Green, Organic and Facile Synthesis of Mesoporous Silica Aerogels for Highly Efficient Pb (II) Removal from Synthetic Wastewater: Isotherm and Kinetic Studies

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Abstract

Two highly mesoporous silica aerogels (SA-Iso and SA-Sim) were synthesized via rapid modified sol-gel aging and ambient pressure drying. Biomass waste of rice husk ash was used as a precursor and sol-gel as facile, green and sustainable method. TEOS, Ethanol and Heptane were used as organic solvents to modify the aerogels. One aerogel (SA-Iso) was aged separately while the other (SA-Sim) was aged simultaneously inside the solvents. This was a novel approach that resulted in reduced synthesis time and enhanced properties. The aerogels were characterized by SEM, FTIR, XRD and BET. As per results, surface area, pore size, pore volume and porosity % for SA-Iso were 312 m^2/g, 9.6 nm, 0.85 cm^3/g, 85%, and for SA-Sim as 298 m^2/g, 9.2 nm, 0.75 cm^3/g, 79.5% respectively. The aerogels were analyzed for Pb (II) removal from aqueous solutions. Both the adsorbents showed excellent removal efficiencies, however, with isolated aging, the variant SA-Iso comparatively showed enhanced removal than SA-Sim. The highest Pb removals by SA-Iso and SA-Sim at 0.1 g of adsorbent dose were 98.45 % and 86.9 %. However, when the adsorbent dose increased to 0.2 g, both the aerogels achieved nearly 100 % removal. Finally, isotherm and kinetic studies were carried out. It was shown that the adsorption data followed well the pseudo-second order (Kinetic Study) and Langmuir isotherm models (Adsorption Isotherm).

Keywords: Sustainable, Mesoporous, Silica Aerogels, Sol-Gel Aging, Pb Removal

1. Introduction

Although the industrial revolution may have contributed to a feasible availability of products and made our lives comfortable yet these are becoming one of the major problems of our society. Every day, various industries release the untreated wastewater into the water streams that contain hazardous contaminants. Among such are different heavy metals that are released by the industries such as metal electroplating, steel, mining operations, fertilizer, tanneries, ceramic, paper, pesticide and battery manufacturing [1]. These heavy metals, directly or indirectly, affect the human health, aquatic life, soil and the environment. Lead (Pb) is one such example that is

released in huge amounts mainly by lead acid battery manufacturing industries. Every day, the Lead acid battery industries release 120,000 liters of wastewater. The presence of Lead in these wastewater can range from 3-9 mg/L, whereas, the permissible limit by WHO in drinking water is only 0.01 mg/L [2, 3]. Excess amounts of Lead can have very lethal effects on the human health, i.e. it can damage nervous system, cause problems in hemoglobin formation, adverse effects on gastrointestinal tract, liver, kidney and is carcinogenic [4]. This shows the danger posed by this heavy metal released in excess amounts by battery industries each day.

Various methods have been employed to treat these heavy metals such as chemical precipitation, ion exchange, electrochemical, membrane separation, solvent extraction and adsorption [1, 5]. However, the adsorption method is shown to be effective due to the possession of remarkable properties by adsorbents that play a significant role such as pore size, pore volume, porosity, surface area and surface chemistry [6]. Moreover, the tunable nature of various adsorbents make it a facile method. Silica aerogel is one such example among these adsorbents. This is a material that is highly porous, has high surface area, low density and show high adsorption capacity. Silica aerogels have already shown great potential for heavy metal adsorption [7–11]. Usually, the synthesis of silica aerogels involves chemical precursors, however to make it sustainable, the present study used rice husk ash as a precursor. Moreover, these materials can be tuned into the desired properties using different modifying agents [9, 12–14]. Also, the final properties of this material depend upon the drying method employed. Usually, silica aerogels are dried at super critical drying, however, various researchers have shown that it is possible to use the ambient pressure drying and achieve the similar properties [15–18].

Herein, we have used a facile and sustainable approach to synthesize 2 silica aerogel variants with slightly different properties. We have used rice husk ash as a precursor, TEOS as co-precursor, Ethanol and Heptane as modifying agents to tune the properties. The synthesized aerogels were dried at ambient pressure and were mesoporous in structure. Finally, the aerogels were investigated for their Pb (II) removal from synthetic wastewater.

2. Materials and Methods

All the reagents were of analytical grade, purchased and used without further purification. TEOS was purchased from Daejung, Korea (99% purity), Ethanol from Merck (98% purity), Heptane from Daejung, Korea (98% purity) and lead nitrate from Merck (99.95 % purity). The de-ionized water was used as a solvent or adsorption media. Prepared silica aerogel variants were characterized by SEM, XRD, FTIR and BET. The samples of metal solution after adsorption were analyzed via Atomic Adsorption Spectrometer (AAS).

2.1 Preparation of Silica Aerogel Variants

Sol-gel method was employed to synthesize silica aerogel variants. However, novel adjustments to aging process were made. The aerogel SA-Sim was aged simultaneously inside the mixture of TEOS, Ethanol and Heptane with 1:1:1. On the other hand, SA-Iso was aged separately inside each of the modifying agents. Although, SA-Iso showed enhanced properties, however, the properties achieved by SA-Sim were significantly comparable as well.

2.1.1   TEOS, Ethanol, Heptane Separately Modified Silica Aerogel (SA-Iso)

5 g of rice husk ash was mixed with 1 M NaOH solution and refluxed for 1 hour on a magnetic stirrer. The solution was filtered through filter paper (Whatman 41 ashless) to separate the sodium silicate solution as a filtrate. The sodium silicate solution was neutralized using 1M HCl solution. The neutralized sol was added with TEOS 1:10 relative to the sol and aged at room temperature for 24 hours. After 24 hours, the hydrogel was washed with de-ionized water 3 times, each time for 4 hours to remove sulfates. After removal of de-ionized water, the hydrogel was aged in ethanol solution (80% v/v ethanol/water) for 24 hours. This aging replaced the water inside the pores of gel with ethanol. After 24 hours, the ethanol was removed and the alcogel was aged inside heptane solution for 24 hours. Then the heptane was removed and the gel was dried at 50 C^0 for 5 hours followed by 120 C^0 until constant weight was achieved [19].

2.1.2   TEOS, Ethanol, Heptane Simultaneous Modified Silica Aerogel (SA-Sim)

5 g of rice husk ash was mixed with 1 M NaOH solution and refluxed for 1 hour on a magnetic stirrer. The solution was filtered through filter paper (Whatman 41 ashless) to separate the sodium silicate solution as a filtrate. The sodium silicate solution was neutralized using 1M HCl solution. The neutralized sol was added with TEOS 1:10 relative to the sol and aged at room temperature for 24 hours. After 24 hours, the hydrogel was washed with de-ionized water 3 times, each time for 4 hours to remove sulfates. After removal of de-ionized water, the hydrogel was aged simultaneously inside TEOS, Ethanol and Heptane solution (1:1:1) for 24 hours. After this, the solution was removed and hydrogel was dried at 50 C^0 for 5 hours followed by 120 C^0 until constant weight was achieved [19].

