



An Extensive Comparative Study of Highly Enhanced Pb²⁺ Adsorption from Synthetic Wastewater by Organically Tailored Silica Aerogels

Faheem Akhter · Abdul Rauf Jamali ·
Munawar Ali Pinjaro · Abdul Samad Shaikh ·
Sobhy M. Ibrahim · Maria del Mar Alguacil

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Abstract The present work is focused on an extensive comparative study of Pb (II) removal by organically tailored silica aerogels with the relevant studies. Briefly, two types of silica aerogels were synthesized using organic solvents, sol–gel method, modified aging techniques, and ambient pressure drying. The aerogels were characterized (SEM, XRD, FTIR) and investigated for Pb (II) removal from synthetic solutions and the results were extensively compared with other studies that synthesized aerogels using inorganic and expensive functionalizing/modifying agents. As per results of the comparative study, aerogel SA-Iso showed excellent removal efficiency and outperforming most of the functionalizing agents used by other studies, whereas SA-Sim showed remarkably decent results. Highest Pb (II) removal by both the aerogels was achieved in adsorbent dose experiments. At adsorbent dose of 0.1 g, pH 6, Pb (II)

concentration of 10 ppm, solution volume of 100 mL, and contact time of 8 h, SA-Iso showed removal of 98.45%, which was higher than the removal achieved by functionalizing agents such as chitosan, bispyrazole, AAPTS, thiamine, glutaraldehyde, TMCS, and hexane. On the other hand, SA-Sim removed 86.9% of Pb (II) ions which was higher than chitosan, bispyrazole, AAPTS, and thiamine, respectively. The comparative study revealed that, under optimum conditions, such organically modified silica aerogels aged using the given aging techniques could result in an enhanced Pb (II) removal than silica aerogels modified with conventional functionalizing/modifying solvents.

Keywords Silica aerogel · Sol–gel · Lead removal · Organic solvents · Aging

F. Akhter (✉)
Department of Chemical Engineering, Quaid-E-Awam
University of Engineering, Science and Technology,
Nawabshah, Pakistan
e-mail: faheemakhtar86@quest.edu.pk

A. R. Jamali
Materials Engineering Department, NED University
of Engineering and Technology, Karachi, Pakistan

M. A. Pinjaro
Department of Mining Engineering, Mehran University
of Engineering and Technology, Jamshoro, Pakistan

A. S. Shaikh
Department of Petroleum and Natural Gas Engineering,
Mehran University of Engineering and Technology, SZAB
Campus Khairpur Mir's, Khairpur, Pakistan

S. M. Ibrahim
Department of Biochemistry, College of Science, King
Saud University, Riyadh, Saudi Arabia

M. del Mar Alguacil
CSIC-Centro de Edafología Y Biología Aplicada del
Segura, Department of Soil and Water Conservation,
Campus de Espinardo, P.O. Box 164, 30100 Murcia, Spain

1 Introduction

Industrial wastewater is becoming a great concern over time. Every day, numerous industries release untreated contaminated wastewater into the water stream, which consequently ends up affecting human health, aquatic life, and the environment. Among such industries are the battery manufacturing industries. These industries are notorious for releasing wastewater that is highly contaminated with heavy metals, specially lead (Pb) (Rahangdale et al., 2012). Besides, it may contain other heavy metals such as zinc, copper, and cadmium. It is estimated that each day, the lead acid battery industries release approximately 120,000 L of wastewater (Ogbodo et al., 2020). After the release, its immediate target is the aquatic life, which is highly affected. Subsequently, this wastewater reaches at the hands of human consumption where this contaminated water is used for domestic purposes or even consumed for drinking. In such case, this wastewater can significantly affect the human health. For example, the WHO recommended permissible limit for lead (Pb) in drinking water is 0.006 mg/L (Rahangdale et al., 2012). However, the wastewater from battery industries may contain lead from 3 to even 10 mg/L, which is far from recommended limit (Macchi et al., 1993). Such a huge concentration in drinking water may cause serious health problems such as kidney failure, damage brain activity, cause serious digestive problems, and affect the nervous system (Jiang et al., 2020). To deal with this problem, researchers have using various techniques and methods such as ion-exchange, electrocoagulation, membrane separation, solvent extraction, and adsorption (Faheem Akhter, Ahmed, et al. 2021; Faheem Akhter, Soomro, & Inglezakis, 2021a, 2021b, 2021c, 2021d; Akhter et al., 2021a, 2021b). Among these techniques, adsorption has remained as efficient, cost effective, and least complicated method. Usually, this method employs an adsorbent which is brought into contact with adsorbate resulting in removal. Adsorption has various types, among such is solid–liquid adsorption. In this type, the adsorbent is usually in solid form while the adsorbate is dissolved in liquid. As soon as the adsorbent is dissolved into liquid, the adsorbate is bound onto the surface as well as the inner

pores of the adsorbent. After the process, the adsorbent is removed (Keshavarz et al., 2021).

