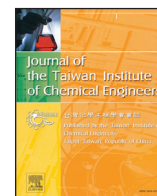




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# An appropriate acid leaching sequence in rice husk ash extraction to enhance the produced green silica quality for sustainable industrial silica gel purpose

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## ABSTRACT

The acid leaching process is believed to increase the silica extracted from rice husk ash (RHA). This study intended to investigate the influence of acid leaching sequences in RHA extraction on green silica purity and surface area. The sequences were RHA leaching (T1), aged gel leaching (T2), dried product leaching (T3), and without leaching as a control. The T1 sequence has been proven to improve the green silica purity and surface area reaching 96.44% and 400.69 m<sup>2</sup>/g, respectively. For T2 and T3 sequences, the green silica has a much lower surface area and purity compared to T1. The values were 87.69% and 55.49 m<sup>2</sup>/g for T2 as well as 62.46% and 11.40 m<sup>2</sup>/g for T3. Without acid leaching, the purity and surface area of green silica significantly reduced to 56.94% and 5.72 m<sup>2</sup>/g. This study concludes that green silica from T1 sequence has successfully obtained high purity, high surface area, and indicates an amorphous and mesoporous particle. It is suitable to be utilized as an industrial silica gel for moisture adsorber which more sustainable and greener process.

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## 1. Introduction

Rice husk ash (RHA) is obtained from rice husk combustion with a wide and huge amount of availability, especially in developing countries [1,2]. Surprisingly, the silica content in RHA is indicated around 80–95%-wt [3–7], which attract to be utilized for industrial interests. Silica extracted from biomass, named green silica, is commonly produced through rice husk calcination and followed by RHA extraction with an alkaline. Acid leaching is usually executed to rice husk before calcination or is carried out to RHA before extraction. When acid leaching is not applied, the silica purity rarely surpasses 95% and the surface area is mostly just below 200 m<sup>2</sup>/g which does not yet meet the criteria for industrial silica gel requirements [4,8–15]. Throughout this fact, acid leaching becomes a serious factor to be considered in RHA extraction.

Acid leaching could be applied to rice husk before it is calcined. An acid leaching study on 50 g rice husk then followed by ash extraction proposed by azat et al. (2019b) was resulted in silica purity of 98.2–99.7% and surface area of 120–980 m<sup>2</sup>/g [8]. An acid leaching

experiment using HCl and citric acid on 15 g rice husk improved the product purity from 82.8% to 99.3% and 98.6%, successively [9]. Rice husk to silica experiment with acid leaching pretreatment showed a greater surface area from 116 m<sup>2</sup>/g to 218 m<sup>2</sup>/g for leaching using HCl and 208 m<sup>2</sup>/g for H<sub>2</sub>SO<sub>4</sub> as well as purity enhancement from 95.77% to 99.58% for leaching using HCl and 99.08% for H<sub>2</sub>SO<sub>4</sub> [15]. Another acid leaching work on 20 g rice husk prior to calcination rectified the silica content from 94.58 to 99.14% [16]. The rice husk acid leaching before calcination not only enhanced the silica purity from 91.5 to 98.8% for leaching using HCl and 99% for citric acid but also increased the surface area from 56.6 to 311.6 m<sup>2</sup>/g for leaching using HCl and 254.1 m<sup>2</sup>/g for citric acid [17]. Meanwhile, another investigation on acid leaching before 10 g rice husk calcination gave an upgraded silica purity to 99.76% and surface area of 234.6 m<sup>2</sup>/g for leaching using HCl and 215.8 m<sup>2</sup>/g for citric acid [18].

Acid leaching could also be carried out to RHA before the extraction process. The research on 10 g RHA alkaline extraction with 1 mol/L HCl-leaching succeeded in achieving 93% of silica purity [19]. A study on acid leaching with 2 mol/L HCl at 90 °C for 2 h before 100 g RHA alkaline extraction could rectify the silica surface area to 169.7 m<sup>2</sup>/g and silica purity from 90.5 to 95.9% [20]. Apart from that,

