity was in the acidic range (pH 4–6), while adsorption decreased in more alkaline conditions (>7). The optimal pH varied depending on the specific adsorbent material used and the preferred interaction mechanism. Pete et al. reported that the adsorption of BPA with polyvinyl pyridine/SiO2/3-aminopropyltrimethoxysilane (APTMS) remained relatively constant across all pH levels [94]. At pH levels above 7.5, some deprotonated BPA interacted electrostatically with APTMS, somewhat improving adsorption. However, at pH 10, all BPA forms were present, but deprotonated APTMS weakened electrostatic and hydrogen bonding, leaving π-π interactions as the main adsorption mechanism. The optimal pH for several studies was determined to be 9.0 [91,93,111], where BPA adsorption was highest. Adjusting the pH to these levels enhanced the adsorption process and improved the overall efficiency of BPA removal from aqueous solutions.

Therefore, for the optimization strategy, one-variable-at-a-time (OVAT) analysis is the most commonly employed approach, while a few studies applied response surface methodology (RSM) strategies such as Box-Behnken design (BBD) and central composite design (CCD). Adsorbent dosages used in optimization ranged from as low as 0.3 mg to 780 mg. On the other hand, the pH of sample solutions ranged from 4 to 11, with neutral to slightly basic conditions (pH 6 to 7.15) being the most commonly applied for efficient BPA adsorption. Besides, contact time varied from less than 1 min to 210 min, with most optimal conditions falling between 30 and 65 min and lastly, temperature was tested within the range of 25°C to 35°C, with 25°C being the most commonly used condition.

Adsorption and separation mechanisms of BPA and its analogues using silica-based adsorbents

Understanding the adsorption mechanism is crucial for designing materials for future applications and developments [65]. There are two primary modes of adsorption: physical adsorption and chemical adsorption. Physical adsorption utilizes intermolecular forces, such as van der Waals forces, to bind contaminants to the surface or pores. Conversely, chemical adsorption involves the creation of chemical bonds between the contaminants and the silica's active sites [32].

The adsorption of bisphenol by silica-based adsorbents is a complex process that primarily involves π-π electron donor-acceptor (EDA) interactions, acid-base interactions, hydrophobic interactions, hydrogen bonding, electrostatic attraction, pore filling, or a combination of these interactions, as shown in **Figure 4**. For EDA interactions, BPA, which possesses aromatic rings, is rich in electrons. Hence, silica surfaces, especially those modified with aromatic groups, can interact with these electron-rich rings through EDA interactions [112]. This strong attraction serves as a primary driving force for BPA adsorption. In addition, for hydrophobic interactions, the hydrophobicity of BPA causes it to repel water. Silica surfaces, even unmodified ones, have hydrophobic regions or can be modified with hydrophobic groups [113]. These hydrophobic regions interact with the hydrophobic parts of BPA, promoting its adsorption [114]. For hydrogen bonding, both BPA and silica surfaces contain functional groups capable of forming hydrogen bonds. These interactions contribute to the overall adsorption process, particularly when the adsorbent is modified with functional groups such as -OH or $-NH_2$ [115]. Electrostatic interactions, on the other hand, involve the charges influences. For instance, the surface charge of silica and the charge of BPA molecules play a role in adsorption. Depending on the specific pH, the adsorbent and BPA might either attract or repel each other due to these charges [114].

The effectiveness of the adsorption process is also significantly influenced by the molecular size, polarity, and concentration of the adsorbate. The mechanism of adsorption is affected by several elements, such as the type of silica, modifications to its surface, and the conditions of the solution. Additionally, pH, ionic strength and temperature affect the relative importance of each mechanism.

Based on Valentini et al., the results have shown that interactions between the π-π conjugate system and the amino group of $CoFe_2O_4@SiO_2$-P4VP were conducive to the adsorption of BPA, which was mainly caused by van der Waals forces and the π-π interactions of the BPA aromatic ring [13]. Besides that, the hydrogen bonding interaction and hydrophobic effect also contribute to the surface adsorption [13]. The effective removal of bisphenol A through adsorption using PES-SiO_2 composite membranes is largely due to the combination of their porous structure and the availability of silanol bonding groups, which act as adsorption sites [68].

