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Optimizing Silica Extraction form Rice Ash: A Comparative Study of Alkaline and Acid Leaching

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ABSTRACT

This research aimed to study the characteristics of silica which had been extracted from rice ash using two methods, i.e. alkaline and acid leaching. There were five types of solutions used; 1M and 2M sodium hydroxide (NaOH) solutions, 1M and 2M potassium hydroxide (KOH) solutions, and a 1M hydrochloric acid (HCl) solution. After the extraction process, the amount of silica was calculated and analyzed using FTIR and XRD. Based on FTIR analysis, the Si-O-Si group was found within the frequencies range of 782 cm^{-1} to 798 cm^{-1} while the Si-O group was found within the 3671 cm^{-1} to 3751 cm^{-1} range and the O-H group was found at 86.4541 cm^{-1}. XRD showed crystallinity index results of 44.0%, 46.4%, 13.0% and 45.1% from the variations of KOH, (1M and 2M) and variations of NaOH (1M and 2M) respectively. The usage of 1M of HCL contributed to the optimum production of silica up to 8.1g with the percentage yield of silica was 40.5%.

Keywords: Rice Husk Ash; Silica; Acid Leaching; Alkaline Leaching

INTRODUCTION

The main element of earth's crust is a silica which has been categorized in a group of minerals composed of oxygen and silicon. Silica can exist in any shape such as in gel, crystalline and amorphous forms and is a wasted material of the earth's crust [1]. Rice husk ash (RHA) is one of the sources of waste which can produce two main types of silica i.e. crystalline silica, which includes forms like quartz and cristobalite that may form under specific high-temperature combustion conditions, and amorphous silica, which is non-crystalline and preferred for its wide range of industrial applications due to its high surface area and versatile properties [2]. In addition, there are a few methods to extract silica from the RHA such as the sol-gel process under ultrasound, alkali and acid leaching, steam explosion, alkali hydrothermal treatment and combustion. These methods involve the use of various chemical and physical processes to obtain high-purity amorphous silica from RHA, which has a wide range of industrial applications [3-5]. Many pharmaceutical industries used silica as an excipient in the form of colloidal silicon dioxide or silica gel. For example, it can be used in the drug formulation of tablets where it acts as a glidant to improve the flow properties of the powder blend during tablet compression [6]. In the manufacturing industry, silica is the raw material for ceramics, zeolite synthesis, glass, electronic insulator materials, and it is also a catalyst. Due to high silica content found in RHA, it can be used as part of a common fabrication method in electric arc and high strength concrete for the construction industry [7,8].

Many researchers studied in synthesized SiO_2 using alkali and acid solvent; however, there still needs to be more literature reporting on the size and characteristic of SiO_2 particles. The alkaline method is characterized by its straightforward process, which entails the dissolution of rice husk ash in an alkaline solution, followed by the precipitation of silica using an acidic solution. This simplicity facilitates its implementation and scalability for industrial production [2]. Then, the alkaline solution's reaction with silica prompts the creation of a gelatinous substance. The gel-like consistency of this substance simplifies subsequent handling and processing required to extract amorphous silica [9]. Additionally, the alkaline leaching method is also a low energy method, which makes it more cost effective compared to the smelting method [10].

Acid leaching method also has many benefits such as being able to produce higher pure silica from rice husk which can be used in the production of silica materials [2]. For the removal of metal impurities, acid leaching is the best method compared to others [11]. Another benefit that we can see is that the acid leaching method can improve silica's purity and surface area [12], which can result in the production of good quality silica. There are several factors that affect the extraction of silica from RHA which can influence the transformation of RHA to silica including the soil type, climatic conditions and agricultural practices [2]. Furthermore, combustion after acid treatment can produce high purity silica which results in the RHA turning from black color to white color, which signifies the change of the RHA to silica [13].

MATERIALS AND METHODOLOGY

Materials

Rice husk ash (RHA) was purchased from D'Syira Enterprise, Taman Seri Serdang, Seri Kembangan. Using the mortar, the RHA was crushed until the sample was fine followed by a sieving process to 250-micron. The chemicals solvent that had been used in this research were hydrochloric acid (HCl), potassium hydroxide (KOH) and sodium hydroxide (NaOH) were purchased from AM Northern Chemicals.

Alkaline leaching method

To make the RHA easy to grind, it was dried in the oven at 100°C for an hour. Then, to get a sample, 500g of this dried ash was crushed using a mortar, followed by a sieving process to get 250-micron size of powder. 20g samples were prepared for the alkaline leaching process with potassium hydroxide (KOH) and sodium hydroxide (NaOH), at a concentration of 1M and 2M. The sample was heated to 80°C for an hour, while stirring at 400 rpm. After the mixture was filtered and left to cool down to room temperature, it was titrated with HCl to form a precipitate. Once the mixture reached a pH of 7 it was stirred further. The precipitate was filtered out and dried out in the oven at 110°C for 1 hour [12].