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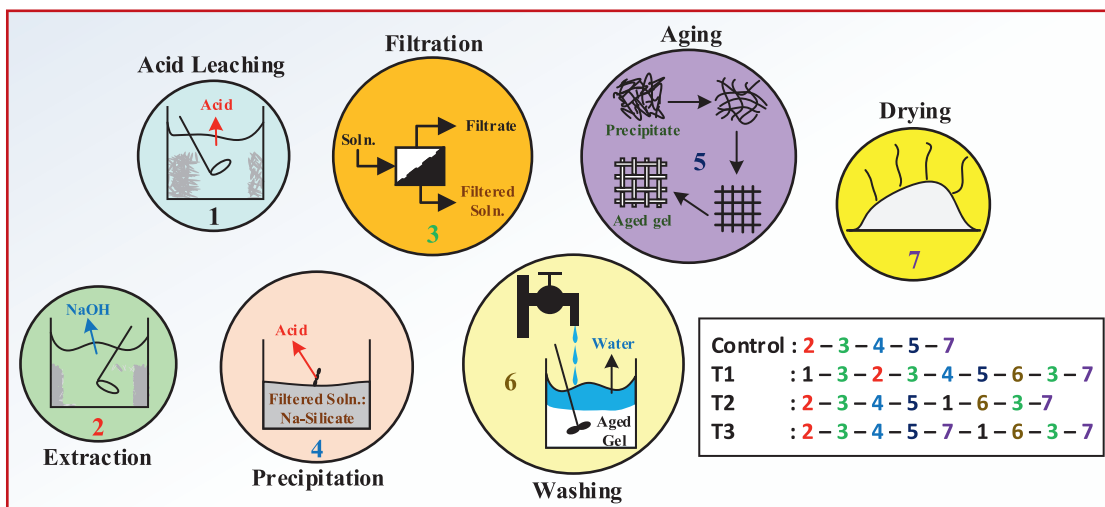


Fig. 1. Experimental setup.

a distinctive study on 10 g rice husk calcination was exploited using oxalic acid with the silica purity change from 92.4% to 99.3% [21].

Many acid leaching works gave satisfying results as aforementioned, but the study on the effects of acid leaching sequence in RHA extraction on green silica purity and surface area seems still to be absent. This present study, hence, is aimed to investigate and discuss it. The acid leaching was carried out in three sequences, those are acid leaching of raw material or RHA (T1), acid leaching of intermediate product or aged gel (T2), acid leaching of dried product (T3). The experiment without leaching was also provided as a control. The observed results were crystallinity and product quality which encompasses mineral composition, surface area, pore diameter, and morphology. Furthermore, the green silica usage as a silica gel for moisture adsorber and its performance compared to the commercial silica gel was reported in the last section of this study.

## 2. Materials and experimental methods

### 2.1. Materials

Rice husk was obtained from Mekarjaya rice milling in Majalaya Regency, West Java, Indonesia. RHA was prepared through rice husk calcination in a muffle furnace at temperature 700 °C for 2 h. A technical grade of sodium hydroxide and hydrochloric acid were procured from the local chemical market. Demineralized water was provided from ITB bioenergy instructional laboratory which using reverse osmosis technology.

### 2.2. Acid leaching procedure

The acid leaching process was assisted with hydrochloric acid. The material was soaked and stirred in 1 mol/L HCl at temperature 100 °C for 1 hour. The leached material was filtered with mesh no. 400 to produce leachate and filtrate. The filtrate was then washed with demineralized water.

### 2.3. Experimental method and variations

There were four sequence variations in this study, namely control, T1, T2, and T3. For control variation, extraction was held without applying acid leaching. The extraction was done for 300 g RHA in 1.2 L NaOH 1 mol/L solution at 120 °C for 2 h under well and intense agitation. Next, the extract solution was filtered with mesh no. 400 to become residue (filtrate) and silica sol (filtered solution). The silica

sol was titrated drop by drop with 1 mol/L HCl until formed gel-like substances. The gel was then aged for 2 h and subsequently dried in an oven at 105 °C for 6 h. For T1 sequence, RHA was leached at first and was then extracted under the same procedure as control variation. But, the aged gel should be washed with demineralized water before drying. For T2 sequence, RHA was extracted as similar as control procedure but acid leaching was subjected to the aged gel. The gel was then washed with demineralized water and was dried. For T3 sequence, RHA was extracted with the same step as the control but the acid leaching was conducted to the dried product. The leached product was washed and then followed by drying again. Whole parameters such as RHA amount, alkaline amount and concentration, extraction time and temperature, aging time, and drying time were identical to control variation. Schematically, the experimental setup is presented in Fig. 1.

### 2.4. Product characterizations

A Bruker D8 Advance XRD instrument using Cu-K $\alpha$  scanned at 2 $\theta$  from 5 to 90° was performed to analyze product crystallinity pattern. Before the measurement was applied, the sample was dried and grinded to a fine powder. The product mineral composition was measured using Rigaku Cartesian Geometry energy dispersive X-ray fluorescent (ED-XRF) equipped with high-performance silicon drift detector, X-ray tube with palladium anode, 50 Watt of power, and 50 kV of voltage. The sample should be grinded and sieved until particle size uniformly reached 200 mesh.

