

## OBTAINING AND CHARACTERIZATION OF AMORPHOUS SILICA FROM RICE HUSK

Kydyr Askaruly<sup>2</sup>, Seitkhan Azat<sup>1,3</sup>, Zhanar Sartova<sup>3</sup>, Mukhtar Yeleuov<sup>1,2</sup>,  
Almagul Kerimkulova<sup>1,3</sup>, Kalampyr Bekseitova<sup>4</sup>

<sup>1</sup> Institute of Combustion Problems, 172 Bogenbay batyr str., Almaty, Republic of Kazakhstan    Received 12 March 2019  
E-mail: mukhtar.yu@gmail.com    Accepted 31 July 2019

<sup>2</sup> Satbayev University, 22a Satpaev str., Almaty, Republic of Kazakhstan  
E-mail: k.askaruly@gmail.com

<sup>3</sup> Al-Farabi Kazakh National University, 71 Al-Farabi ave., Almaty, Republic of Kazakhstan  
E-mail: seytkhan.azat@gmail.com

<sup>4</sup> LTD «ZHALYN», Almaty, Republic of Kazakhstan  
E-mail: bekalsu@mail.ru

---

### ABSTRACT

When processing the harvested rice grain, a large amount of waste remains - straw, bran and husks. The mass fraction of the latter in the total volume can reach ~20 % (the total share of the waste is about ~30 %). This paper addresses the production of pure porous silica ( $\text{SiO}_2$ ) from the rice husk (RH) obtained in the south region of Kazakhstan by pre-treatment with HCl at different temperatures. It was found that pre-treatment of the RH with HCl can give  $\text{SiO}_2$  with purity ~90.1 - 99.5 wt. %. In this work are shown X-ray diffraction and TG analyses and morphology of RH before and after direct incineration (Scanning Electron Microscopy). TGA-DTG curves were divided into three regions: I - for drying and evaporation of light components (50°C - 250°C), II - for devolatilization of hemicellulose and cellulose components (250°C - 400°C) and, III - for lignin decomposition (400°C - 900°C).

**Keywords:**  $\text{SiO}_2$ , acid treatment, rice husks, hydrochloric acid, silica, RH ash.

---

### INTRODUCTION

Silicon (Si) is a non-metal, standing on the 2<sup>nd</sup> place after oxygen in terms of reserves and location on Earth (~28.8 % in the Earth's crust) [1]. In its pure form, it almost does not occur, mainly presenting on the planet in the form of compounds, e.g. as  $\text{SiO}_2$ . This compound is known for its hardness since antiquity because of very strong silicon-oxygen covalent bonds have to be broken throughout the structure before the melting occurs and crystalline silicon has the same structure as diamond. To turn it into silicon dioxide, all you need to do is to modify the silicon structure by including some oxygen atoms.  $\text{SiO}_2$  is most commonly found in nature as sand or quartz and in rice husk (RH).  $\text{SiO}_2$  is used in all directions in the life. For example,  $\text{SiO}_2$  is the primary ingredient in

the production of most glasses [2], crystalline  $\text{SiO}_2$  is used in hydraulic fracturing of formations which contain tight oil and shale gas,  $\text{SiO}_2$  is a common additive in food production, hydrated  $\text{SiO}_2$  is used in toothpaste as a hard abrasive to remove tooth plaque [3], in microchips, etc. If silicon is separated from oxygen, silicon can be used in lithium-ion batteries [4] as well as in other semiconductor electronics [5].

As said before  $\text{SiO}_2$  is found in many substances in nature and one of them is RH [6]. The RH, also called rice hull, is the coating on a seed or grain of rice. It is formed from hard materials, including  $\text{SiO}_2$  and lignin, which protect the seed. Each kilogram of milled white rice results in roughly 0.28 kg of RH as a by-product of rice production during milling [7]. RH was long considered a waste from the rice milling process and was

often dumped and/or burned. But because it can be easily collected and is cheap, some amount of RH has always been used as an energy source for small applications, such as for brick production, for steam engines and gasifiers used to power rice mills, and for generating heat for rice dryers. The high SiO<sub>2</sub> content of RH ash makes it a good additive for the steel and concrete industries and also for carbonized nanomaterials [8], for clean the biostimulator produced from germinated wheat seeds [9].

At 758.8 million tonnes (503.6 million tonnes, milled basis), world paddy production in 2017 would stand 0.8 percent, or 5.8 million tons, above a revised estimate for 2016 [10]. On average 28 % of the rice paddy is husk, giving an annual total global production of 212.5 million tonnes. Proximate analysis of a husk sample gives about 75 % organic volatile matter and 25 % of the weight of this husk is ash and commonly known as RH ash (RHA). This RHA in turn contains about 85 - 90 % amorphous SiO<sub>2</sub>.

## STAGES OF OBTAINING SILICA FROM RH

Obtaining pure SiO<sub>2</sub> from RH requires several methods. At the moment there are many methods for obtaining SiO<sub>2</sub> from RHs with different purities of SiO<sub>2</sub>. For example Bakar et al. [11] obtained SiO<sub>2</sub> with purity 99 %, Azat et al. [12] - 99.67 %, Kongmanklang et al. [13] - 99.29 %, Şımşek [14] - 98 % and Yunusa et al. [15] - 96.1 %.

