

Highly Enhanced Pb(II) Removal By Mercaptopropyl Trimethoxysilane (MPTMS) Surface-Modified Silica Aerogel: Synthesis, Characterization and Isotherm Studies

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In the present work, the surface modification of silica aerogel was done using mercaptopropyl trimethoxysilane (MPTMS). The synthesized aerogel was characterized using FTIR, TGA and BET. The performance of the modified silica aerogel for Pb(II) removal from aqueous solution was investigated using pH, metal concentration, and adsorbent dose experiments. According to the results, the respective aerogel performed excellent with maximum adsorption capacity (q_m) of 240 mg g⁻¹. Comparing to the 14 other relevant studies, our results was the second best in terms of significantly higher q_m ; the value was greater than organic ligand, AFSG, diethylenetriaminopropyl, ethylenediaminopropyl, aminopropyl, PEI, AAAPTS, rGO, thiamine, bispyrazole, DETA, and 2-furoyl functionalized silica gels. According to the isotherm studies, the adsorption data best fitted the Freundlich isotherms with R² value of 0.99.

Keywords: Silica aerogel, MPTMS, Pb(II), Wastewater

INTRODUCTION

The silica aerogel material has attracted considerable interest in diverse scientific and engineering areas due to its exceptional characteristics. Silica aerogel is a material that is recognized for its remarkably low density and outstanding thermal insulation properties. It is commonly referred to as "frozen smoke" or "blue smoke" owing to its translucent visual aspect [1]. The aforementioned material is comprised of a porous network of silica nanoparticles, which yields a structure with a composition of more than 90% air. Silica aerogel, despite its fragile appearance, possesses exceptional mechanical robustness, rendering it a versatile substance suitable for a diverse array of uses [2]. The field of silica aerogels has witnessed a fascinating development

through the incorporation of MPTMS (3-mercaptopropyl-trimethoxysilane) to modify these substances. The use of MPTMS modification in silica aerogels results in improved characteristics and customized capabilities in contrast to unmodified silica aerogels. The incorporation of MPTMS, a silane compound, into the aerogel network during the synthesis process leads to a modified structure [3-5]. Incorporating MPTMS into the aerogel matrix results in distinctive chemical characteristics, including heightened hydrophobicity, superior chemical durability, and augmented adsorption potential. The use of MPTMS-modified silica aerogels in the elimination of heavy metals has garnered considerable interest owing to the escalating apprehensions regarding environmental contamination resulting from heavy metal pollutants. The presence of heavy metals, namely lead, cadmium, mercury, and arsenic, constitutes a significant hazard to both human health and the environment. The efficacy, cost-effectiveness, and reusability of conventional heavy metal removal techniques, such as precipitation, ion

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exchange, and activated carbon adsorption, are constrained. The utilization of MPTMS-modified silica aerogels presents a potentially viable resolution to this issue, owing to their capacity for high surface area, adjustable porosity, and exceptional adsorption characteristics. These attributes render them highly suitable for the purpose of heavy metal remediation [6]. The introduction of thiol (-SH) functional groups onto the surface of silica aerogel nanoparticles is achieved through the modification process utilizing MPTMS. Thiol groups exhibit a high binding affinity for heavy metal ions, thereby enabling efficient removal of mentioned contaminants from aqueous solutions. The substantial surface area exhibited by silica aerogels results in a heightened quantity of active binding sites, thereby augmenting the adsorption capacity for heavy metals [7]. In addition, the hydrophobic characteristics of silica aerogels modified with MPTMS serve to inhibit the adsorption of water molecules onto the active sites, thereby augmenting the efficacy and specificity of heavy metal elimination. The distinctive characteristics of silica aerogels modified with MPTMS can be further customized to suit particular applications involving the removal of heavy metals. The manipulation of MPTMS concentration during the synthesis procedure enables regulation of the density and pore architecture of the aerogels, thereby enhancing the adsorption efficacy for diverse heavy metals^[7]. Furthermore, the integration of alternative functional groups or substances into the aerogel framework has the potential to augment the specificity towards distinct heavy metal ions [8]. The incorporation of chelating ligands has the potential to enhance the binding strength of particular heavy metal ions, thereby facilitating the targeted elimination of distinct pollutants from intricate blends. The potential for regeneration and reusability is an additional benefit of utilizing MPTMS-modified silica aerogels for the purpose of heavy metal removal [9]. Upon adsorption of heavy metal ions, aerogels can be regenerated through desorption techniques, including thermal regeneration or acid treatment. The aforementioned procedure facilitates the retrieval of heavy metals and the recycling of aerogels, rendering the approach more economically viable and ecologically sustainable in contrast to conventional techniques. To sum up, the utilization of MPTMS-modified silica aerogels exhibits significant potential for the purpose of eliminating heavy metals from various sources. The distinctive

