

Efficient Lead(II) Adsorption onto Mesoporous Silica Aerogels Synthesized via Green Method, Organic Solvents, and Modified Aging

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(Received 26 November 2022, Accepted 7 January 2023)

Green method, organic solvents, modified aging, and ambient pressure drying were used to synthesize two types of silica aerogels (SGTEH-Iso and SGTEH-Sim). The gels were characterized using scanning electron microscopy, Fourier-transform infrared spectroscopy, and Brunauer-Emmett-Teller. The results indicated that both aerogels possessed remarkable properties with a highly mesoporous structure. Such properties were significantly associated with the novel modified sol-gel aging method employed for each aerogel. The prepared silica aerogels were investigated for their Pb(II) removal from aqueous solution. Among the two aerogels, SGTEH-Iso performed better than SGTEH-Sim, which can be attributed to isolated aging, versus simultaneous aging, with solvents. A comparison of the results of this study to those of previous similar studies revealed that the removal% by both aerogels were higher.

Keywords: Sol-Gel, Mesoporous, Silica aerogel, Organic solvents, Lead removal

INTRODUCTION

Silica aerogel is a highly porous material with high surface area, pore size, pore volume, and thermal insulation. Due to its remarkable properties, this material has received significant attention from the scientific community. Already, researchers have successfully utilized this material as an insulator, a catalyst, a biosensor, a composite, and for various other applications [1]. The synthesis of silica aerogel usually involves the sol-gel process, which is a facile and viable process. The major components of this method are the precursors, solvents, aging method, and drying method. Choosing any of these components can have a major impact on the end product. Several factors, such as cost estimation, sustainability, availability, and properties of the final product, play a significant role in finalizing each component of the sol-gel. For example, conventional chemical

precursors, such as TMOS, MTES, and PEDS, have their own advantages, but their expensive cost and hazardous nature have made researchers use biomass-based precursors that are low-cost, sustainable, and environment friendly [2]. One such biomass-based precursor is rice husk, which is usually leached and calcined to transform into its ash. This ash is then reacted with NaOH and filtered to separate sodium silicate solution, which acts as a precursor [3]. After choosing the component, the second phase involves choosing the right solvents. These solvents act as modifying agents to bring about some major changes inside the structure of the gel network [4]. There are a number of solvents used for silica aerogel synthesis; however, their cost is considered a major hurdle. Hence, researchers are looking for solvents that are sustainable and low-cost and that can lead to enhanced properties in silica aerogel. Similarly, the aging procedure inside the solvents is very important. It can take from hours to days. Therefore, researchers are continuously tuning the aging procedures to reduce the time so that the aerogel may have efficient properties. Finally, the drying method has a

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significant impact on the structure of silica aerogels [5]. In fact, silica aerogels are among those types of gels that are usually dried by supercritical drying. However, cost and environmental concerns have led to the use of silica aerogels dried by ambient pressure. It has been reported that silica aerogels can gain comparatively similar or even enhanced properties by ambient pressure drying instead of supercritical drying [6-9].

Therefore, the present work used the green method to utilize rice husk ash to prepare sodium silicate solution and used it as a precursor. The solvents used were tetraethyl orthosilicate (TEOS), ethanol, and n-heptane, which are all sustainable, low-cost, and organic. The aging of the hydrogel inside the solvents was tuned to reduce time and achieve better properties, which can be considered the novelty of the present work. Finally, the gels were dried using ambient pressure drying and showed excellent properties. Based on the preceding methods, two highly mesoporous silica aerogels were synthesized. The aerogels were characterized using scanning electron microscopy (SEM), Fourier-transform infrared spectroscopy (FTIR), and Brunauer-Emmett-Teller (BET) and indicated excellent properties. The synthesized gels were analyzed for Pb removal from aqueous solutions and showed more efficient and enhanced removal compared to similar previous studies.

MATERIALS AND METHODS

The reagents used in the present work were of analytical grade and used without further purification. The sorption studies were conducted in batch mode. The metal solutions for sorption studies were prepared using lead nitrate. First, 1000 ppm stock solution was prepared and used to make 5, 10, 20, 30, and 40 ppm solutions. The solutions after adsorption were analyzed with an atomic absorption spectrometer (AAS).