Kalapathy et al. [16] obtained 93 % SiO<sub>2</sub> from RHA using a chemical method. 10 grams of RHA samples were dispersed in 60 ml of distilled water, and the pH was adjusted to 1, 3, 5 or 7 using 6 N and 1 N HCl. These dispersions were stirred for 2 h, filtered through filter paper and then the RHA residues were washed with 100 ml of water. The residues were used for SiO<sub>2</sub> extraction. The filtrate and washings at each pH were collected and dried in an evaporating dish. 60 ml portions of 1N NaOH were added to the washed and unwashed RHA samples and boiled in covered 250 ml Erlenmeyer flasks for 1 h with constant stirring to dissolve the SiO<sub>2</sub> and to produce a sodium silicate solution. The solutions were filtered through ashless filter paper, and the carbon residues were washed with 100 ml of boiling water. The filtrates and washings were allowed to cool to room temperature and were titrated with 1N HCl with constant stirring to pH 7. SiO<sub>2</sub> gels started to precipitate when the

pH decreased to < 10. The SiO<sub>2</sub> gels formed were aged for 18 h. Deionized water (100 ml) was added to gels and then the gels were broken to make a slurry. Slurries were then centrifuged for 15 min at 2500 rpm, the clear supernatants were removed and the washing step was repeated. The gels were transferred into a beaker and dried at 80°C for 12 h to produce xerogels.

For receiving xerogel SiO<sub>2</sub>, Rambo et al. [17] use RHA samples dried in a kiln for 24 h at 110°C. The SiO<sub>2</sub> was prepared with the aid of NaOH and KOH extracting solutions with the respective bicarbonates being used as catalyst. Micronised RHA (20 g) was placed in a 250 mL round-bottom flask and mixed with the corresponding alkaline solution and catalyst, in concentrations predicted by the Response Surface Methodology (RSM) for sodium-based SiO<sub>2</sub>, using as independent variables the pH (1, 3, 5, 7 and 9), base concentration (2.0, 3.5, 5.0, 6.5 and 8.0 mol L<sup>-1</sup>), catalyst concentration (0.6, 1.3, 2.0, 2.7 and 3.4 mol L<sup>-1</sup>) and time (1, 2, 3, 4 and 5 h). The mixture was then kept under reflux for 3 h. Following this, the solution passed through a filter paper and the filtered material was acidified to pH predicted by the RSM with a solution of 5.5 N H<sub>2</sub>SO<sub>4</sub> which formed a slightly pink SiO<sub>2</sub> precipitate. It was filtered again in the same way as mentioned before, now adding 20 mL of 3 % H<sub>2</sub>O<sub>2</sub>. The discoloured SiO<sub>2</sub> xerogel was dried in a kiln for 24 h; after that, it was washed with distilled water to remove the excess acid (pH~6) and put back in the kiln for a further drying period of 24 h.

Batteggazzore et al. [18] in their experiment have taken 40 g of milled RH and placed in a becher containing 386 g of distilled water and 14 g of sulfuric acid under stirring for 3 h at 80°C. Such operation was used as previous step to a further calcination in order to prevent the formation of black particles due to the presence of impurities. The solid residue was separated by filtration, purified in deionized water and dried. Subsequently, the residue was calcined in a muffle furnace at 800°C for 1 h to obtain 15 % of the original material weight as SiO<sub>2</sub>.

The RH was converted into RHA by incinerating the husk at 650°C in a muffle furnace. All chemicals were of analytical grade and used without any purification. RHA was pre-treated with acid to remove metallic impurities and enhance SiO<sub>2</sub> purity. pH of RHA samples dispersed in water, was adjusted to 7 using an acid (HCl, HNO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>). The samples were stirred for 2 hours, filtered and the residues were washed with water. After the pre-treat-

ment, the residues were dispersed in NaOH and boiled for 1 hour. The solutions were filtered and the carbon residues were rinsed with boiling water. The washings were collected and cooled to room temperature, before titrating with HCl until pH 7. SiO<sub>2</sub> precipitation started at pH below 10. The precipitated SiO<sub>2</sub> gels were aged for at least 12 hours. The gels were crushed to form a slurry with deionized water, which was then centrifuged at 2500 rpm. The supernatant was discarded and the washing was repeated, if required. The gels were then dried in a beaker at 80°C [19].

To produce SiO<sub>2</sub>, Moosa et al. [20] have taken 100 g of the cleaned RHA which was refluxed with 800 ml 2N HCl at 70°C for one hour. After the reaction is completed, the acid was removed from the RHA by washing with distilled water many times using filter paper with pore size 0.45 µm. It was then dried in an electric oven at 50°C for 6 hrs. The treated RHA was then heated in an electrical box furnace a heating rate of 10°C/min at 650°C for 3 hours. The rice husk ash treated at 400°C has a black colour, while the heat treated rice husk ash at 650°C has a white color. A 10 g of the obtained silica was refluxed with 80 ml, 6N HCl at 65°C for 3 hours. After the reaction is completed, the acid was removed from the silica by washing with distilled water several times and then filtered at vacuum using filter paper with pore size 0.2 µm. The silica was dried in an electric oven at 50°C for 6 hrs. The silica was then magnetically stirred with 2.5N NaOH solution for 14 hours to form sodium silicate. H<sub>2</sub>SO<sub>4</sub> was then added drop wise to sodium silicate solution with constant stirring until the pH equal to 8 and then a nanosilica gel was formed. The gel was then washed with double distilled water until pH 7 and then dried in an oven at 50°C for 48 hours to form a silica nanoparticles powder.

The pure nano silica powder was extracted from RHA by controlling different process parameters. RH was washed thoroughly with distilled water to remove any adhering impurities. The washed RH was air-dried at room temperature and then burnt at 973K for 3 h in a muffle furnace under an inert atmosphere. The obtained RHA was washed with distilled water to neutralise the pH in order to remove the sand, dust, light empty grains and fine dirt. The neutralised RHA was refluxed with 6 N HCl (Merck GR) for 1.5 h and then filtered in order to remove metallic impurities and to extract pure nano silica. The filtered RHA was thoroughly rinsed with hot

water repeatedly and then boiled with different concentration of NaOH (Merck GR) solutions at 353K for 1.5 h, while being stirred magnetically, and was then filtered to obtain sodium silicates. The obtained residue was thoroughly washed with hot water for complete extraction of sodium silicate. The obtained pH of the filtrate (sodium silicate) was reduced to 2.0 using controlled addition of concentrated H<sub>2</sub>SO<sub>4</sub>, while being stirred magnetically, to extract the nano silica precipitates. The obtained precipitate was washed thrice in warm distilled water and then filtered. The obtained residues were sintered at 973 and 1373 K for 3 h in a muffle furnace. To obtain the desired grain size, the obtained silica powder was pulverised using a milling process [21].