As reported in [69], the mechanism involved can be predicted based on the interaction energy by obtaining a low value of ΔG_{bind}. Hence, ZIF-8 (Zn) was predicted to have a van der Waals interaction with BPA. On the other hand, the same interactions can be seen in literatures [70], [55] and [28] with hydrophobic interactions and π-π interactions. BPA anion adsorption happens when it interacts with the positively charged 'head' of the cetyltrimethylammonium bromide (CTAB) surfactant within the layer. The BPA benzene rings orient themselves towards the CTAB bilayer's interior, facilitating hydrophobic interactions between the BPA rings and the CTAB's 16-carbon tail, thereby boosting BPA adsorption [44]. The imprinted silica presented high recognition ability to bisphenols.

For instance, Yu et al. [71] and Chen et al. [72] conducted studies involving dummy molecularly imprinted polymers (DMIPs) using different analogues of BPA as templates for the determination of BPA. When BPA is introduced to the imprinted material, it readily enters the BPAF-imprinted cavities and forms hydrogen bonds with the -NH_2 groups within them [71].

Magnetic dodecyl chitosan was chosen to be synthesized and characterized for the enrichment of emerging bisphenol contaminants, and in this case, the high pH value plays an important role in strengthening the formation of hydrogen bonding, which improves the adsorption performance. The repulsive interaction between BPA and the composite's negatively charged surface is intensified when the solution's pH is near BPA's pK_a value due to the partial dissociation of BPA's hydroxyl groups [6]. Hydrogen bonds, van der Waals forces, and ionic bonds are responsible for the negative enthalpy change in work done by Xu et al. (2023) [73]. The hydrogen bonds between BPA and CaAlg@SiO_2 also played a major role in the adsorption of BPA.

The study by Choong et al. used a low-cost adsorbent from waste material, which involved hydrogen bonding and electrostatic attraction [51]. The authors stated that the Si-O group is attracted to the hydroxyl group of BPA via electrostatic attraction and that the hydroxyl groups of calcite sludge-aluminum hydroxide attract BPA molecules to form hydrogen bonds.

Recently, a study on the removal of BPA using $Fe_3O_4@SiO_3$@PANI-GO indicated that at higher pH, along with π–π interactions, hydrogen bonding between BPA-GO, another interaction called Lewis acid-base interactions (Fe–O coordination) between BPA and Fe_3O_4 occurred [64].

Another work by Wang et al. synthesized three functionalized silica adsorbents with imidazole, phenyl, and hexadecyl groups (Sil-(HDTMS-MPS)-VIm, Sil-(HDTMS-MPS)-S and Sil-MPS-(S-VIm)) to understand their impact on bisphenol A (BPA) adsorption [76]. Results showed alkaline imidazole was key, and its synergy with hexadecyl yielded the highest BPA adsorption (89.6 mg/g) between pH 4.0-9.0, attributed to BPA's high K_{ow}. These adsorbents-maintained performance in high salt and followed pseudo-second-order kinetics and the Freundlich model, suggesting that combining hydrogen bond-forming alkaline groups with hydrophobic chains enhances BPA removal from water.

The varying effectiveness of the three adsorbents in capturing BPA can be primarily explained by the different functional groups present on their surfaces. Specifically, the imidazole groups, characterized by their five-membered aromatic ring containing nitrogen, can interact with BPA through two mechanisms: hydrogen bond formation between the imidazole nitrogen and BPA's hydroxyl groups, and π–π interactions between the imidazole ring and the phenyl groups of BPA [76]. Sil-(HDTMS-MPS)-VIm and Sil-MPS-(S-VIm) are strong π-acceptor because of their N-heteroaromatic ring and the BPA molecule is a strong π-donor due to the hydroxyl groups on benzene rings. The π–π interactions between π-acceptor and π-donor are the strongest among acceptor–acceptor, donor–donor and acceptor–donor interactions. Therefore, π–πelectron acceptor–donor interactions would explain the significant adsorption differences of the three adsorbents for BPA [76].