Acid leaching method

The sample was prepared with 20g of rice husk ash and 500 mL of 1 M HCl. This mixture was then stirred for 90 minutes on a magnetic stirrer at room temperature. After the stirring process, the mixture was filtered using a filter paper. The filtration was washed with 100mL of distilled water at room temperature using a magnetic stirrer for swirling. The wash was filtered again to remove the water and was dried in an oven at 110°C for 1 hour [6].

Dried precipitate of silica

Eq. (1) was used to determine the percentage yield of silica for each of the samples from alkaline and acid leaching process.

$$Yield\ (\%) = \frac{The\ amount\ of\ silica\ obtained}{The\ amount\ of\ rice\ husk\ ash} \times 100 \tag{1}$$

Each sample was then tested with XRD, which revealed the crystallographic structure, chemical composition, and physical properties of each sample, whilst, FTIR was used to determine the types of groups band, i.e. Si-O, Si-O-Si, and O-H in the silica.

RESULTS AND DISCUSSION

The percentage yield of SiO_2

Table 1 shows the amount of silica extracted from the alkaline and acid leaching process.

Table 1: Percentage yield of silica in rice husk ash

No.	Solvent types	Rice husk ash (g)	Silica (g)	Yield (%)
1	1M KOH	20	5.5	27.5
2	2M KOH	20	6.9	34.5
3	1M NaOH	20	7.7	38.5
4	2M NaOH	20	7.9	39.5
5	1M HCl	20	8.1	40.5

The weight of rice husk ash (RHA) used for each of the leaching process was 20g. For the 1M of KOH, it produced 5.5g of silica which had a percentage yield of silica 27.5%, whilst, for 2M of KOH, it produced 6.9g of silica and the percentage yield was 34.5%. Additionally, 1M of NaOH could produce 7.7g of silica and the percentage yield of silica was 38.5%. 2M of NaOH could produce 7.9g of silica and the percentage yield was 39.5g. As can be seen in Table 1, the higher the concentration of alkaline used as a solvent, the higher amount of silica can be produced.

However, the higher amount of silica produced was achieved by using 1M of HCL which resulted in 8.1g of silica produced and this had a higher percentage yield of silica which was 40.5%. Acid leaching significantly useful and efficient compared to alkaline solution to remove impurities and increased the purity of silica. Leaching with hydrochloric acid (HCl) can dissolve inorganic impurities such as metals contained in rice husk up to enlarge the pores. When the pore is increased, the surface area of the material increases as well. This contributes to the higher extraction of silica compared to the alkaline leaching process [13].

Several previous studies have shown increasing the concentration of NaOH affects the yield of SiO_2 from RHA [14]. The above results are in accordance with the SiO_2 extraction process from fly ash, there is an increase in SiO_2 yield along with the increase in the concentration of NaOH. This is due to increasing NaOH concentration, increase the OH^- ions in solution and more SiO2 is bound from RHA [15].

Fourier Transform Infrared Spectroscopy (FTIR) analysis

To clarify the composition and functional groups of SiO2 produced by various alkali and acid solvents, the interpretation of the FTIR spectrographs and sample observation are depicted in Figure 1. It shows that the commercial SiO_2 and SiO2 with varying KOH and NaOH concentrations were dominated by silanol (Si-O-H) and siloxane (Si-O-Si) group bands which gave wavelength at 798 cm^{-1} for 1M KOH, 1M NaOH and 2M NaOH, 782 cm^{-1} for 1M HCl, and 796 cm^{-1} for 2M KOH, respectively. However, the presence of silanol (Si-OH) groups resulted in peaks in the spectrum at 1588 cm^{-1} for 1M HCl, 1656 cm^{-1} for 1M KOH, 1635 cm^{-1} for 2M KOH, 1633 cm^{-1} for 1M NaOH, and 1646 cm^{-1} for 2M NaOH.

The wavelength at 3751 cm^{-1} for 1M HCl, 3744 cm^{-1} for 1M KOH, 3671 cm^{-1} for 2M KOH, 3745 cm^{-1} for 1M NaOH, 3751 cm^{-1} for 2M NaOH attribute to the hydroxyl (O-H) group.

