ISSN NO. 2320-5407

Journal homepage: http://www.journalijar.com

INTERNATIONAL JOURNAL OF ADVANCED RESEARCH

RESEARCH ARTICLE

Production of Activated Carbon and Precipitated White Nanosilica from Rice Husk Ash

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Manuscript Info

Manuscript History:

Received: 15 December 2014
Final Accepted: 21 January 2015
Published Online: February 2015

Key words:

Rice husk ash, Precipitated white nanosilica, Activated carbon

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Abstract

This work presented the preparation of rice husk ash (RHA) by Open-field burning and by combustion at 1000 ^{o}C for 4 hrs. X-ray diffraction studies of produced RHA revealed that increase in temperature of burning will increase the crystal growth rate with the occurrence of different varying degrees of quartz (Q), Cristobalite (C) and Tridymite (T). The purity concentration of silica in RHA samples were measured by X-Ray Fluorescence and found to be in the range of 82.7-91.6 % with major impurities of K_2O, P_2O_5 and Cr_2O_3. RHA produced via open-field burning was treated with an activating reagent sodium hydroxide (3.5 mol/L), the activated rice husk so obtained was heated at a temperature of 900°C to get activated carbon. Silica was precipitated from sodium silicate by acidification using sulphuric acid with a percent yield 97% of extracted silica. Infrared spectral data supported the presence of hydrogen bonded silanol group (Si–O–H) and the siloxane group (Si–O–Si) in RHA and precipitated silica and reflects the high purity of precipitated white nanosilica (PWNS).

INTRODUCTION

Agricultural wastes in Egypt amount range from 30-35 million tons a year of which only 7 million tons are used as animal feed and 4 million tons as organic manure. Burning of agricultural wastes not only is considered an economic loss but also has harmful effects on the environment. These harmful effects are emission of poisons gases to the air and reducing the microbial activities in the soil thus causing environmental problems [1, 2].

Rice husk, which is considered as an agricultural waste, has a high ash content varying from 18 – 20 %. Due to its high silicon content rice husk ash has become a source for preparation of elementary silicon and a number of silicon compounds, especially silica [3-9], silicon carbide [10] and silicon nitride [11].

The rice husk ash so formed may contain several metallic impurities like Fe, Mn, Na, K, Ca etc. These impurities can decrease its surface area and purity. Several attempts have been made to synthesize pure rice husk silica by eliminating these metallic impurities. The major methods of treatments are acid or alkaline leaching of rice husk followed by calcination at higher temperature [12, 13].

The manufacturing cost of commercial activated carbon is in fact rather high. As such, there is a need to produce activated carbon with high adsorption capacity from alternative material that is cheaper and renewable.

Many studies have been carried out to prepare low cost activated carbons from lignin by chemical activation [14] and from agricultural wastes such as oil palm empty fruit bunch [15, 16], sugarcane bagasse [17] and rice husk [18-20].

In this work, an attempt was made to optimize the preparation conditions of nanosilica and activated carbon from rice husk (RH) waste. Utilizing RH in preparing activated carbon and nanosilica will decrease the cost of waste disposal and also convert this waste into value-added products.

2. Experimental

2.1 Material

Rice husk (RH) as precursor was obtained from the local rice processing mill. It was washed with distilled water to get rid from dust and impurities and then dried for 24 hrs at 100ºC. The dried sample was then subjected to two types of treatment to produce rice husk ash.

2.2 Production of rice husk ash

2.2.1 (Open-field burning)

A known weight of rice husk sample was put in stainless steel reactor and ignited at atmospheric pressure. Incomplete or partial burning of rice husk produced porous and bulky ash with uniform intact black particles RHA (OF).

2.2.2 (Muffle furnace)

A known weight of rice husk sample was heated in muffle furnace (Nabertherm, Germany) at 1000 ^{o}C for 4 hrs. A grey color ash rich crystalline silica was produced RHA (MF).