2.1.3   Preparation of Metal Solutions (Pb) Lead Nitrate

First, 1 liter stock solution of lead nitrate was prepared with 1000 ppm by dissolving 1.6 g of lead nitrate in 1000 mL de-ionized water. This stock solution was then used to prepare 5, 10, 20, 30 and 40 ppm metal solutions.

2.1.4   Adsorption Studies

The adsorption performance of silica aerogel variants was evaluated for Pb(II) ions through batch experiments with various parameters including pH, initial metal concertation, contact time and adsorbent dose. The Pb(II) ion concentrations in the solutions after adsorption were analyzed by atomic adsorption spectrometer (AAS). The removal efficiency (R %) and adsorption capacity (q_e, mg/g) were calculated using the following equations:

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

Adsorption capacity$(q_e$, mg/g$) = [(C_0 - C_e)V]/m$ (2)

Where q_e is the equilibrium capacity of silica aerogel variant toward selected Pb(II) ions (mg/g), C_0 and C_e are the initial and equilibrium concentrations of the selected ions in solution (ppm), V is the volume of the solution (L) and m is the weight of the adsorbent (g).

3. Results and Discussion
3.1 Characterization of Silica Aerogel Variants

The synthesized aerogels were characterized using Scanning Electron Microscopy (SEM), Fourier-transform infrared spectroscopy (FTIR), X-ray Powder Diffraction (XRD) and Brunner Emission Teller (BET) respectively. The results are presented and discussed below:

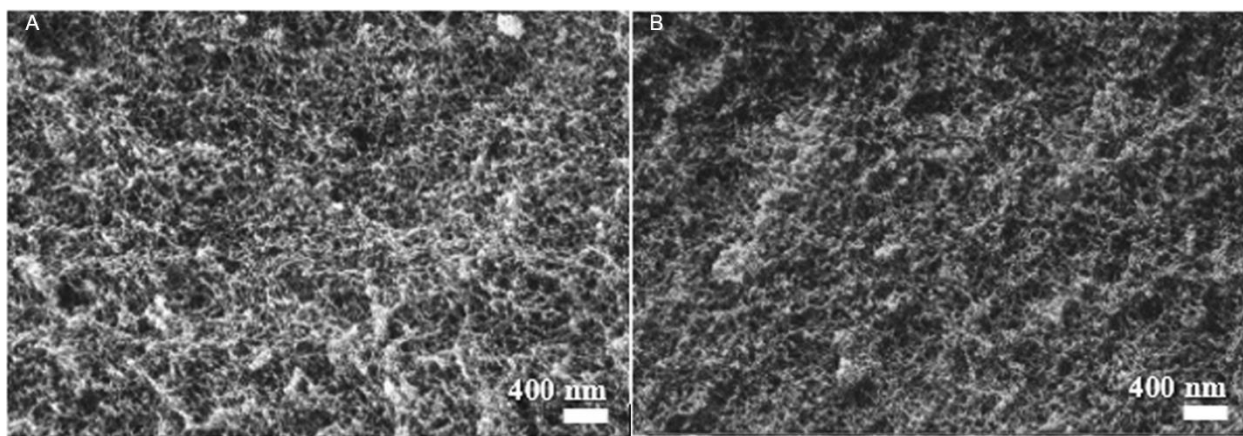### 3.1.1 Effect of Aging Techniques over Morphology of Silica Aerogels (SEM)

Figure 1. SEM Images of (A) SA-Iso (B) SA-Sim

Figure 1 shows the morphology of 2 silica aerogel variants used as adsorbents in the present study. It is indicated that both the adsorbents are highly porous in nature, as also confirmed by the BET results. Moreover, the figure shows that the pore structure in both the adsorbents is irregular and non-uniform. However, when the results are compared, the adsorbent SA-Iso indicates a bigger pore size, uniformity and porosity than SA-Sim. This can be attributed to the separate treatment and modification with TEOS, Ethanol and Heptane instead of simultaneous treatment with SA-Sim respectively.

3.1.2 Effect of Aging Techniques over Structure of Silica Aerogels (XRD)
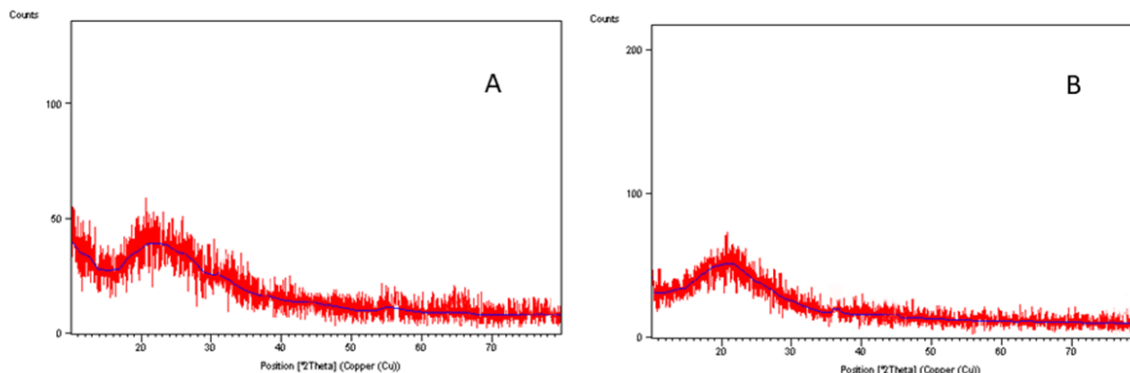
Figure 2. XRD Results (A) SA-Sim (B) SA-Iso

Figure 2 shows the XRD results of silica aerogel variants synthesized from rice husk ash samples calcined at 600 C^0 for 4 hours. This temperature and time are known to produce amorphous rice husk ash. Hence, as indicated by figure 2, the silica aerogel variants are amorphous in nature with peaks lying between 21° and 22° respectively. Besides these, no other sharp peaks are observed, thereby indicating the amorphous structure of both the adsorbents.