To remove lead (Pb) from the wastewater, various researchers have employed solid–liquid adsorption technique where they have used silica gels as adsorbents (Delacour et al., 1999; Joshi & Srivastava, 2019; Kushwaha et al., 2017; Liu et al., 2021; Verwilghen et al., 2004). Silica gel is a porous material that can have high surface area, porosity, and low thermal conductivity (Sarawade et al., 2010). Such properties also enable this material to have efficient adsorption capabilities. Usually, silica gel are 3 types: cryogel, xerogel, and aerogel. The major difference between them is the material inside the pores and the drying technique used. Cryogels are dried using freeze drying method, xerogel contains alcohol and are dried via evaporation whereas aerogel has pores filled with gas/air and dried with supercritical drying (Okay, 2014). In this, various researchers have successfully synthesized aerogels via ambient pressure drying with remarkable properties and efficient removal capabilities (Du et al., 2020; Guo et al., 2021; Hirashima, 2011). Such aerogels are synthesized using inexpensive materials and facile method but possess remarkably decent properties in comparison to aerogels synthesized via supercritical drying method. Besides the drying technique, the modifying agents play a key role in determining the properties to be achieved by the final product. Various researchers have used a number of functionalizing/modifying agents to achieve the desired properties; however, the present work has used inexpensive and sustainable agents such as TEOS, ethanol, and heptane to equip aerogels with decent properties.

The present study has successfully synthesized 2 variants of silica aerogels using TEOS, ethanol, and heptane as solvents. The aerogels showed remarkable properties such as high surface area, mesoporous structure, high pore volume, and porosity %. Moreover, when these aerogels were used for Pb (II) adsorption from artificial wastewater, they showed excellent removal capabilities. The excellent removal achieved in the present work was validated by comparing these results with other silica gels used for adsorption of lead. In conclusion, the present study emphasized that it was possible to synthesize ambient pressure dried silica aerogels with remarkably decent properties and excellent Pb (II) removal capabilities from artificial wastewater.

2 Materials and Methods

2.1 Synthesis of Organic Silica Aerogels

Initially, the rice husk was washed with distilled water to remove adhering dirt and impurities. The washed rice husk was dried and then leached with 1 M HCl. The leached rice husk was washed with distilled water, dried, and calcined at 600 °C for 4 h. The calcined rice husk ash was further processed for synthesis of silica aerogels.

Present study employed sol–gel technique to synthesize the respective aerogel variants. However, the aging procedures were slightly modified for each variant. In this, SA-Iso was aged inside the solvents separately whereas SA-Sim was aged simultaneously.

2.1.1 SA-Iso (Separate Aging)

This variant was synthesized by first mixing 5 g of calcined rice husk ash with 150 mL of 1 M NaOH solution and stirred for 1 h at 60 degree centigrade. The mixture was filtered with filter paper and the filtrate, known as sodium silicate solution, was separated. The initial pH of the silicate solution was measured to be 12.5, which was neutralized to 7 with 1 M HCl. The neutralized sol was added with TEOS (1:10) relevant to the sol and left to age for 24 h at room temperature. After the aging, the hydrogel was soaked in de-ionized water 3 times, each time for 4 h to remove sulfates. Removal of de-ionized water was followed by aging the hydrogel in an ethanol solution (80% v/v ethanol/water) for 24 h. Subsequently, ethanol solution was removed and the gel was aged inside heptane solution for 24 h. Volume of the heptane solution was 1:1 relative to the gel. Finally, heptane solution was removed and the gel was dried inside the drying oven at 50 °C for 5 h followed by 120 °C until the constant weight (F. Akhter et al., 2023).

2.1.2 SA-Sim (Simultaneous Aging)

To prepare this variant of silica aerogel, all the steps were followed the same as of SA-Iso until after the removal of de-ionized water from the hydrogel; it was then aged simultaneously inside the mixture of TEOS, ethanol, and heptane with ratio 1:1:1 for 24 h. This was done to simultaneously enhance the pore size and strengthen gel network. Finally, the mixture

was removed and the gel was dried at 50 °C for 5 h followed by 120 °C until the constant weight (F. Akhter et al., 2023).

2.1.3 Preparation of Metal Ion Solutions

Lead nitrate was used to prepare the Pb (II) metal solutions. 1.6 g of lead nitrate was dissolved in 1 L de-ionized water to make 1000 ppm solution. This stock solution was then used to prepare metal solutions with concentrations of 5, 10, 20, 30, and 40 ppm, respectively.

2.1.4 Adsorption Studies

The adsorption studies were conducted via batch experiments. The parameters that were analyzed were pH, initial Pb (II) concentration, contact time, and adsorbent dose. Final concentrations of the solutions were analyzed through atomic adsorption spectrometer (AAS). Following equations were used to estimate removal ($R\%$) and adsorption capacity (q_e), respectively:

$$\text{Adsorption efficiency } (R, \%) = (C_0 - C_e)/C_0 \times 100 \quad (1)$$

$$\text{Adsorption capacity } (q_e, \text{mg/g}) = [(C_0 - C_e)V]/m \quad (2)$$

Herein, q_e represents equilibrium capacity of the respective aerogel variant towards Pb (II) ions (mg/g), V is the solution volume in liter (L), m is the adsorbent dose in grams (g), and C_0 and C_e are the initial and equilibrium concentrations of metallic ions in solution indicated as parts per million (ppm), respectively.