2.3 Preparation of activated carbon (AC)

RHA (OF) was treated with an activating reagent, sodium hydroxide (3.5 mol/L), for about 2 hour at 90°C. The activated rice husk and the solution containing the activating agent were then filtered. The activated rice husk so obtained was heated at a temperature of 900°C to get activated carbon.

2.4 Preparation of precipitated white nanosilica (PWNS)

The initial step is extraction of silica from RHA (OF) as sodium silicate using aqueous sodium hydroxide (3.5 mol/L). This reaction was carried out in an open stainless steel reactor for about 2 hrs at 99 ^{o}C and at atmospheric pressure. RHA contains mostly silica which reacts at around 90-100 ^{o}C with NaOH solution to yield sodium silicate. Transparent and colorless sodium silicate solution was obtained after filtration of the reacted slurry. The residue (on the filter medium) consisted of the unburnt carbon.

In the second step of the process, silica was precipitated from sodium silicate by acidification using sulphuric acid. The addition of the acid was done very slowly till a pH of 7 is reached.

Activation mechanism between alkali hydroxide and rice husk is complicated. The proposed reaction mechanism approved may be as follows [20]:

$$4NaOH + C \rightarrow Na_2CO_3 + Na_2O + 2H_2 \quad (1)$$

$$2NaOH \rightarrow Na_2O + H_2O \quad (2)$$

$$C + H_2O \rightarrow H_2 + CO \quad (3)$$

$$CO + H_2O \rightarrow H_2 + CO_2 \quad (4)$$

$$Na_2O + CO_2 \rightarrow Na_2CO_3 \quad (5)$$

$$Na_2O + H_2 \rightarrow 2Na + H_2O \quad (6)$$

$$Na_2O + C \rightarrow 2Na + CO \quad (7)$$

$$Na_2CO_3 + 2C \rightarrow 2Na + 3CO \quad (8)$$

$$Na_2O + SiO_2 \rightarrow Na_2SiO_3 \quad (9)$$

Precipitation of Silica

$$Na_2SiO_3 + H_2SO_4 \rightarrow SiO_2 + Na_2SO_4 + H_2O \quad (10)$$

A precipitate of white silica was obtained according to the above reaction (10). The silica obtained above was filtered and subjected to successive washings with demineralized water to remove any residues. The precipitate was finally dried in an oven for 24 hrs at 110 ^{0}C. XRD chart shows that particle size of nanosilica to be about 7 nm.

2.5 Scanning electron microscope (SEM)

Philips XL 30 scanning electron microscope was employed to study the microstructure distribution of produced rice husk ash, precipitated silica and activated carbon.

2.6 XRD measurements

X-ray diffraction profiles (XRD) were recorded on ARL X'TRA a diffractometer (Cu-K*a* radiation, 40 kV/300 mA). Spectra were observed from 5 to 65 degree. Average particle size had been estimated by using Debye-Scherrer formula:

$$D = \frac{0.9\lambda}{\beta \cos\theta} \qquad (11)$$

where λ is wave length of X-Ray (0.1541 nm), β is FWHM (full width at half maximum),

θ is the diffraction angle and D is particle diameter size.

2.7 X-Ray Fluorescence (XRF)

The chemical composition of rice husk ash and precipitated silica was determined by X-ray fluorescence (XRF) fischer XDV-SDD, at a tube current of 1000 μA with an acquisition lifetime of 60 s. Data values were calculated following the theoretical formula and fundamental parameters.

2.8 Fourier Transform Infrared Spectroscopy (FTIR)

Functional groups present in RHA and precipitated silica were confirmed using Fourier Transform Infrared (FTIR) spectroscopy using KBr pellet technique on JASCO model 4100. Approximately 1 mg of sample was mixed with 100 mg of KBr and then grounded and pressed to prepare the pellets. FT-IR spectra were obtained in the range of 400 to 4000 cm^{-1}.