3.1.3   Effect of Aging Techniques over Functional Groups of Silica Aerogels (FTIR)
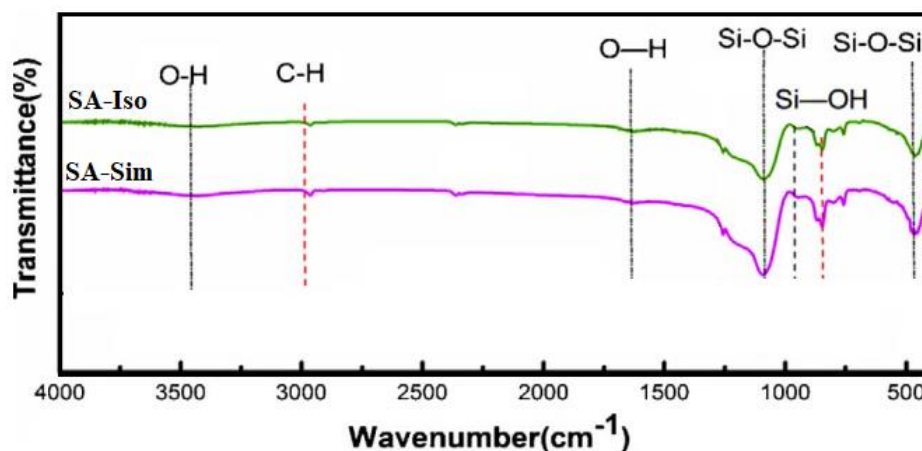

Figure 3. FTIR of Silica Aerogel Variants

Figure 3 shows the FTIR of 2 silica aerogel variants used as adsorbents in the present work. As indicated by the figure, both the adsorbents consist silica gel network. Notable peaks at 1091 cm^{-1}, 794 cm^{-1} and 468 cm^{-1} may be ascribed to siloxane (Si-O-Si) network vibration modes, suggesting the presence of a silica network. The peaks at 966 cm^{-1} indicate the alcoholic presence as a result of aging with ethanol. The peaks at 3455 cm^{-1} and 1635 cm^{-1} are attributed to adsorbed water (O-H) which is very low as a result of pore fluid exchange and ambient pressure drying of the respective silica aerogels. The 1250 cm^{-1} peaks may be ascribed to C-O stretching.

3.1.4 Effect of Aging Techniques over Surface Area and Pore Features of Silica Aerogels (BET)
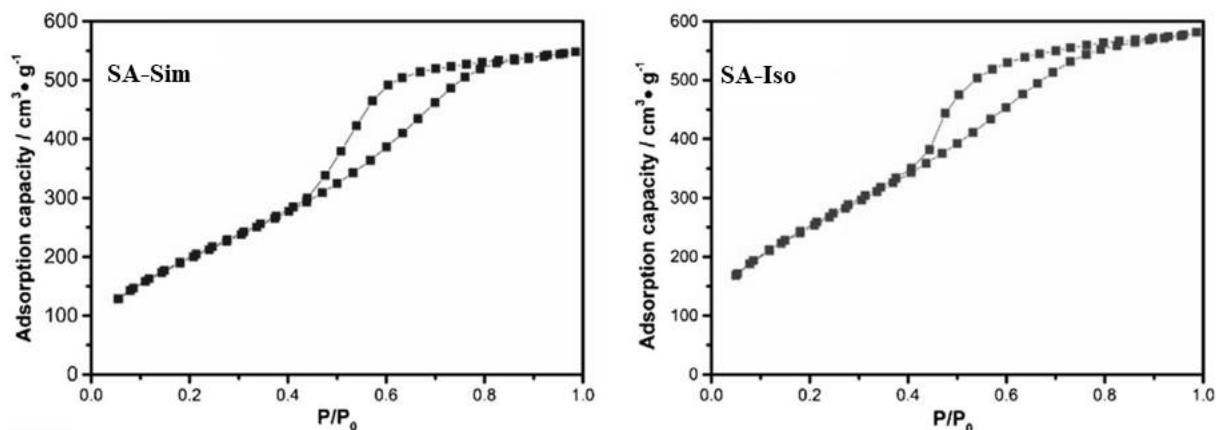

Figure 4. N2 Adsorption Desorption Isotherms of Silica Aerogel Variants

Table 1. Properties of Silica Aerogel Variants

Silica Gels	Surface Area (m²/g)	Average Pore Size (nm)	Average Pore Volume (cm³/g)	Porosity %
SA-Sim	298	9.2	0.75	79.5
SA-Iso	312	9.6	0.85	85

Figure 4 shows the N_2 adsorption-desorption isotherms while the table 1 reflects the properties of silica aerogel variants. As can be seen from figure 4, both of the adsorbents exhibit H1 hysteresis loop thereby indicating that the pores are cylindrical in shape. Furthermore, the table 1 shows that the silica aerogel variant (SA-Iso) possesses improved properties than SA-Sim. This can clearly be attributed to the separate treatment of the respective sample with TEOS, Ethanol and Heptane. TEOS is known to improve the silica purity, Ethanol enhances the pore features whereas Heptane strengthens the gel network so as so sustain the crack formation during ambient pressure drying [19, 20], which is clearly reflected by the properties of the variants. Comparatively, the separate treatment and aging has resulted SA-Iso to achieve 4.48% improved surface area, 4.16% improved average pore size, 11.76% improved average pore volume and 6.47% improved porosity % than SA-Sim. Finally, both the adsorbents were mesoporous (pore size between 2 and 50 nm) in structure.

3.2 Adsorption Studies

The adsorption studies were carried out in batch experiments. The metal solutions after adsorption were analyzed using Atomic Adsorption Spectrometer (AAS) for their final Pb(II) concentrations.
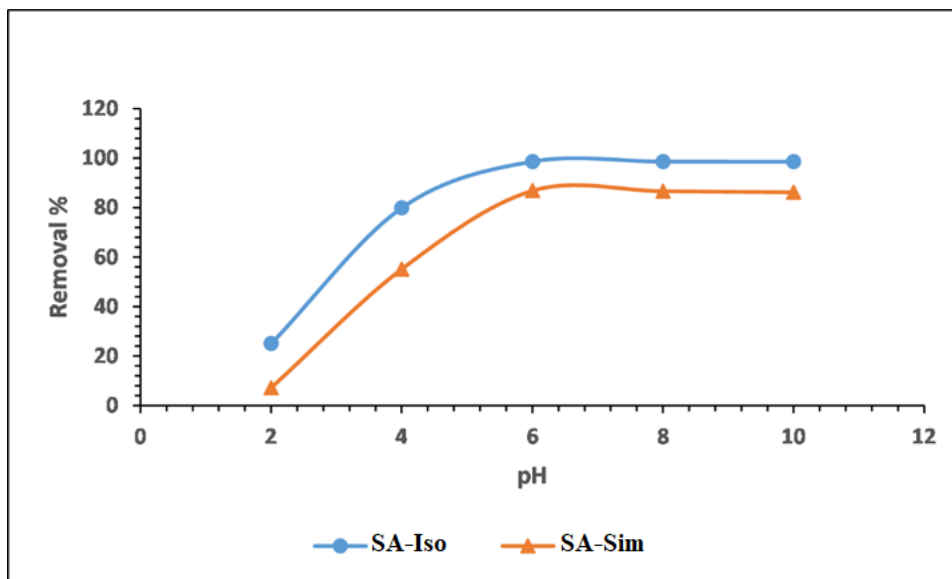
3.2.1 Effect of pH