Sorption Experiments

To conduct sorption studies, the following three sets of experiments were carried out: pH, initial metal concentration, and adsorbent dose. To carry out pH experiments, the removal% was analyzed at the pH range of 2, 4, 6, 8, and 10. Constant parameters were adsorbent dose (0.1 g), solution volume (100 ml), initial Pb concentration (10 ppm),

contact time (8 h). Experiments with different initial metal concentrations were carried out without pH adjustment at varying concentrations (5, 10, 20, 30, and 40 ppm). Other parameters were kept constant. The effect of adsorbent dosage was studied by carrying out experiments using varying doses of 0.01, 0.05, 0.1, 0.2, and 0.3 g. Other parameters were kept constant, as mentioned above. Equations 1 and 2, given below, were used to calculate the removal% and adsorption capacities of the respective adsorbents.

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

$$\text{Adsorption capacity (q}_e, \text{ mg g}^{-1}) = [(C_0 - C_e)V]/m \quad (2)$$

Synthesis of Silica Aerogels

Two types of silica aerogels were synthesized using the sol-gel method, sodium silicate solution, sustainable solvents, and ambient pressure drying. The sol-gel aging method was modified for each of the two aerogels. It was shown that the modified aging method had a significant impact on the properties of aerogels. Silica aerogels were synthesized as described below.

Synthesis of Silica Aerogel with Isolated Aging

To prepare this type of silica aerogel (SGTEH-Iso), 5 g of rice husk ash was reacted with 150 ml 1M NaOH solution for 1 h under constant stirring and a temperature of 60 °C. After the stirring, the mixture was filtered and the filtrate was separated, the result of which is called sodium silicate solution. Then, the pH of the silicate solution was neutralized. In the next stage, TEOS was added to the neutralized hydrosol with a ratio of 1:10 relevant to the sol, and the resultant neutralized hydrosol was aged for 24 h under ambient pressure and temperature. This aging resulted in the conversion of hydrosol to hydrogel. After 24 h, the hydrogel was washed with deionized water several times to remove sulfates. Then, the hydrogel was aged in an ethanol/water mixture with 80% ethanol and 20% water for 24 h. After 24 h, the ethanol mixture was removed and the gel was aged inside the heptane solution for 24 h. Afterward, the heptane solution was removed and the gel was dried in a drying oven at ambient pressure drying at 50 °C for 5 h, followed by 120 °C until a constant weight was achieved

[10].

Synthesis of Silica Aerogel with Simultaneous Aging

To synthesize this variant of silica aerogel (*i.e.*, SGTEH-Sim), the same steps described above for SGTEH-Iso were followed up to the stage where hydrosol was washed with deionized water. After the washing, the gel was aged simultaneously inside a mixture of TEOS/ethanol/heptanes with a ratio of 1:1:1 relevant to the gel for 24 h. This was a novel approach that resulted in comparable properties. After 24 h, the gel was dried for 5 h at 50 °C. This was followed by drying at 120 °C until a constant weight was reached [10].

RESULTS AND DISCUSSION

The synthesized silica aerogels (*i.e.*, SGTEH-Iso and SGTEH-Sim) were characterized using SEM, FTIR, and BET. The results are presented below.

The Impact of Solvents and Drying on Morphology of Silica Aerogels

Figure 1 shows the morphology of the synthesized silica aerogels. It is clear that both aerogels were highly porous in their structure. However, there was apparently a difference between the pore sizes, pore volumes, and pore distribution.