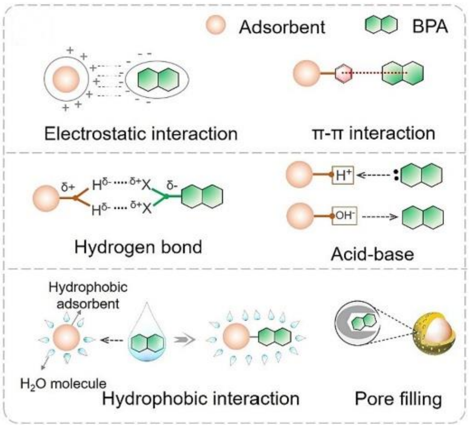

Figure 4. Diagram of possible mechanisms of interactions for the adsorption of BPA by silica-based adsorbents. Reprinted from Ref. [65] Copyright (2024) with permission from Elsevier

On the other hand, adding amino groups to mesoporous silica increases its chemical adsorption capacity for contaminants, as this modification utilizes the silica's large surface area to create numerous active sites, enhancing both adsorption and selectivity [32]. The introduction of amino groups strengthens the interaction between the silica and contaminants, leading to more efficient adsorption. Apart from that, they also suggested that adsorbents with structural configurations such as alkaline imidazolium groups are crucial for BPA adsorption. Hence, **Table 4** summarizes the possible analyte-adsorbent interactions for the adsorption of bisphenols in environmental matrices.

As a summary, the interactions of bisphenols towards silica-based adsorbents are mainly governed by the π–π interactions, hydrogen bonding, hydrophobic interactions, van der Waals forces, and in some cases, electrostatic or Lewis acid-base interactions. The π–π interactions commonly formed through the interactions between the aromatic rings of bisphenols and aromatic or any conjugated structures in the silica-based adsorbents, while hydrogen bonding usually formed through the hydroxyl group of bisphenols and functional groups on the silica surface. Hydrophobic interactions are the common interaction enhanced by long alkyl chair or any hydrophobic polymer coatings which improved the affinity toward bisphenol molecules. Van der Waals forces usually contributing to physisorption behaviour in adsorbent such as ZIF-8 or functionalized mesoporous silica. In addition to these interactions, electrostatic interaction also plays an important role to the adsorbent with charged functional groups and Lewis acid-base interactions and ionic bonding usually present in adsorbents containing metal ions or charged sites such as Fe_3O_4@SiO_2@PANI-GO, PP-g-CaAlg@SiO_2.

Conclusions and future perspectives

Significant enhancements in adsorption capacity and selectivity were achieved through the modification or functionalization of silica surfaces with specific functional groups such as amino, thiol, carboxylic acid, imidazole, and phenyl. It is worth noting that these modifications have improved the interactions between the adsorbents and analytes through hydrogen bonding, hydrophobic interactions, and π-π interactions. The utilization of natural silica sources offered cost-effective alternative adsorbents, while mesoporous silica materials such as MCM-41, MCM-48, and SBA-15 exhibited superior adsorption performance due to their advantages in controlled pore structures and higher surface areas. In order to improve the synthesis and functionalization of silica-based adsorbents, scientists could use greener methods, for example using water or green solvents such as deep eutectic solvents or ionic liquids. These solvents can be further explored to reduce the environmental impact. A better control of particle size and porosity can be achieve using templating or flow-based techniques which may also led to enhance adsorption efficiency. Additionally, developing dual-functional or multi-functional silica-based adsorbents capable of removing various types of endocrine-disrupting chemicals (EDCs). This approach may increase the effectiveness of the treatment and allow these adsorbents to be used in a wider range of applications. To support this, environmentally friendly materials such as ionic liquids and deep eutectic solvents can be used as green functionalizing agents. Utilizing agricultural waste or industrial by-products could also be an option to reduce costs and minimize environmental impact. New methods like click chemistry or using responsive materials (e.g., magnetic or pH-sensitive) can also make the adsorbents more effective and easier to reuse. Advanced techniques such as detailed material