Figure 5. Effect of pH over Pb(II) Removal by Silica Aerogel Adsorbents

Table 2. Highest Removal % of Adsorbents

Silica Aerogel Variant	pH	Highest Removal %
SA-Iso	6	98.58
SA-Sim	6	86.8

PH of the solution carries an important role in heavy metal removal. It can affect both the surface charge as well as metal ion distribution [21]. The removal by the adsorbents was investigated at pH ranging from 2 to 10. Metal removal beyond 10 may not give accurate results due to the precipitation of ions [1]. The constant parameters were as adsorbent dose 0.1g, solution volume 100 mL, contact time 8 hours and initial Pb(II) concentration of 10 ppm respectively. As indicated by the figure 5, the lower pH shows the lower removal. On the other hand, as the pH increases, the removal is increased. This is due to the fact that at lower pH, the protonation occurs, which is the presence of H^+ ions in the solution. Such ions exhibit electrostatic repulsion with positively charged metal specie, hence resulting in lower removal [1]. As the pH is increased, the deprotonation occurs, which results in lowering of H^+ ions and increase in negatively charged OH^- ions. These negatively charged ions make the active sites on the adsorbents to remove the positively charged metal ions, hence improved removal. The removal increases until the equilibrium is reached. In the present case, the highest removal for SA-Iso and SA-Sim were achieved at pH 6 with 98.58% and 86.8% respectively (table 2). After

this pH, the equilibrium was reached, hence pH 6 can be attributed as optimum value. Comparatively, SA-Iso showed 11.94% higher removal than the sample SA-Sim. This can be attributed to the enhanced properties achieved with separate treatment and aging of SA-Iso.

3.2.2 Effect of Initial Metal Concentration

Effect of Initial Pb (II) concentration over removal was investigated by selecting the metal concentration range as 5, 10, 20, 30 and 40 ppm respectively. The constant parameters were as adsorbent dose 0.1 g, solution volume 100 mL and contact time 72 hours. The experiments were carried out without adjusting the pH.
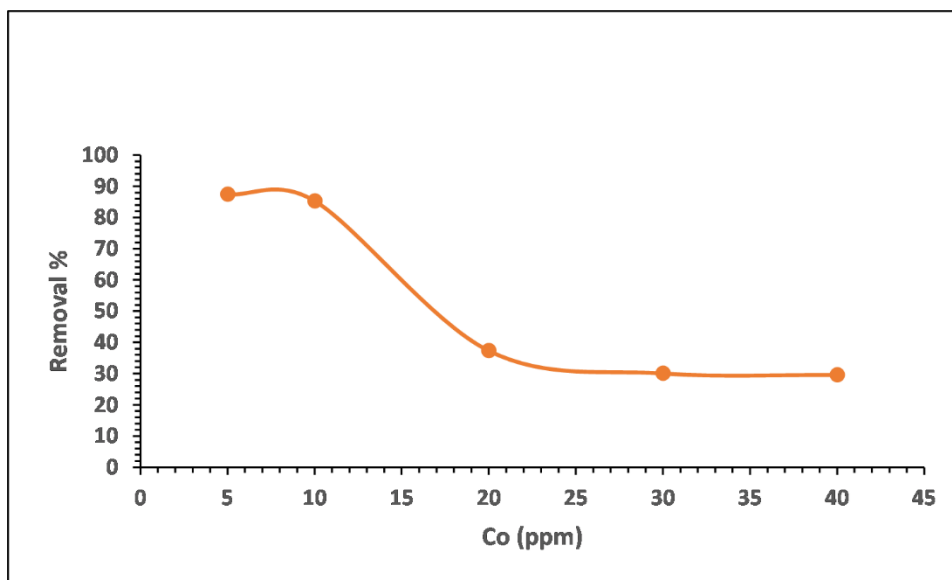

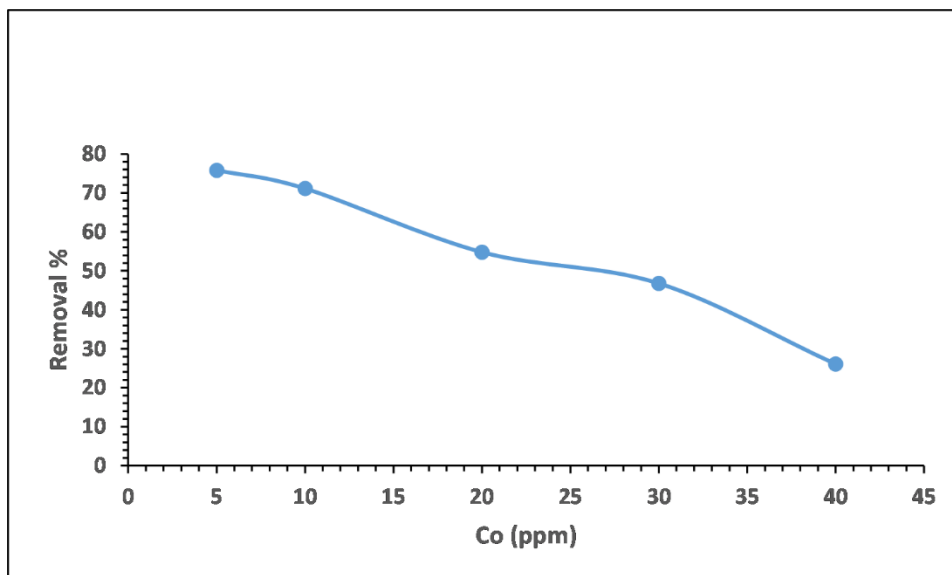Figure 6. Effect of Initial Pb (II) Concentration over Removal (SA-Iso)

Figure 7. Effect of Initial Pb (II) Concentration over Removal (SA-Sim)

Table 3. Effect of Initial Pb(II) Concentration over Removal (SA-Iso vs SA-Sim)

Initial Pb Conc. (ppm)	SA-Iso			SA-Sim		
	Final Pb Conc. (C_e)	Removal %	q_e	Final Pb Conc. (C_e)	Removal %	q_e
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

Figures 6, 7 and table 3 show the removal % achieved by SA-Iso and SA-Sim at the mentioned metal concentration range. As can be seen, both the adsorbents showed highest removal (87.4 and 75.8 %) at the lowest concentration of 5 ppm. As the concentration increased, the removal decreased. Moreover, for both the adsorbents, the significant drop in removal is observed between the concentrations 10 and 20 ppm respectively. For adsorbent SA-Iso, the decrease in removal after 20 ppm is slow in comparison with SA-Sim. The adsorption reduces at higher

