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by Rosalyza Hasan

Synthesis of KCC-1 using rice husk ash for Pb removal from aqueous solution and petrochemical wastewater

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ABSTRACT

A silica-rich rice husk ash (RHA, 95.44% SiO_2) was used as a silica precursor in the synthesis of KCC-1 for Pb(II) removal. The extraction of silica was carried out under several extraction methods (alkali fusion (AF), reflux (RF) and microwave heating (MW)) and extraction parameters (NaOH/RHA mass ratio, fusion temperature and H_2O/NaOH-fused RHA mass ratio). The highest silica content was obtained using AF method at extraction conditions of NaOH/RHA mass ratio = 2, fusion temperature = 550 °C, and H_2O/NaOH-fused RHA mass ratio = 4, with silica concentration of 85,490 ppm. TEM, FTIR, and BET analyses revealed the synthesized KCC-1 has fibrous morphology with surface area of 220 m^2/g. The synthesized KCC-1 showed good performance in removal of Pb(II) from aqueous solution (74%) and petrochemical wastewater (73%). The analyses of petrochemical wastewater revealed that the adsorption process using synthesized KCC-1 effectively decreased the concentration of COD (489 mg/L to 106 mg/L), BOD (56 mg/L to 34 mg/L) and Pb(II) (22.8 mg/L to 0.5 mg/L). This study affirmed that KCC-1 was successfully synthesized using RHA as silica precursor and applied as an efficient adsorbent for Pb(II) removal.

Keywords: Rice husk ash (RHA), Alkali fusion, Lead, Adsorption.

1. Introduction

The issue of disposal of various pollutants into wastewater has been emerged with the rapid industrialization over these decades, with the heavy metals contributed to the largest sorts and even hardest to be treated. Lead (Pb) undoubtedly as the most problematic element which brought adverse impacts to this ecosystem [1]. Even in a mild quantity, Pb can resulted in severe neurological and physiological consequences to human beings [1]. Once this element has been ingested exceeded the allowable concentration, it tends to accumulate in the human body, and consequently cause serious health disorders [2]. The widespread of Pb in the ecosystem devoted to the efforts of investigating the most efficient technique to eliminate Pb from contaminated water. There are several well-documented and widely applied heavy metals removal techniques such as precipitation stabilization [3], ion exchange [4], coagulation-flocculation [5], and adsorption [6–9]. Generally, the simplicity design, technical user friendly and cost effectiveness would be the key considerations for treatment selection [10].

In recent years, a considerable number of researches have been devoted the application of mesoporous silica materials as adsorbent due to their well-ordered structure and high surface area [11,12]. In 2010, a novel mesoporous silica, namely, fibrous silica nanosphere (KCC-1) with unique spherical shape and high surface area was discovered [13]. Unlike the typical pore-based silica materials, KCC-1 is surrounded by vast amount of dendrimer, thus form fibrous morphology on it [14]. The unique morphology of KCC-1 renders abundant accessible active sites, which subsequently enhance its performance towards several applications [15]. The impressive physical properties of KCC-1, induced the attempt to remove Pb(II) by using KCC-1 as adsorbent. Nonetheless, the involvement of relatively expensive commercial silica precursor during KCC-1 preparation such as tetraethyl orthosilicate ($SiC_8H_{20}O_4$, TEOS) and sodium silicate (Na_2SiO_3), gave the idea of seeking alternative silica source.

In the meantime, low cost and high availability agricultural by-products which owned high silica content would be a good choice. Rice husk, which known to be invaluable agro-based waste accounts for 545 million metric tons (roughly one-fifth of the annual yield of rice around the world) [16]. Factories tend to combust the rice husk due to the overwhelming availability of this rice husk to form rice husk ash (RHA) and dumped by landfilling. Since these materials was disposed in an uneconomical way, utilization of the material can achieve win-win situation by extracting silica content in it and applied in KCC-1 preparation. Thus, in this study, RHA was used as silica precursor for KCC-1 and the performance of synthesized KCC-1 was analyzed for Pb(II) removal from aqueous solution and wastewater.

3.0 Result and discussion

3.1 Pre-treatment of Rice Husk Ash

Table 1 shows the oxide groups in RHA with and without acid leaching treatment. From the result shown in Table 1, silica (SiO_2) is the major component in both RHA indicating the high suitability of RHA as silica precursor for KCC-1 synthesis. By comparing the percentage of SiO_2 in RHA and A-RHA, it was clearly observed that the pre-treatment of RHA with HCl was improved the purity of SiO_2 by removing the metallic impurities in RHA. The presence of chloride ion (Cl^-) in HCl attracted the metallic element to form salts, which can be easily dissolved and removed by filtration [21]. The positive role of acid leaching in removing the metallic impurities of RHA was also reported by Bakar et al. with an increase in SiO_2 purity from 95.77 % to 99.58% [22].

