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Review

Recent Advances in Heavy Metal Adsorption via Organically Modified Mesoporous Silica: A Review

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Abstract: Despite the significant advancements in wastewater treatment technologies, the field still lacks a comprehensive synthesis of recent developments in heavy metal removal using functionalized mesoporous silica materials. Adsorption has emerged as a preferred method due to its versatility, efficiency, and cost-effectiveness. Mesoporous silica stands out as a leading adsorbent due to its porous structure and surface modification potential, yet there remains a critical need to systematically analyze and evaluate recent innovations in this rapidly evolving field. This review addresses this knowledge gap by consolidating and critically analyzing the recent literature on functionalized mesoporous silica for heavy metal ion removal from wastewater. Through a detailed examination of the maximum adsorption efficiencies of organically modified mesoporous silica-based materials for various metal ions, with particular emphasis on recent trends in removing heavy metal ions, this work provides a crucial foundation for understanding current capabilities and limitations. Additionally, by identifying patterns in synthesis methods, surface functionalization strategies, and performance metrics across different studies, this review offers valuable insights into optimization opportunities and unexplored research directions. The systematic compilation and analysis of these data will serve as an essential resource for researchers working to advance the design and application of modified mesoporous silica for diverse water pollutant removal applications.

Keywords: mesoporous silica; functional groups; heavy meals; water treatment

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1. Introduction

Water, the foundation of life, is increasingly threatened by worldwide pollution and scarcity, mostly as a result of unregulated industrial growth, rapid population rise, and widespread environmental degradation [1,2]. Chemical-induced water pollution has grown to be a significant issue and a top priority for the industrial world as a whole, as well as for society and government agencies [3]. There are numerous ways to define water pollution. Water pollution happens when one or more substances that have a harmful impact on the water are released into it. These compounds have the potential to damage the environment and all living organisms, as well as their ecosystems.

It is possible to view the two primary sources of water pollution as point and non-point. Pollutants from a single source, such as those released into the water by industries, are referred to as the first type; pollutants from several sources are referred to as the second type [4]. Because of their toxicity and availability, heavy metals pose a major threat to the environment. It has been established that the presence of heavy metals in aquatic environments can lead to a number of health issues for both humans and animals [5]. Because they are found in sludge, fertilizers, pesticides, municipal garbage, and mine residues, heavy metals—the primary class of inorganic pollutants—contaminate vast land areas and, as a result, water bodies [6]. Due to their non-degradable, persistent, and accumulative nature, the majority of heavy metals are known to cause cancer and may pose a major risk to the living population [7–9]. However, some heavy metals are necessary for life and have an irreplaceable function in the human metabolic system, but when they are present in excess, they become toxic [10].

The ability to sustain basic living standards and participate in productive activities is hampered by the severe freshwater shortages that more than 15% of the world's population are facing [11,12]. Therefore, it is crucial to purify contaminated water before releasing it into the environment. Numerous conventional methods, including filtration, reverse osmosis, coagulation, and ozone treatment, have been suggested to eliminate these organic contaminants from water. Despite their relative effectiveness, these techniques have drawbacks, including significant energy consumption and the possibility of secondary contamination [13–15].

Adsorption, on the other hand, is considered an efficient method to remove contaminants, such as heavy metals. In addition to their performance simplicity and reasonable price, adsorption processes are considered effective in removing heavy metals from aqueous media, particularly when they are present in trace amounts [16]. Desorption, which is the inverse process, eluted adsorbate ions from the adsorbent surface to the solution and frequently occurs in conjunction with adsorption. The reversibility of adsorption can be assessed based on the amount of adsorbate that is desorbed from the adsorbent; the more adsorbate that is desorbed, the more reversible the adsorption process is [17]. There are two types of adsorption: chemical (chemisorption), which is brought on by chemical reactions between the adsorbent and the adsorbate that form covalent or ionic bonds, and physical, where the increase in the adsorbate concentration at the interface is caused by non-specific (i.e., not dependent on the substance nature) van der Waals forces [18,19]. To successfully remove the target contaminants from wastewater effluents, adsorbent materials should have a high adsorption interaction with the contaminants. Activated carbons, zeolites, clay minerals, industrial byproducts, agricultural waste, biomass, and polymeric materials are examples of adsorbents that can have a mineral, organic, or biological origin [20].

Nanotechnology has been used in practically every field of science and technology over the last 20 years. Currently, a variety of nanostructure-based materials, including carbon nanotubes, fullerenes, and mesoporous silica, have been created and applied to the removal of the pollutants in question from water [21–23]. Because of their wide surface area, narrow pore-size distributions, and regulated pore sizes, mesoporous silicas, such as MCM-48, MCM-41, HMS, and SBA-15, are regarded as preferable for these kinds of applications [24]. MCM-41, MCM-48, HMS, and SBA-15 are all considered excellent candidates for heavy metal adsorption due to their shared characteristics of high surface area, ordered pore structures, and tunable pore sizes. These features provide abundant binding sites, minimize diffusion limitations, and allow for selective adsorption of target metal ions. Furthermore, the chemical stability of their silica framework and the ease with which their surfaces can be functionalized to introduce specific binding sites contribute to their effectiveness in removing heavy metals from wastewater. In essence, the combination of high capacity, selectivity, and stability makes these mesoporous materials highly desirable for heavy metal remediation [23,24].