characteristics of these materials, such as their substantial surface area, customizable porosity, resistance to water, and chemical durability, render them highly suitable for proficient and discerning adsorption of heavy metal pollutants. Through continued investigation and advancement, it is plausible that MPTMS-modified silica aerogels could significantly transform the domain of heavy metal remediation, thereby playing a crucial role in safeguarding both human well-being and the natural world [10,11].

In the present work, silica aerogel was synthesized using sodium silicate solution as a precursor and sol-gel method. The surface of the obtained aerogel was then modified with MPTMS. The material was characterized via TGA, FTIR and BET. The surface-modified silica aerogel is then used for Pb(II) removal; the removal efficiency was tested by considering PH, initial metal concentration and adsorbent dose experiments. Finally, the adsorption data was used to investigate the isotherm studies.

MATERIALS AND METHODS

Characterization of the MPTMS-Silica Aerogel

The synthesized MPTMS-modified silica aerogel was characterized using FTIR, TGA and BET. FTIR indicated the functional groups that validated the modification. TGA was used to study the weight loss of the respective sample at different temperatures. The N₂ adsorption/desorption isotherms and the properties of the synthesized aerogel were evaluated by BET. The synthesized modified aerogel was used for Pb(II) removal from aqueous solution. The solutions were prepared using lead nitrate salt and de-ionized water. The samples were analyzed after adsorption through AAS for their final Pb(II) concentrations.

Synthesis of the MPTMS-Modified Silica Aerogel

Initially, the silica aerogel was synthesized using sodium silicate solution as a precursor. The silicate solution was neutralized to pH 7. This promoted the hydrolysis and condensation of the precursor. Next, the neutralized sol was left to age for 24 h at room temperature and pressure. The aged gel was then modified using MPTMS. First, 11.3 g of MPTMS was mixed with 56.85 g of n-hexane. The mixture was then added to the synthesized gel and stirred for 3 h. The

silica aerogel was then separated and washed with n-hexane. Finally, the aerogel was dried in a n-hexane rich environment at room temperature for 48 h followed by 5 h at 50 °C.

RESULTS AND DISCUSSION

Characterization Results

FTIR. Figure 1 displays the FT-IR spectra of the modified silica aerogels. The spectral analysis revealed a broad peak exhibiting high intensity within the range of 1,000 to 1,100 cm^{-1} . Additionally, a less intense peak was observed in the vicinity of 800 cm^{-1} , which was attributed to the asymmetric and symmetric bending of Si-O-Si bonds, respectively. These findings are consistent with previous studies [16-18]. The prominent spectral peak observed at approximately 470 cm^{-1} could be attributed to the bending of the O-Si-O bonds. The spectra analysis of the modified silica aerogel indicated the presence of stretching vibrations of S-H and C-H at 2,564 and 2,932 cm^{-1} , respectively. This observation confirmed the surface modification of silica aerogel using mercaptopropyl trimethoxysilane (MPTMS).

TGA. Figure 2 displays the TGA curve of the modified silica aerogel. A reduction in weight was observed at a temperature of 320 °C, while the degradation of aerogel, which resulted in a weight loss of 20.2%, took place at a higher temperature of up to 800 °C. The primary cause of the weight loss observed during the low-temperature range was attributed to the elimination of physically adsorbed water or solvent molecules. Subsequently, a notable reduction in weight was frequently observed, which could be attributed to the breakdown of organic moieties located on the aerogel's surface. The thermal gravimetric analysis (TGA) curve of MPTMS-modified silica aerogels exhibited a weight loss peak, which could be attributed to the decomposition of MPTMS. The point at which the maximum weight loss was observed served as an indicator of the thermal stability of the MPTMS modification. Elevated decomposition temperatures indicated the enhanced thermal stability, rendering the material more suitable for practical applications that require resistance to high temperatures.