3. Results and Discussion

3.1 Scanning electron microscope

The results presented in Fig. 1(a) reveal that burning of rice husk in open environment will produce fibrous intact black particles; however it appears that silica particles will form at interface. Micrograph (b) of RHA formed by combustion at 1000 ºC reveals that there is abundant amount of silica. Micrograph (c) shows the disordered layer of amorphous carbon composed of carbon sheets oriented in a considerably random fashion, it appears that there are some inorganic impurities, traces illustrated by XRF of K_2O and other impurities. Micrograph (d) represents the precipitated white nanosilica it appears as a highly agglomerated pure particle which indicates the efficiency of extraction by acid deposition technique.

3.2 XRD Measurements

The results indicated by Fig. 2 (a) reveal that open field burning of rice husk produces regions of crystallographic phases of silica i.e. crystoballite, tridymite and quartz in a matrix of amorphous silica. Temperature is an important factor for the crystallization of silica in (RHA). The increase in temperature will increase both the nucleation rate and the crystal growth rate. Thus, higher growth rates and larger crystals have been obtained by combustion at 1000°C. The crystallographic phases of silica in RHA are presented in table 1. As is evident from the diffractograms, the sample produced by combustion at 1000°C for 4 hrs is crystalline and contains mostly cristobalite phase, while by burning RH in open environment amorphous silica is produced with some crystalline nature. Fig. 3 represents different crystalline planes of SiO_2 with different symmetry, various crystalline silica forms can transform into different crystalline forms when subjected to high heat. The presence of trace elements in the silica affects transformation rates. Also heating rate is important to bring out the required organization [21, 22].

Fig. 2 (c) represents XRD pattern of activated carbon and shows two broad peaks at around 24 .6º and at around 44.2º attributed to amorphous carbon. XRD pattern of precipitated silica shown in Fig. 2 (d) reveals broad peak appears at 23.2º with d – spacing 3.876 which coincide well with the studied diffractograms of commercial silica.

3.3 XRF characterization

The purity concentration of silica in RHA samples and PWNS were measured by XRF and shown in table 3 to be in the range of 82.79-97.00 % with major impurities of K_2O, P_2O_5 and Cr_2O_3. The purity of produced silica seems to be affected by processing condition including heat history, activation technique and time of extraction.

3.4 FT-IR Characterization

FTIR spectrums of RHA (OF), RHA (MF) &PWNS as shown in Fig. 4 show a wide O-H stretching band around 3450 cm^{-1} due to freely vibrating OH groups and a band at 1640 cm^{-1} due to O-H bending vibration. The peak around 1095 cm^{-1} is due to Si–O-Si asymmetric stretching vibration. The peak at 967 cm^{-1} is due to Si–OH stretching vibration. The peaks at 819 cm^{-1} and 441 cm^{-1} are due to Si–O–Si symmetric stretching and bending vibrations respectively. We can observe that intensity of peak at 1090 cm^{-1} is small for RHA (OF), while it increases in case of RHA (MF) and shows maximum broad peak in case of PWNS. This indicates that silica content would vary by method of treatment and reflects the highest purity of PWNS.

Table 1: XRD data of rice husk ash silica phases.

d-spacing	Intensity	2 theta	Silica phases
RHA (OF)			
7.35	57.47	12.01	Q
3.62	100.00	24.54	T
3.15	76.65	28.28	C
2.83	98.20	31.52	---
7.35	57.47	12.01	---
2.002	23.51	40.3	C
2.002	30.57	45.25	Q
RHA (MF)			
3.13	100.00	28.44	C
2.98	34.35	29.88	T
2.89	49.51	30.91	C
2.49	5.35	35.92	---
2.21	65.95	40.60	C
1.81	12.59	50.20	Q

Table 2: Elemental analysis of rice husk, rice husk ash and precipitated white nanosilica.

Composition	RH	RHA (OF)	RHA (MF)	PWNS
Na_2O	-----	0.021	0.027	0.03
MgO	0.084	0.490	0.410	0.33
Al_2O_3	0.073	0.400	0.230	0.100
SiO_2	16.780	82.790	91.600	97
P_2O_5	0.236	3.240	2.400	0.02
K_2O	0.543	3.300	2.500	1.180
CaO	0.122	1.600	0.900	0.500
Cr_2O_3	0.002	0.700	0.5	0.07
MnO	----	0.044	0.036	0.022
Fe_2O_3	----	0.380	0.340	0.220
ZnO	----	0.500	0.300	0.02
LOI*	82.16	6.535	0.757	0.508

LOI*: Loss on ignition.