concentrations due to the presence of more metal ions than the adsorption sites on the adsorbent [1]. At 5 ppm, the adsorbent SA-Iso has showed an improved removal of 13.27 % than SA-Sim, which is due the enhanced properties. Therefore, based on the results, the optimum metal concentration for both the adsorbents is 5 ppm followed by 10 ppm respectively. When the adsorption capacities (q_e) are compared, SA-Iso showed the highest adsorption capacity (q_e) of 11.83 mg/g at 40 ppm, and SA-Sim showed 14.03 mg/g at 30 ppm respectively. In case of SA-Iso, based on these results, it can be stated that SA-Iso can efficiently and quickly remove Pb (II) ions at lower metallic concentrations of 5 and 10 ppm, however, when the concentration is increased to 20, 30 and 40 ppm, we see a significant drop. This can be attributed to two reasons; first, the presence of higher number of adsorption sites readily available on the surface than its inter-porous structure; and second, the initial adsorption only taking place on the surface. Higher adsorption sites on the surface result in immediate and quick adsorption of metallic ions. Next, when all the sites are occupied and the adsorbent reaches its maximum surface adsorption, the rest of the adsorption takes place in the inter-porous structure of the adsorbent which slows down the mass transfer and results in significant drop. In case of SA-Sim, although the removal % is lower than its companion adsorbent at lower concentrations of 5 and 10 ppm, however, we see a higher removal % and adsorption capacities at 20 and 30 ppm. At 40 ppm, we again see the lower removal % by SA-Sim than SA-Iso. This indicates that SA-Sim can adsorb and remove higher number of Pb (II) ions from the solution at higher ppm than 10 up to 30 ppm. The only plausible reason for this can be a comparatively higher inter-porous mass transfer than SA-Iso, resulting in higher removals at higher concentrations. The most explicit reason is the simultaneous aging of the aerogel with solvents, which might have resulted in an enhanced inter-porous adsorption of the metallic ions. It should also be noted that these experiments were carried out without pH adjustments. In brief, without pH adjustment, the SA-Iso can effectively and efficiently adsorb higher Pb (II) ions at lower concentrations of 5 and 10 ppm, whereas SA-Sim can show higher removal % at 20 and 30 ppm of metallic concentrations.

3.2.3   Effect of Contact Time

Effect of contact time over removal was investigated by selecting the contact time range of 0.25, 0.5, 1, 2, 4, 8, 24, 48 and 72 hours. The parameters set at constant were adsorbent dose 0.1 g, solution volume 100 mL, initial Pb(II) concentration at 10 ppm while the pH was unadjusted.
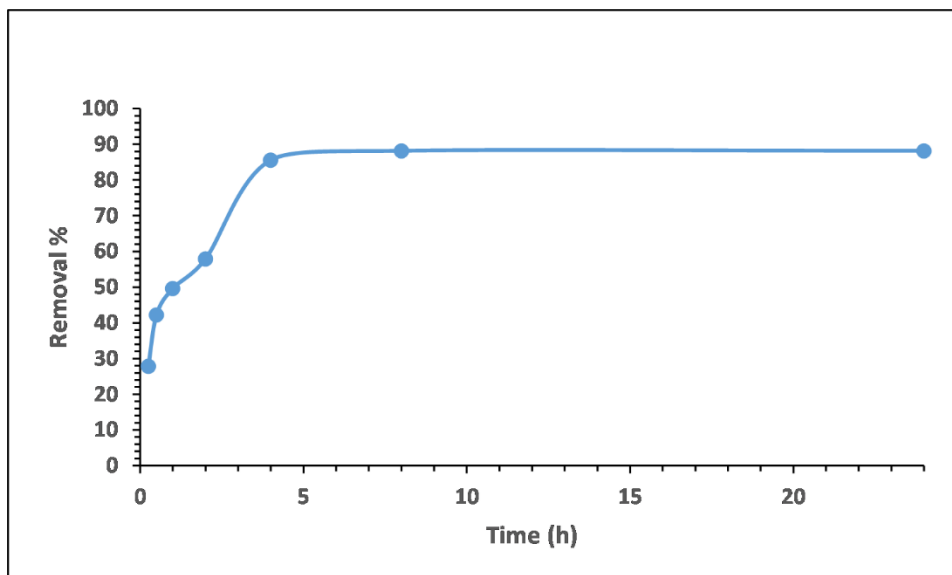
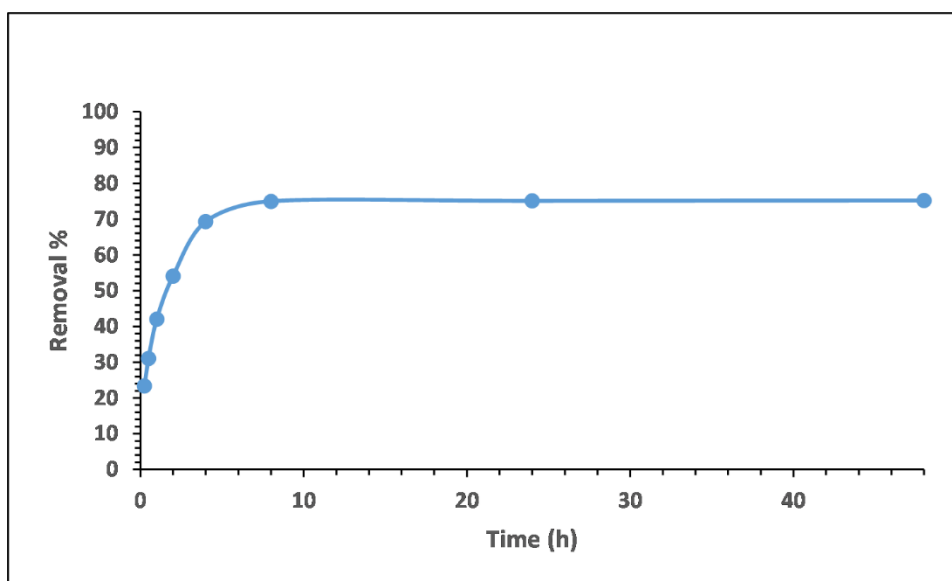Figure 8. Effect of Contact Time over Pb(II) Removal (SA-Iso)

Figure 9. Effect of Contact Time over Pb(II) Removal (SA-Sim)

Figures 8 and 9 show the results of contact time over removal by SA-Iso and SA-Sim respectively. As indicated by the results, increase in contact time results in an increased removal. This continues until the equilibrium is reached at 8 hours after which there is no major removal change observed. Initially, with time range from 0.25 to 4 hours, there is an abrupt and continuous increase in removal for both the adsorbents as the absorption sites are ready to receive the metal ions. At this point, most of the adsorption occurs on the surface of the adsorbents. Then, as the contact time increases from 4 to 8 hours, the removal % becomes slow. This is a result of surface adsorption sites being occupied with adsorbate and further adsorption taking place inside the pores of the material, thereby slowing the mass transfer rate. Moreover, as the adsorption reaches the optimum contact time, the mass transfer rate slows. Finally, the

adsorption reaches equilibrium. For both the adsorbents, from 0.25 to 4 hours, we see a significant increase in the removal. From 4 to 8 hours, the adsorption is slow and finally, at 8 hours, both the adsorbents reach equilibrium with highest removal of 88.18 % (SA-Iso) and 75 % (SA-Sim) respectively.