3 Results and Discussion

3.1 Characterization

3.1.1 Morphology (SEM)

Figure 1 shows the morphology of the silica aerogel adsorbents used in the present study. The figure shows that both the adsorbents are porous in their structure. However, there is the difference of the pore size and uniformity among the adsorbents. The adsorbent SA-Iso apparently contains bigger

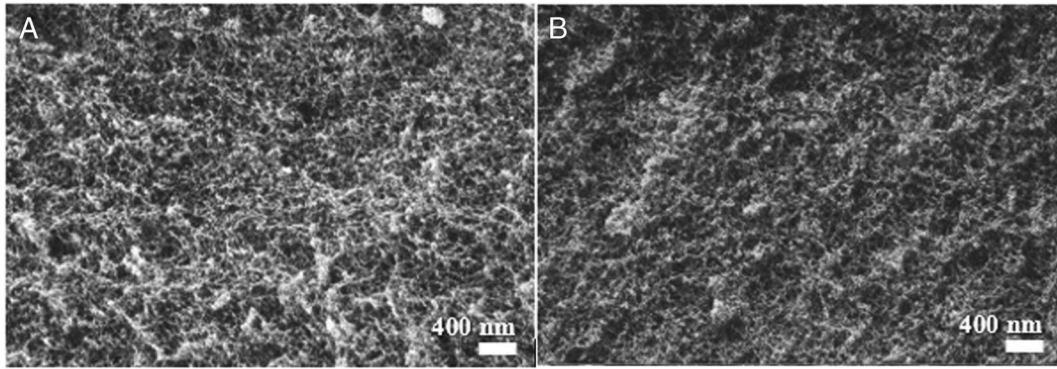


Fig. 1 SEM of silica aerogel adsorbents **A** SA-Iso and **B** SA-Sim

pores and uniform distribution than SA-Sim. This could be attributed to separate treatment and aging of the respective aerogel with organic solvents. The enhanced pore size and uniform distribution of SA-Iso can be attributed to isolated aging inside modifying agents (TEOS, ethanol, and heptane). During isolated aging, each of the solvents fully impacted over the morphology of the respective aerogel. This indicates that the isolated aging promotes enhanced pore features than simultaneous aging.

3.1.2 Structure (XRD)

Figure 2 shows the XRD results of the adsorbents. As can be seen in the figure, the peaks for both the adsorbents lie between 21° and 22° , respectively. No other sharp peaks are observed. This indicates that both the adsorbents are amorphous.

3.1.3 Functional Groups (FTIR)

FTIR is a technique used to determine the functional groups present inside a material. Figure 3 shows the FTIR results of the adsorbents. As can be seen, both the adsorbents contained silica network indicated by the notable peaks at 794 and 1063 cm^{-1} , respectively. Other peaks at 1250 cm^{-1} and 3200 cm^{-1} can be attributed to C-O and OH stretching, respectively. The peaks at 3600 cm^{-1} and 1635 cm^{-1} are attributed to surface-absorbed water hydroxyl groups.

3.2 Adsorption Studies

3.2.1 Effect of pH

pH experiments were carried out in batch with pH ranges as 2, 4, 6, 8, and 10. The constant