Table 1: XRF Analysis of RHA and A-RHA

Parameter	RHA (%)	A-RHA (%)
SiO_2	88.52	95.44
K_2O	5.783	2.43
P_2O_5	2.76	1.02
CaO	1.35	0.54
MgO	0.46	0.18
Cl	0.40	0.18
SO_3	0.24	0.00
Fe_2O_3	0.16	0.10
MnO	0.14	0.08
Al_2O_3	0.08	0.00
ZnO	0.04	0.01
Rb_2O	0.02	0.01
SrO	0.01	0.01
CuO	0.01	0.01
NiO	0.01	0.00

3.2 Sodium silicate preparation from rice husk ash (Na_2SiO_3-RHA)

Figure 1 shows the influence of extraction method on the concentration of extracted silica. The amount of the silica content in Na_2SiO_3-RHA in a sequence of AF > RF > MW, indicating the excellent performance of AF method in the extraction process. An excellent performance of AF method was also reported by Shoppert [23], owing to its high silica concentration, efficient and energy-saving. Since AF method was found as a best method for extraction process, this method was used to identify the best extraction conditions.

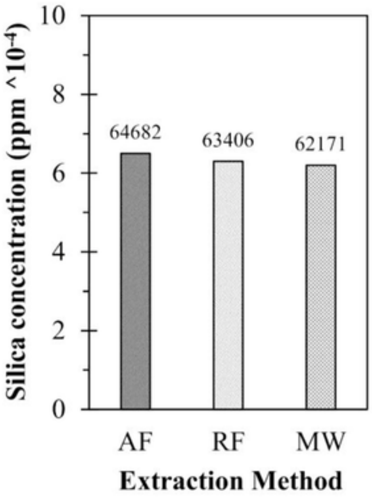

Figure 1: The silica concentration versus extraction method. Condition: NaOH/RHA = 1:1; *T* = 550°C, and H_2O/NaOH-fused RHA = 2:1.

Several extraction parameters including NaOH/RHA mass ratio (5:1, 1:1, 2:1 and 3:1), fusion temperature (500, 550, 600 and 700 ºC) and H_2O/NaOH-fused RHA mass ratio (1:1, 2:1, 4:1 and 5:1) were studied to identify the highest amount of silica concentration. Figure 2(A) shows the effect of NaOH/RHA mass ratio towards extracted silica concentration. An increase in the mass ratio of NaOH/POFA resulted to an increase in extracted silica concentration, passing through the optimum at 2, and decreased at elevated mass ratio NaOH/RHA. This result might be related with the changes in the pH of solution, and thus altering the silica dissolution process in aqueous solution which predominantly caused by the hydrolysis of Si-O-Si bonds [24]. Similar trend was reported by Keawthun et al. [25] for conversion of waste glasses into sodium silicate solutions.

Figure 2(B) shows the effect of the AF temperature towards extracted silica concentration. According to the plot, it was observed that increasing in AF temperature up to 550 °C, increased the concentration of extracted silica and slightly decreased at elevated temperature. The similar optimum fusion temperature (550 ºC) was also reported by Yilmaz and Piskin [17] for the extraction of silica from tailings slurry of gold mine treatment plant by AF method. They reported that the most suitable fusion temperature is 550 ºC and higher fusion temperature will lead to the degradation of chemical compounds and consequently decrease the silica extraction efficiency.

Figure 2(C) displays the effect of mass ratio of H_2O/NaOH-fused RHA towards the concentration of extracted silica. An increase in the mass ratio of H_2O/NaOH-fused RHA resulted to an increase in extracted silica concentration, passing through the optimum at 4, and decreased at elevated mass ratio. Silva et al. [26] reported that the lower ratio of H_2O/NaOH-fused RHA would decrease the concentration of sodium, which can be claimed on the highly viscous solution worsen the leaching process. Similar trend was also reported by Shelke et al. [27] for the extraction of silica from RHA.

From the results observed in Figure 2, the best extraction conditions were achieved at mass ratio of 2, fusion temperature of 550 ºC and H_2O/NaOH-fused RHA mass ratio of 4.