BET. Figures 3 presents the N_2 adsorption-desorption isotherms of modified silica aerogel. The data obtained from these experiments are shown in Table 1. The distinct step observed in the type IV isotherm curves is a clear indication

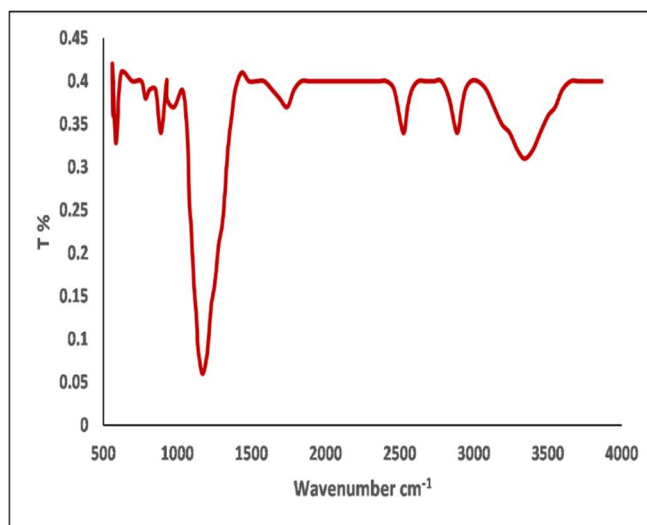


Fig. 1. FTIR of the MPTMS-modified silica aerogel.

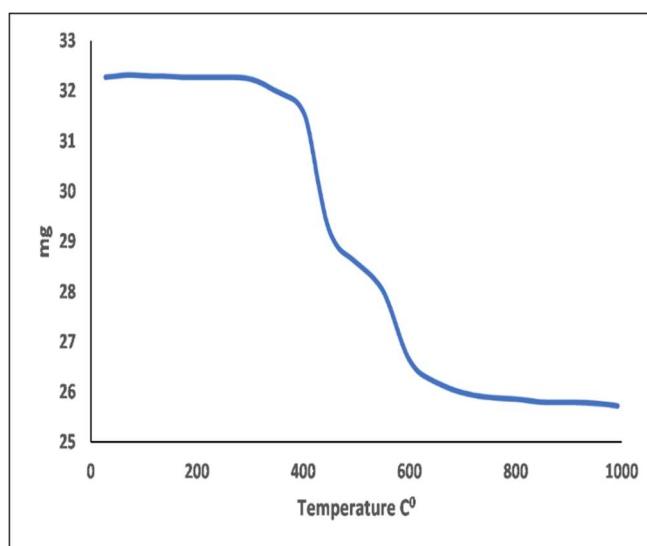


Fig. 2. TGA of the MPTMS-modified silica aerogel.

Table 1. Properties of the MPTMS-modified Silica Aerogel (BET)

Surface area ($\text{m}^2 \text{g}^{-1}$)	Pore volume ($\text{cm}^3 \text{g}^{-1}$)	Pore size (nm)
380	0.46	3.90

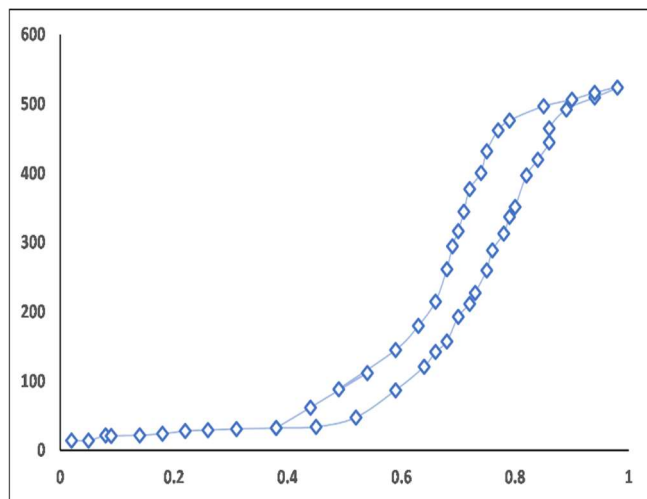


Fig. 3. N₂ Adsorption-Desorption Isotherms of the MPTMS-modified silica aerogel.

of the presence of mesoporous structure in this material. The nitrogen adsorption isotherms demonstrated a sudden rise at a P/P₀ value of roughly 0.70, indicating the occurrence of capillary condensation in the homogeneous mesopores of the substances. The results indicated a significant reduction in the specific surface area, pore volume, and pore diameter of silica aerogel following its modification with MPTMS, as presented in Table 1. The mean pore size of altered silica aerogel was 3.9 nanometers, correspondingly. The filling of micropores in the substrate with the modifier resulted in a decrease in the pore volume of the modified silica aerogel.