Mesoporous materials are generated using a template method that generates surfactant micelles or block copolymers in a solvent–silica mixture. A material composed of a variety of regular pores is obtained after the silica is polymerized and the organic micelles are removed. Through the combination of nitrogen adsorption–desorption tests, X-ray diffraction, and transmission electronic microscopy (TEM), the properties of mesoporous micelle templated silicas have been thoroughly investigated. According to IUPAC nomenclature, mesoporous materials can be defined as nanostructures with pore diameters ranging from 2–50 nm. Additionally, adding appropriate functional groups to the surface improves its affinity for the target metal ions [25]. The significance of mesoporous materials stemmed from their pore sizes, which, depending on pore shape, permit controlled accessibility for big molecules. In addition, the open pore structure of ordered mesoporous materials facilitates better target molecule access and transfer than bottleneck pores of amorphous materials, which impede molecular passage to the adsorption sites [26].

Attempts have been undertaken to address a number of water treatment concerns with mesoporous silica, taking into consideration the significance of water quality and the new advantages of nanotechnology. In this sense, these nanomaterials might offer chances to develop innovative solutions to the issue of water pollution. Taking into account the aforementioned points, this research reviews the utilization of different types of mesoporous silica materials (MCM, HMS, and SBA materials) as adsorbents to remove heavy metals from wastewater effluents. Moreover, a comparison of these materials' adsorption capacities for the pollutants under consideration is given.

This review distinguishes itself through its comprehensive and critical analysis of recent advances in this rapidly evolving field. While previous reviews may have focused on specific types of mesoporous materials or individual heavy metals, this work offers a broader perspective by systematically examining the diverse range of modifications employed to enhance heavy metal adsorption. The novelty lies in its integrated approach, critically comparing the efficacy of various modification strategies—including surface functionalization, and nanocomposite formation—across a spectrum of mesoporous materials. Furthermore, this review delves into the underlying mechanisms governing adsorption, providing insights into the structure–activity relationships that dictate performance. By synthesizing the latest research findings and identifying key trends and challenges, this review not only provides a valuable resource for researchers but also highlights promising future directions in the development of next-generation adsorbents for heavy metal remediation.

2. Modification of Mesoporous Silica

Mesoporous silica materials possess unique properties, such as a high surface area, tunable pore size, and chemical inertness, making them attractive for various applications.

However, their surface silanol groups can limit their compatibility with certain substances. Organic modification of mesoporous silica aims to address this by introducing organic functional groups onto the silica surface, thereby tailoring its properties for specific applications.

The two primary methods that have been thoroughly researched for modifying the mesoporous materials via covalent bonding of the organic functionality and the silica are co-condensation (one-pot synthesis) and grafting (post-synthesis). Furthermore, periodic mesoporous organosilicates—mesoporous materials with functional groups inside the framework—have also been employed to remove heavy metals [27]. Grafting is a widely used method where organic molecules are attached to the silica surface through covalent bonds. This is typically achieved by reacting surface silanol groups (Si-OH) with organosilanes, which are molecules with both organic and silicon-containing groups. The reaction results in the formation of a Si-O-Si bond, linking the organic molecule to the silica framework.

In the co-condensation method, the organic molecules are incorporated into the silica framework during the synthesis of the mesoporous material itself. This is achieved using a mixture of silica precursors (such as tetraethyl orthosilicate, TEOS) and organosilanes as the starting materials. The co-condensation process leads to the formation of a silica network with organic molecules distributed throughout.

A variety of functional groups can be incorporated into the walls of the pre-synthesized mesoporous silica using the post-synthesis grafting technique (Figure 1). The structural features and chemical makeup of the functionalized materials made via grafting affect their properties [28]. Mesoporous silicas can be modified by grafting using a variety of organic functionalities. The proper selection of the organic group is a significant factor influencing the performance of grafted adsorbents. Grafting's primary drawback is that it shrinks the materials' pores, particularly when large or numerous functional groups are added [29–33]. This limits the diffusion to the adsorption sites. The amount of Si-OH bonded to the surface typically limits modification, which is another drawback of adding functional groups by grafting [34].