The Impact of Solvents and Drying on Morphology of Silica Aerogels

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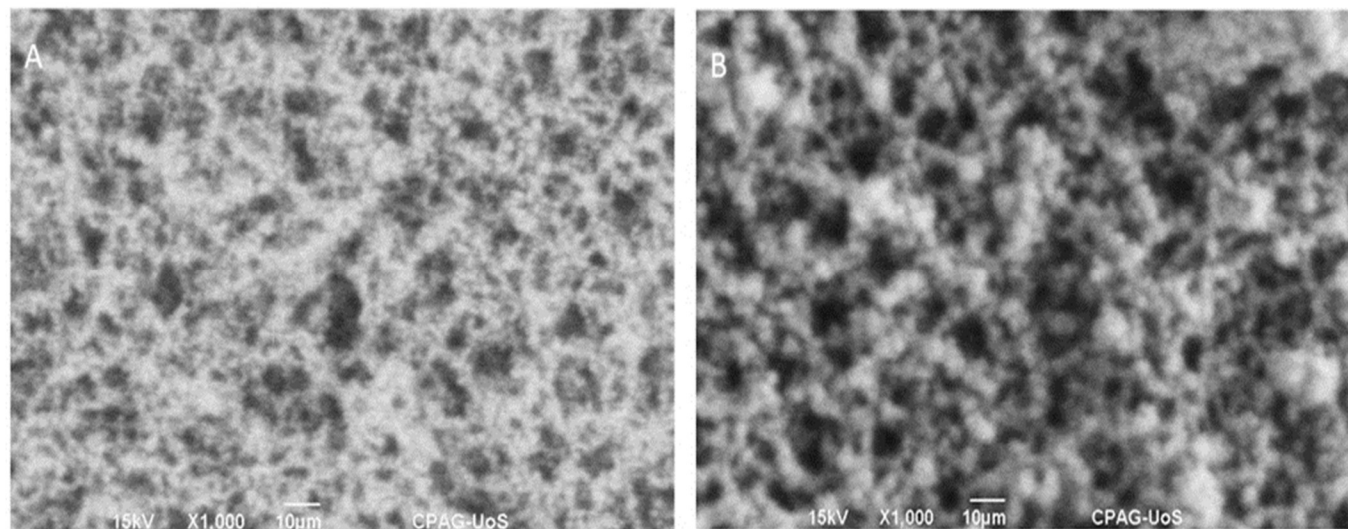


Fig. 1. SEM of silica aerogel adsorbents (A) SGTEH-Sim (B) SGTEH-Iso.

Table 1. Properties of Synthesized Silica Aerogels

Silica gels	Average pore size (nm)	Average pore volume (cm ³ g ⁻¹)	Porosity (%)	Surface area (m ² g ⁻¹)
SGTEH-Iso	9.6	0.85	85	312
SGTEH-Sim	9.2	0.75	79.5	298

ethanol as it is known for enhancing the pore properties of aerogels. Larger pore size and higher pore distribution, in turn, lead to higher removal efficiency by providing more adsorption sites on the surface as well as inside the pore structure. The gel network of both aerogels appeared to be stronger without any cracks during drying. This is the result of heptane aging. Heptane is a solvent that strengthens the gel network, thereby suppressing crack formation during ambient pressure drying. However, SGTEH-Sim indicated slight cracks whereas SGTEH-Iso showed no cracks with a stronger structure. This is likely due to the isolated aging with heptane.

The Impact of Solvents and Drying on Functional Groups of Silica Aerogels

Figure 2 shows the presence of different functional groups indicated by peaks. It can be seen that a silica network was formed in both aerogels, as reflected in sharp peaks at 794 and 1063 cm^{-1} , respectively. The peak at 1250 cm^{-1} can be attributed to C-O stretching whereas the peak at 3200 cm^{-1} represents OH stretching. Peaks at 1635 and 3600 cm^{-1} show water hydroxyl groups [11].