## EXPERIMENTAL

For this experiment we used the following substances and equipment: RH from Kyzylorda, tap water, distilled water, conc. HCl, sodium hydroxide, filter paper (Whatman™ filter paper, Grade 41), vacuum pump, muffle furnace (SNOL 8,2/1100 LSM01), drying oven (SNOL 20/300 LSN11 ST) and magnetic stirrer (Four E's 5 Inch, 100-1500RPM,5L). The whole process from the beginning to the end takes about 28 h.

RH has been selected from Kyzylorda region of Republic of Kazakhstan. Sample of RH with a length of about 5 - 10 mm was pre-washed with tap water, and then was washed with hot distilled water to remove other substances and dried in a drying oven at 10°C for 12 h (for evaporation of the water in the composition).

A sample (50 g) of the raw material was treated with 2M HCl solution (500 ml) and fried at 90°C for 2 h. The prepared sample was continuously washed with distilled water to 7 pH and separated from solution via filtration. RH was dried at 105°C for 12 h and then calcinated at different temperature for 4 h in a muffle furnace to produce white RH ash (WRHA). The color of the resulting ash was different depending on the temperature. At 400°C - 500°C black ash was obtained and at 750°C - 900 °C - grey ash, due to the high combustion efficiency resulting in a high residual carbon. WRHA was obtained at 550°C - 700°C for 4 h. The optimal temperature was accepted to be 600°C. After the end of the process, the mass of WRHA obtained after the combustion process was about ~9 g.

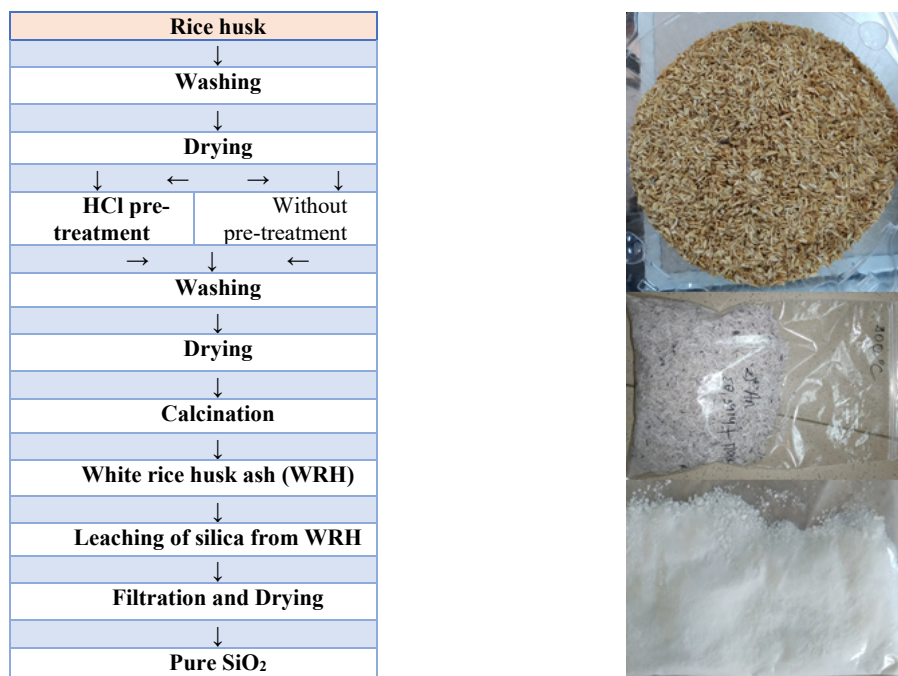
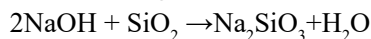


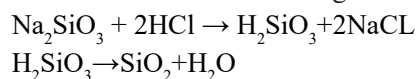
Fig. 1. Scheme RH treatment methods and obtaining pure SiO<sub>2</sub> from RH.

Subsequently the WRHA was mixed with 100 ml of 2M NaOH at 90°C under continuous vigorous stirring for 2 h in order to extract the solid SiO<sub>2</sub> into water-soluble silicate through a chemical reaction:



The water-soluble sodium silicate solution (SSSS) was filtered via the vacuum pump to remove insoluble

residues (Fig. 2). After filtration, the SSSS is converted into insoluble silicic acid by reaction with concentrated HCl under continuous stirring:



The final product is passed through the filter and washed with hot water to remove by-products (NaCl