A Quantachrome Touchwin v1.2 analyzer that follows Brunauer-Emmett-Teller (BET) and Barrett-Joyner-Halenda (BJH) methods were employed to exploit the product surface area and pore diameter. The sample was first degassed at temperature 300 °C for 3 h at a vacuum condition to guarantee the moisture and impurities being removed. Nitrogen was then injected at temperature 77.35 K with relative pressure, P/P<sub>0</sub>, ranging from 0.022 – 0.3 to meet the Langmuir adsorption isotherm mechanism [22,23]. The product morphology was evaluated under Phenom™ ProX desktop scanning electron microscope (SEM) with 12.5 kV voltage, working distance 9.6 mm, and under 2500 x magnification.

### 2.5. Green silica quality enhancement calculation

The green silica purity enhancement percentage ( $E_p$ ) was evaluated using Eq. (1) and the surface area enhancement percentage ( $E_s$ ) was obtained using Eq. (2).

$$E_p = \frac{p' - p}{p} \times 100\% \quad (1)$$

$$E_s = \frac{S' - S}{S} \times 100\% \quad (2)$$

$p'$  is green silica purity with acid leaching,  $p$  is green silica purity without acid leaching (control),  $S'$  is green silica surface area with acid leaching, and  $S$  is green silica surface area without acid leaching (control). The  $p'$  and  $p$  values are obtained from ED-XRF data whereas  $S'$  and  $S$  values are acquired from BET data.

### 2.6. Green silica usage experiment procedure

The commercial silica gel, green silica with the poorest quality, and green silica with the highest quality, with the initial amount of 100 g each were intended for a humid room moisture adsorber. The room temperature and humidity were first measured using a hygrometer. The silica mass change was measured every day for 12 days and the experiment was carried out in duplicate. The daily cumulative moisture adsorbed amount ( $A_{M,cum}$ ) for each product was calculated follows Eq. (3).

$$A_{M,cum} \left[ \frac{mg}{g} \right] = \frac{m_{s,i} - m_A}{m_A} \times 1000 \quad (3)$$

$m_{s,i}$  is the silica mass at  $i^{\text{th}}$  day [g] and  $m_A$  is the initial silica mass [g].

## 3. Results and discussion

### 3.1. The considerations of rice husk ash acid leaching with HCl

Several researchers have compared leaching performance with citric acid, oxalic acid,  $H_2SO_4$ , and HCl. The leaching is all done in the first sequence as pretreatment with a negligible difference of product purity and surface area [9,15,17,18,21]. It requires a larger amount of chemicals with a factor of 5.4 for making 1 mol/L of citric acid, 2.5 for oxalic acid, and 2.8 for  $H_2SO_4$ , compared to HCl as a result of their higher molecular weight. As a consequence, leaching with citric acid, oxalic acid, or  $H_2SO_4$  will produce a larger leachate amount than HCl. In the meantime, the leachate treatment either through biological process, neutralization, or oxidation prior to discharge is equally a must [24]. Therefore, using HCl could reduce the operational cost in terms of chemicals and leachate treatment costs.

The rice husk has a rigid and cylindrical shape as shown in Fig. 2a. Within these properties, acid is difficult to infiltrate the inner structure of rice husk [1,2,5,25]. Thus, rice husk acid leaching should be

taken at 6–8 bar of pressure, extra vigorous mixing, and temperature 200 °C to crack the particle, which guarantees the complete acid diffusion to the rice husk inner structure and drag the impurities out [2,6]. On the other hand, RHA is a rice husk combustion product that has a more brittle structure. Consequently, most of the cylindrical shape of rice husk is cracked and transformed into more spherical. Some of the cylindrical shapes are standstill but with a lesser amount (Fig. 2b).

Due to the brittle structure of RHA, leaching could be performed in atmospheric pressure, mild mixing condition, and lower temperature with a shorter time. Apart from that, RHA leaching requires an acid about  $1/5$  times than rice husk leaching, because RHA occupies nearly 20%-wt from rice husk. For a closure, RHA acid leaching has an eminence to suppress the acid amount and leachate treatment cost.

### 3.2. Acid leaching effects on extraction process

It is found that in absence of acid leaching, extraction with 1.2 L NaOH was unsuccessful, even at a higher temperature and longer extraction time. This phenomenon occurs due to RHA is not perfectly soaked in the NaOH solution as can be seen in Fig. 3a, so the agitator could not work properly. Without agitation, the RHA is difficult to be dissolved in the NaOH solution due to the low solid-liquid mass transfer [26,27] and those prevent the pulverization of RHA structure which obviously hinders the extraction process.