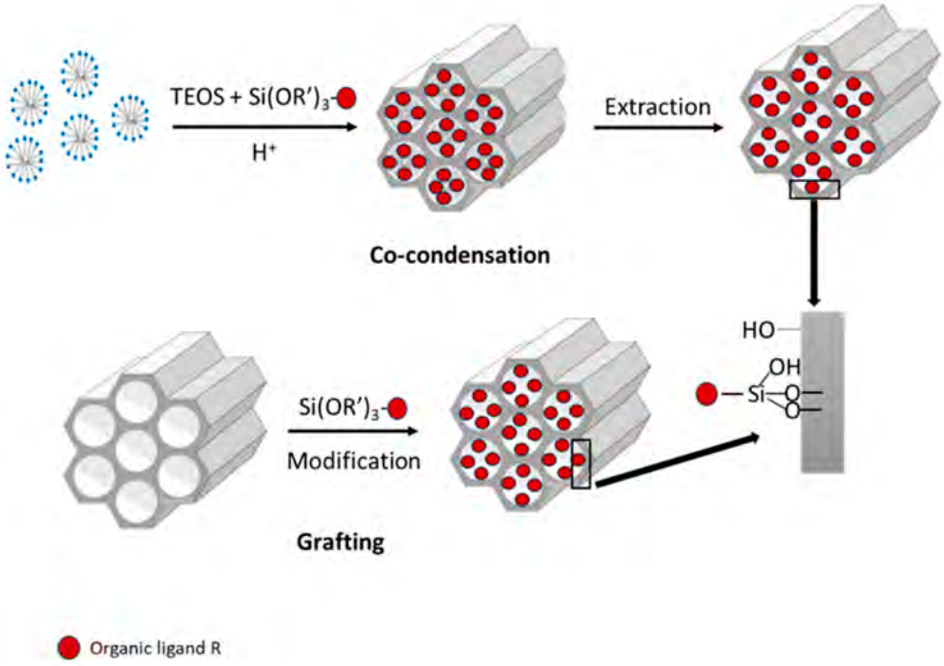

Figure 1. Modification routes of mesoporous silica: co-condensation versus grafting.

Co-condensation, in which the organosilane and silica precursor are condensed together, is another important method for adding functional moieties (Figure 1). Faster synthesis and the uniform distribution of functional groups are advantages of co-condensation. Additionally, it immobilizes a large number of functional groups onto the mesoporous silica surface with remarkable efficiency. Higher performance would probably come from increasing the number of functional groups while keeping the pore structure open. However, the co-condensation process has a number of drawbacks. In general, the degree of mesoscopic order of the products declines with increasing concentration of $(R'O)_3SiR$ in the reaction mixture, eventually leading to completely disordered products. The organic functionalities in modified silica phases are typically less than 40 mol%. Furthermore, the amount of terminal organic groups absorbed into the pore–wall network is often lower than what would be expected given the reaction mixture's starting concentration. Furthermore, increasing the loading of integrated organic groups can reduce pore width, pore volume, and specific surface area [35]. Another downside of the co-condensation process is that significant care must be taken to protect organic functionality during surfactant removal, necessitating the employment of only extractive methods [36].

Heavy metal adsorption on organically modified mesoporous silica occurs through a combination of complex physical and chemical interactions. Electrostatic interactions, including charge attraction between metal cations and negatively charged modifiers, and ion exchange, where metal ions replace other bound ions, play a significant role. Complexation or chelation, involving the formation of coordinate bonds between metal ions and ligand-containing organic modifiers, contributes significantly to adsorption, with chelation, facilitated by modifiers with multiple binding sites, resulting in stronger binding. Physical adsorption, driven by weaker forces, like van der Waals and hydrophobic interactions, along with pore filling within the mesoporous structure, also contributes. Additional interactions, such as hydrogen bonding and hydrophobic interactions between metal ions, water, and modifiers, can further influence the process [37]. The specific adsorption mechanism is affected by various factors, like the type of organic modifier, metal ion properties, pH, ionic strength, and temperature. A thorough understanding of these mechanisms is crucial for designing effective adsorbents, optimizing adsorption conditions, and developing efficient regeneration strategies.

3. SBA-15 and SBA-16

These two types of mesoporous materials are formed using nonionic triblock copolymer surfactants of the type $EO_xPO_yEO_x$ [38,39]. Because it is affordable, easily separated, harmless, and biodegradable, this kind of surfactant is extremely interesting. When using these surfactants in the synthesis process, the interaction usually takes place in low-pH ($pH \leq 2$) solutions using the $S^0H^+X^-I^+$ mechanism, where S^0H^+ is the surfactant hydrogen-bonded to a hydronium ion, X^- is the chloride ion, and I^+ is the protonated silica. SBA materials as SBA-15 and SBA-16 are, thus, obtained. The SBA-15 has long 1D channels (*P6mm* plane group) with hexagonal pores in a 2D array. The *Im3hm* space group is represented by the 3D cubic arrangement of mesopores in SBA-16. High-resolution electron microscopy demonstrates that every mesopore in this body-centered cubic structure is connected to its eight adjacent pores [40]. Both materials have thick walls (up to 9 nm) [41] and big pores (up to 30 nm for SBA-15) [38,39], the latter of which increase the stability of these structures [42].

The creation of heavy metal adsorbents requires merging appropriate organic compounds with strong reactivity toward the target metal ions with the open pore structure of mesoporous silica. The proper selection of the organic group is one important component influencing the performance of grafted adsorbents. Several studies (Table 1) investigated the efficiency of functionalization of SBA-15 and ABS-16 with organic groups for heavy metals removal. The adsorption conditions employed are clearly presented in the table, including details of pH, initial metal concentration, and adsorbent dose. The specific metals adsorbed in the study are explicitly named. Furthermore, the adsorption capacity is consistently reported in mg/g, providing a standardized unit for comparison. The method used to determine this adsorption capacity is also detailed; it was derived from experimental isotherm data. These data provide a comprehensive and transparent description of the adsorption process and results, strengthening the validity of the presented data.

Table 1. Heavy metals adsorption on modified SBA-15 and SBA-16.