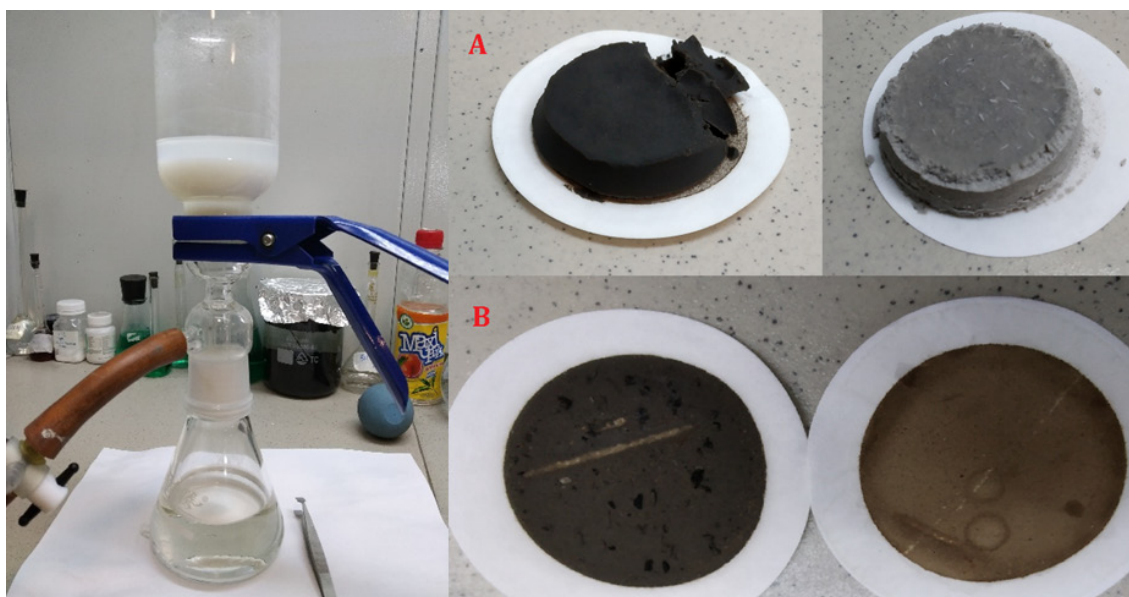


Fig. 2. The filtration stage of water-soluble sodium silicate solution via the vacuum pump to remove insoluble residues. A) Insoluble residues without acid treatment. B) Insoluble residues after acid treatment.



residue). Finally, the obtained pure silicon dioxide was placed in a laboratory-drying oven at 120°C for a night. After drying, the white porous powder was obtained (SiO<sub>2</sub>) (Fig. 1).

## RESULTS AND DISCUSSION

Different grades of silica with different yields can be obtained based on the calcination or combustions temperature and duration of calcinations or combustions in different treatment temperature [22 - 24] (Table 1, Fig. 3). The concentration of the activating reagent, temperature, time of heating, the pre-treatment influenced the yield of silica. The white RH ash (WRHA) yields obtained at the specified incineration conditions (600°C for 4 h) were found to be in the range of 14,9 % (Table 2, Figs. 4 and 5).

The yield of silica from the RH and its purity also depends on the production scheme. The highest yield of final pure product (11,5 %) is formed by preliminary acid treatment (WRH+HCl) (Table 3). The silica content varies in the range ~90,1 - 99,5 % depending on the purity of reagents used for washing.

The elemental composition of RH, WRHA is determined by X-ray fluorescence analysis (XRF). The obtained data indicate a rich and diverse elemental composition of the investigated RH (Table 4, 5).

### X-ray diffraction and TG analysis

X-ray diffraction patterns for silica samples were recorded using the Rigaku powder diffractometer at a

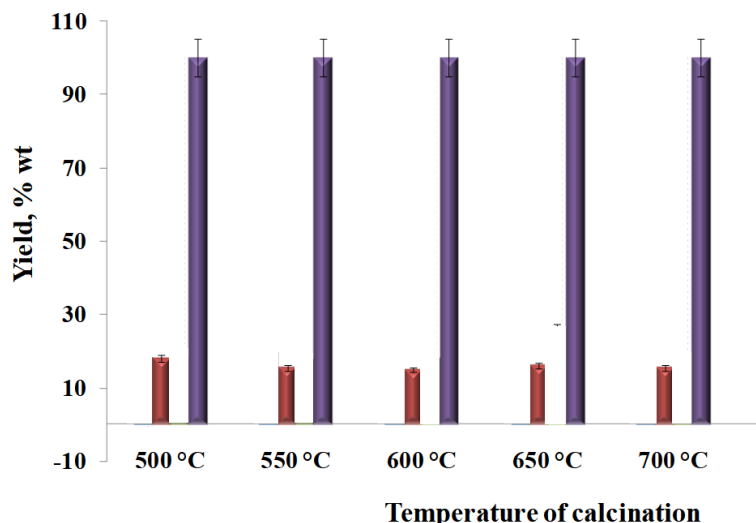


Fig. 3. Yield of WRH extracted from the RH (without HCl).

Table 1. Yield of WRH Silica from RH depending on treatment methods and without treatment (thermally assisted).

Methods	Yield, wt. %	
	WRH	SiO <sub>2</sub>
Pre-treatment with HCl	16	11,5
Thermally assisted	14,9	11,6

Table 2. Yield of WRH Silica from RH during direct incineration in the muffle furnace.

Temperature of calcination (τ=4 hours)	Yield of m□ss %	
	RH	Raw RH
500°C	17,9	100
550°C	15,4	100
600°C	14,9	100
650°C	16	100
700°C	15,5	100
750°C	15,5	100
800°C	15	100

Table 3. Yield of WRHA, silica from RH during pre-treatment with HCl.

Yield, %	WRHA	SiO <sub>2</sub>	RH
RH	16	11,5	100

scanning speed of 0.02 2 θ/min, using nickel-filtered Cu-Kα radiation in the angular range from 10 to 90 of 2 θ.

The obtained silica powder is in the X-ray amorphous state, which is confirmed by X-ray phase analysis. On X-ray diffraction patterns is observed one diffuse

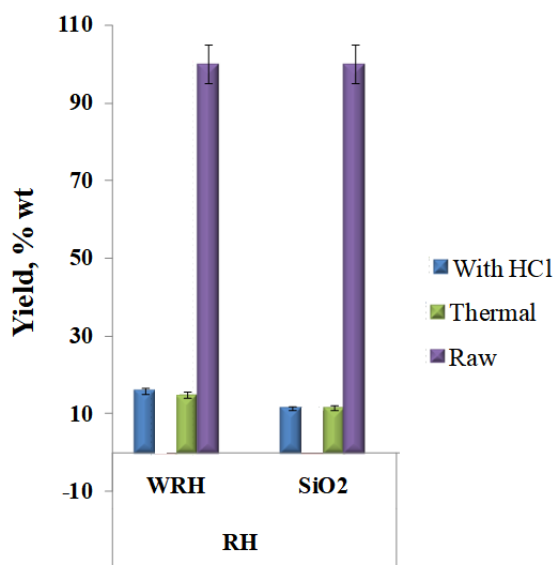


Fig. 4. Yield of WRH and RH silica extracted from the RH by different methods.