Fig. 3b shows that when acid leaching is applied, the previous extraction is possible to be held because leached RHA is perfectly soaked in the NaOH solution. Acid leaching allows pulverization of RHA particle (Fig. 3c). Pulverized RHA decreases the void fraction, which in line with reducing bulk volume. Agitation occurs and this will aid the extraction process. From this exposition, the control sequence could not be performed. To overcome this matter, all extraction variations are then carried out using 1.5 L of NaOH.

### 3.3. Acid leaching effects on product quality

RHA and green silica were first analyzed with XRD. Actually, RHA itself is a silica source because it has a similar  $2\theta = 22^\circ$  peak appearance with amorphous silica [21], while another recorded peak indicates other minerals compound which is absorbed from soil and fertilizer. The green silica from this study has the same XRD pattern as amorphous silica, which indicates a disordered structure of silica [9,17,28–32], as shown in Fig. 4.

The amorphous percentage for RHA is 61.7%, while for green silica from control, T1, T2, and T3 are 51.6%, 99.9%, 99.9%, and 62.5%, in a respective term. More impurities impact on higher crystallinity. Sharp narrow peak appearance in Fig. 4 is not caused by silica

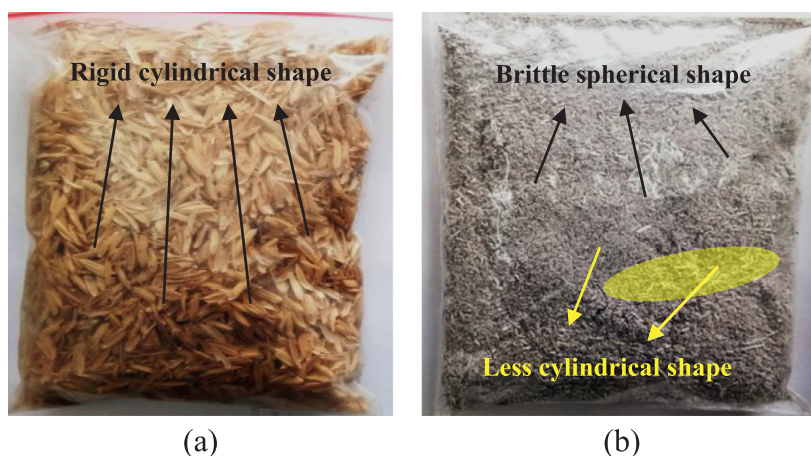


Fig. 2. Rice husk (a) and rice husk ash particle (b).

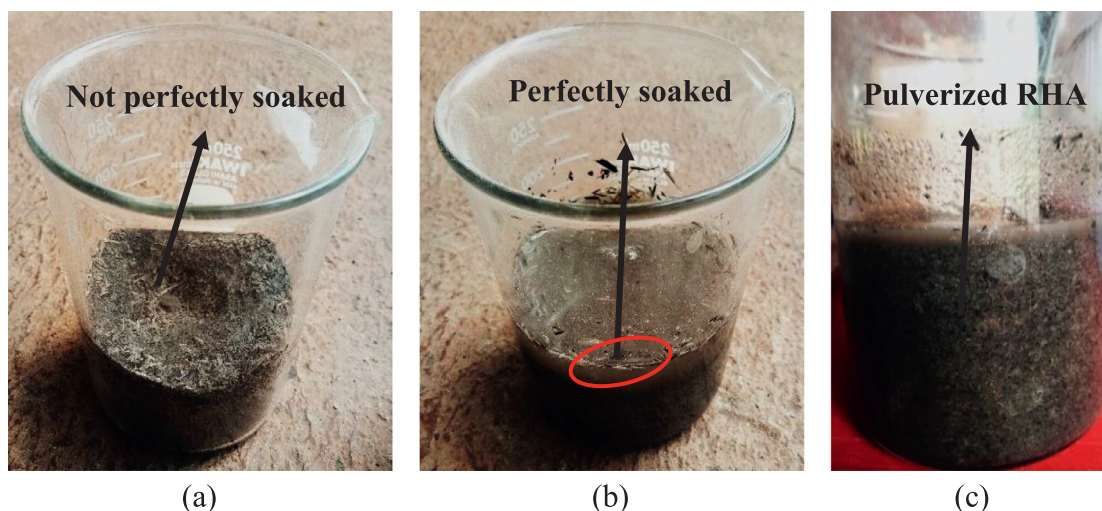


Fig. 3. RHA in NaOH solution without acid leaching (a); with acid leaching (b); Pulverized RHA particle.