Material	Chelating Agent	Adsorption Conditions	Metals Adsorbed	Adsorption Capacity (mg/g)	Ref.
SBA-15	N-Propylsalicylaldimino	C_i: 4 mmol/L, 0.05 g adsorbent, pH 4.8	Cu^{2+}, Zn^{2+}, Ni^{2+}, and Co^{2+}	46, 22, 19, and 26	[43]
SBA-15	Amine	C_i: 10 mg/L, 0.2 g adsorbent, pH 5	Pb^{2+} and Cd^{2+}	39 and 41	[44]
SBA-15	Melamine-based dendrimer amines	C_i: 100 mg/L, 1 g/L adsorbent, pH 4	Pb^{2+}, Cu^{2+}, and Cd^{2+}	94.8, 90.3, and 71.1	[45]
SBA-15	Trimethoxysilylpropyl diethylenetriamine	C_i: 11 mg/L, 10 g/L adsorbent, pH 4	Pb^{2+}, Cu^{2+}, and Cd^{2+}	1.1, 1, and 1.1	[46]
SBA-15	2-mercaptopyrimidine	C_i: 0.01 M, 0.2 g, adsorbent, pH 6	Cd^{2+}	111.3	[47]
SBA-15	Amine	C_i: 100 mg/L, 0.1 g adsorbent, pH 5	Pb^{2+}, Cr^{3+}, Cu^{2+}, Cd^{2+}, Ni^{2+}, and Co^{2+}	0.12, 0.06, 0.11, 7.9, 16.7, and 22.4	[48]
SBA-15	Cyanex 272	C_i: 100 mg/L, 0.15 g adsorbent, pH 4.8	Co^{2+}, Cu^{2+}, and Zn^{2+}	31.8, 100, and 61.4	[49]
SBA-15	Amine	C_i: 10 mg/L, 0.03 g adsorbent, pH 5	Pb^{2+}, Cd^{2+}, and Ag^{+}	29.4, 111.1, and 100	[50]
SBA-15	Schiff base	C_i: 100 mg/L, 1 g/L adsorbent, pH 5	Cu^{2+}, Pb^{2+}, and Ni^{2+}	128, 175, and 72	[51]
SBA-15	Amine	C_i: 100 mg/L, 0.1 g adsorbent, pH 5	Ni^{2+}	64.5	[52]
SBA-15	Urea	C_i: 100 mg/L, 2.5 g/L adsorbent, pH 2.5, 4, 5	Cr^{6+}, Cd^{2+}, and Pb^{2+}	26.83, 30.53, and 43.85	[53]
SBA-15	Imidazole-derivatized	C_i: 100 mg/L, 0.05 g adsorbent, pH 5	Cr^{6+}	113	[54]
SBA-15	N-propylsalicylaldimine	C_i: 10 mg/L, 0.04 g adsorbent, pH 2.5	Cr^{6+}, As^{5+}, and Hg^{2+}	79.58, 20.63, and 7.59	[55]
SBA-15	2-acetylthiophene	C_i: 10 mg/L, 0.05 g adsorbent, pH 6	Cr^{3+}	114.2	[56]
SBA-15	Pyridyl Schiff base	C_i: 100 mg/L, 0.04 g adsorbent, pH 5	Cu^{2+} and Pb^{2+}	48.26 and 106.62	[57]
SBA-15	EDTA	C_i: 0.47 mmol/L, 0.02 g adsorbent, pH 6	Cu^{2+}, Ni^{2+}, Cd^{2+}, and Pb^{2+}	88.2, 58.6, 123.6, and 232	[58]
SBA-15	Bis-schiff	C_i: 100 mg/L, 0.05 g adsorbent, pH 6	Pb^{2+}	151.5	[59]
SBA-15	Thiazole Schiff base	C_i: 100 mg/L, 1 mg/mL adsorbent, pH 6	Pb^{2+}	152.9	[60]
SBA-15	EDTA	C_i: 30 mg/L, 0.02 g adsorbent, pH 6	Zn^{2+}	107	[22]
SBA-15	Bis-salicylaldehyde Schiff base	C_i: 100 mg/L, 0.04 g adsorbent, pH 5	Pb^{2+} and Cu^{2+}	60.9 and 36.6	[61]
SBA-15	2,4 dinitrophenylhydrazine	C_i: 45 mg/L, 0.02 g adsorbent, pH 5	Pb^{2+}, Cr^{3+}, Cd^{2+}, and Co^{2+}	242.50, 214.72, 187.86, and 166.46	[62]
SBA-15	1-(2-thiazolylazo)-2-naphthol	C_i: 100 ng/mL, 0.05 g adsorbent, pH 6	Ni^{2+}	Detection limit of 1.8 ng mL^{-1}	[63]
SBA-16	Guanine	C_i: 100 mg/mL, 20 mg for Pb^{2+} and Hg^{2+}, and 25 mg for Cd^{2+}, pH 5 for Hg^{2+} and Cd^{2+}, 8 for Pb^{2+}	Pb^{2+}, Hg^{2+}, and Cd^{2+}	289.9, 259.9, and 228.8	[64]
SBA-16	–SH groups	C_i: 127 mg/mL, 0.05 g adsorbent, pH 6	Cu^{2+}	36.38	[65]
SBA-16	Maltodextrin	pH 6	Cd^{2+}, Zn^{2}, and Cu^{2+}	575.53, 564.67, and 509.73	[66]
SBA-16	Aminopropyl groups	C_i: 200 mg/mL, 0.1 g adsorbent, pH 5	Pb^{2+}	90	[67]
SBA-16	EDTA	C_i: 30 mg/L, 0.02 g adsorbent, pH 6	Zn^{2+}	184.1	[22]

Note: C_i: Initial metal concentration.