Table 4. Possible analyte-adsorbent interaction for the adsorption of bisphenols in environmental matrices

No.	Adsorbent	Possible Analyte-Adsorbent Interaction	Advantages	Ref
1	$CoFe_2O_4$@SiO_2-P4VP)	van der Waals interactions, π-π interactions, hydrogen bonding, hydrophobic interaction	BPA adsorption was optimized with good correlation ($R > 0.88$), magnetic recovery, good reusability	[13]
2	PES–SiO_2 composite membranes	π-π interactions, hydrophobic interactions	High adsorption capacity, good reusability, membrane-based separation	[68]
3	ZIF-8 (Zn)	van der Waals interactions	High adsorption capacity, reusable	[69]
4	GAC with different silicate binders	π-π interactions, hydrogen bonding, hydrophobic interactions	Improved mechanical strength and stability compared to powdered activated carbon	[70]
5	Zeolite modified with CTAB	Hydrophobic interactions, electrostatic interactions	Enhanced adsorption capacity compared to unmodified zeolite	[55]
6	Renewable (MSN) from sugarcane waste ash, functionalized with CTAB	Hydrophobic interactions, π-π interactions	High adsorption capacity, fast kinetics, derived from renewable source	[44]
7	D-MIMS	Hydrogen bonding, π-π interactions	High selectivity and adsorption capacity	[71]
8	m-DMI-MSNP	π-π interactions, hydrogen bonding	High selectivity, fast adsorption kinetics, magnetic separation	[72]
9	PP-g-CaAlg@SiO_2	van der Waals interactions, ionic bonds, hydrogen bonding	High adsorption capacity, good mechanical stability, environmentally friendly	[73]
10	Magnetic dodecyl chitosan/silica composite (MDCS)	Hydrophobic interactions, hydrogen bonding, π-π interactions	High adsorption capacity, magnetic separation, good reusability	[6]
11	ILFP	Synergistic effect of hydrogen bond, π-π and electrostatic interactions	High adsorption capacity	[74]
12	MS-Aluminum hydroxide	Hydrogen bonding, electrostatic attraction	Excellent removal, low-cost adsorbent from waste material, good reusability	[51]
13	SiO_2 modified biobased hydrolyzed hollow polymer particles	Hydrogen bonding	High adsorption capacity, biodegradability, low-cost	[75]
14	Graphene Oxide Assimilated Magnetic Silica Polyaniline Composite (Fe_3O_4@SiO_2@PANI-GO)	π-π interactions, hydrogen bonding, Lewis acid–base interactions	High adsorption capacity, magnetic recovery, good reusability	[64]
15	Three functionalized silica adsorbents (with imidazole, phenyl, and long-chain alkyl groups)	π-acceptor and π-donor interaction, hydrogen-bonding and hydrophobic interaction	High adsorption capacity, selective adsorption, good stability in high salt solutions	[76]
16	Fe_3O_4@SiO_2-C18	Adsorbent offers both hydrophilic interactions, such as hydrogen bonding, and hydrophobic interactions with BPA molecules, owing to the presence of hydroxyl groups and a long hydrocarbon chain (C18)	High adsorption capacity good reusability	

characterization and computer modelling could assist researchers better understand how adsorption works at the molecular level. These insights could support the design of more selective and effective adsorbents. It is also important to study how different BPA analogues interact with silica-based and other types of adsorbents, as they share similar structures and pose potential health risks. Overall, silica-based adsorbents have shown strong potential in removing BPA and its analogues. Future progress in this field can be achieved by focusing on low-cost, sustainable, and efficient materials for environmental clean-up efforts.

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