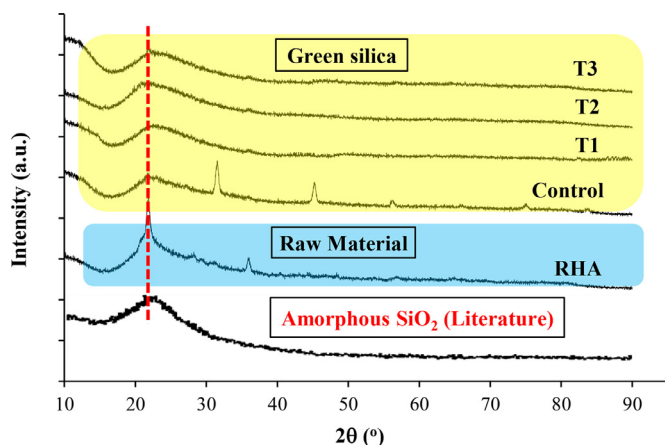


Fig. 4. RHA and green silica XRD profile.

crystalline but is attributed to the mineral crystal of  $\text{NaCl}$ ,  $\text{Na}_2\text{SO}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{K}_2\text{O}$ ,  $\text{Na}_2\text{O}$ , and  $\text{Al}_2\text{O}_3$  as major impurities [30,33–36]. In line with the amorphous percentage, impurities were significantly found in control variation and then followed by T3 variation.

Subsequently, the explanation is achieved from a mineral assay through ED-XRF. Table 1 implies that the major impurities recorded are sodium oxide, potassium oxide, and sulfur trioxide. The acid leaching on RHA reduces the impurities and increases the silica content only by 6.22%. Meanwhile, in T1 sequence, acid leaching and gel washing successfully removed much amount of impurities content [32,37,38] and thus promotes the highest green silica purity, 96.44%.

Table 1  
ED-XRF result for RHA and green silica.

Mineral	Composition (% mass)					
	RHA	Leached-RHA	Control	T1	T2	T3
$\text{SiO}_2$	79.63	85.85	56.94	96.44	87.69	62.46
$\text{Na}_2\text{O}$	7.04	3.31	31.42	1.38	5.85	21.30
$\text{K}_2\text{O}$	3.83	2.65	2.23	0.51	0.49	3.44
$\text{SO}_3$	2.16	1.62	8.34	0.79	0.94	2.57
Cl	–	0.61	0.32	0.28	0.62	1.21
$\text{P}_2\text{O}_5$	1.87	0.94	0.28	0.27	2.82	4.39
$\text{Al}_2\text{O}_3$	1.86	1.73	0.31	0.18	1.01	1.99
CaO	2.48	2.35	0.13	0.10	0.49	2.50
$\text{Fe}_2\text{O}_3$	1.04	0.92	0.01	0.01	0.08	0.10
ZnO	0.09	0.02	0.01	0.04	0.01	0.04

The previous explanation is then strengthened with BET result. RHA from this experiment has a surface area of  $46.78 \text{ m}^2/\text{g}$ , slightly higher than RHA obtained from Fernandes et al. (2016) [3]. The green silica from control variation has the lowest surface area,  $5.72 \text{ m}^2/\text{g}$  due to a large amount of sodium hydroxide from RHA and alkaline solvent still adheres to the aged gel. After gel drying, the mineral from impurities and sodium mineral then plug into the pore structure and this phenomenon will provoke the lowest surface area [39–42]. Sodium oxide content was 31.42% in control variation, which supported the aforementioned argument.

It is found that impurities in the extract solution are far lesser for T1 and this will contribute to the highest surface area, reaching  $400.69 \text{ m}^2/\text{g}$ . Aside from 96.44% purity, sodium oxide content is significantly reduced to 1.49%. It can be said that pretreatment with acid leaching is able to reduce earth alkaline impurities and gel washing with water is proven to be effective for sodium oxide removal [19,33,43].

Interestingly, T2 sequence also resulted in the green silica with quite low purity and surface area. Impurities removal through T2 seems not satisfying compared to T1. However, sodium oxide mineral was found at 5.85%. Again, sodium oxide is still able to be reduced but the remaining impurities still exist because they have already been trapped in the established gel networks which are difficult to be dragged out by acid leaching [41,42,44]. These trapped impurities block the silica pore and lead to a surface area of  $55.49 \text{ m}^2/\text{g}$ , just slightly higher than RHA.

For T3 sequence, leaching was employed on dried green silica, which has a tougher structure than RHA. Drying segregates the solvent in the gel networks porous matrix and complicates the trapped impurities to free indeed [42]. Moreover, it also induces the impurities to sinter tightly and collapses in the capillary structure [39–41]. This result has a significant amount of sodium oxide, 21.30%. The green silica from T3 sequence has the lowest surface area compared to T1 and T2 but still higher than that of the control,  $11.40 \text{ m}^2/\text{g}$ . The explanation for rice husk ash extraction phenomena under various acid leaching sequences is illustrated briefly in the graphical abstract.