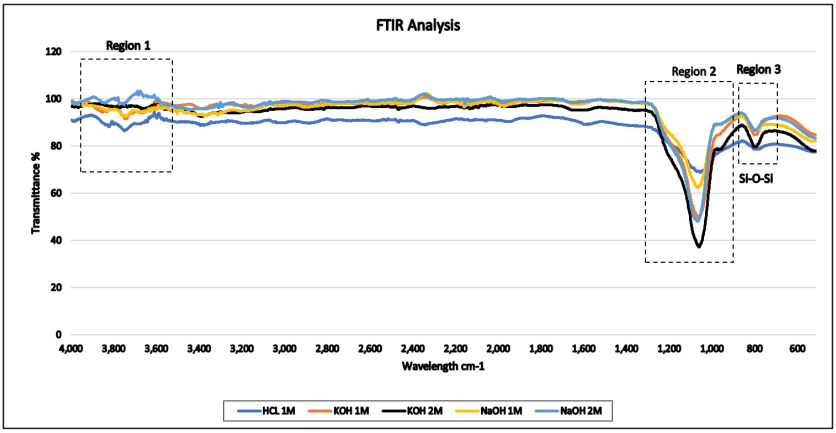

Figure 1: FTIR spectra of silica from acid and alkaline leaching extraction

As could be seen in the FTIR result above, the silanol (Si-OH) group in alkaline leaching samples had higher wavelength than the acid leaching sample. However, for hydroxyl (O-H) groups, the 2M NaOH had the same wavelength as the 1M HCl. A sample that showed a peak at 782 cm^{-1}, corresponding to the siloxane (Si–O–Si) group and possessing another peak at 1646 cm^{-1} for the Si–OH group signified that these functional groups were heavily prevalent [15]. The hydroxyl (O-H) group at 3751 cm^{-1} was also significant and seemed to be extremely sharp without excessive broadening, promising a balanced and solid structure of silica. In addition, the consistent presence of key functional groups suggested that the extraction process with 2M NaOH was reliable and reproducible, making it a suitable choice for high-quality silica extraction from rice husk ash [16,17].

The process of forming the Si-OH group occurs when KOH is dissolved in HCl, while the Si-O-Si group is formed during condensation [18]. Based on this research, it is concluded that SiO_2 from RHA extracted with varying concentrations of KOH has the same peak similarity as commercial SiO_2.

X-ray Diffraction (XRD) analysis

XRD was performed to analyze the data to determine whether the phase of the sample was either amorphous or crystalline. With regards to its functional group, physical makeup, and amorphous character, the resulting silica powder was comparable to commercial silica for acid leaching process. Numerous other investigations [19,20] have demonstrated that acid leaching can increase the amorphousness of silica. This proven through FTIR analysis which stated the peaks in the spectrum at 1588 cm^{-1}. The phase change of silica can be caused by alkali metals found in rice husks, which can encourage the synthesis of cristobalite [21]. These alkaline metals can be eliminated by acid leaching, which stops the silica's phase change. In another study by Bakar *et al.* [14], high-purity amorphous silica was synthesized through combustion following acid treatment of rice husks (RH). This acid leaching process led to a notable increase in silica purity, elevating it from an initial level of approximately 95 % to around 99 %.

Figure 2 and Figure 3 show the XRD analysis for 1M KOH and 2M KOH respectively. The diffraction characteristics of SiO_2 powder formed by increasing the concentration of KOH are illustrated in Fig. 2 and 3. The position of the diffraction pattern at 28.42◦ confirm the crystalline SiO_2, which is also confirmed by FTIR with the peak present at 1635 cm^{-1}. It has been reported by Hanum *et. al.* [15], the peaks at 27.75◦, 27.81◦, 27.95◦, 28.15◦, and 28.35◦ form sharp peaks indicating that SiO_2 from RHA has a crystalline structure. This peak appears due to impurities in the form of KOH and NaCl. Impurities in the form of KOH may still be included because of the excessive concentration of KOH so that it is left behind when the reaction takes place. While impurities in the form of NaCl are still included because of the titration time using HCl. This sharp XRD pattern also shows the same pattern reported by [21]. In addition, the 1M KOH solution would normally be enough to dissolve a significant portion of the silica which was present in the rice husk ash. The siloxane (Si-O-Si) bonds were attacked by the hydroxide ions (O-H) in the solution to dissolve the silica network thereby creating silicate ions [20].

