Available online at http://www.ajol.info/index.php/njbas/index
Nigerian Journal of Basic and Applied Science (June, 2016), 24(1): 41-45
DOI: http://dx.doi.org/10.4314/njbas.v24i1.7

ISSN 0794-5698

Full Length Research Article

Preparation of High Grade Silica from Rice Husk for Zeolite Synthesis

***[1]S. Yunusa, [1]A.S. Ahmed, [1]S.G. Bawa, [2]J.F. Iyun and [3]M. Dauda**
[1] Department of Chemical Engineering, Ahmadu Bello University, Zaria, 810261, Nigeria
[2]Department of Chemistry, Ahmadu Bello University, Zaria, 810261, Nigeria
[3]Department of Mechanical Engineering, Ahmadu Bello University, Zaria, 810261, Nigeria
[*Corresponding author: slmnyunus@gmail.com: ☎: +2348038951472]

ABSTRACT: Silica powder with approximately 96% purity was extracted from rice husk (RH) and used as a silica source for the synthesis of zeolite Y by hydrothermal process**.** The effect of synthesis parameters such as pH, temperature of burning the rice husk, acid leaching prior to formation of rice husk ash (RHA) on the properties of final product were studied. The acid-leached rice husk calcined at 650 °C for 6 h produced rough powder of rice husk silica, light brown in colour. The silica purity of the rice husk ash (RHA) calcined at 400, 450 and 500^{0}C were 95.6wt%, 96.1wt% and 95.89 wt% respectively. The X-Ray Diffraction (XRD) pattern of the silica obtained show that the silica was amorphous with traces of crystalline phase. The amorphous nature of the RHA and the relatively high purity of silica content in it make it a suitable source of silica for zeolite synthesis.
Keywords: Silica; RHA; Zeolite; Synthesis; Hydrothermal; Temperature

INTRODUCTION

Rice Husk (RH) is a solid waste product from rice milling industries (Kordatos *et al*., 2008). Rice Husk Ash (RHA) derived from burning RH is one of the most silica rich raw materials containing about 90-98% silica after complete combustion, highly porous and lightweight, with a very high external surface area. Its absorbent and insulating properties are useful to many research studies (Mittal, 1997; Kordatos *et al*., 2008). About 20% of the rice husk ash residues left after the combustion cause environmental problems due to the pollution of both water and air (Kordatos *et al*., 2008, Rahman *et al.,* 2009). Although, hosts of synthetic silica are produced commercially, the ones produced from plant origins such as rice husks have been noted to have some significant advantages over those from mineral and synthetic origins. In particular, the processing steps are relatively simple and require no elaborate infrastructure or consumption of costly reagents as in the case of the synthetic processes. In addition the silica powder produced from plant sources contains a narrow range of metal oxide impurities (Edson *et al.,* 2006) which makes them exceptionally desirable in applications where high purity silica at modest cost is a necessary prerequisite. Rice husk is largely composed of silica with small amounts of inorganic compounds. In practice, the type of ash from rice husk varies considerably according to the burning technique. Previous research had shown that pure silica can be obtained from rice husk by acid leaching procedure and calcination at 600 °C under atmospheric pressure. This process is considered expensive, due to the energy consumption (Edson *et al.,* 2006). Hydrochloric acid is most often used for acid leaching (Rahman *et al*., 2009, Johan *et al.,* 2011). The extraction of silica from rice husk by acid treatment provides more than 95% by weight of amorphous silica (Suyanta and Kuncaka *et al.,* 2011).