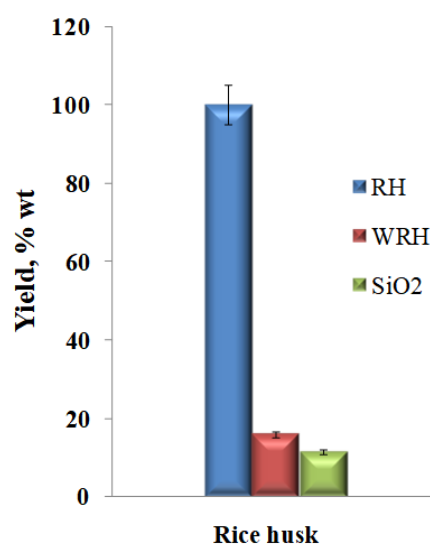


Fig. 5. Yield of the WRH and RH silica extracted from the RH (with HCl).

peak in the  $2\theta = 24^\circ$  region typical for the amorphous structure of the RH silica, while for amorphous silicon dioxide the maximum of the diffuse peak is at  $2\theta = 30^\circ$ .

intensity at  $2\theta = 20 - 24^\circ$ , corresponds to the d-spacing value of 0.36 nm and confirms an amorphous and some quarts structure the obtained SiO<sub>2</sub>.

The observed broad halo (Fig. 6) with maximum of

The amorphous structure is very porous and has high

Table 4. The elemental analysis of the amorphous silica (600°C).

Sample name	The yield of final product, %	Content, %							
		SiO <sub>2</sub>	Na <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	SO <sub>3</sub>	Cl	K <sub>2</sub> O	Fe <sub>2</sub> O <sub>3</sub>	ZnO
WRH+HCl	11,5	95.87	3.578	0.032	0.018	0.279	0.021	0.014	0.006

Table 5. The elemental analysis of the amorphous silica (600 °C).

Elemental composition	Samples name		
	RH (Raw) %	WRH	WRH+HCl
Na <sub>2</sub> O	0.365	3.578	0.079
MgO	0.311	n/d	0.03
Al <sub>2</sub> O <sub>3</sub>	0.23	0.032	0.134
SiO <sub>2</sub>	90.194	95.871	99.506
SO <sub>3</sub>	2.021	0.018	0.021
Cl	0.278	0.279	0.041
K <sub>2</sub> O	2.949	0.021	0.074
CaO	2.618	n/d	0.036
MnO	0.138	n/d	n/d
Fe <sub>2</sub> O <sub>3</sub>	0.570	0.014	0.071
CuO	0.071	n/d	n/d
ZnO	0.037	0.006	0.007
Tm <sub>2</sub> O <sub>3</sub>	0.172	n/d	n/d
PtO <sub>2</sub>	0.047	n/d	n/d
P <sub>2</sub> O <sub>5</sub>	n/d	n/d	n/d
NiO	n/d	n/d	n/d

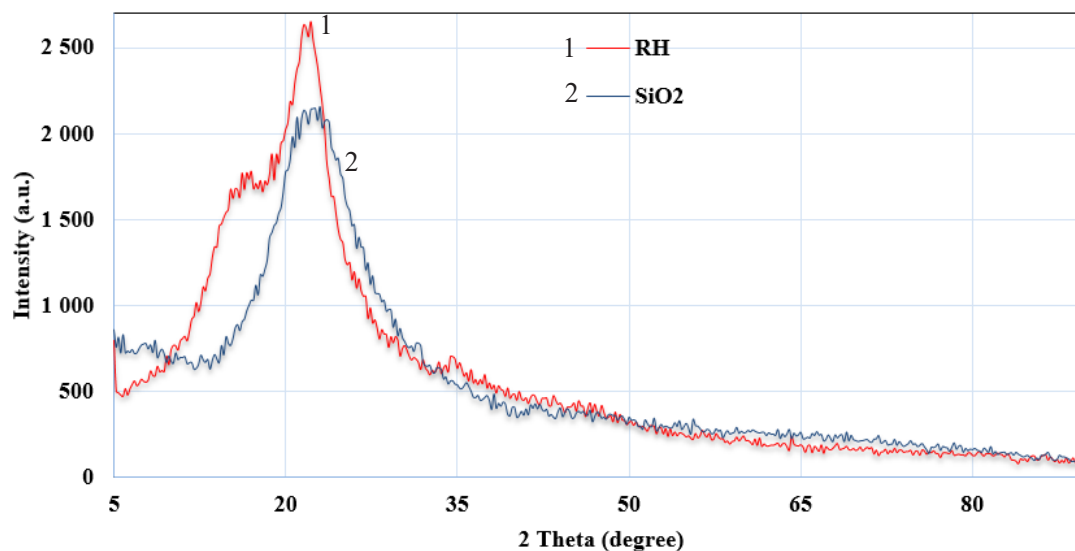
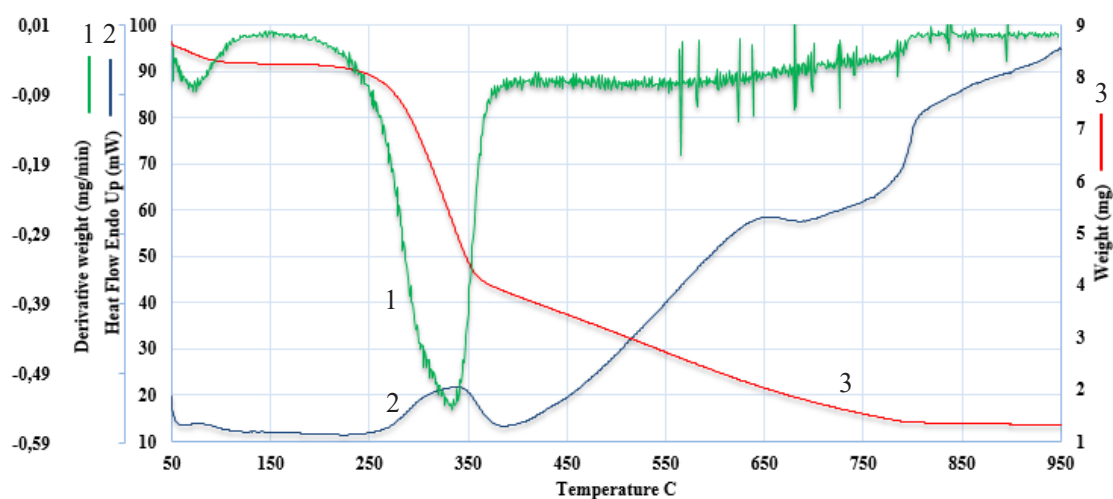
Fig. 6. X-ray diffraction pattern of SiO<sub>2</sub> (2) obtained from RH (1).