The Impact of Solvents and Drying on Properties of Silica Aerogels

Figure 3 and Table 1 show the N_2 adsorption/desorption isotherms and properties of silica aerogels. The BET isotherms indicated that both aerogels followed the H1

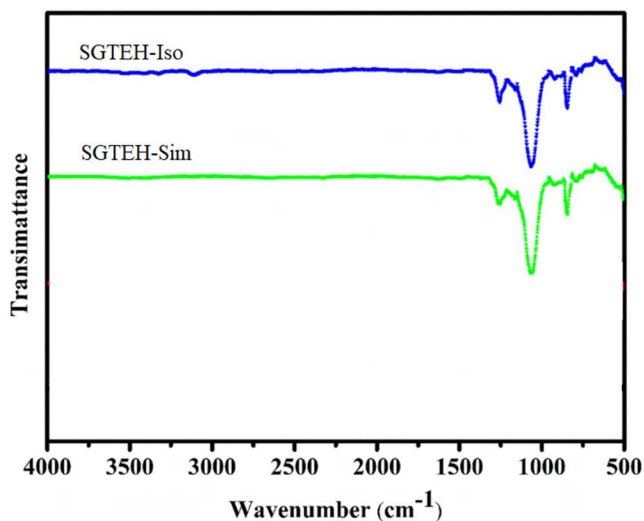


Fig. 3. FTIR of silica aerogels.

hysteresis loop. This is the result of the pore structure being cylindrical in shape. Furthermore, when the resultant properties of aerogels were compared (Table 1), it was found that SGTEH-Iso had enhanced surface, pore size, pore volume, and porosity% compared to SGTEH-Sim. This can be attributed to the isolated aging of SGTEH-Iso with TEOS, ethanol, and heptane compared to the simultaneous aging of SGTEH-Sim. This finding shows that isolated aging leads to enhanced properties. However, it must be noted that the properties achieved through simultaneous aging were also comparatively good.

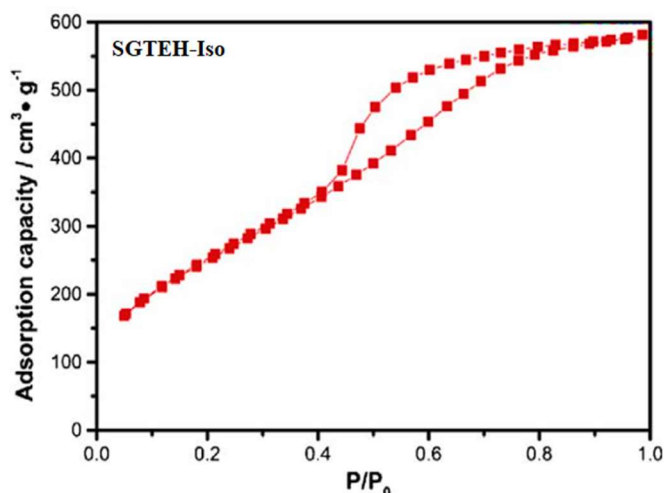
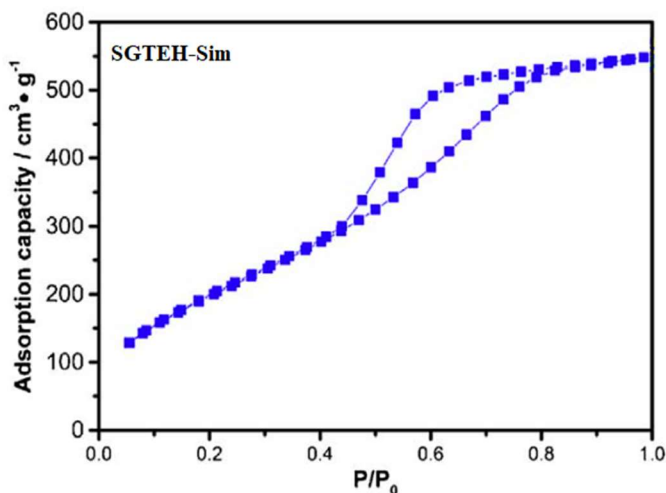


Fig. 2. N_2 adsorption-desorption isotherms of silica aerogels.