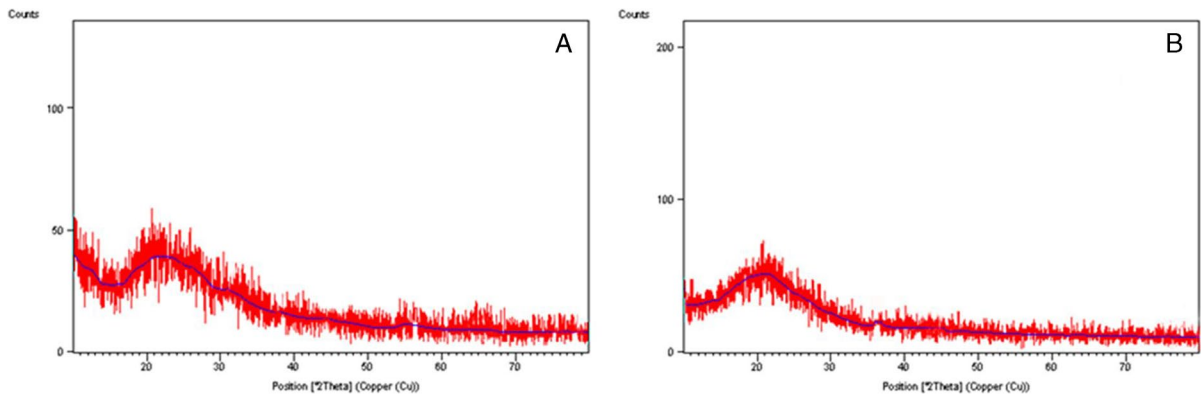


Fig. 2 XRD results **A** SA-Sim and **B** SA-Iso

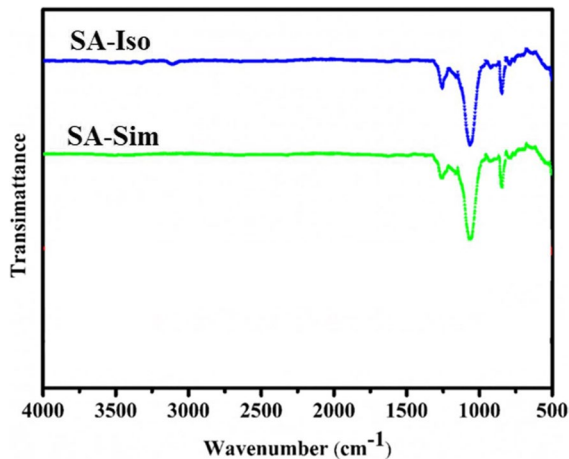
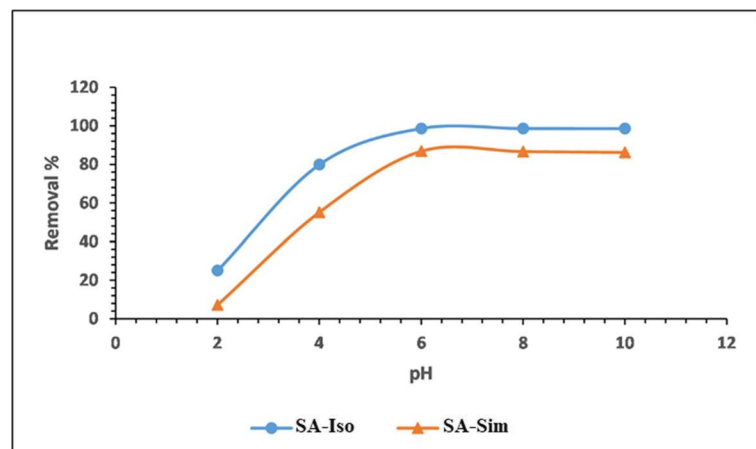


Fig. 3 FTIR results of the adsorbents

parameters were as adsorbent dose 0.1 g, solution volume 100 mL, initial Pb (II) concentration of 10 ppm, and contact time of 8 h, respectively.

Figure 4 shows the adsorption of Pb (II) by the adsorbents at different pH values. As can be seen from the figure, for both the adsorbents, as the pH increases, the removal % also increases until it reaches equilibrium. Both adsorbents showed lowest removal at pH 2, which significantly increased when the pH increased to 4. This can be the result of electrostatic repulsion between positively charged metal and H^+ ions. As the pH increases, the negatively charged ions increase, which results in increased adsorption due to electrostatic attraction of oppositely charged adsorbent surface and metal ions (Du et al., 2020). This continues

Fig. 4 Pb (II) removal by the adsorbents at varying pH



until the equilibrium is reached. The equilibrium indicates the full capacity of the adsorbent after which no major change is observed. The highest removal by SA-Iso was achieved as 98.58% and for SA-Sim as 86.8%, which are excellent when compared with other studies presented in the upcoming section. Both the adsorbents reached equilibrium at the pH of 6, hence the optimum pH value. Moreover, the adsorbent SA-Iso showed 11.94% higher removal efficiency than SA-Sim. This is attributed to isolated aging with modifying agents TEOS, ethanol, and heptane.

Comparison with Other Studies As can be seen from Table 1, the adsorbent SA-Iso has performed excellent with higher removal than studies no. 1–14 and very close to studies no. 16 and 17. On the other hand, the adsorbent SA-Sim has performed remarkably decent with higher removal than studies no. 1–7, respectively.

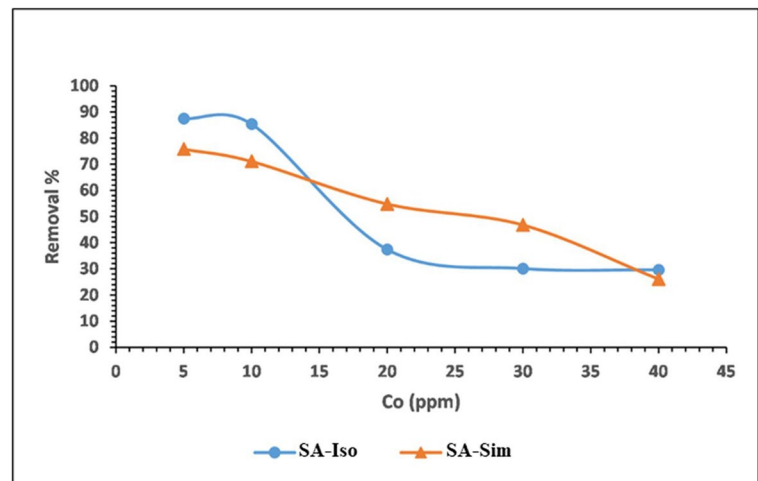
3.2.2 Effect of Initial Metal Concentration

Initial metal concentration experiments were carried out with Pb (II) concentration range as 5, 10, 20, 30, and 40 ppm. The constant parameters were as adsorbent dose 0.1 g, solution volume of 100 mL, and contact time of 72 h, respectively. The pH of the experiments was not adjusted.