Fig. 7. TGA and DTG of rice husk.

surface area so this also gives a high activity for chemical treatment and absorption. In general silica in this condition has far more potential for commercial utilization than mineral sources of silica which are characterized by higher temperature crystalline forms.

TG and DTG analysis was used to determine the existence of organic components in the RH (Fig. 7). In addition these TGA-DTG curves were divided into three phases of thermal degradation of rice husk as follows: phase I for drying and evaporation of light components (50°C - 250°C), phase II for devolatilization of hemicellulose and cellulose components (250°C - 400°C) and phase III for lignin decomposition (400 - 900°C). Meanwhile, the leftover after 700°C was labelled as

solid residues. It can be seen that initial weight loss occurs within the range of 50°C - 250°C. The second stage reveals a rapid and large weight loss at temperature between 300°C - 400°C. This is due to the thermal decomposition of hemicellulose and cellulose as a major organic component in the RH.

The third stage shows a weight loss of about 25 - 30 % that could be due to lignin, a thermally more stable aromatic polymer which undergoes gradual decomposition between 300°C and 600°C. The ash residual is mainly the noncombustible silica (~15 %, > 600°C).

The functional groups present in RHs and SiO<sub>2</sub> precipitate were identified by FTIR-ATR spectra (Fig. 8).

The broad peak in Fig. 8 at about 3500 cm<sup>-1</sup> corresponds

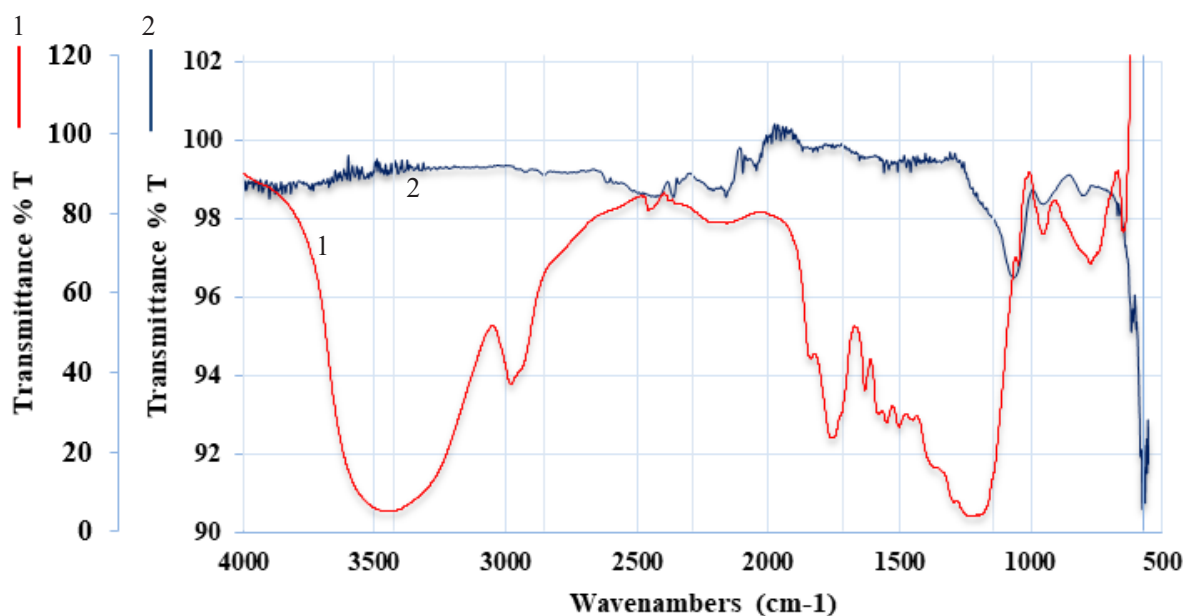


Fig. 8. FTIR spectra of RH (1) and SiO<sub>2</sub> (2).