ADSORPTION RESULTS

Effect of PH

The pH experiments were carried out with adsorbent dose of 0.05 g, contact time of 30 min and metal concentration of 50 ppm. Figure 4 illustrates the impact of the pH of aqueous samples on the adsorption of Pb(II) ions by the modified silica aerogel. It was observed that the adsorption capacity of the adsorbent increased with an increase in the pH of the samples. The chelation mechanisms and the relatively minor competitive adsorption of hydrogen ions at elevated pH levels contributed to the enhancement of the removal efficiency. At alkaline pHs, the adsorption of OH⁻ ions onto the SH groups took place via a hydrogen bond. At lower pH, it involved the complex formation of M²⁺. The protonation of

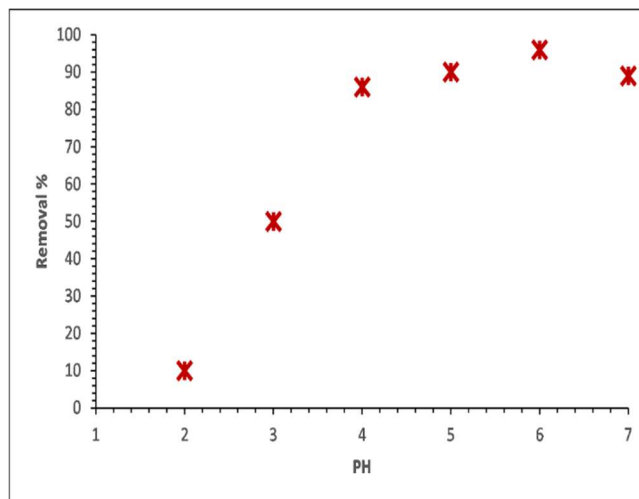


Fig. 4. Effect of PH on adsorption of Pb(II) by the MPTMS-modified silica aerogel.

SH groups resulted in a decrease in the number of available adsorbent sites for complex formation. Conversely, the electrostatic repulsion existing between the positively charged ions and the surface of the altered silica aerogel was increased as a result of the protonation of sulfhydryl (SH) groups. The decrease in pH of the aqueous sample resulted in a reduction of cation adsorption. The reduction of SH group protonation and subsequent increase in the number of SH sites on the surface of modified silica aerogel for cation adsorption were observed upon elevating the pH of solutions. This resulted in an increase in the adsorption capacity of the aerogel. However, the adsorption capacity of adsorbent was reduced at an upper pH of 6 for Pb(II). The increasing adsorption of Pb(II) with increasing pH showed the ion exchange mechanism for these ions. A decrease in the removal yield at the upper optimum pH for each cation was due to the formation of soluble hydroxy complexes. The variation in the adsorption characteristics of distinct heavy metal ions could be attributed to their diverse ion exchange capacity on the surface, which is dependent on their charge density, degree of hydrolysis, and solubility of hydrolyzed metal ions in the solution under the current experimental circumstances.

Initial Metal Concentration

The metal concentration experiments were carried out with adsorbent dose of 0.05 g, pH of 6 and contact time of

30 min. The influence of the initial concentration of cations on the amount of cation that may be adsorbed is shown in Fig. 5 and Table 2. When the starting concentration of Pb(II) ions was increased from 50 to 400 mg l⁻¹, a decrease in the removal yield was observed. This was the case across the whole concentration range. Adsorption of cations might take place at starting concentrations that were lower because there were adequate adsorption sites available. Additionally, the starting concentration of an ion did not affect the amount of ion that might be adsorbed as a percentage. At increasing concentrations of the adsorbate, however, there was a decrease in the number of adsorbent sites that were accessible. The maximum adsorption capacity (q_m) by the respective aerogel was 240 mg g⁻¹ which is remarkable.