Figure 5 and Table 2 represent the effect of initial Pb (II) concentration over removal by the adsorbents. As can be seen, with increase of Pb (II) concentration, the removal % decreases. The highest removal for both the adsorbents is achieved at 5 ppm followed

Table 1 Comparison of pH results with other studies

Effect of pH					
S. no	Material	Functionalizing/modifying agents	Optimum pH	Highest removal %	Ref
1	Silica gel	Chitosan	6	32.4	Rajiv Gandhi and Meenakshi (2012)
2	Silica gel	Bispyrazole	6	35.26	Radi et al. (2017)
3	Silica gel	AAAPTS	5	36	Fan et al. (2014)
4	Silica gel	PEI	6	51	Delacour et al. (1999)
5	Silica gel	rGO	5.5	55	Gao et al. (2020)
6	Silica gel	Organic ligand	6.5	75	Pranudta et al. (2021)
7	Silica gel	PEI	6	81	Ghoul et al. (2003)
8	Silica gel (SA-Sim)	TEOS, ethanol, heptane	6	86.8	This study
9	Silica gel	Glutaraldehyde	5	89.78	Joshi and Srivastava (2019)
10	Silica gel	TMOS, MTMS	6	90	Tadayon et al. (2012)
11	Silica gel	Thiamine	6	90	Deniz et al. (2017)
12	Silica gel	L-proline	5.5	95	Kushwaha et al. (2017)
13	Silica gel	TMCS, hexane	8	96	Falsafi et al. (2020)
14	Silica gel	AFSG	4	98.32	Huang et al. (2020)
15	Silica gel (SA-Iso)	TEOS, ethanol, heptane	6	98.58	This study
16	Silica gel	TEOS, MPTS, HAD	4	99	Li et al. (2021)
17	Silica gel	TEOS, 2-aminothiazole	5	99	Tzvetkova and Nickolov (2012)

Fig. 5 Effect of initial Pb (II) ion concentration over removal

by 10 ppm. As the concentration is increased from 10 to 20 ppm, the removal significantly drops. This may be due to the adsorption sites being occupied with metal ions and adsorbents reaching their maximum adsorption capacities (q_e) as shown in Table 2. After the adsorbent reaches its maximum adsorption capacity, it does not adsorb further when the concentration

is increased. The highest removal achieved by SA-Iso is 87.4% at 5 ppm and 85.32% at 10 ppm, respectively. On the other hand, SA-Sim achieved the highest removal of 75.8% at 5 ppm and 71.1% at 10 ppm, respectively. When the results of both the adsorbents at 5 ppm are compared, SA-Iso showed enhanced removal by 13.27% than SA-Sim.

Table 2 Effect of initial Pb (II) ion concentration over removal

Initial Pb (II) conc. (ppm)	SA-Iso			SA-Sim		
	Equilibrium Pb (II) conc. (C_e)	Removal %	q_e (mg/g)	Equilibrium Pb (II) conc. (C_e)	Removal %	q_e (mg/g)
5	0.63	87.4	4.37	1.21	75.8	3.79
10	1.468	85.32	8.532	2.89	71.1	7.11
20	12.53	37.35	7.47	9.04	54.79	10.95
30	20.98	30.06	9.02	15.96	46.78	14.03
40	28.168	29.58	11.83	29.57	26.07	10.43

Comparison with Other Studies Table 3 shows the comparison of initial metal concentration results with other studies. As can be seen from the table, although the removal by both the adsorbents is lower than other studies, yet it is remarkably decent in comparison.

3.2.3 Effect of Contact Time

Kinetic study was carried out with time range as 0.25, 0.5, 1, 2, 4, 8, 24, 48, and 72 h, respectively. The constant parameters were as adsorbent dose 0.1 g, solution volume 100 mL, initial Pb (II) concentration of 10 ppm, and unadjusted pH.

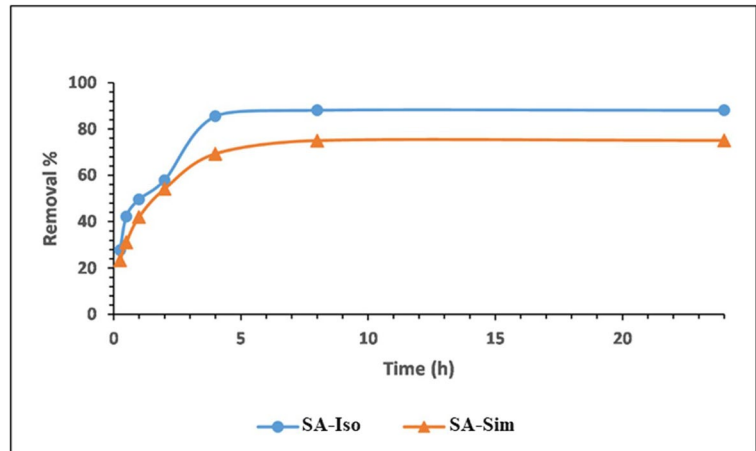
Kinetic adsorption occurs in 3 phases; first, there is a rapid increase in adsorption due to the empty sites of the adsorbent, second, the adsorption slows down because the surface adsorption sites get occupied and adsorption taking place only inside the pores, and finally, it reaches equilibrium (El-Feky et al., 2022). As can be seen in Fig. 6, with the increase of contact time, the adsorption increases for both the adsorbents

until equilibrium. The first phase of adsorption can be attributed to time range between 0.25 and 4 h when the adsorption is rapid and continuous. Then, the second phase is reached with the time range between 4 and 8 h when the adsorption is slow. Finally, third phase reaches when both the adsorbents reach equilibrium at 8 h. Overall, the SA-Iso has performed better with highest removal of 88.18% at 8 h. The adsorbent SA-Sim achieved highest removal of 75% at the same contact time. The removal by both the adsorbents is excellent, as will be shown in the comparison. The optimum time for both the adsorbents was shown to be 8 h, which is decent.