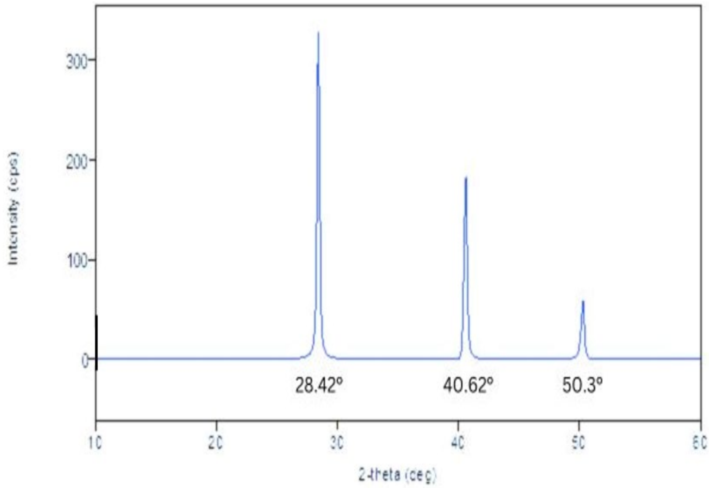

Figure 2: XRD analysis for 1M KOH

Figure 3 shows the XRD diffraction pattern of 1M KOH in the extraction silica. Silicon (Si), one of the other components being tracked, was present at the peak intensity of 28.48°. During the extraction procedure, RHA was subjected to a 2M KOH solution, which was two times as concentrated as the previously described 1M KOH solution. With the high concentration of KOH in this sample, the impurities present, such as residual organic matter and inorganic contaminants, could be removed from the ash which would lead to higher purity of silica. However, excessively high concentrations of KOH can sometimes lead to new impurities or unwanted side reactions which would have negative effects on the silica produced. The lower KOH concentrations would result in incomplete dissolution of silica which would lead to lower yield [18]. Purity and crystal structure of the silica pointed to the removal of non-crystalline components and contaminants, resulting in quartz silica [20]. The findings highlighted the efficiency of the alkaline treatment in dissolving and purifying silica, with the concentration of KOH playing a pivotal role in the quality and yield of the extracted material.

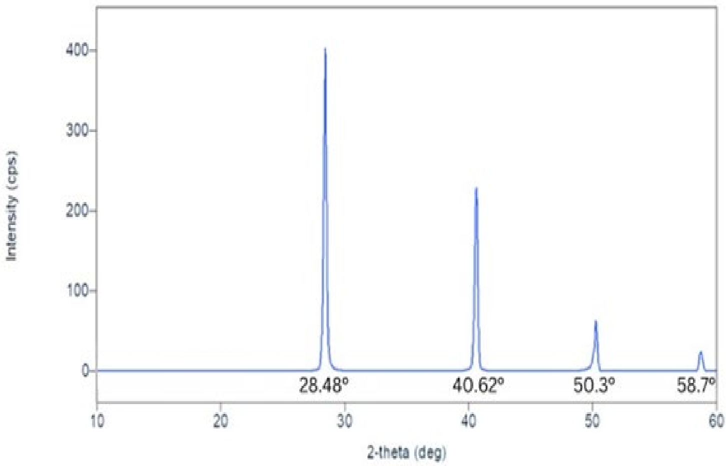

Figure 3: XRD analysis for 2M KOH

The intensity of the crystalline peaks in Figure 3 indicated that the extraction process using 2M KOH was effective in preserving the crystallinity of silica. The higher concentration of KOH likely enhanced the dissolution of silica from RHA, leading to a more pronounced crystalline phase compared to the 1M KOH extraction [20]. Alongside the crystalline peaks, a broad peak indicative of amorphous silica was also observed, reflecting the presence of both crystalline and amorphous phases in the extracted material. This dual-phase nature is common in silica extracted from natural sources like RHA, where amorphous silica typically coexists with crystalline forms such as quartz [21].

Figure 4 and Figure 5 show the XRD analysis for 1M NaOH and 2M NaOH respectively.

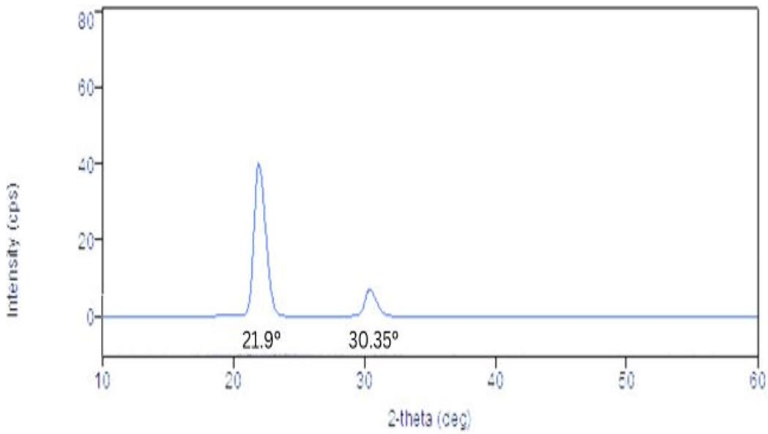

Figure 4: XRD analysis for 1M NaOH

The diffraction characteristics of SiO_2 powder formed by increasing the concentration of NaOH are illustrated in Figure 4 and Figure 5. The position of the diffraction pattern at 21.9º confirm the amorphous SiO_2, which is also confirmed by FTIR with the peak present at 798 cm^{-1}. This result can be related to the lower crystallinity index in Table 2.