Sorption Studies

PH. PH plays an important role in heavy metal adsorption. It can significantly affect the surface charge of the adsorbent. Figure 4 shows the removal% of Pb by silica aerogels. As it can be seen in Fig. 4, in both aerogels, as the pH increased, the removal% increased as well until it reached equilibrium. At lower pH levels, the removal% was lower due to the electrostatic repulsion between H^+ ions and metal positive ions. As the pH increased, the number of H^+ ions decreased and negatively charged OH^- ions increased. This led to an increase in the adsorption and hence resulted in an increased removal. Finally, when all the adsorption sites of the adsorbent were filled, it reached equilibrium. The highest removal% by both silica aerogels was achieved at a pH of 6, with 98.58% for SGTEH-Iso and 86.8% for SGTEH-Sim. The aerogel SGTEH-Iso showed enhanced higher removal% compared to SGTEH-Sim, which can be attributed to isolated aging with solvents.

Initial Metal Concentration

The initial metal concentration has a significant effect on adsorption and indicates the overall capacity of the adsorbent. Figure 5 and Table 2 show the removal% by silica aerogels at varying Pb concentrations. As shown in Fig. 5, in both aerogels, the highest removal% was achieved at the lowest metal concentration of 5 ppm. This was due to the fact that the adsorption sites were ready to adsorb the metal ions and that most of the adsorption took place on the surface of aerogels. As the metal concentration increased, the removal% decreased. This may be due to the fixed adsorbent dosage, which, in turn, meant that the adsorption sites were filled with metal ions and that the rest of the ions were left in the

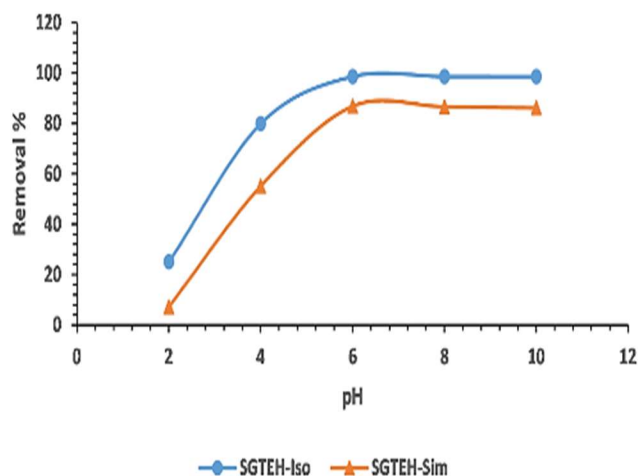


Fig. 4. Pb removal by silica aerogels.

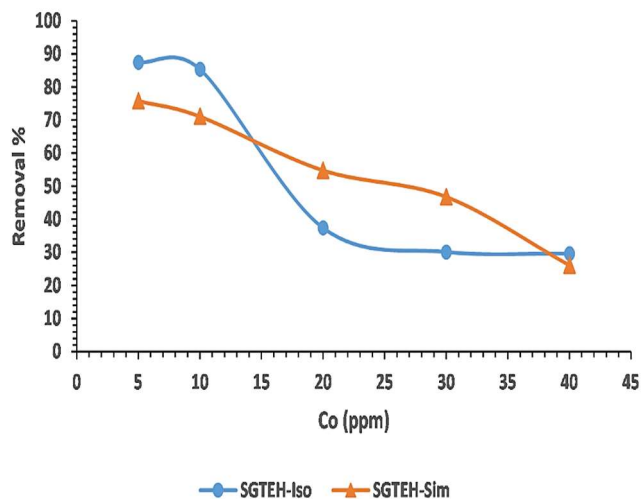


Fig. 5. The effect of initial Pb concentration on removal% by silica aerogels.

Table 2. The Effect of Initial Pb Concentration on Removal% by Silica Aerogels

Initial Pb conc. (ppm)	SGTEH-Iso			SGTEH-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	30.06667	9.02	15.96	46.78	14.03
40	28.168	29.58	11.83	29.57	26.07	10.43

solution. Furthermore, the highest removal% by both aerogels was achieved at 5 ppm, with 87.4% for SGTEH-Iso and 75.8% for SGTEH-Sim. The removal% achieved at 10 ppm by both aerogels was relatively similar, with 85.32% for SGTEH-Iso and 71.1% for SGTEH-Sim. Overall, it can be stated that at 5 and 10 ppm, the removal% by SGTEH-Iso was considerably high while that of SGTEH-Sim was moderate. The high removal% by SGTEH-Iso can be attributed to its isolated aging with solvents.