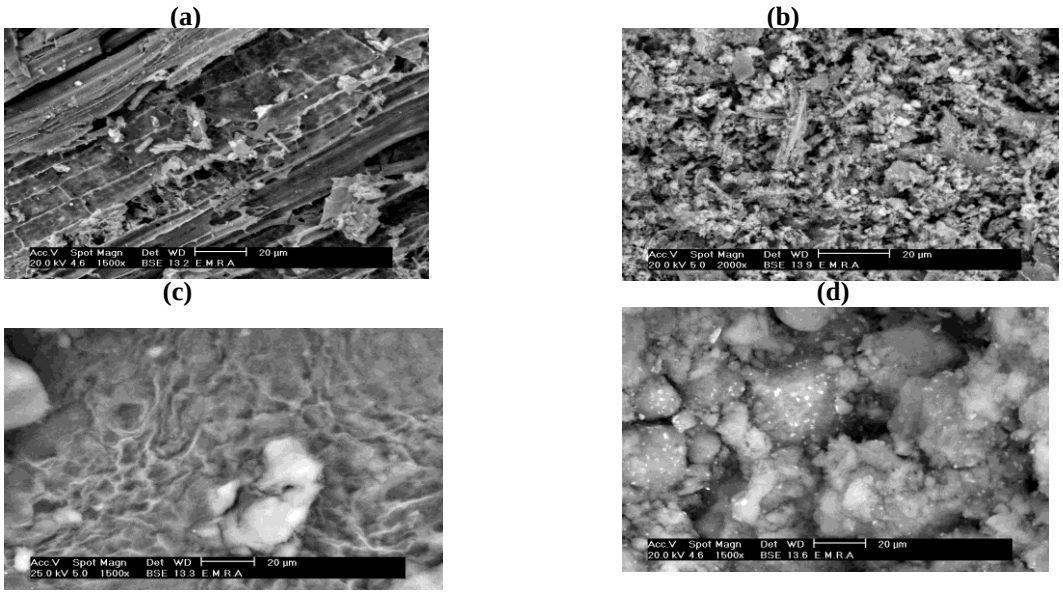

Fig. 1: SEM micrographs of: RHA (OF) (a); RHA (MF) (b); AC (c); PWNS (d).