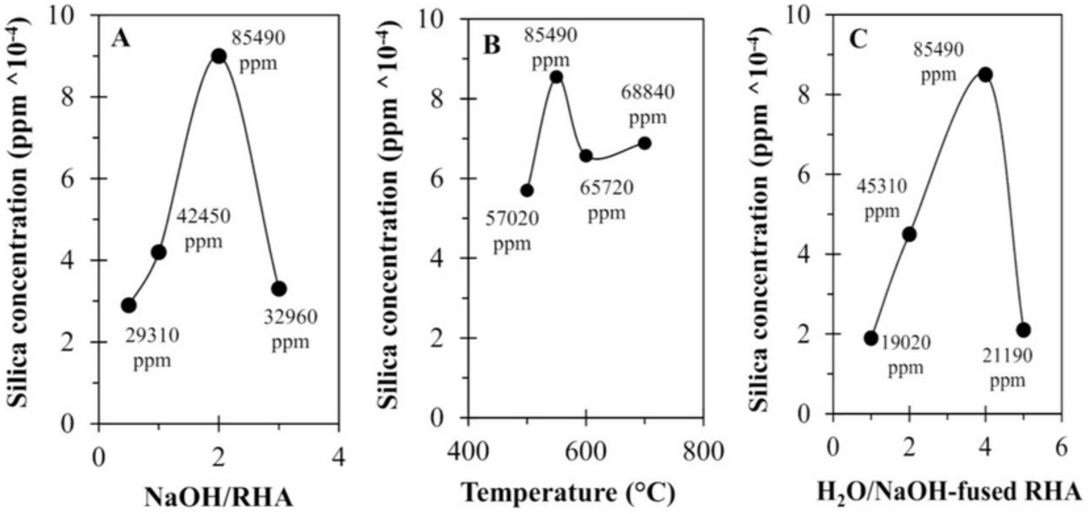

Figure 2: Concentration of extracted silica at different effect of (A) NaOH/RHA mass ratio; (B) fusion temperature; and (C) H_2O/NaOH-fused RHA mass ratio.

3.3 Characterization of KCC-1 synthesized from RHA

Figure 3 shows the TEM image of KCC-1 synthesized from RHA. As illustrated from the figure, the sample was spherical in shape and covered with the fibrous morphology. The TEM image of the synthesized KCC-1 was in conformity with literature [15,28].

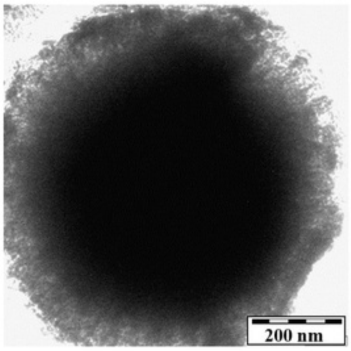

Figure 3: TEM image of KCC-1 synthesized from RHA.

The N_2 physisorption analysis revealed that the synthesized KCC-1 possesses BET surface area (S_{BET}) of 220 m^2/g, pore size (d_p) of 17.37 nm, and pore volume (V_p) of 0.94 cm^3/g. As compared to the literature [29], the synthesized KCC-1 has relatively lower S_{BET} as compared to the literature (641 m^2/g) that synthesized using TEOS, might be caused by the impurities that existing in RHA, thus, affected the KCC-1 structure [30]. This study was in agreement with Wang et al. [30] whom reported that the textural properties of SBA-15 synthesized using coal gangue (S_{BET} = 552 m^2/g, d_p = 7.0 nm, and V_p = 0.54 cm^3/g) was lower than that of SBA-15 synthesized using commercial sodium silicate (S_{BET} = 567 m^2/g, d_p = 7.2 nm, and V_p = 0.68 cm^3/g), due to the impurities present in coal gangue.

The functional groups present in KCC-1 synthesized from RHA were analysed using FTIR, as shown in Figure 4. The spectra shows several bands at approximately 3423 cm^{-1}, 1058 cm^{-1}, 800 cm^{-1}, and 450 cm^{-1}, which ascribed to the O-H stretching vibration of Si-OH, Si-O asymmetrical stretching, Si-O symmetrical stretching, and bending of Si-O, respectively [31]. Similar peaks were also reported by Dong et al. (2015) for modified fibrous silica nanospheres [32], signifying the successful formation of KCC-1 structure from RHA as silica precursor.