The diverse array of chelating agents listed, while seemingly comprehensive, requires more than mere enumeration. Schiff bases, formed by the condensation of amines and aldehydes/ketones, offer versatile metal coordination through their imine nitrogen and oxygen atoms, finding applications in extraction, catalysis, and environmental remediation. Amines, including specific examples, like aminopropyl groups, chelate via their nitrogen lone pairs and are often used for surface modification. While urea possesses some chelating ability through its carbonyl and amine groups, it is less potent than others. Imidazole derivatives, including the generalized "imidazole-derivatized" term, coordinate through their ring nitrogens and are crucial in biological systems and catalysis. Other organic ligands, like 2-acetylthiophene, 2,4-dinitrophenylhydrazine, 1-(2-thiazolylazo)-2-naphthol, guanine, and thiol (-SH) groups, offer diverse coordination sites (S, N, O) and varying selectivity for metal ions. EDTA, a ubiquitous and strong chelating agent with multiple carboxylate and amine groups, forms stable complexes with a wide range of metals. Even polysaccharides, like maltodextrin, though weaker chelators via their hydroxyl groups, can be useful in specific contexts. It is crucial to move beyond simply listing these agents and to delve into comparative analyses of their strengths, weaknesses, target metal specificities, and relevant applications within each chapter's context, providing a meaningful discussion of structure–activity relationships, mechanisms, and trends [22,43–67].

4. MCM-41 and MCM-48

MCM-41 and MCM-48 are mesoporous silica materials renowned for their high surface areas, uniform and tunable pore sizes, and ordered pore structures, making them attractive for diverse applications. MCM-41 features a hexagonal arrangement of cylindrical pores, resembling a honeycomb, while MCM-48 boasts a more complex body-centered cubic arrangement of interconnected cages, leading to even higher surface areas and pore volumes. Although both materials share applications in adsorption, catalysis, and drug delivery, their distinct pore architectures influence their performance; MCM-48's intricate structure may offer advantages for larger molecules or complex reactions, while MCM-41 synthesis is generally more straightforward, and producing defect-free MCM-48 can be more challenging. Ultimately, the choice between these materials hinges on the specific application requirements, including pore size, arrangement, and synthetic feasibility.

The synthesis of adsorbents with tailored properties is one of the main goals of the M41S family of ordered mesoporous adsorbents, which have garnered a lot of attention since their discovery by Mobil Oil scientists [68]. The pore widths of M41S materials range from 2 to 10 nm, and their pore walls are amorphous [69]. The key to creating such adsorbents is the use of organic templates, which form organic–inorganic mesophases. The two most studied materials, MCM-41 (Mobil composition of matter) with a 2D hexagonal structure and MCM-48 with a 3D cubic structure, are synthesized using n-alkylammonium salts as templates [70]. Whereas the MCM-41 surface is concave only, the MCM-48 surface is composed of both concave and convex regions. MCM-48 is made up of short cylindrical pores that are joined together, while MCM-41 is made up of long cylindrical pores [71]. Several studies (Table 2) investigated the efficiency of functionalization of MCM-41 and MCM-48 with organic groups for heavy metals removal.

A variety of chelating agents are employed for metal binding, each with unique properties and applications. Molybdophosphate, a complex inorganic compound, can interact with metal ions, while polyethyleneimine, a polymer with numerous amine groups, offers multiple binding sites. Long-chain amines, like N,N-dimethyldodecylamine (DMDDA) and dodecylamine (DDA), often used in combination, can enhance metal removal through certain mechanisms, like micelle formation. TiO_2, a metal oxide, exhibits surface hydroxyl groups that can bind metal ions. Organosilanes, like aminopropyl-Si and 3-[2-(2-aminoethylamino)ethylamino]-propyl-trimethoxysilane (DTES), along with simpler amines (NH2, amine, amino, diamino), provide amine functionalities for metal coordination. Thiol (-SH, thiol, sulfhydryl) groups demonstrate a strong affinity for heavy metals. Titanium (Ti) in various forms can act as a metal-binding site. Polymers, like β-cyclodextrin polymer and chitosan (CH) combined with β-cyclodextrin (CD), offer multiple binding sites and can enhance selectivity. Dendrimer amines based on [1,3,5]-triazines provide a branched structure with numerous amine groups for efficient metal binding. Finally, benzoylthiourea, containing both amine and thiourea functionalities, offers versatile metal coordination [72–97].

Table 2. Heavy metals adsorption on modified MCM-41 and MCM-48.