Silica (SiO_2) is one of the valuable inorganic multipurpose chemical compounds. It can exist in gel, crystalline and amorphous forms. Amorphous silica is a naturally occurring or synthetically produced oxide of silicon characterized by the absence of a pronounced crystalline structure and whose X-ray diffraction patterns have no sharp peaks (Larbi, 2010). This type of silica may be anhydrous or have a significant water of hydration in its structure. In general, rice husk ash (RHA) might well be considered slightly impure silica. The content of silica and all impurities in RHA varies, depending on the variety, climate and geographic location (Larbi, 2010). Amorphous silica is well known and commonly used as a silica source in the synthesis of micro-porous materials such as zeolites, silica gel etc (Adam *et al*., 2006). However, to obtain pure amorphous silica is energy intensive as it requires high furnace temperature (Mittal *et al.,* 1997). Previous research work (Rahman *et al*., 2009) had shown that pure silica can be obtained from RH by an acid leaching procedure and pyrolysis in an oxygen atmosphere at 500^{0}C for 5 hrs. This paper is aimed at preparation of high grade amorphous silica in an oxygen environment at lower temperature.

EXPERIMENTAL

Purification Treatment of Rice Husk (RH)

Leaching of RH

Rice husk was sieved to eliminate clay particle. This was washed with distilled water and dried in an oven at 100°C for 24hrs. Acid leaching of the rice husk was carried out to remove soluble elemental impurities such as iron, magnesium, calcium etc. Leaching was carried out at 10 wt% solids in 10wt% HCl. The HCl solution was prepared from a standard HCl stock of mean concentration of 37wt% and density of 1.19 g/ml. The dried RH was soaked in the prepared solution of 10wt% HCl for 24hrs. The treated rice husk was washed again thoroughly with distilled water until pH became 7 and then dried at 100^{0}C for 24hrs.

Pyrolysis of RH

The dried RH was pyrolyzed in an oxygen atmosphere at 500^{0}C, 450^{0}C and 400^{0}C respectively for 6 hours. The RHA obtained was characterized using Empyreal PANalytic Diffractometer, using Cu Kα radiation and Munipal 4 Energy Dispersed- X ray fluorescence (ED-XRF) machine for mineralogy and chemical compositions respectively.

Zeolite Synthesis

The three major Steps involves in synthesis of zeolite Y are Seeding gel, Feedstock gel and over all gel.

Preparation Seed Gel

About 1.7g sodium hydroxide pellets were dissolved in 7.5ml deionized water and stirred until clear and homogenous solution appeared. 2 ml of the aqueous NaOH were added to 0.75g sodium aluminate and stirred with heating until a homogenous mixture was formed. 1.5g silica source (RHA, cab-o-sil and sodium metasilicate) were added separately to 5.5 ml sodium hydroxide aqueous and stirred while heating on the magnetic stirrer until homogenously mixed (Rahman *et al.*, 2009). The two solutions were then mixed simultaneously in a 150 ml propylene bottle and stirred with heating for 30 minutes. The gel formed was then age for 24 hrs.

Preparation of Feedstock Gel

About 7.8g NaOH pellets were dissolved in 142 ml distilled water and stirred until a clear solution formed. 42.5 ml of NaOH solution were added to 13.7711g sodium aluminate and stirred with heating gently on the hot plate until clear solution appeared then 28.1463g silica source (RHA, cab-o-sil and sodium metasilicate) were added to 100ml of NaOH solution in a polypropylene bottle. The mixture was then stirred and heated on the hot water bath. The aluminate and silicate solutions formed were then mixed together in a polypropylene bottle and stirred for 2hrs.

Preparation of Overall Gel

The seed gel and feed stock gel were then mixed. The seed gel was added slowly into the feed stock gel and the mixture was continuously stirred with magnetic stirrer for 2hrs at room temperature. The mixture was then transferred into 150 ml propylene bottle and was aged for 24hrs at room temperature. After ageing, the mixture was inserted into oven at 100^{0}C for 22hrs. The propylene bottle was removed after 22 hrs in the oven and the cap was quickly opened and left to cool to room temperature. After cooling the mixture was, filtered and washed with hot de-ionized water, then dried overnight in the oven at 100^{0}C.