to the O–H stretching vibrations of water molecules. The absorption peak at 2926 cm<sup>-1</sup> can be attributed to the symmetric and asymmetric stretching vibrations of the aliphatic C–H bonds in -CH<sub>3</sub> and CH<sub>2</sub> groups in the structures cellulose, hemicellulose and lignin, respectively, and 2114 cm<sup>-1</sup>, 1423 cm<sup>-1</sup> and 1383 cm<sup>-1</sup> are related to the –C–H stretching vibrations of the methylene groups. The peak at 1632 cm<sup>-1</sup> can be attributed to the –C=O stretching vibrations of the carbonyl groups in aldehydes and ketones. The peak at 1515 cm<sup>-1</sup> corresponds to the –C–O groups stretching of carboxylates. The broad peaks at 898 cm<sup>-1</sup>, 796 cm<sup>-1</sup>, 662 cm<sup>-1</sup> and 471 cm<sup>-1</sup> are attributed to the stretching vibrations of the siloxane groups. The IR band at 1190 cm<sup>-1</sup> shows the beginning of crystalline silica (quartz, cristobalite and tridymite). The notable absorption peak in Fig. 8 (blue) at 1055 cm<sup>-1</sup> can be attributed to the siloxane (Si–O–Si) network vibration modes, indicating a highly condensed

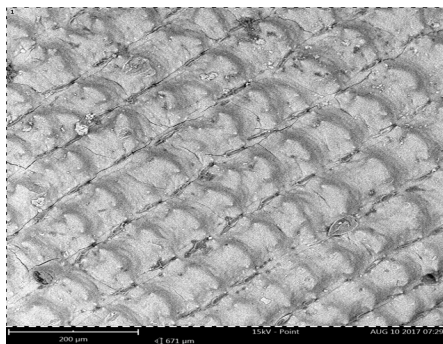
silica network [25 - 26] and is in agreement with the literature [27]. The spectra of the produced SiO<sub>2</sub> show no other band attributed to organic or inorganic impurities.

#### ***Morphology of RH before and after direct incineration (Scanning Electron Microscopy)***

Comparisons of the surface structures of RH and the resulting silicon oxide were represented by SEM Quanta 3D (FEI company, USA).

Fig. 9 shows SEM images of the original RHs before direct incineration. As can be seen the samples in the initial form are with very dense surfaces.

Fig. 9 (a) illustrates the morphology of outer surfaces of RH, which is uneven and highly roughened. Fig. 9 (b) is a low-magnification SEM image of the silica sample. The nano-scale roughness comes from the morphology of silica nanoparticles, dispersed within the bulk.



(a)



(b)

Fig. 9. SEM images of RHs before heat treatment (a) and SEM micrograph of the SiO<sub>2</sub>.



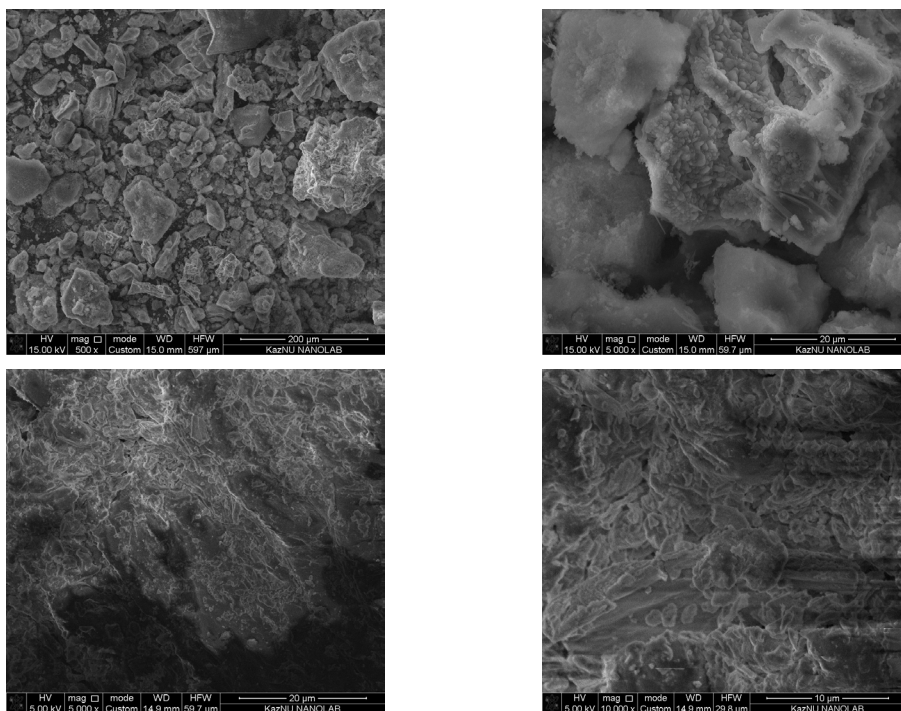


Fig. 10. SEM micrograph of the SiO<sub>2</sub> obtained from the RH ash.

### Morphology of RH silica after pre-treatment with hydrochloric acid (Scanning Electron Microscopy)

The morphology of the particles of the obtained silica powder was studied using the scanning electron microscope of the brand Quanta 200i 3D (FEI Company, USA) (National Nanotechnology Laboratory of Al-Farabi Kazakh National University, Almaty, Kazakhstan) (Fig. 9).

The low-magnification SEM image of the silica sample (Fig. 10) represents a nano-scale roughness, which comes from the morphology of silica nanoparticles, dispersed within the bulk.

### CONCLUSIONS

Highly pure silica is synthesised from RH using a simple precipitation technique. The elemental analysis of the RHs indicated an increase in the percentage of SiO<sub>2</sub> for the RHs treated with HCl, and there was a decrease in the other oxides/impurities with the use of the acid treatment. The XRD profiles showed that the RHs are crystalline; however, the RHAs have amorphous diffraction patterns, due to the presence of amorphous silica and purity reaches 99,5 %. The silica has a small particle size making it suitable for use in rubber and ink industries. The mesoporous structure of silica makes it a potential catalytic (or catalyst support) material.