Material	Chelating Agent	Adsorption Conditions	Metals Adsorbed	Adsorption Capacity (mg/g)	Ref.
MCM-41	Molybdophosphate	C_i: 10 mg/L, 0.02 g adsorbent, pH 5	Pb^{2+}, Cu^{2+}, and Cd^{2+}	63.17, 60.53, and 4.05	[72]
MCM-41	Polyethyleneimine		Cu^{2+}, Cd^{2+}, and Ni^{2+}	39.30, 21.10, and 33.67	[73]
MCM-41	N-N dimethyldodecylamine (DMDDA) and dodecylamine (DDA)	C_i: 100 mg/L, 0.1 g adsorbent, pH 5	Cu^{2+}, Pb^{2+}, Cd^{2+}, and Co^{2+}	DMDDA: 259.2, 267, 191.1, and 145.1 DDA: 249.5, 219.4, 153.9, and 125.1	[74]
MCM-48	DMDDA and DDA	C_i: 100 mg/L, 0.1 g adsorbent, pH 5	Cu^{2+}, Pb^{2+}, Cd^{2+}, and Co^{2+}	DMDDA: 259.1, 238, 175.34, and 136.3 DDA: 222.9, 194.6, 148.36, and 121.5	[75]
MCM-41	TiO_2	C_i: 100 mg/L, 0.05 g adsorbent, pH 5.5	Cr^{6+}	120.64	[76]
MCM-41	Aminopropyl–Si	C_i: 10 mg/L, 1 g adsorbent, pH 4	Ni^{2+} and Cu^{2+}		[77]
MCM-41	NH_2	C_i: 100 mg/L, 0.05 g adsorbent, pH 4.5 to 5.2	Co^{2+}, Ni^{2+}, Cu^{2+}, and Zn^{2+}	37.76, 41, 48.3, and 47.72	[78]
MCM-41	Thiol	C_i: 100 mg/L, 0.1 g adsorbent, pH 5	Cu^{2+}, Pb^{2+}, Ag^{+}, and Cr^{3+}	63.2, 123.36, 90.3, and 15.71	[79]
MCM-48	Amine	C_i: 50 mg/L, 0.1 g adsorbent, pH 6	Cu^{2+} and Mn^{2+}	33.02 and 44	[80]
MCM-41	-	C_i: 50 mg/L, 0.1 g adsorbent, pH 7	Cr^{6+} and Zn^{2+}	392 and 554	[81]
MCM-41	Amino	1 g, adsorbent, pH 2	Cr^{6+}	86.4	[82]
MCM-41	EDTA	0.1 g, adsorbent, pH 3.1	Cu^{2+}, Zn^{2+}, and Ni^{2+}	79.36, 74.07, and 67.56	[83]
MCM-48	Ti	C_i: 50 mg/L, 0.05 g adsorbent, pH 8	Cd^{2+}	83.57	[84]
MCM-48	Aminopropyl	C_i: 10 mg/L, 0.05 g adsorbent, pH 7	Mn^{2+}, Ni^{2+}, Cu^{2+}, Cd^{2+}, and Pb^{2+}	63.3, 74.1, 96.2, 158.7, and 322.6	[85]
MCM-48	Amine	C_i: 50 mg/L, 0.5 g adsorbent, pH 6	Cd^{2+} and Pb^{2+}	82.7 and 119.24	[86]
MCM-48	-	C_i: 2 mg/L, 0.1 g adsorbent, pH 4.5	Pb^{2+}	386.84	[87]
MCM-48	β-cyclodextrin polymer	C_i: 100 mg/L, 0.5 g adsorbent, pH 6	As^{5+} and Hg^{2+}	265.6 and 207.9	[88]
MCM-48	Amino- and thiol	C_i: 100 mg/L, 0.5 g adsorbent, pH 5.5	Pb^{2+}	MCM-48-NH_2:75.2 and MCM-48-SH: 31.2	[89]
MCM-48	Chitosan (CH) and β-cyclodextrin (CD)	C_i: 50 mg/L, 0.02 g adsorbent, pH 7	Cd^{2+}	CH/MCM: 122.4 and CD/MCM: 152.2	[90]
MCM-48	Sulfhydryl	C_i: 80 mg/L, 0.1 g adsorbent, pH 2	Cr^{6+}	17.9	[91]
MCM-48	Ti	C_i: 10 mg/L, 0.05 g adsorbent, pH 8	Co^{2+} and Ni^{2+}	9.870 and 22.94	[92]
MCM-48	Dendrimer amines based on [1,3,5]-triazines	C_i: 100 mg/L, 1 g adsorbent, pH 5	Ag^{+}	169.49	[93]
MCM-48	Diamino	C_i: 100 mg/L, 0.05 g adsorbent, pH 2.5	Cr^{6+}	166.67	[94]
MCM-48	Benzoylthiourea	0.05 g adsorbent	Hg^{2+}	1343.9	[95]
MCM-48	Amine	pH: 6 for Zn^{2+} and 4 for Ni^{2+}	Zn^{2+} and Ni^{2+}	36 and 25.24	[96]
MCM-48	3-[2-(2-aminoethylamino) ethylamino]-propyl-trimethoxysilane (DTES)	C_i: 10 mg/L, 0.03 g adsorbent, pH 7	Zn^{2+}	83.33	[97]