RESULTS AND DISCUSSION

XRD Results

Figures 1 (a-c) present the XRD patterns of the resulting RHA obtained at calcination temperatures of 400, 450 and 500˚C respectively. From the XRD patterns the broad peaks at 2θ angle range of 18-30° and low intensity counts confirmed the RHA to be largely amorphous silica, which is suitable for zeolite synthesis than the crystalline phase because amorphous silica is very soluble in alkaline medium. However, a sharp peak appears at 2θ value of about 26° which indicates the presence of quartz, a crystalline phase. The presence of quartz might have resulted from the fine particles of it that could not be trapped during the sieving of the rice husk at the treatment stage. The RHA obtained at 400^{0}C and 450^{0}C are more amorphous as shown in Figures 1(a) and (b) respectively, than the sample obtained at 500°C presented in Figure1 (c). This observation agrees with literature, that at higher temperatures, an orderly arrangement of the silica molecules occurs which makes it crystalline.

X-Ray Fluorescence (XRF) Analysis

A chemical composition of RHA was determined using XRF technique as shown in Table 1. The chemical compositions of RHA sample in oxides form with silica content of 95.6 wt%, 96.1wt% and 95.89wt% at calcination temperatures of 400°C, 450°C and 500°C respectively. Furthermore, it was observed that the silica obtained at calcination temperatures of 400°C and

450°C is alumina free. The silica obtained at calcination temperature of 400°C when used as silica source for the synthesis of zeolite Y (Figure 2 (a)) matched well when compared to the XRD pattern of standard zeolite Y (Figure 2(b)).