Comparison with Other Studies Table 4 shows the comparison of kinetic study results with other studies. It can be seen that the adsorbent SA-Iso has performed excellent with highest removal among all the studies. On the other hand, SA-Sim has performed remarkably decent with higher removal than studies no. 1–6, respectively. However, when the contact time is compared, both the adsorbents showed

Table 3 Comparison with other studies

Effect of initial metal concentration					
S. no	Material	Functionalizing/modifying agents	Initial Pb (II) conc. (ppm)	Highest removal %	Ref
1	Silica gel (SA-Sim)	TEOS, ethanol, heptane	5	75.8	This study
2	Silica gel (SA-Sim)	TEOS, ethanol, heptane	10	71.1	This study
3	Silica gel (SA-Iso)	TEOS, ethanol, heptane	5	87.4	This study
4	Silica gel (SA-Iso)	TEOS, ethanol, heptane	10	85.32	This study
5	Silica gel	Glutaraldehyde	20	89.78	Joshi and Srivastava (2019)
6	Silica gel	TMCS, hexane	20	96	Falsafi et al. (2020)
7	Silica gel	L-proline	5	96	Kushwaha et al. (2017)
8	Silica gel	TMOS, MTMS	10	99	Tadayon et al. (2012)

Fig. 6 Kinetic results of adsorbents**Table 4** Comparison with other studies

Effect of contact time					
S. no	Material	Functionalizing/modifying agents	Contact time (h)	Highest removal %	Ref
1	Silica gel	rGO	2	30	Gao et al. (2020)
2	Silica gel	Chitosan	0.3	32.2	Rajiv Gandhi and Meenakshi (2012)
3	Silica gel	Bispyrazole	0.4	35.26	Radi et al. (2017)
4	Silica gel	AAAPTS	1	36.6	Fan et al. (2014)
5	Silica gel	PEI	2	61	Ghoul et al. (2003)
6	Silica gel	Thiamine	1	72.8	Deniz et al. (2017)
7	Silica gel (SA-Sim)	TEOS, ethanol, heptane	8	75	This study
8	Silica gel	TEOS, MPTS, HAD	1	84	Li et al. (2021)
9	Silica gel	PEI	15	87	Kang et al. (2021)
10	Silica gel (SA-Iso)	TEOS, ethanol, heptane	8	88.18	This study

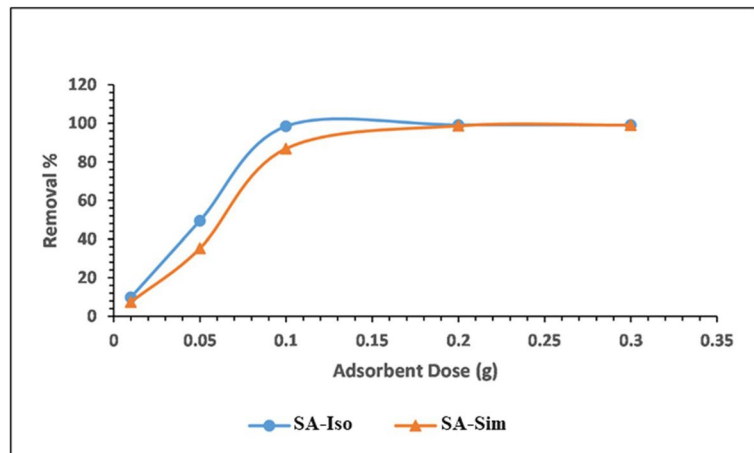
lower contact time than study no. 9, but higher than the remaining studies. Therefore, it can be stated that the removal efficiencies of both the adsorbents were excellent with decent contact time.

3.2.4 Effect of Adsorbent Dose

Adsorbent dose has a significant impact over the adsorption of metal ions. Experiments were carried out with varying adsorbent dose range of 0.01, 0.05, 0.1, 0.2, and 0.3 g, respectively. The other parameters were kept to their optimum values: initial Pb concentration 10 ppm, solution volume 100 mL, pH 6, and contact time 8 h.

In Fig. 7, the Pb (II) removal by the adsorbents is demonstrated at various doses. The results reveal that the lowest removal rate for both adsorbents is seen at the lowest dose of 0.01 g. As the dose increases, so does the removal rate. A significant increase in removal is observed for both adsorbents when the dose is raised from 0.01 to 0.1 g, which is due to the availability of more adsorption sites at higher doses. The highest removal rates achieved by SA-Iso and SA-Sim at 0.1 g are 98.45% and 86.9%, respectively, which are impressively high. As the dose is increased from 0.1 to 0.2 and 0.3 g, the adsorbents reach equilibrium, and the removal rate almost reaches 100%. When comparing the removal rates of both

Fig. 7 Effect of adsorbent dose on Pb (II) removal by the adsorbents



adsorbents, SA-Iso outperforms SA-Sim with higher removal rates. This may be attributed to improved morphological properties attained through isolated aging with modifying agents. Overall, at 0.1 g, SA-Iso exhibited an 11.73% enhanced removal rate compared to SA-Sim.