3.2.4 Effect of Adsorbent Dose

To investigate the effect of adsorbent dose on removal of Pb(II) ions, the dose range of 0.01, 0.05, 1, 2 and 3 g was selected respectively. Other parameters were kept at their optimum values; initial Pb(II) concentration of 10 ppm, solution volume of 100 mL, pH 6 and contact time of 8 hours.
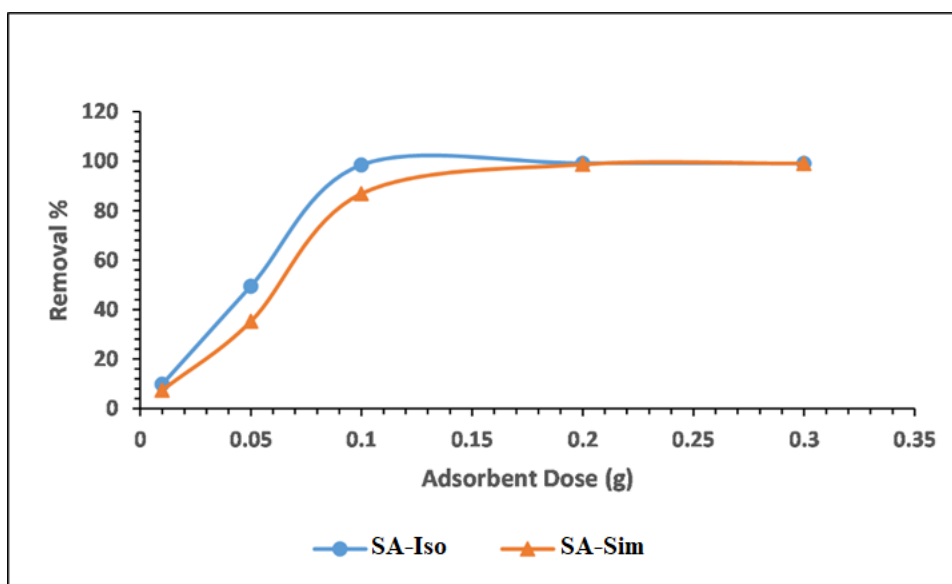

Figure 10. Effect of Adsorbent Dose on Pb (II) Removal

Table 4. Effect of Adsorbent Dose

Adsorbent Dose	SA-Iso Removal %	SA-Sim Removal %
0.01	9.9	7.4
0.05	49.5	35.2
0.1	98.45	86.9
0.2	99.13	98.7
0.3	99.05	99.12

As shown in figure 10 and table 4, for both the adsorbents, increase in adsorbent doses result in increase in removal %. The lowest removal is observed at adsorbent dose of 0.01 g. This is due to the insufficient availability of adsorption sites for the metal ions at 10 ppm. As the adsorbent dose is increased from 0.01 to 0.5 g, a significant rise in removal is achieved in both the adsorbents. This continues until there is nearly 100 percent removal at 0.3 g. With higher dose of

adsorbent, there is availability of more adsorption sites resulting in higher removal. At the optimum dose of 0.1 g, the SA-Iso showed 98.45 % removal for Pb(II) ions whereas the SA-Sim showed 86.9 %.

3.3 Synthesis Time

When we consider the fact that green approach, organic solvents and ambient pressure drying were followed to synthesize silica aerogels, both the aerogels were comparatively rapid [22, 23], however, the SA-Sim took the least time in preparation. This was due to the novel modification via simultaneous aging where the hydrogel was soaked inside the mixture of all three solvents. In this, from refluxing with NaOH to final drying, SA-Iso took 100 hours, whereas SA-Sim took 76 hours, which is 24 hours less than its companion aerogel. Moreover, when the properties are compared, although SA-Iso indicated enhanced properties, the SA-Sim showed significantly comparable properties. Similarly, when we look at the Pb removal efficiencies, SA-Sim showed remarkably decent results. Hence, when all these factors are taken into account, the SA-Sim could be advantageous than SA-Iso.

3.4 Isotherm Studies

Isotherm studies are important for any sorption system. The common isotherm models are Langmuir and Freundlich. Langmuir isotherm supposes that all the adsorption sites of the adsorbent possess the same binding energy and every site joins to only one adsorbate ion. On the other hand, Freundlich isotherm model is used to describe the adsorption characteristics of heterogenous surface [21]. Linearized form of the Langmuir isotherm is given by the following equation:

$$\frac{C_e}{q_e} = \frac{b}{q_m} + \frac{1}{q_m} C_e \tag{1}$$

Where q_e is the equilibrium adsorption capacity of the adsorbent in mg/g, C_e is the equilibrium concentration of Pb ions in ppm, q_m is the maximum adsorption capacity of the adsorbent in mg/g and b is the Langmuir constant.

Moreover, the essential characteristics of Langmuir isotherm can be expressed by dimensionless constant called equilibrium parameters, R_L, defined by the following equation:

$$R_L = \frac{1}{1 + bC_0} \tag{2}$$

The equilibrium data of both the adsorbents was analyzed using Langmuir and Freundlich isotherms. It was found that for both adsorbents, the Langmuir Isotherm best fitted with R^2 values of 0.93 and 0.95 respectively (figures 11, 12). Table 5 shows the constants for the relevant Langmuir isotherms.
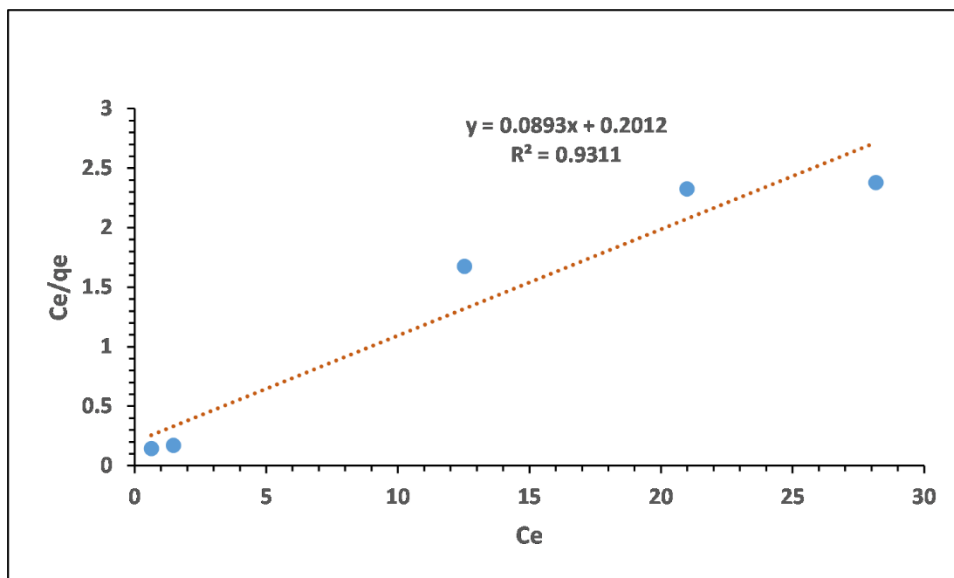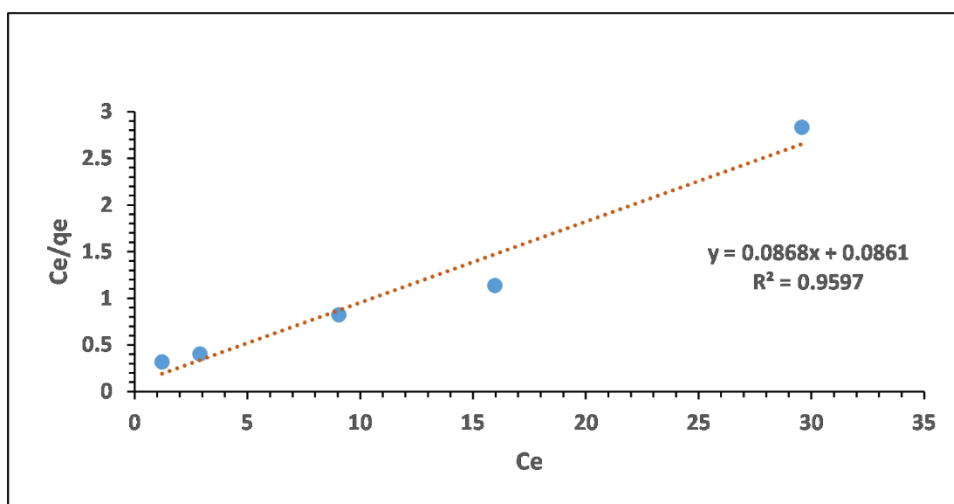
Figure 11. Langmuir Isotherm (SA-Iso)