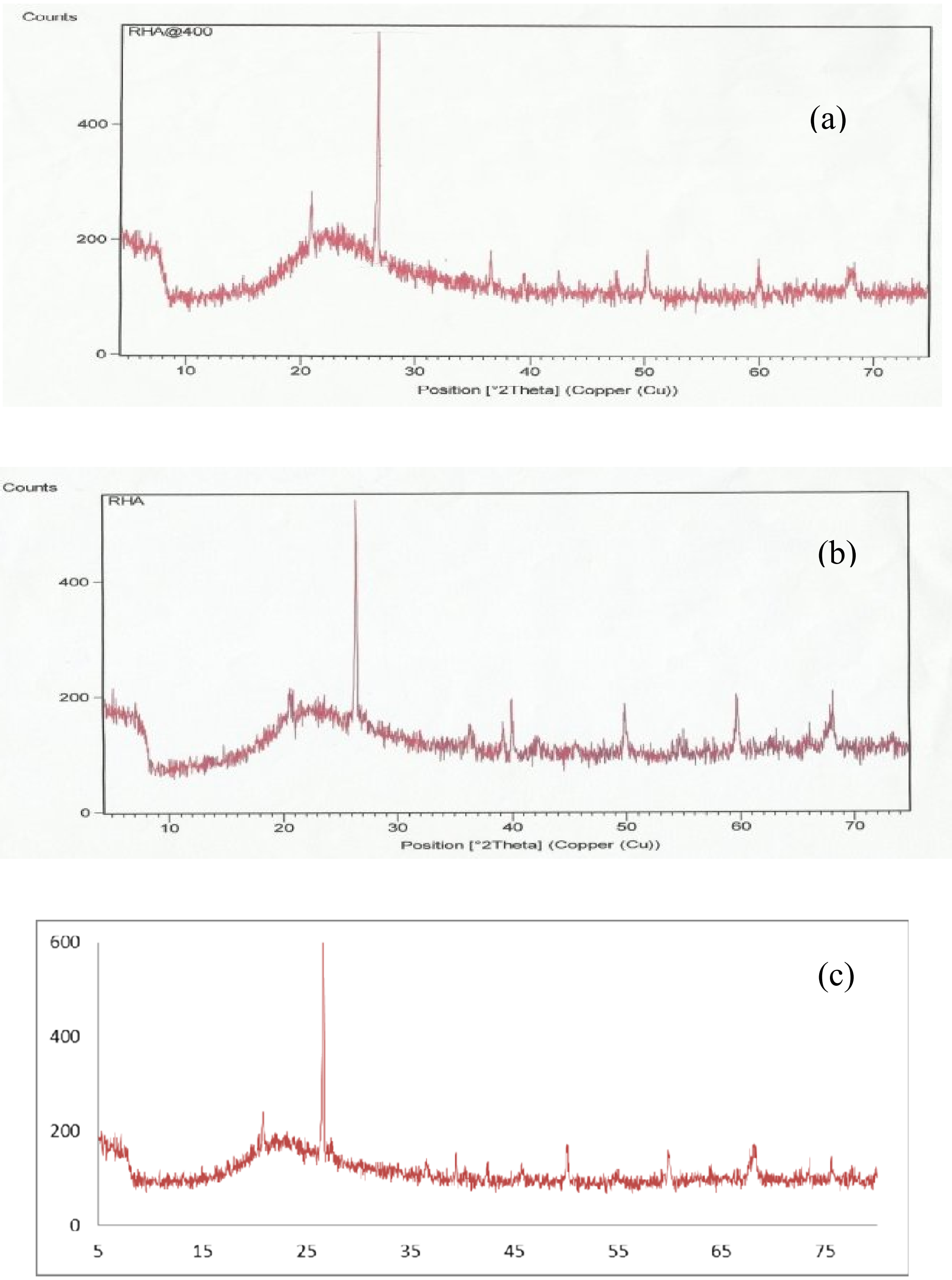

Figure 1: Rice Husk Ash produced at different temperatures (a) 400^0C, (b) 450^0C and (c) 500^0C