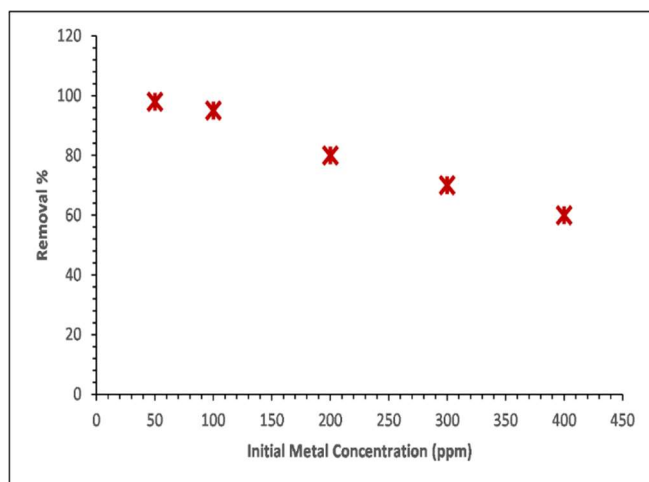


Fig. 5. Effect of Pb(II) Metal Concentration on Pb(II) adsorption.

Table 2. Adsorption data of Modified Silica Aerogel at Different Pb(II) Concentrations

Initial Pb Conc. (ppm)	Final Pb Conc. (Ce)	Removal (%)	q_e (mg g ⁻¹)
50	1	98	49
100	5	95	95
200	40	80	160
300	90	70	210
400	160	60	240

Isotherm studies. The Langmuir and Freundlich isotherms are widely recognized models utilized in physical chemistry and surface science to elucidate the adsorption phenomenon. Adsorption is a complex process that involves the interaction between adsorbent and adsorbate molecules. The insights gained from studying this interaction are highly valuable and aid in the understanding and quantification of the adsorption process. The adsorption data was analyzed through Langmuir and Freundlich isotherms. As can be seen from the Figs. 6 and 7, the obtained data best fitted the Freundlich isotherm with R^2 of 0.99.

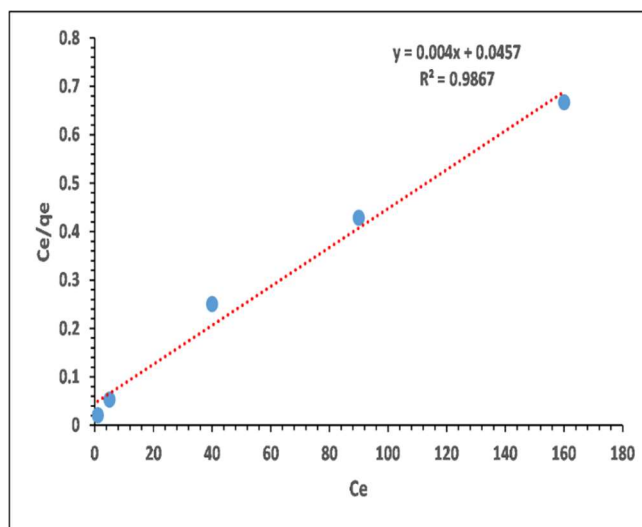


Fig. 6. Langmuir isotherm for the Pb(II) adsorption.

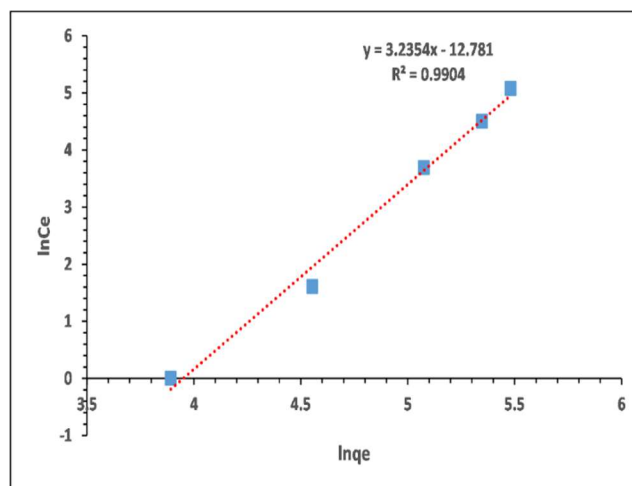


Fig. 7. Freundlich isotherm for the Pb(II) adsorption.