Comparison with Other Studies Table 5 shows the comparison of adsorbent dose results with other studies. As the table shows, SA-Iso has performed excellent with removal of 98.45%. This adsorbent has shown enhanced removal than studies no. 1–9. However, its removal is lower but very close to studies no. 11 and 12. It should be noted that studies no. 11 and

12 used adsorbent doses higher (0.2 and 1.6 g) than the present work, hence, in this respect SA-Iso has performed better. Similarly, SA-Sim has performed quite decent with higher removal than studies no. 1–4. Overall, it can be stated that SA-Iso performed excellent and SA-Sim performed quite decent with 0.1 g of adsorbent dose.

4 Discussion

The present work is mainly focused on investigating the Pb (II) removal by organically tailored silica aerogels and their comparison with the relevant studies

Table 5 Comparison with other studies

Effect of adsorbent dose					
S. no	Material	Functionalizing/modifying agents	Adsorbent dose (g)	Highest removal %	Ref
1	Silica gel	Chitosan	0.1	32	Rajiv Gandhi and Meenakshi (2012)
2	Silica gel	Bispyrazole	0.1	35.26	Radi et al. (2017)
3	Silica gel	AAAPTS	0.1	36	Fan et al. (2014)
4	Silica gel	Thiamine	0.1	78.8	Deniz et al. (2017)
5	Silica gel (SA-Sim)	TEOS, ethanol, heptane	0.1	86.9	This study
6	Silica gel	Glutaraldehyde	1.5	90	Joshi and Srivastava (2019)
7	Silica gel	TEOS, APTMS, MTMS	0.1	90	Lamy-Mendes et al. (2019)
8	Silica gel	L-proline	2	95	Kushwaha et al. (2017)
9	Silica gel	TMCS, hexane	0.3	96	Falsafi et al. (2020)
10	Silica gel (SA-Iso)	TEOS, ethanol, heptane	0.1	98.45	This study
11	Silica gel	TMOS, MTMS	0.2	99	Tadayon et al. (2012)
12	Silica gel	TEOS, APTES	1.6	100	Faghihian et al. (2012)

to highlight their adsorption potential. In brief, two types of silica aerogels were synthesized using green method, organic solvents, and modified aging. Major difference in preparation of both the samples was the aging techniques. One sample (SA-Iso) was aged separately inside the solvents while the other (SA-Sim) was aged simultaneously inside the mixture. The aerogels were investigated for their Pb (II) removal from synthetic solutions and the results were extensively compared with other relevant studies that used different organic/inorganic solvents to tailor the properties of the acquired aerogels. The synthesized aerogels were characterized using SEM, XRD, and FTIR. As per SEM results, both the aerogels were apparently highly porous; however, the sample SA-Iso indicated a higher pore size, porosity, and uniformity than SA-Sim. This is an explicit result of using organic solvents via separate modification and aging technique. Separate aging inside the solvents resulted in enhanced pore features and stronger gel network. For example, the ethanol is used to enhance the pore features of an aerogel (Meador et al., 2009); therefore, the acquired hydrogel was aged inside the ethanol. Similarly, heptane is known to strengthen the gel network, thereby suppressing the crack formation of the gel during ambient pressure drying (Fidalgo et al., 2007). Hence, with separate aging technique, the sample SA-Iso indicated higher porous structure and lesser cracks than SA-Sim. This is in agreement with the previous studies (Chen et al., 2021; Fidalgo et al., 2007; Jung et al., 2019; Sarawade et al., 2010; Soleimani Dorcheh & Abbasi, 2008). Next, the aerogels were characterized using XRD. This was done to investigate either the prepared aerogels possessed amorphous or crystalline structure. As per results, both the aerogels formed sharp peaks lying between 21° and 22° which indicates their amorphous structure. No other sharp peak was observed in the samples. This can be attributed to several factors that influenced the aerogels during their preprocessing and processing steps, such as leaching of rice husk with acid to remove metallic impurities, calcination of rice husk at 600°C for 4 h, and sol-gel method (Abbas et al., 2019; Mohamad et al., 2020; Nayak & Bera, 2009). The synthesized aerogels were then characterized using FTIR. The results indicated the notable peaks of silica network by both the aerogels at 794 and 1063 cm^{-1} . Moreover, other peaks at 1250 and 3200 cm^{-1} indicate C-O and OH stretching.