Apart from this, T3 sequence requires two steps of the drying process. This is considered a weakness due to the high-cost process. The result for RHA and green silica surface area are tabulated in Table 2. Hereafter, the Langmuir adsorption isotherm plot for T1, T2, and RHA show a concave curve towards relative pressure whereas T3 and control exhibit almost linear lines, as depicted in Fig. 5. Based on the qualitative view, the more concave curvature implies the mesoporous product was obtained [22,45,46]. Based on BJH analysis, pore

**Table 2**  
RHA and green silica surface area.

Sample Code	Surface Area (m <sup>2</sup> /g)
RHA	46.78
Control	5.71
T1	400.69
T2	55.49
T3	11.40

diameter for green silica from T1 sequence was obtained at 41 Å and categorized as a mesoporous material [45,47–49], which supports the qualitative explanation.

According to XRD, ED-XRF, and BET results analysis, the production of high purity and high surface area of amorphous green silica from RHA should apply acid leaching as a pretreatment or in the first sequence. The green silica surface area from T1 is found higher than the untreated one as a control, but the purity has not yet exceeded 99%. This might be due to 300 g of RHA used in this study, whereas other researchers worked only on 10–50 g. Furthermore, the green silica purity from control variation is 56.94%, which is far lower than other researchers (82.80–95.77%) and this is supposed to be the cause of difficulty reaching purity of 99%. Nevertheless, RHA extraction for this study (through T1 sequence) gives a very significant purity and surface area enhancement percentage, 69% and 6905%, respectively, as summarized in Table 3.

Fig. 6 observes that green silica particle morphology from T1 sequence is indicated an unlaminated structure, irregular shape, and non-uniform particle size with mostly smaller than 10 μm. It is realized that alkaline extraction method mostly produced irregular particle shape due to not easy to control their formation [38]. The non-uniform particle size was also obtained from Setyawan et al. (2019) [31], Al-maamori et al. (2015) [33], and Phoohinkong and Kitthawee (2014) [36]. Several bigger size silica is found due to the aggregation tendency that might be happened for a reason of drying [39,40]. Likewise, manual grinding is the cause for particle non-uniformity and still allows the large particle to remain [21].

**Table 3**  
Acid leaching effects on product purity and surface area enhancement percentage.

Acid	P (%)	P' (%)	E <sub>p</sub> (%)	S (m <sup>2</sup> /g)	S' (m <sup>2</sup> /g)	E <sub>s</sub> (%)	Reference
HCl	–	98.20	–	–	120	–	[8]
	–	99.10	–	–	980	–	
	–	99.70	–	–	150	–	
	82.80	99.30*	19.93	–	–	–	[9]
	95.77	99.58	3.98	116	218	87.93	[15]
Citric	91.50	98.80	7.98	56.6	311.6	451.50	[17]
	–	99.76	–	–	234.60	–	[18]
	–	93.00	–	–	–	–	[19]
	90.50	95.90	5.97	–	169.70	–	[20]
	82.80	98.60*	19.08	–	–	–	[9]
Oxalic	94.58	99.14	4.82	–	–	–	[16]
	91.50	99.00	8.20	56.6	254.1	349.73	[17]
	–	99.77	–	–	215.80	–	[18]
H <sub>2</sub> SO <sub>4</sub>	92.40	99.30*	7.47	–	–	–	[21]
HCl	95.77	99.08	3.46	116	208	79.31	[15]
HCl	56.94	96.44**	69.37	5.72	400.69	6905.07	This study

\* Highest value from the experiment results.

\*\* T1 sequence.

### 3.4. Green silica usage performance

The green silica is preliminary proposed for industrial silica gel as a moisture adsorber and it fulfills some requirements as compared in Table 4. In order to strengthen the qualitative explanation, the study on silica usage performance in a humid room was conducted on commercial silica gel, the poorest green silica quality (control), and the highest quality (T1). The commercial silica gel has specifications of blue color, spherical, purity 97.71%, and surface area 676.67 m<sup>2</sup>/g. The humid room has an average temperature of 27.9 °C (± 0.4) and relative humidity of 64.1% (± 3.1), while the outside environment has an average relative humidity of 40.1% (± 1.3). For commercial silica gel and green silica, the daily cumulative moisture adsorbed amount from the humid room is continued to increase day by day, as seen in Fig. 7.