Figure 12. Langmuir Isotherm (SA-Sim)

Table 5. Constants for the Langmuir isotherms for the adsorption of Pb (II) ions on SA-Iso and SA-Sim

Silica Gel Variant	Solution (mL)	Parameters			
		R^2	$1/q_m$	$1/q_mK_L$	RL (0 < RL < 1 = favorable)
SA-Iso	100	0.9311	0.0893	0.2012	0.007-0.596
SA-Sim	100	0.9597	0.868	0.861	0.003 - 0.201

3.5 Adsorption Kinetic Studies

Adsorption kinetic studies provide an important information on the mechanism of adsorption process, which is essential for practicality of the process [1]. The mechanism and rate of adsorption were proposed by fitting the kinetic data to pseudo-first and pseudo-second order. The pseudo-first order can be represented by the following equation:

$$\log(q_e - q_t) = \log q_e - k_1/2.303t \tag{3}$$

Where q_e and q_t are the adsorption capacities at equilibrium and time t in mg/g, k_1 is the rate constant of the first order kinetics.

On the other hand, the pseudo-second order can be represented by the following equation:

$$t/q_t = 1/k_2 q_e^2 + t/q_e \tag{4}$$

Where q_e and q_t are the adsorption capacities at equilibrium and time t in mg/g, k_2 is the rate constant of second order kinetics. The values of k_1 and k_2 can be determined from the above equations.

As shown in figures 13 and 14, it was found that the mechanism of kinetic data for both the adsorbents fitted best with Pseudo-Second order. The value of R^2 for both adsorbents was 0.99, hence valid.
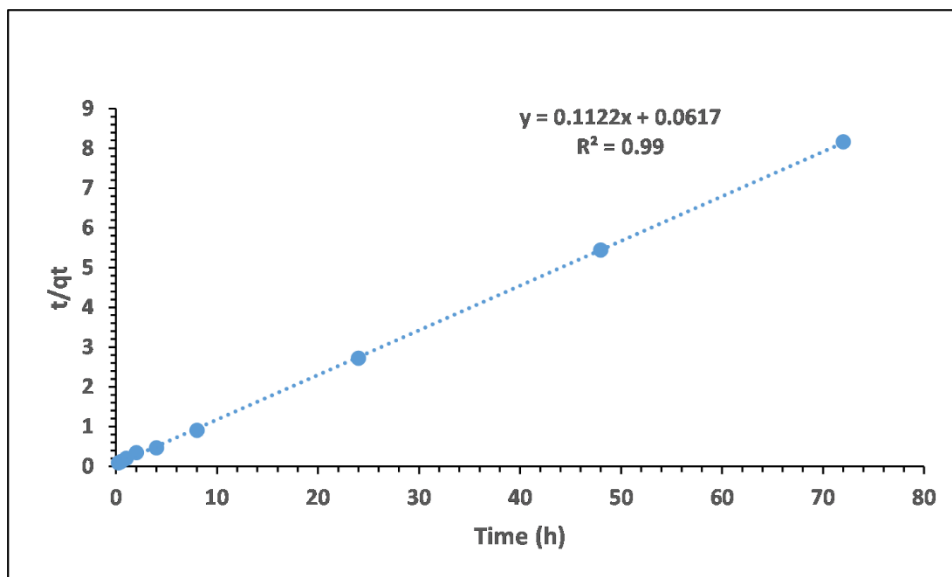

Figure 13. Pseudo-Second Order plots for adsorption of Pb(II) ions onto SA-Iso
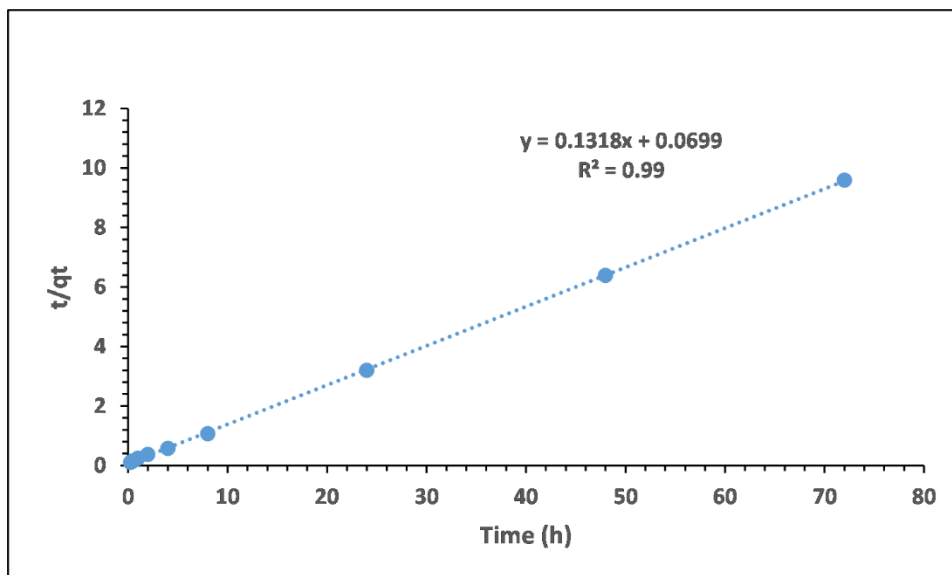
Figure 14. Pseudo-Second Order plots for adsorption of Pb (II) ions onto SA-Sim

4. Conclusion

A green and sustainable approach was used to synthesize two highly mesoporous silica aerogels, SA-Iso and SA-Sim, using rice husk ash as a precursor. Both variants were modified with TEOS, Ethanol, and Heptane using isolated and simultaneous aging techniques. The materials were characterized using SEM, FTIR, XRD, and BET, and it was found that the SA-Iso aerogel modified through the separate method had superior material properties compared to the SA-Sim aerogel modified simultaneously. The aerogels were analyzed for their ability to remove Pb (II) from aqueous solutions, and the results showed efficient performance by both adsorbents. The highest removal percentages were achieved at pH 6, with SA-Iso achieving 98.5% removal and SA-Sim achieving 86.6% removal. At an initial metal concentration of 5 ppm, the highest removal percentages were 87.4% for SA-Iso and 75.8% for SA-Sim. In kinetic studies, the highest removal percentages were achieved after 8 hours of exposure, with 88.18% removal for SA-Iso and 75% removal for SA-Sim. Moreover, when the adsorbent dose was increased from 0.1 g to 0.2 g, SA-Iso showed 99.13% removal, while SA-Sim removed 98.7%. Both adsorbents followed the Langmuir isotherm, and the kinetic data for both the adsorbents best fitted the Pseudo-Second order model. Overall, these findings suggest that both SA-Iso and SA-Sim aerogels could be promising adsorbents for removing Pb (II) from aqueous solutions.