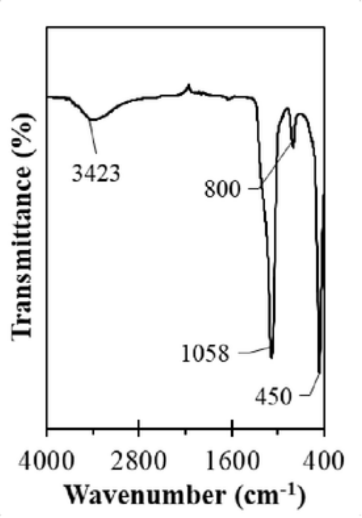

Figure 4: FTIR spectra of KCC-1 synthesized from RHA.

3.4 Adsorption performance of KCC-1 synthesized from RHA

The adsorption performance of synthesized KCC-1 on Pb(II) removal was evaluated using aqueous solution and petrochemical wastewater and the results are shown in Figure 5. KCC-1 was proven to have high feasibility as adsorbent for Pb(II) removal as demonstrated in its high removal percentage (74% (aqueous), 73% (wastewater)), owing to its favourable structural properties. In addition, the analyses of wastewater (Table 2) clearly showed that the synthesized KCC-1 effectively reduced the COD, BOD and heavy metals concentration of the wastewater, indicating an excellent performance of KCC-1 in adsorption process. An excellent performance of synthesized KCC-1 in Pb(II) removal (74%, 37.26 mg/g) can be proved by the comparison of the KCC-1's performance with other reported adsorbent as listed in Table 3.

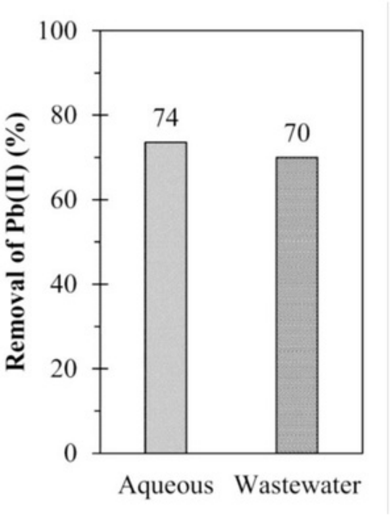

Figure 5: Removal percentage of Pb(II) from aqueous solution and petrochemical wastewater using KCC-1 synthesized from RHA. Conditions: $C_{o,aqueos} = 50$ mg/L, $m_{KCC\text{-}1} = 1$ g/L, pH = 6, and time = 80 min.

Table 2: Petrochemical wastewater analysis before and after the adsorption process using KCC-1 synthesized from RHA.

Parameter	**Before (mg/L)**	**After (mg/L)**
COD	489	106
BOD	56	34
Sodium (Na)	60.9	23.6
Potassium (K)	21.9	Not detected
Calcium (Ca)	4.2	3.02
Magnesium (Mg)	0.3	0.17
Copper (Cu)	0.0852	Not detected
Aluminium (Al)	0.0049	Not detected
Vanadium (V)	0.0031	Not detected
Manganese (Mn)	0.032	Not detected
Nickel (Ni)	0.0072	Not detected
Chromium (Cr)	< 0.0005	Not detected
Arsenic (As)	<0.0005	Not detected
Selenium (Se)	<0.0005	Not detected
Molybdenum (Mo)	0.936	Not detected

Silver (Ag)	<0.0005	Not detected
Cadmium (Cd)	0.0352	Not detected
Antimony (Sb)	0.0017	Not detected
Barium (Ba)	< 0.0005	Not detected
Lead (Pb)	22.8	0.5

Table 3: Comparison of KCC-1 performance with other reported adsorbents.

Adsorbent	Adsorption capacity (mg/g)	Ref
KCC-1 synthesized from RHA	37.26	This study
MCM-41-S9	34.33	[33]
Diatomite	26.00	[34]
Silica Gel	14.42	[35]
Phenol-formaldehyde/silicon dioxide resin (PFSR)	13.74	[36]
Mercapto-modified silica particles	10.42	[37]
Anionic layered double hydroxide (LDH)	6.81	[38]

Figure 6 shows the regeneration and reusability of synthesized KCC-1 during four consecutive adsorption–desorption cycles. KCC-1 shows excellent performance during adsorption–desorption cycles with moderate decreased in the percentage of Pb(II) removal. The declined in the adsorption performance with increasing in the number of cycles might be due to the blockage of some active sites available on the KCC-1's surface, owing to the partial desorption of Pb(II) molecules during the regeneration process [39]. The decreased in the adsorption performance of the synthesized KCC-1 (20 % for aqueous solution, 34 % for wastewater, 4 cycles) during consecutive adsorption–desorption cycles was lower as compared to the reported literatures for modified potato starch-magnetic nanoparticles (MPS-MNPs) (50%, 4 cycles) [40] and nanoscale zero-valent iron immobilized in alginate microcapsules (60%, 4 cycles) [41], implying good performance of synthesized KCC-1 in Pb removal.