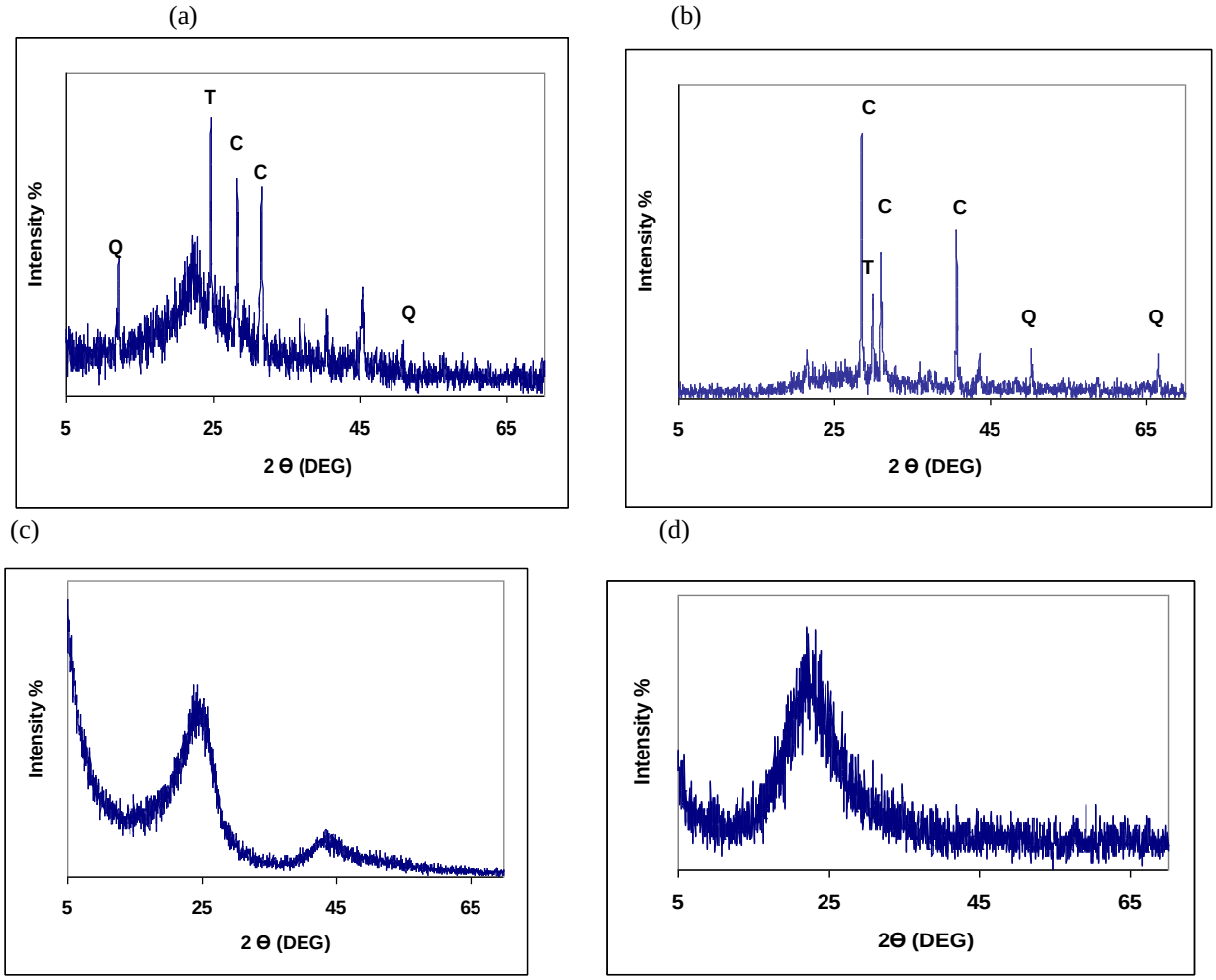

Fig. 2: XRD patterns of: RHA (OF) (a); RHA (MF) (b); AC (c); PWNS (d).