Adsorbent Dose

The adsorbent dose experiments were carried out at metal concentration of 50 ppm, contact time of 30 min and pH of 6. The data presented in Fig. 8 illustrates the relationship between the removal yield of cations and the adsorbent dose within the range of 0.01 to 0.12 g. An escalation in the adsorption capacity for Pb(II) was noted as the adsorbent dose was raised from 0.01 to 0.05 g, after which the adsorption reached equilibrium. Initially, with least dose of adsorbent, few sites were available for the metallic ions; hence, adsorption was reduced. As the dose increased, the availability of adsorption sites was increased, thereby promoting the adsorption of more ions. Finally, when the adsorbent dose reached its maximum capacity and all of its sites adsorbed the adsorbate, the adsorption reached the equilibrium. Here, the adsorbent reached equilibrium at 0.05 g; hence, it can be stated that the optimal dosage is 0.05 g for MTPMS-modified silica aerogel. The increase in the functional groups on the surface area of the adsorbent with an increase in adsorbent dose could be attributed to the corresponding increase in removal percentage. The variation in elimination efficiency of distinct cations under identical conditions, including initial concentration and adsorbent dosage, could be attributed to the disparity in their chemical affinity towards SH groups on the adsorbent's surface.

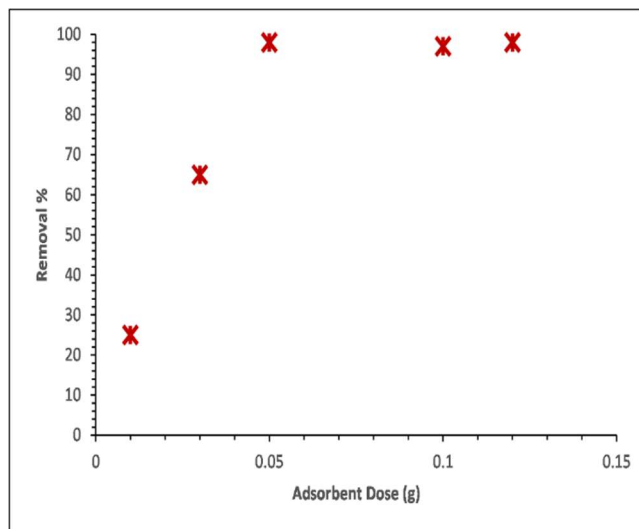


Fig. 8. Removal of Pb(II) ions at different adsorbent doses.

COMPARATIVE STUDY

Table 3 compares the maximum adsorption capacity achieved by the MTPMS-modified silica aerogel (this work) with that previously reported in the literature (14 studies). As can be seen, MPTMS-modified silica aerogel showed enhanced removal capacity compared to that reported in the studies no. 3-15, which is quite remarkable. However, only

Table 3. Comparison of the Maximum Adsorption Capacity of MPTMS-modified Silica Aerogel with Literature Data

S. No.	Material	Functionalizing/Modifying agents	Maximum adsorption capacity (Q_m) mg g^{-1}	Ref.
1	Silica gel	Chitosan	316	[12]
2	Silica gel	MPTMS	240	This work
3	Silica gel	Organic Ligand	175	[13]
4	Silica gel	AFSG	140.23	[14]
5	Silica gel	Diethylenetriaminopropyl	90.5	[15]
6	Silica gel	Ethylenediaminopropyl	98.2	[15]
7	Silica gel	Aminopropyl	98.5	[15]
8	Silica gel	PEI	82.64	[16]
9	Silica gel	AAPTS	61.9	[17]
10	Silica gel	rGO	58.04	[18]
11	Silica gel	Thiamine	39.4	[19]
12	Silica gel	Bispyrazole	35.26	[20]
13	Silica gel	DETA	31	[21]
14	Silica gel	2-Furoyl	22.22	[22]
15	Silica gel	Chitosan	19.86	[23]

chitosan-modified silica gel (study no.1) had higher removal capacity than the present work. This indicates the potential application of MPTMS-modified silica aerogel in the Pb(II) removal from aqueous solution.