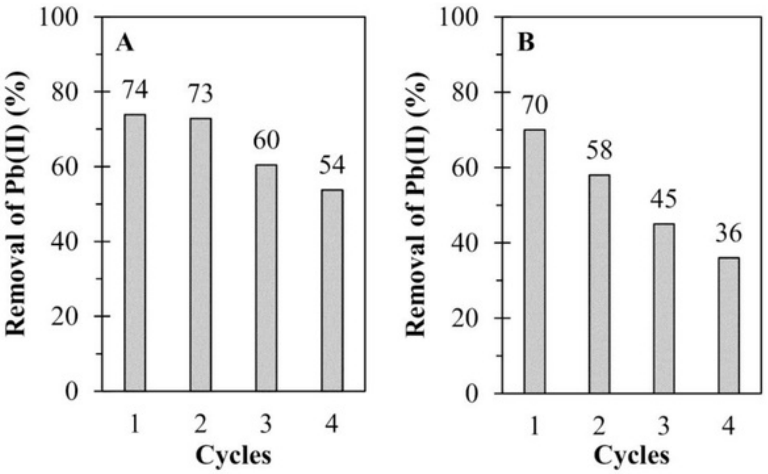

Figure 6: Reusability of KCC-1 synthesized from RHA during Pb(II) removal from (A) aqueous solution and (B) petrochemical wastewater. Conditions: $C_{o,\text{aqueos}}$ = 50 mg/L, $m_{KCC\text{-}1}$ = 1 g/L, and time = 80 min.

The spent KCC-1 was analysed using FTIR spectroscopy and was compared with the fresh KCC-1 for identifying the functional groups that responsible for the adsorption process (Figure 7). It was observed that the FTIR peaks of the synthesized KCC-1 (3423 cm^{-1}, 1058 cm^{-1}, 800 cm^{-1}, and 450 cm^{-1}) were shifted after the adsorption process due to the electrostatic and chemical interaction of functional groups with metal ions [42]. The significant alteration of 3423 cm^{-1}, in terms of the wavenumber and intensity might be related to the Pb molecules' interaction with Si-O-H of KCC-1 to form the Si-O-Pb.

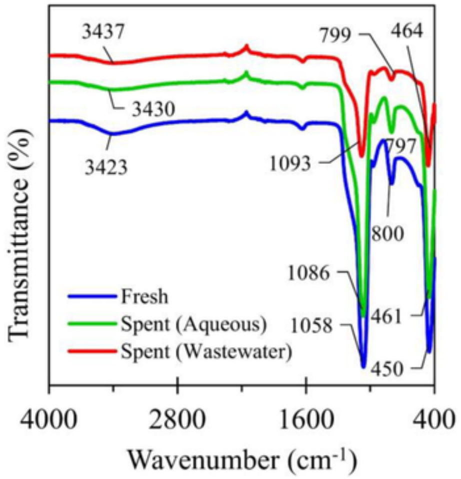

Figure 7: FTIR spectra of fresh and spent KCC-1 synthesized from RHA.

Conclusion

The potential of KCC-1 synthesized from rice husk ash (RHA) was tested on Pb(II) removal from aqueous and petrochemical wastewater. Prior to the extraction process, acid treatment was executed to enhance the purity of silica, SiO_2 by elimination the metallic impurities in RHA. The study of extraction methods (alkali fusion (AF), reflux (RF) and microwave heating (MW)) and extraction parameters (NaOH/RHA mass ratio, fusion temperature and H_2O/NaOH-fused RHA mass ratio) revealed that the highest silica content was obtained using AF method at extraction conditions of NaOH/RHA mass ratio = 2, fusion temperature = 550 °C, and H_2O/NaOH-fused RHA mass ratio = 4, with silica concentration of 85,490 ppm. The characterization analyses (TEM, BET and FTIR) of KCC-1 in conformity with literature, signifying the successful formation of KCC-1 structure from RHA. The performance studies showed that the synthesized KCC-1 has a good performance in Pb removal from aqueous (74%) and petrochemical wastewater (73%), with moderate reduction in the percentage of Pb(II) removal during adsorption–desorption cycles.

The wastewater analyses (COD, BOD and ICP-MS) revealed an excellent performance of KCC-1 as demonstrated by a significant decrease in the COD (489 mg/L to 106 mg/L), BOD (56 mg/L to 34 mg/L), and Pb(II) concentration (22.8 mg/L to 0.5 mg/L).

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