Adsorbent Dose

The adsorbent dosage plays a key role in the adsorption process. Higher doses provide a higher number of adsorption sites to remove metal ions. Figure 6 shows the removal of Pb ions by silica aerogels at different doses. As can be seen, the lowest removal% was obtained at lower doses. As the adsorbent dosage was increased, the removal% increased as well. This trend continued until nearly 100% removal was achieved. Initially, the lowest removal for both aerogels was observed at 0.01 g, with 9.9 for SGTEH-Iso and 7.4% for SGTEH-Sim. As the dose was increased from 0.01 to 0.5 g, a significant was observed in the removal% to 49.5% for SGTEH-Iso and 35.2% for SGTEH-Sim. Similarly, a significant rise in the removal% was observed for both aerogels as the adsorbent dosage increased from 0.5 to 0.1 g. At 0.1 g, the removal% reached 98.45% for SGTEH-Iso and 86.9% for SGTEH-Sim. Finally, when the adsorbent dose was increased beyond 0.1 g, nearly 100% removal was

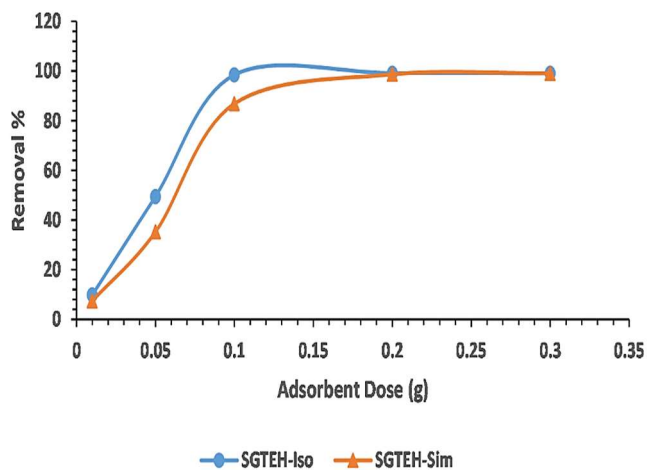


Fig. 6. The effect of adsorbent dose on Pb removal by the silica aerogels.

obtained by both adsorbents, which can be due to the high number of adsorption sites available as the adsorbent dosage increased. Overall, at 0.1 g of adsorbent dose, the removal% by SGTEH-Iso was 11.73% more than that of SGTEH-Sim. Table 3 presents a comparison of the results of this study with those of other studies.

As can be seen in Table 3, SGTEH-Sim resulted in a removal% that was slightly higher than that reported in studies 1 and 2. However, SGTEH-Iso showed a higher removal%, which was markedly higher than that reported in studies 1-8. Although studies 10 and 11 reported higher

Table 3. A Comparison of Results with Those of Other Studies

S. No.	Material	Functionalizing/Modifying agents	Highest removal%	Ref.
1	Silica gel	rGO	55	[12]
2	Silica gel	Organic ligand	75	[13]
3	Silica gel	PEI	81	[14]
4	Silica gel (SGTEH-Sim)	TEOS, Ethanol, Heptane	86.8	Present work
5	Silica gel	TMOS, MTMS	90	[15]
6	Silica gel	Thiamine	90	[16]
7	Silica gel	TMCS, Hexane	96	[17]
8	Silica gel	AFGS	98.32	[18]
9	Silica gel (SGTEH-Iso)	TEOS, Ethanol, Heptane	98.58	Present work
10	Silica gel	TEOS, MPTS, HAD	99	[19]
11	Silica gel	TEOS, 2-aminothiazole	99	[20]

removal% than that achieved by SGTEH-Iso, the difference in their removal% was marginal. Therefore, it can be stated that synthesizing silica aerogels with the green method, organic solvents, and modified sol-gel aging can lead to a remarkably high Pb removal%.