5. Hexagonal Mesoporous Silica HMS

Hexagonal mesoporous silica (HMS) is a mesoporous material characterized by a hexagonal arrangement of pores, though its pore structure and connectivity can differ from other similar materials, like MCM-41. Distinguished by its amorphous silica walls, HMS often exhibits larger pore sizes, making it well-suited for adsorbing larger molecules, while still maintaining the high surface area and pore volume crucial for applications in adsorption, catalysis, and drug delivery. Synthesized typically under basic conditions using a sol–gel method with structure-directing agents, HMS offers tunable pore sizes adaptable to specific needs. While its amorphous walls can lead to lower thermal stability compared to some crystalline mesoporous materials, its ease of synthesis and large pore size remain advantageous, particularly for applications involving bulkier molecules. HMS with wormhole-like framework structures can be created by hydrogen bonding interactions and self-assembly between neutral amine surfactants (S°) or nonionic PEO-based surfactants (N°) and neutral inorganic precursors (I°). HMS silica has homogeneous mesoporous channels and a large surface area, just like MCM-41 [98]. However, HMS channels were constructed over a much shorter range in a hexagonal-like form than the long-range hexagonal framework structures of MCM-41. It has been demonstrated that these channels have sponge-like or wormhole-like framework structures [99]. Pinnavaia and his colleagues [100] have investigated the effects of preparation conditions on the characteristics of HMS silicas assembled through the S°I° pathway in water–alcohol cosolvent systems, using dodecylamine (DDA) as S° and TEOS as I° [101,102]. The results showed that the ratio of textural (interparticle) porosity to framework pore volume of HMS could be controlled by varying the water–ethanol ratio in the solvent systems. HMS silica, like MCM-41, has a high surface area and uniform mesoporous channels, but they were arranged over a much shorter range in a hexagonal-like fashion and have been demonstrated to possess wormhole-like or sponge-like framework structures. Several studies (Table 3) investigated the efficiency of functionalization of HMS with organic groups for heavy metals removal.

Table 3. Heavy metals adsorption on modified HMS.

Material	Chelating Agent	Adsorption Conditions	Metals Adsorbed	Adsorption Capacity (mg/g)	Ref.
HMS	5-mercapto-1-methyl-1-H-tetrazol	C_i: 43.5 mg/L, 0.1 g adsorbent, pH 8	Pb^{2+}	29	[103]
HMS	Thiol	C_i: 35 mg/L, 0.01 g adsorbent	Hg^{2+}	300.9	[104]
HMS	Schiff base (C16—hexadecylamine)	C_i: 100 mg/L, 0.025 g adsorbent, pH 5	Pb^{2+}	181.81	[105]
HMS	Amino	C_i: 41.44 mg/L, pH 4	Pb^{2+}	90.7	[106]
HMS	N-(2-aminoethyl)-3-aminopropyl methyldimethoxysilane	C_i: 50 mg/L, 1 g adsorbent, pH 5	Pb^{2+}	119	[107]
HMS	Polyaniline/polypyrrole	C_i: 50 mg/L, 0.04 g adsorbent, pH 8	Cd^{2+}	384.61	[108]

Several chelating agents demonstrate effectiveness in metal binding. 5-mercapto-1-methyl-1-H-tetrazole and thiol groups offer strong affinity for various metal ions through their sulfur atoms. A Schiff base derived from C16-hexadecylamine provides a nitrogen and oxygen coordination site, potentially enhancing selectivity due to the long alkyl chain. Amino groups, including those present in N-(2-aminoethyl)-3-aminopropyl methyldimethoxysilane, offer nitrogen-based coordination. Conductive polymers, like polyaniline and its composite with polypyrrole, can complex metal ions through their nitrogen atoms and other functional groups, offering the potential for electrochemical sensing and removal applications [105–109].

6. Adsorbents Regeneration

Adsorbent regeneration is a crucial aspect of adsorption processes, enabling the reuse of spent adsorbents and making the overall process more economically and environmentally sustainable. Various regeneration methods exist, each tailored to the specific adsorbent and adsorbate, such as chemical regeneration, which utilizes solvents or solutions to remove the adsorbed species. The choice of regeneration method depends on various factors, like the strength of the adsorption bond, the stability of the adsorbent and adsorbate, and economic considerations. Effective regeneration restores the adsorbent's initial adsorption capacity, extending its lifespan and minimizing waste generation.

The adsorbent must have a consistent metal adsorption capacity across a number of adsorption/desorption cycles and be structurally stable for commercial uses in an adsorption system. Desorption provides crucial information regarding the reversibility of the adsorption process in addition to reusing adsorbents. Numerous researchers reported recycling the adsorbents they used, primarily by treating them with complexing agents, like EDTA [110,111], to elute the metal ions, washing them with HCl solution [43,65,112–118], or using HNO_3 solution [119,120]. The neutral amine should be recovered using a $NaHCO_3$ solution following metal desorption with acids. The adsorbent surface becomes protonated as a result of acid treatment, increasing the positive surface charge density [121]. Structural instability of the adsorbent, which may lose its structural order and adsorption sites accessible within the pores after regeneration treatment, is another potential cause of adsorption capacity loss [114]. It was shown that while the pore size remained constant, the surface area and pore volume of the SBA-15(SH) and SBA-15(NH_2) adsorbents slightly (by 5%) decreased. Although MCM-41(SH) and MCM-41(NH_2) adsorbents showed a significant decrease in surface area and pore volume, his result was in perfect agreement with earlier findings that SBA-15 is more stable than MCM-41 [122] and could account for the lower regeneration performance of the MCM-41-based adsorbent in comparison to SBA-15.