DISCUSSION

The compound known as 3-mercaptopropyl-trimethoxysilane (MPTMS) is frequently employed as a coupling agent or surface modifier for a range of materials, including those composed of silica such as silica aerogel. Upon encountering the surface of silica aerogel, MPTMS engages in chemical reactions that lead to the formation of covalent bonds between the MPTMS molecule and the silica surface. The chemical interaction mechanism between MPTMS and silica aerogel is facilitated by a hydrolysis and condensation reaction. Upon exposure to moisture, MPTMS undergoes a reaction wherein the methoxy groups (-OCH₃) present on the molecule react with water molecules (H₂O), leading to the formation of silanol groups (Si-OH) and methanol (CH₃OH) moieties on the MPTMS molecule. The MPTMS molecule's silanol groups (-Si-OH) have the potential to undergo additional reactions with the silanol groups that are already present on the surface of the silica aerogel. The process of condensation results in the establishment of covalent bonds between the MPTMS molecule and the silica surface, ultimately leading to the attachment of MPTMS to the aerogel. The process of condensation reaction persists as additional MPTMS molecules undergo hydrolysis and subsequently react with the silica surface, resulting in the formation of a covalent bond network. The aforementioned network facilitates the formation of a robust and enduring layer atop the silica aerogel, thereby inducing alterations in its characteristics.

When it comes to Pb(II) adsorption, 3-mercaptopropyl-trimethoxysilane (MPTMS) groups are integrated into the silica framework. This modification has been shown to increase the aerogel's affinity towards metal ions such as lead (Pb). The chemical interaction between silica aerogel modified with MPTMS and Pb(II) in aqueous solutions is a subject of significant interest within this particular context. The introduction of thiol (-SH) groups onto the surface of silica aerogel is facilitated by the MPTMS modification. The thiol groups are of paramount importance in the adsorption

process due to their ability to establish robust coordination bonds with Pb(II) ions. The thiol group's sulphur atom exhibits Lewis's basicity by contributing its unshared electron pair to establish a dative bond with the Pb(II) cation. The coordination bonding mechanism facilitates the sequestration and confinement of Pb(II) ions within the aerogel matrix. The phenomenon of MPTMS-modified silica aerogel and Pb(II) interaction is governed by multiple chemical forces, such as electrostatic attractions, coordination bonds, and surface complexation. The inclusion of additional functional groups on the surface of silica aerogel, such as silanol (-Si-OH) groups, may also play a role in the overall adsorption mechanism *via* hydrogen bonding and electrostatic interactions. The adsorption capacity and kinetics for Pb(II) ions are influenced by the specific surface area, pore size, and surface chemistry of the silica aerogel that has been modified with MPTMS. The augmentation of surface area facilitates the interaction between aerogel and Pb(II) ions, whereas regulation of pore size distribution can optimize the availability of thiol groups to the metal ions. Additionally, the efficacy of adsorption can be influenced by various factors, including the pH of the solution, the initial concentration of metal ions, and the temperature. The surface charge of aerogel and the speciation of Pb(II) ions are impacted by the pH of the solution, which in turn influences the electrostatic attraction and coordination bonding. Elevated concentrations of metal ions at the outset can result in the saturation of the adsorption sites that are available, thereby causing a decrease in the efficiency of uptake. Changes in temperature can impact both the thermodynamics and kinetics of the adsorption phenomenon. To summarize, the chemical interaction that occurs between MPTMS-modified silica aerogel and Pb(II) in aqueous solutions is attributed to the coordination bonding that takes place between the thiol groups present on the surface of the aerogel and the metal ions. Comprehending the aforementioned interactions holds significant importance in enhancing the efficacy of MPTMS-modified silica aerogels in their design and implementation as proficient adsorbents for eliminating Pb(II) from polluted water.

CONCLUSION

In the present study, silica aerogel was synthesized using

sodium silicate solution as a precursor and sol-gel method. The surface of the aerogel was modified using mercaptopropyl trimethoxysilane (MPTMS). The synthesized aerogel was characterized using FTIR, TGA and BET. According to results, the aerogel had mesoporous structure (3.90 nm) and high surface area (318 m² g⁻¹). The surface-modified silica aerogel was used for Pb(II) removal at different pH, metal concentration and adsorbent dose. Remarkable performance of the adsorbent was reflected by maximum adsorption capacity of 240 mg g⁻¹. Comparing the capacity values reported in 15 relevant studies, our result showed the second-best value. Finally, the adsorption data was analyzed using isotherms and the data best fitted the Freundlich isotherm.

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