### REFERENCES

1. M.Sommer, D. Kaczorek, Y. Kuzyakov, J. Breuer, Silicon pools and fluxes in soils and landscapes - a review, *Journal of Plant Nutrition and Soil Science*, 169, 3, 2006, 310-329.
2. M. Tomozawa, D.L. Kim, V. Lou, Preparation of high purity, low water content fused silica glass, *Journal of non-crystalline solids*, 296, 1-2, 2001, 102-106.
3. I. M.C. Camargo, M. Saiki, M.B.A. Vasconcellos, D.M. Avila, Abrasiveness evaluation of silica and calcium carbonate used in the production of dentifrices, *Journal of cosmetic science*, 52, 3, 2001, 163-168.
4. K. Feng, M. Li, W. Liu, A.G. Kashkooli, X. Xiao, M. Cai, Z. Chen, Silicon Based Anodes for Lithium Ion Batteries: From Fundamentals to Practical Applications, *Small*, 14, 8, 2018, 1702737.
5. W. Heywang, K.H. Zaininger, Silicon: the semiconductor material, In *Silicon* (pp. 25-42), Springer, Berlin, Heidelberg, 2004.
6. D. Geetha, A. Ananthiand, P.S. Ramesh, Preparation and characterization of silica material from rice husk ash—an economically viable method, *Research & Reviews: Journal of Pure and Applied Physics*, 4,3, 2016, 20-26.
7. Rice Knowledge Bank, 2016, Retrieved from <http://www.knowledgebank.irri.org/step-by-step-production/postharvest/rice-by-products/rice-husk>.

8. S. Azat, V.V. Pavlenko, A.R. Kerimkulova, Z.A. Mansurov, Synthesis and structure determination of carbonized nano mesoporous materials based on vegetable raw materials, In *Advanced Materials Research*, v. 535, pp. 1041-1045, Trans Tech Publications, 2018.
9. A. R. Kerimkulova, S. Azat, Z.A. Mansurov, M.K. Gilmanov, S.A. Ibragimova, S.M. Adekenov, B.B. Rachimova, Mesoporous nano carbon sorbents for separating different biomolecules, In *Advanced Materials Research*, v. 535, 2012, pp. 284-288, Trans Tech Publications.
10. Food and Agriculture Organization Rice Market Monitor (FAO RMM), 2018, Retrieved from <http://www.fao.org/economic/RMM>.
11. R.A. Bakar, R. Yahya, S.N. Gan, Production of high purity amorphous silica from rice husk, *Procedia Chemistry*, 19, 2016, 189-195.
12. S. Azat, A.V. Korobeinyk, K. Moustakas, V.J. Inglezakis, Sustainable production of pure silica from rice husk waste in Kazakhstan, *Journal of Cleaner Production*, 2019.
13. C. Kongmanklang, K. Rangriwatananon, Hydrothermal synthesis of high crystalline silicalite from rice husk ash, *Journal of spectroscopy*, 2015.
14. Y. Şimşek, Preparation and Characterization of High Purity Silica Obtained from Rice Husks, *Acta Physica Polonica A*, 132, 3, 2017, 1002-1005.
15. S. Yunusa, A.S. Ahmed, S.G. Bawa, J.F. Iyun, M. Dauda, Preparation of High Grade Silica from Rice Husk for Zeolite Synthesis, *Nigerian Journal of Basic and Applied Sciences*, 24, 1, 2016, 41-45.
16. U. Kalapathy, A. Proctor, J. Shultz, An improved method for production of silica from rice hull ash, *Bioresource Technology*, 85, 3, 2002, 285-289.
17. M.K. Rambo, A.L. Cardoso, D.B. Bevilaqua, T.M. Rizzetti, L.A. Ramos, G.H. Korndorfer, A.F. Martins, Silica from rice husk ash as an additive for rice plant, *Journal of Agronomy*, 10, 3, 2011, 99-104.
18. D. Battezzore, S. Bocchini, J. Alongi, A. Frache, Rice husk as bio-source of silica: preparation and characterization of PLA–silica bio-composites, *RSC Advances*, 4, 97, 2014, 54703-54712.
19. V. Tyagi, S. Pandit, A. Sharma, R.K. Gupta, Extraction and characterization of silica from rice husk for use in food industries, *Extraction*, 2, 4, 2017.
20. A. Moosa, B. Saddam, Synthesis and Characterisation of Nanosilica from Rice-Husk with Applications to Polymer Composites, *American Journal of Materials Science*, 7, 6, 2017, 223-231.
21. R. Yuvakkumar, V. Elango, V. Rajendran, N. Kannan, High-purity nano silica powder from rice husk using a simple chemical method, *Journal of experimental nanoscience*, 9, 3, 2014, 272-281.
22. Madrid, Rosario, C.A. Nogueira, F. Margarido, Production and characterisation of amorphous silica from rice husk waste, *WasteEng'2012: Proceedings of the 4th International Conference on Engineering for Waste and Biomass Valorisation*, 2012.
23. Ma, Xiaoyu, et al., A recyclable method for production of pure silica from rice hull ash, *Powder Technology*, 217, 2012, 497-501.
24. Costa, José Arnaldo Santana, Caio Marcio Paranhos, Systematic evaluation of amorphous silica production from rice husk ashes, *Journal of Cleaner Production*, 192, 2018, 688-697.
25. H.I. Meléndez-Ortiz, Y. Perera-Mercado, J.A. Mercado-Silva, Y. Olivares-Maldonado, G. Castruita, L.A. García-Cerda, Functionalization with amine-containing organosilane of mesoporous silica MCM-41 and MCM-48 obtained at room temperature, *Ceramics International*, 40, 7, 2014, 9701-9707.
26. S. Music, N. Filipovic-Vincekovic, L. Sekovanic, Precipitation of amorphous SiO<sub>2</sub> particles and their properties, *Brazilian journal of chemical engineering*, 28, 1, 2011, 89-94.
27. J. Umeda, K. Kondoh, High-purification of amorphous silica originated from rice husks by combination of polysaccharide hydrolysis and metallic impurities removal, *Industrial crops and products*, 32, 3, 2010, 539-544.