7. Challenges and Future Perspectives

While organically modified mesoporous silica materials hold great promise for heavy metal adsorption, several challenges remain. The organic groups grafted onto the silica surface can be susceptible to degradation under certain conditions (e.g., extreme pH, high temperatures, or in the presence of oxidizing agents). This can lead to a decrease in adsorption capacity over time. Some organic groups might leach from the silica matrix into the solution, potentially causing secondary pollution and reducing the adsorbent's effectiveness. If the organic modifiers are too bulky or densely packed, they might hinder the access of heavy metal ions to the adsorption sites within the pores of the mesoporous silica. While some modifications enhance selectivity for specific heavy metals, achieving high selectivity for a particular metal in a mixture of contaminants can be challenging. Regenerating organically modified mesoporous adsorbents can be more complex than regenerating unmodified silica materials. The regeneration process must not damage the organic modifiers or alter their properties. The synthesis and modification of mesoporous silica can be more expensive than using other adsorbents. The cost-effectiveness of these materials needs to be considered for large-scale applications. Scaling up the production of organically modified mesoporous silica with consistent properties and performance can be challenging.

Addressing these challenges is crucial for the successful implementation of organically modified mesoporous silica materials in heavy metal removal applications. Research efforts are focused on developing more stable organic modifiers, improving synthesis methods to minimize leaching, optimizing the structure and arrangement of organic groups, enhancing selectivity, developing efficient regeneration strategies, and reducing the overall cost of these materials.

Future research in heavy metal adsorption on organically modified mesoporous silica should prioritize developing novel, more stable, and selective organic modifiers tailored for specific heavy metal targets. Exploration of stimuli-responsive materials that facilitate easy regeneration and minimize leaching is crucial for sustainable applications. Advanced characterization techniques should be employed to gain deeper insights into the interactions between heavy metals and the modified silica surface, guiding the design of more effective adsorbents. Computational modeling can play a significant role in predicting adsorption behavior and optimizing material design. Furthermore, research should focus on cost-effective and scalable synthesis methods to enable widespread implementation. Finally, evaluating the long-term performance and stability of these materials under real-world conditions, including complex matrices and varying environmental factors, is essential for their practical application in heavy metal remediation.

To sum up, organically functionalized mesoporous silica holds great promise for heavy metal remediation, but realizing its full potential requires addressing key challenges. For a real technological breakthrough, functionalized mesoporous materials must exhibit a synergistic combination of properties: high selectivity for target heavy metals in complex matrices, robust stability under varying pH and redox conditions to prevent leaching or degradation of the organic ligands, high adsorption capacity to minimize material usage, and efficient regenerability for cost-effectiveness. Furthermore, facile and scalable synthesis routes are essential for practical applications. The earliest commercial implementations are likely to emerge in specialized water treatment applications where the high performance justifies the potentially higher cost. These could include developing selective adsorbing cartridges for removing specific heavy metals from industrial wastewater or creating highly sensitive sensors for rapid on-site detection of heavy metal contamination. However, several obstacles currently hinder industrial-scale deployment. The cost of functionalized mesoporous silica, especially for high-performance materials, remains a significant barrier. Scaling up synthesis to produce consistent, high-quality material at a competitive price is crucial. Long-term stability and performance under real-world conditions, including complex water matrices and varying flow rates, require further investigation. Finally, public perception and regulatory approval play a role; demonstrating the safety and environmental benignity of these materials is essential for widespread adoption.

8. Conclusions

Organically functionalized mesoporous silicas hold considerable promise as adsorbents for heavy metal removal due to their ordered porous structures, which provide access to functional groups and large surface areas. This inherent potential has driven significant research interest in their application for heavy metal adsorption, with modifications tailored to target ions to enhance binding strength and selectivity. However, despite numerous studies and promising results, these materials have yet to achieve widespread practical application. Two primary challenges hinder their commercialization: first, developing high-selectivity adsorbents that also exhibit robust stability and effective recyclability; and, second, designing mesoporous materials with sufficient adsorption capacity for large-volume discharges, particularly in continuous flow systems. The practical implementation of these materials is further complicated by difficulties in handling powdered adsorbents, both in column packing and post-adsorption filtration in batch processes, necessitating research into particle size management. The limited commercial availability of specialized organosilane reagents needed for targeted selectivity presents another obstacle, demanding the design and synthesis of application-specific functionalized reagents. Enhancing the long-term durability of the adsorbents to withstand structural degradation during adsorption and regeneration cycles is also crucial. Finally, the economic viability of these materials, particularly regarding the cost of newly developed, high-performance mesoporous silica compounds at scale, remains a key question that must be addressed for the successful and sustainable implementation of modified mesoporous silica-based water treatment technologies.

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