On the twelfth day, the average cumulative moisture adsorbed value reached 240.12 mg/g for commercial silica gel, 206.72 mg/g for

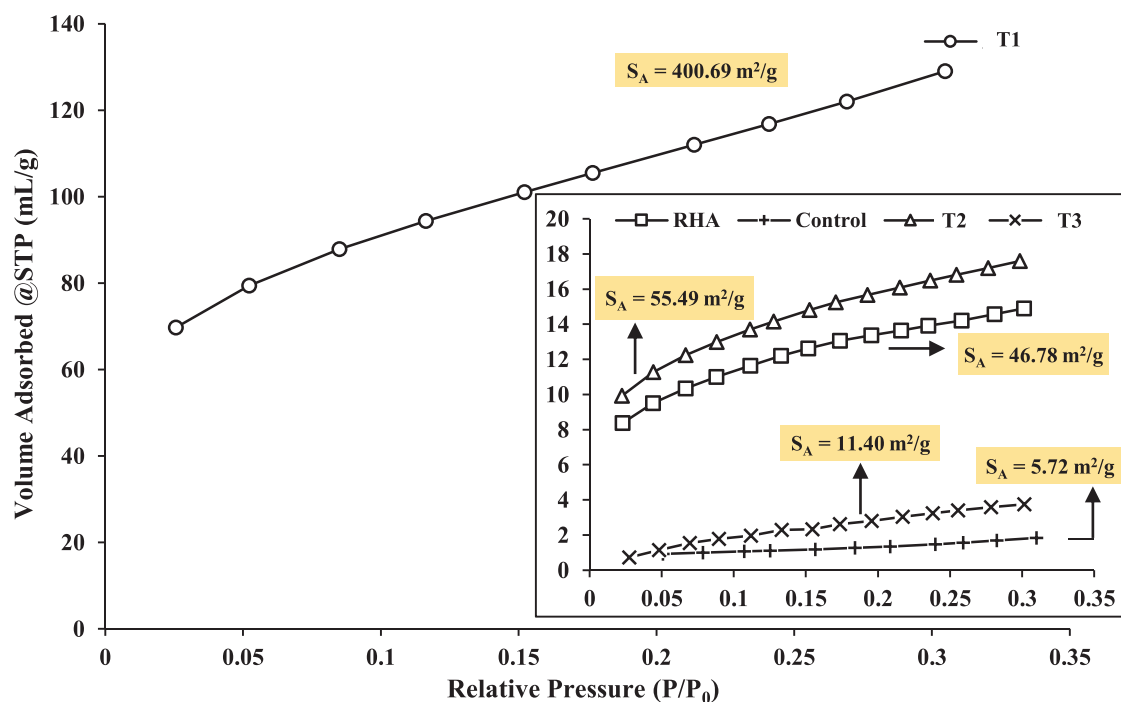


Fig. 5. Low-temperature N<sub>2</sub> adsorption isotherm curves for RHA and green silica.

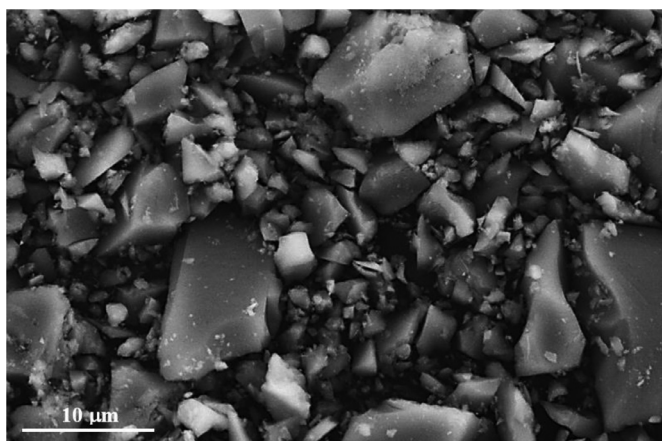


Fig. 6. Green silica morphology from T1 sequence.

**Table 4**  
Industrial silica gel specifications comparison.

Surface Area (m <sup>2</sup> /g)	Pore Diameter (nm)	Reference
136–285	8.42 – 19.80	[50]
142–357	13.00	[11]
201	–	[51]
254–262	10.97 – 13.36	[52]
610	2.40	[12]
686–827	3.19 – 3.24	[10]
528–863	–	[13]
400.69	4.10	This Study