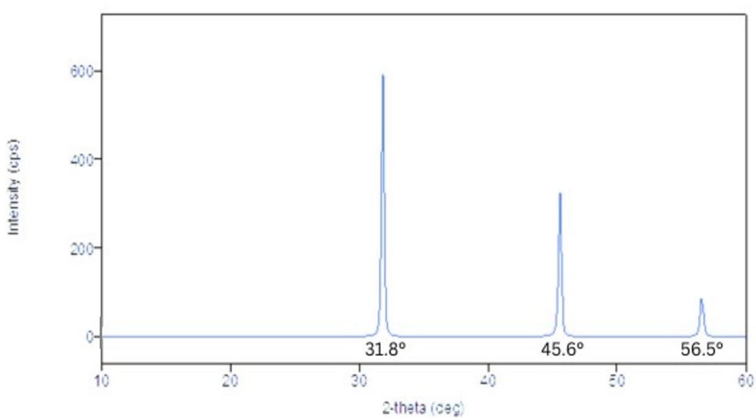

Figure 5: XRD analysis for 2M NaOH

The intensity and sharpness of these peaks suggested a high degree of crystallinity in the extracted material. The process of silica dissolution, when catalyzed by NaOH, happens when several water molecules attack a neutral silica surface in the presence of an NaOH molecule. The trick is to make sure that each water molecule approaches the silicon atom and that the Si-O bonds on the surface. This bond is made stable by the presence of the NaOH molecule. So basically, the NaOH helps the water molecules break down the silica [15]. The neutral silica surface interacts with water molecules during the NaOH-catalyzed silica dissolving process. NaOH is essential to this process because it helps water molecules assault the silica surface by stabilizing the Si-O bonds. This is how the mechanism may be explained where NaOH interaction with the Si-O bonds is stabilized by the interaction between the NaOH molecule and the silica surface [17].

Crystallinity Index (%)

Table 2 shows the crystallinity index of the silica extracted from both leaching processes. Based on Table 2, it shows that the concentration of KOH and NaOH affects the crystallinity development of SiO_2. When a higher concentration of KOH and NaOH were used, the formation of crystallization will be increased. In addition, the melting and crystallization of particles may occur more quickly in higher molarity of KOH and NaOH. The crystallinity index for 1M NaOH shows a lower percentage parallel with the peak present in Figure 4. This is due to the presence of other impurities in the product, thereby reducing the degree of crystallinity.

Table 2: Results of crystallinity index %

No	Alkaline Solvent	Crystallinity Index, %
1.	1M KOH	44.0
2.	2M KOH	46.4
3.	1M NaOH	13.0
4.	2M NaOH	45.1

This difference can be attributed to the stronger alkaline nature and higher ionic potential of potassium ions (K^+) compared to sodium ions (Na^+). The stronger alkalinity facilitated more effective dissolution and leaching of amorphous silica from the RHA matrix [15]. This means that 2M NaOH is the best solvent to use to extract silica from rice ash. Additional refinement of leaching parameters, such as temperature, duration, and solid-to-liquid ratio, may enhance the purity of the extracted silica and the crystallinity index. Furthermore, adding purification processes such as ion-exchange may aid in the removal of any remaining contaminants from the silica product [22].

CONCLUSION

In this research, the extraction of the silica from the RHA was successfully achieved using the alkaline leaching method (KOH solution and NaOH solution) and acid leaching method (HCL solution). The 2M NaOH was selected as the optimum solvent for the extraction process due to the higher yield percentage of the silica and crystallinity index. In addition, it also had higher peaks for groups of silica like siloxane, silanol and hydroxyl based on the Fourier Transform Infrared Spectroscopy (FTIR) analysis. In the XRD results, the 2M NaOH silica sample showed the crystalline silica in tridymite phase which makes it a suitable choice for extracting high-purity silica from rice husk ash. It also had a high crystallinity. It has been discovered that acid leaching results in a better purity and specific surface how well they remove organic materials and metallic contaminants. The concentration, reaction time, and temperature of alkaline and acid solutions affect their capacity to dissolve organic and metallic contaminants from rice husk.

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