DISCUSSION

The present work successfully synthesized highly mesoporous silica aerogels using sustainable solvents and modified sol-gel aging. Two types of silica aerogels were synthesized using the same solvents but different sol-gel aging methods. SGTEH-Iso was aged separately inside each of the solvents whereas SGTEH-Sim was aged simultaneously. The SEM results indicated that both aerogels were highly porous. However, SGTEH-Iso showed a larger pore size and greater uniformity in pore distribution compared to SGTEH-Sim. This can be attributed to the isolated aging with solvents. All solvents contributed to the overall improvement of properties, but the pore properties were particularly enhanced when ethanol was used [21]. Furthermore, compared to SGTEH-Sim, SGTEH-Iso was found to have a stronger gel network without any crack formation during ambient pressure drying. This is likely due to the isolated aging with heptane, which is known to strengthen the gel network [10]. The FTIR results showed that both aerogels had a silica network with peaks at 794 and 1063 cm^{-1} , respectively. This was the result of using a biomass-based precursor (rice husk ash) for sol-gel as the rice husk ash is rich in silica content, which was transferred to silica aerogels. Moreover, peaks at 1635 and 3600 cm^{-1} showed the presence of water hydroxyl groups, which is likely the result of aging in ethanol [22]. The BET analysis revealed that both aerogels were within the mesoscale dimension, hence highly mesoporous. The surface area, pore size, pore volume, and porosity of SGTEH-Iso was higher than those of SGTEH-Sim. This is clearly the result of isolated aging with solvents. PH results indicated that both aerogels achieved the highest removal% at pH 6, but SGTEH-Iso showed a comparatively higher removal%. This can be attributed to the higher surface area, pore size, pore volume, and porosity of SGTEH-Iso, which, in turn, provided more adsorption sites for the metal ions to attach to SGTEH-Iso [23]. The pH removal percentages by both silica aerogels

were higher than those reported in previous studies that had used silica gels with different modifying agents [12-13,24-26]. The metal concentration results showed that SGTEH-Iso and SGTEH-Sim aerogels achieved the highest removal% at 5 and 10 ppm, respectively. Beyond this level of concentration, the removal% decreased. This suggests that beyond 10 ppm, the adsorption sites on the aerogels could not attach to metal ions anymore, but this does not imply that all adsorption sites were filled. Although it is quite possible that the sites on the surface of aerogels were filled with metal ions, the sites on the inner pores were still empty and any further mass transfer was slowed or even stopped on surface sites [13]. Based on the results concerning different adsorbent doses, both aerogels achieved considerably high removal% at 0.1 g and nearly 100% removal at 0.2 g. This can be attributed to the higher number of adsorption sites with higher doses of adsorbents, which, in turn, could attract more metal ions. When compared to previous studies that used silica gels modified with different agents and solvents, SGTEH-Sim was found to have higher removal% than those reported in studies 16, 24, and 25] whereas SGTEH-Iso showed higher removal% than those reported in studies 17 and 27-29].

CONCLUSIONS

In the present study, the green method was employed to synthesize two silica aerogels. Rice husk ash was used to extract water glass solution as a precursor for sol-gel. Two novel approaches were followed during the sol-gel process. First, sustainable and cost-effective solvents, including TEOS, ethanol, and heptane, were used. Secondly, each of the silica aerogels was synthesized using the modified aging method. SGTEH-Iso was aged separately in solvents while SGTEH-Sim was aged simultaneously. Although the characterization results showed that isolated aging led to better properties, simultaneous aging was also found to produce a comparatively significant aerogel. Both silica aerogels were investigated for Pb removal from aqueous solution. As per sorption results, SGTEH-Iso performed better than SGTEH-Sim and showed a higher removal%. Moreover, compared with the removal% reported in previous studies, SGTEH-Iso performed perfectly well whereas SGTEH-Sim showed quite good results. The present study showed that it is possible to use sustainable solvents,

modified aging, and ambient pressure drying to synthesize silica aerogels to obtain aerogels with remarkable properties and efficient Pb removal.

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