The indicated peaks are in agreement with the previous studies (Fidalgo et al., 2007; Prabhu et al., 2015; Sarawade et al., 2010). Next, the synthesized aerogels were investigated for Pb (II) removal from synthetic solutions. The parameters involved pH, initial metal concentration, contact time, and adsorbent dose. In pH results, the highest removal by SA-Iso was 98.58% and by SA-Sim was 86.8%, respectively. When these results were extensively compared with similar studies, SA-Iso showed significantly higher removal than studies that used chitosan (Rajiv Gandhi & Meenakshi, 2012), bispyrazole (Radi et al., 2017), AAPTS (Fan et al., 2014), PEI (Delacour et al., 1999), rGO (Gao et al., 2020), organic ligand (Pranudta et al., 2021), glutaraldehyde (Joshi & Srivastava, 2019), TMOS/MTMS (Tadayon et al., 2012), thiamine (Deniz et al., 2017), and proline (Kushwaha et al., 2017). Similarly, SA-Sim indicated higher removal than chitosan (Rajiv Gandhi & Meenakshi, 2012), bispyrazole (Radi et al., 2017), AAPTS (Fan et al., 2014), PEI (Delacour et al., 1999), rGO (Gao et al., 2020), and organic ligand (Pranudta et al., 2021). This is a remarkably significant removal when considered the fact that the present work used organic and inexpensive solvents, sol-gel technique, ambient pressure drying, and modified aging. Moreover, various solvents used by other studies are expensive and inorganic in nature, thereby emphasizing the potential of this work. Even though other researchers did attempt to utilize the organic approach by using organic ligand (Pranudta et al., 2021), yet the achieved removal was significantly lower than the present work. After pH, the aerogels were investigated for initial metal concentration. As per results, the highest removals achieved by SA-Iso and SA-Sim at 5 ppm were 87.4% and 75.8%. Similarly, the highest removals by SA-Iso and SA-Sim at 10 ppm were 85.32% and 71.1%, respectively. When these results were compared with other studies, the removals by SA-Iso at 5 and 10 ppm (87.4% and 85.32%) are competitive with proline (96% at 5 ppm) (Kushwaha et al., 2017) and TMOS/MTMS (99% at 10 ppm) (Tadayon et al., 2012). Furthermore, the aerogels were investigated for their removal at different contact time. The results showed that both the aerogels achieved highest removal with the contact time of 8 h which is lower than PEI (Kang et al., 2021). However, when the removal % is taken into account, SA-Iso removed higher Pb (II) than rGO (Gao et al., 2020), chitosan

(Rajiv Gandhi & Meenakshi, 2012), bispyrazole (Radi et al., 2017), AAPTS (Fan et al., 2014), PEI (Ghoul et al., 2003), thiamine (Deniz et al., 2017), MPTS, HAD (Li et al., 2021), and PEI (Kang et al., 2021), respectively. Similarly, SA-Sim showed higher removal than rGO (Gao et al., 2020), chitosan (Rajiv Gandhi & Meenakshi, 2012), bispyrazole (Radi et al., 2017), AAPTS (Fan et al., 2014), PEI (Ghoul et al., 2003), and thiamine (Deniz et al., 2017). Lastly, the aerogels were investigated to see their removal at various adsorbent doses. As per results, 0.1 g of SA-Iso was able to remove 98.45% of Pb (II) ions which is higher than chitosan (Rajiv Gandhi & Meenakshi, 2012), bispyrazole (Radi et al., 2017), AAPTS (Fan et al., 2014), thiamine (Deniz et al., 2017), glutaraldehyde (Joshi & Srivastava, 2019), APTMS/MTMS (Lamy-Mendes et al., 2019), proline (Kushwaha et al., 2017), and TMCS/hexane (Falsafi et al., 2020). On the other hand, SA-Sim indicated highest removal of 86.9% at 0.1 g of adsorbent dose, which is higher than chitosan (Rajiv Gandhi & Meenakshi, 2012), bispyrazole (Radi et al., 2017), AAPTS (Fan et al., 2014), and thiamine (Deniz et al., 2017). The results of the comparative study indicate a significantly higher removal than various other solvents used by similar studies. This shows that by using organic solvents, green approach, ambient pressure drying, and sol-gel method, it is possible to synthesize silica aerogels with significantly enhanced Pb (II) removal from synthetic solutions.

5 Conclusion

The present study is focused on an extensive comparative study of Pb (II) removal by organically tailored silica aerogels. Two aerogels (SA-Iso and SA-Sim) were synthesized using organic solvents, sol-gel method, ambient pressure drying, and modified aging techniques. The variant SA-Iso was aged separately inside the solvents whereas SA-Sim was aged simultaneously. Both the aerogels were investigated for their Pb (II) removal from synthetic solutions and the results were extensively compared with the relevant studies. The comparison indicated that SA-Iso showed enhanced removal than most of the studies while SA-Sim showed higher removal than various relevant studies. In conclusion, it was shown that

SA-Iso possessed excellent removal efficiency, while SA-Sim was remarkably decent.

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Author Contribution Faheem Akhter, Abdul Rauf Jamali, Munawar Ali, Maria del Mar: Introduction, Materials and Methods; Faheem Akhter, Abdul Samad Shaikh, Sobhy M. Ibrahim: Results and Discussion, Comparison with Other Studies, Conclusion.

Data Availability The data in this manuscript is available with the corresponding author and can be provided on reasonable request.

Declarations

Ethical Approval Not applicable.

Conflict of Interest The authors declare no competing interests.

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