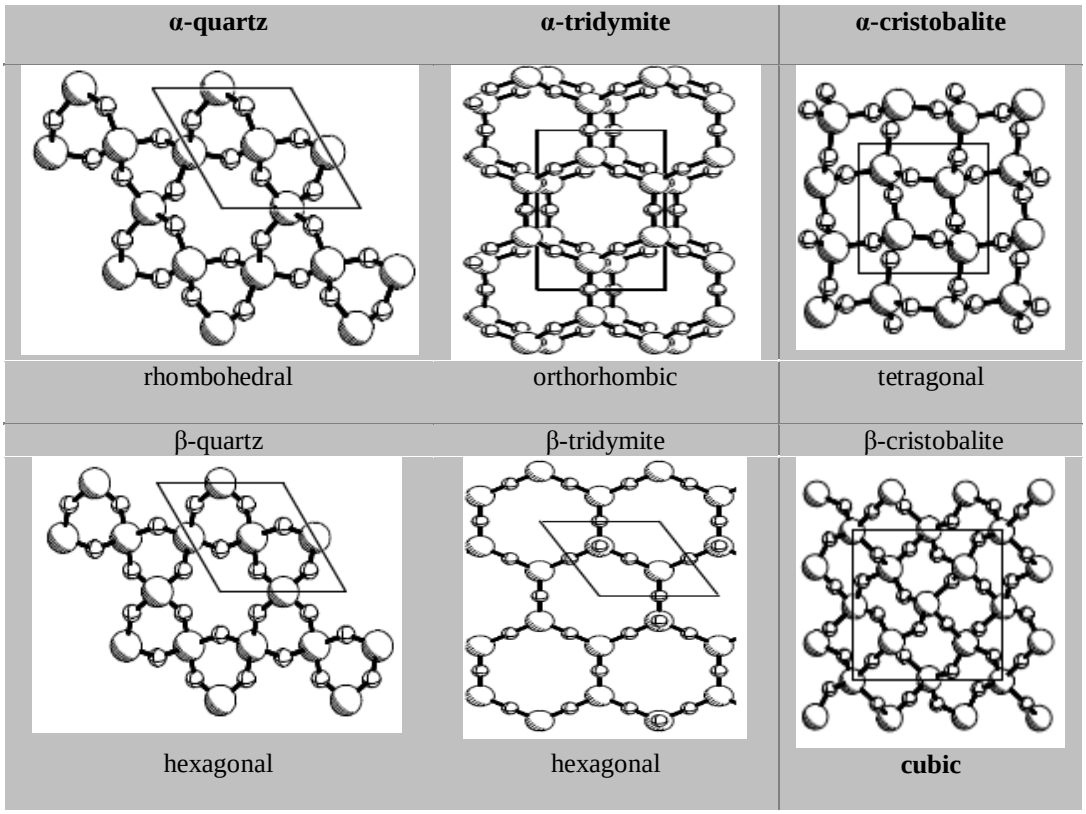

Fig. 3: Different phases of crystalline SiO_2 [21-23].
Large spheres represent Si; small spheres represent O atoms.

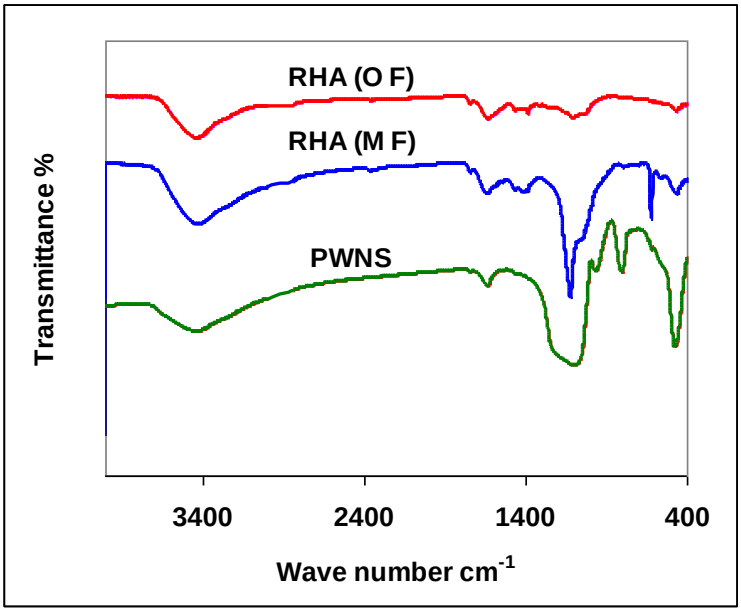

Fig. 4: FT-IR spectrum of RHA (OF), RHA (MF) &PWNS.

4. Conclusions

RHA prepared by open field burning and by combustion at 1000 °C was analyzed. The results revealed that higher growth rates and larger crystals had been obtained by combustion at 1000 °C and the occurrence of polymorphs of silica. Activation of RHA (OF) with 3.5 N sodium hydroxide followed by acid deposition technique resulted in high silica in the agglomerate form with dimension of 7 nm and the particle shape distribution was found to be uniform. The diffraction pattern of the particles showed a diffuse amorphous phase. The value-added activated carbon of rice husk can be produced by simple carbonization-chemical activation of RHA at 900 °C for 2 hr and XRD diffractogram showed two broad peaks at around 24 .6º and at around 44.2º attributed to amorphous carbon sheets.

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