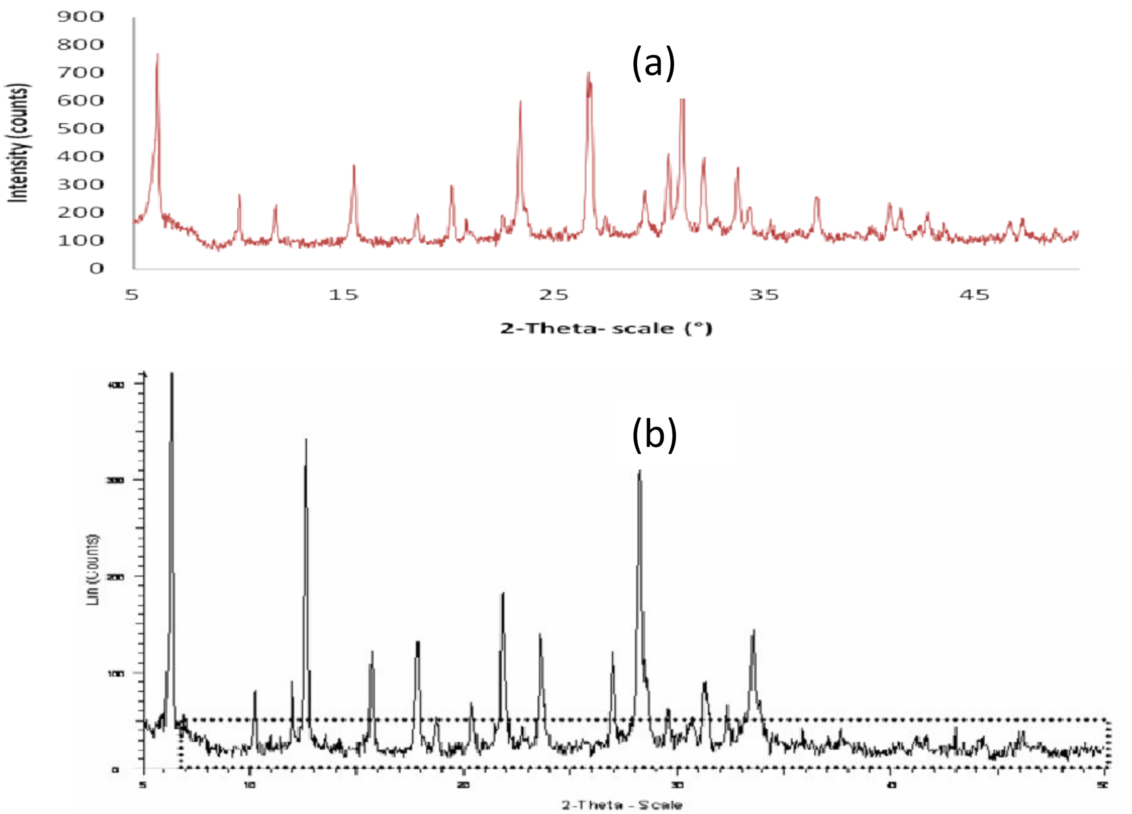

Figure 2: XRD pattern of (a) RHA based Zeolite Y and (b) standard Zeolite Y

Table 1: Chemical composition of Rice Husk Ash

	Oxides (%)							
Temperature	SiO_2	Al_2O_3	CaO	K_2O	Na_2O	LOI	Other component	Total
400°C	95.6	nd	0.59	0.65	0.04	nd	2.36	99.24
450°C	96.1	nd	0.41	0.57	0.31	0.98	2.54	99.93
500°C	95.89	0.91	0.39	0.27	0.11	0.99	2.41	99.98

nd = Not detected

CONCLUSION

It may be concluded from the investigations carried out that a simple and low energy method has been developed to produce highly amorphous silica of 96.1wt% purity from rice husk pyrolysed in an oxygen atmosphere at 450^{0}C. The silica obtained could be suitable for chemical synthesis such as preparation of zeolite. The zeolite Y prepared from RHA compares favorably with standard zeolite Y.

ACKNOWLEDGMENT

The authors gratefully acknowledge the Petroleum Technology Development Fund (PTDF), Abuja-Nigeria, for supporting this research work.

REFERENCES

Adam, F., Balakrishnan, S. and Wong, P. (2006). Rice Husk Ash Silica as a Support Material for Ruthenium based Heterogenous Catalyst. *Journal of Physical Science*, **17(2):** 1-13.

Edson, L.F., Ederson, G.L., Hadlich de, O. and Sérgio, L.J. (2006). Conversion of rice hull ash into soluble sodium silicate. *Materials Research*, **9(3):** 335-338

Jezreel J., Saceda F. and Leon R. (2011). Properties of Silica from Rice Husk and Rice Husk and their Utilization for Zeolite Y Synthesis. *Quim Nova*, **34(8):** 1394-1397.

Khemthong, P., Prayoonpokarach, S and Wittayakun, J. (2007). Synthesis and Characterization of Zeolite

NaY from Rice Husk Silica. *Korean Journal of Chemical Engineering*, **25(4):** 861-864.

Kordatos K., Gavela S., Ntziouni A., Pistiola K.N., Kyritsi A. and Kasselouri- Rigopoulou V. (2008). Synthesis of highly siliceous ZSM-5 zeolite using silica from rice husk ash. *Microporous and Mesoporous Materials*, **115(1&2):** 189-196.

Larbi, K.K. (2010). Synthesis of High Purity Silicon from Rice Husks; Master of Applied Science, Department of Materials Science and Engineering, University of Toronto. https://tspace.library.utoronto.ca/bitstream/1807/24595/1/Larbi_Kingsley_K_MASc_thesis.pdf.

Mittal, D. (1997). Silica from ash a valuable product from waste material. *Resonance,* **2(7):** 64-66.

Johan, E., Ogami, E., K, Abidin Z., Matsue N. and Henmi T. (2011). Synthesis of Zeolite MFI from Rice Husk Ash and its Ability for VOCs Adsorption.http://www.ibrarian.net/navon/page.jsp?paperid=18285525.

Rahman M. M., Hasnida N. and Wan Nik W.B. (2009) Preparation of Zeolite Y Using Local Raw Material Rice Husk as a Silica Source, *Journal of Scientific Research,* **1(2):** 285-291.

Suyanta, S. and Kuncaka, A. (2011). Utilization of Rice Husk as Raw Material in Synthesis of Mesoporous Silicates MCM-41 *Indonesian* Journal of *Chemistry*, **11(3):** 279 – 284.