Acknowledgment

Declarations

Ethical Approval

Not Applicable

Competing Interests

The authors declare no conflict of interests

Author's Contributions

Faheem Akhter, Suhail Ahmed: Abstract, Introduction, Materials and Methods; Faheem Akhter Vassilis J Inglezakis: Results and Discussion, Conclusion

Funding

Not Applicable

Availability of Data and Materials

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

References

1. Faghihian H, Nourmoradi H, Shokouhi M (2012) Performance of silica aerogels modified with amino functional groups in PB(II) and CD(II) removal from aqueous solutions. Polish Journal of Chemical Technology 14:50–56. https://doi.org/10.2478/v10026-012-0059-4

2. Rahangdale R v, Kore S v, Kore VS, et al (2012) Waste Management in Lead-Acid Battery Industry: A Case Study . World Journal of Applied Environmental Chemistry 1:7–12

3. Gu S, Fu B, Ahn JW (2020) Simultaneous removal of residual sulfate and heavy metals from spent electrolyte of lead-acid battery after precipitation and carbonation. Sustainability (Switzerland) 12:. https://doi.org/10.3390/su12031263

4. Yang J, Li X, Xiong Z, et al (2020) Environmental pollution effect analysis of lead compounds in China based on life cycle. Int J Environ Res Public Health 17:. https://doi.org/10.3390/ijerph17072184

5. Falsafi M, … MM-J of A, 2020 undefined Removal of heavy metals from synthetic wastewater using silica aerogel-activated carbon composite by adsorption method. arww.razi.ac.ir

6. Oladoja NA, Unuabonah EI, Amuda OS, Kolawole OM (2017) Operational Principles and Material Requirements for Coagulation/Flocculation and Adsorption-based Water Treatment Operations. Springer, Cham, pp 1–11

7. Hagers DL (1999) Performance evaluation for heavy metal ion removal using silica-polyamine composite materials made with different silica gels and polyamines

8.   Huang X, Wang L, Chen J, et al (2020) Effective removal of heavy metals with amino-functionalized silica gel in tea polyphenol extracts. Journal of Food Measurement and Characterization 14:2134–2144. https://doi.org/10.1007/s11694-020-00460-x

9.   Tzvetkova P, Nickolov R (2012) Modified and unmodified silica gel used for heavy metal ions removal from aqueous solutions. Journal of the University of Chemical Technology and Metallurgy 47:498–504

10.   Shahata MM (2016) Adsorption of some heavy metal ions by used different immobilized substances on silica gel. Arabian Journal of Chemistry 9:755–763. https://doi.org/10.1016/j.arabjc.2011.12.010

11.   Kang JK, Lee SC, Jang HY, Kim SB (2021) Synthesis of poly(ethyleneimine)-functionalized mesoporous silica gel with dual loading of host ion and crosslinking for enhanced heavy metal removal in multinary solutions. Microporous and Mesoporous Materials 311:. https://doi.org/10.1016/j.micromeso.2020.110698

12.   Purcar V, Rădițoiu V, Rădițoiu A, et al (2021) Preparation and characterization of some sol-gel modified silica coatings deposited on polyvinyl chloride (Pvc) substrates. Coatings 11:1–13. https://doi.org/10.3390/coatings11010011

13.   Zhang D, Hegab HE, Lvov Y, et al (2016) Immobilization of cellulase on a silica gel substrate modified using a 3-APTES self-assembled monolayer. Springerplus 5:. https://doi.org/10.1186/s40064-016-1682-y

14.   Okhlobystin A, Eremenko I, … VS-A, 2021 undefined Removal of acidic-sulfur-containing components from gasoline fractions and their simulated analogues using silica gel modified with transition-metal carboxylates. ACS Publications

15.   Sarawade PB, Kim JK, Hilonga A, Kim HT (2010) Production of low-density sodium silicate-based hydrophobic silica aerogel beads by a novel fast gelation process and ambient pressure drying process. Solid State Sci 12:911–918. https://doi.org/10.1016/j.solidstatesciences.2010.01.032

16.   Du D, Jiang Y, Feng JJ, et al (2020) Facile synthesis of silica aerogel composites via ambient-pressure drying without surface modification or solvent exchange. Vacuum 173:109117. https://doi.org/10.1016/j.vacuum.2019.109117

17.   Pornchuti B, Pongpattananurak B, Sutthiard D, Singtothong P (2020) Adsorption of copper, nickel and chromium ions using silica aerogel synthesized by ambient-pressure drying and modified with EDTA. IOP Conf Ser Mater Sci Eng 778:. https://doi.org/10.1088/1757-899X/778/1/012133

18.   Aravind PR, Shajesh P, Soraru GD, Warrier KGK (2010) Ambient pressure drying: A successful approach for the preparation of silica and silica based mixed oxide aerogels. J Solgel Sci Technol 54:105–117. https://doi.org/10.1007/s10971-010-2164-2

19. Akhter F, Soomro SA, Jamali AR, Inglezakis VJ (2023) Structural, Morphological and Physiochemical Analysis of SiC8H20O4/C2H5O/C7H16 Modified Mesoporous Silica Aerogels. Physical Chemistry Research 11:1–8. https://doi.org/10.22036/PCR.2022.332609.2044

20. Akhter F, Soomro SA, Inglezakis VJ (2021) Silica aerogels; a review of synthesis, applications and fabrication of hybrid composites. Journal of Porous Materials 28:1387–1400. https://doi.org/10.1007/s10934-021-01091-3

21. El-Feky HH, Behiry MS, Amin AS, Nassar MY (2022) Facile Fabrication of Nano-sized SiO2 by an Improved Sol–Gel Route: As an Adsorbent for Enhanced Removal of Cd(II) and Pb(II) Ions. J Inorg Organomet Polym Mater 32:1129–1141. https://doi.org/10.1007/S10904-021-02214-8

22. Nayak JP, Bera J (2009) Preparation of silica aerogel by ambient pressure drying process using rice husk ash as raw material. Transactions of the Indian Ceramic Society 68:91–94. https://doi.org/10.1080/0371750X.2009.11082163

23. Feng Q, Chen K, Ma D, et al (2018) Synthesis of high specific surface area silica aerogel from rice husk ash via ambient pressure drying. Colloids Surf A Physicochem Eng Asp 539:399–406. https://doi.org/10.1016/j.colsurfa.2017.12.025