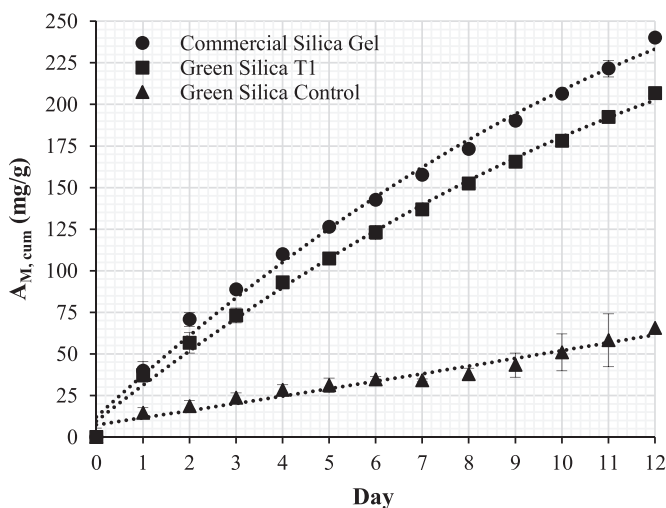


Fig. 7. Cumulative moisture adsorbed amount for commercial silica gel and green silica (T1 and control).

green silica T1, and 65.53 mg/g for green silica control. From this study, the green silica from T1 has a very close performance to the commercial silica gel. On the other hand, green silica from control exhibits the lowest performance. The moisture adsorption into the silica pores is hindered due to its low purity and surface area which is caused by pore-clogging by a large number of impurities. This is implied that high surface area and mesoporous green silica from T1 is meet the criteria and proven to be utilized as industrial silica gel with another superiority comprises sustainability and greener process.

#### 4. Conclusions

An amorphous and mesoporous green silica with 96.44% of purity and 400.69 m<sup>2</sup>/g of surface area is successfully produced from RHA

extraction under T1 sequence. This study confirmed that acid leaching should be carried out at first, as a biomass pretreatment. In absence of acid leaching, as well as acid leaching on the other sequences, will induce lower quality of green silica. Green silica production following T1 sequence is aimed for product quality improvement, which encompassed higher purity, greater surface area, has an amorphous and mesoporous particle. The improved green silica product from T1 sequence could be combined with future utilization for sustainable industrial silica gel with the moisture adsorber performance that is somewhat similar to the commercial spherical silica gel blue.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### References

- Chen G, Du G, Ma W, Yan B, Wang Z, Gao W. Production of amorphous rice husk ash in a 500kW fluidized bed combustor. *Fuel* 2015;144:214–21. doi: 10.1016/j.fuel.2014.12.012.
- Rufai IA. *Biosilica from rice husk ash as a new engineering raw material in Nigeria*. Natl. Eng. Conf. Exhib. Annu. Gen. Meet. 2015.10.086.
- Fernandes JJ, Calheiro D, Kieling AG, Moraes CAM, Rocha TLAC, Brehm FA, et al. Characterization of rice husk ash produced using different biomass combustion techniques for energy. *Fuel* 2016;165:351–9. doi: 10.1016/j.fuel.2015.10.086.
- Azat S, Korobeinyk AV, Moustakas K, Inglezakis VJ. Sustainable production of pure silica from rice husk waste in Kazakhstan. *J Clean Prod* 2019;217:352–9. doi: 10.1016/j.jclepro.2019.01.142.
- Rozainee M, Ngo SP, Salema AA, Tan KG, Ariffin M, Zainura ZN. Effect of fluidising velocity on the combustion of rice husk in a bench-scale fluidised bed combustor for the production of amorphous rice husk ash. *Bioresour Technol* 2008;99:703–13. doi: 10.1016/j.biortech.2007.01.049.
- Mittal D. Silica from ash. *Resonance* 1997;2:64–6. doi: 10.1007/bf02838592.
- Bangwar D, Saand A, Keerio M, Soomro M, Bhatti N. Development of an amorphous silica from rice husk waste. *Eng Technol Appl Sci Res* 2017;7:2184–8. doi: 10.5281/zenodo.1118285.
- Azat S, Sartova Z, Bekseitova K, Askaruly K. Extraction of high-purity silica from rice husk via hydrochloric acid leaching treatment. *Turkish J Chem* 2019;43:1258–69. doi: 10.3906/kim-1903-53.
- Zarib NSM, Abdullah SA, Jamil NH. Extraction of silica from rice husk via acid leaching treatment. *Eur. Proc. Soc. Behav. Sci.* 2019:1–6.
- Mohammed RH, Mesalhy O, Elsayed ML, Hou S, Su M, Chow LC. Physical properties and adsorption kinetics of silica-gel/water for adsorption chillers. *Appl Therm Eng* 2018;137:368–76. doi: 10.1016/j.applthermaleng.2018.03.088.
- Prasad R, Pandey M. Rice husk ash as a renewable source for the production of value added silica gel and its application: an overview. *Bull Chem React Eng Catal* 2012;7:1–25. doi: 10.9767/bcrec.7.1.1216.1-25.
- Rong A, Liu M, Pang L, Yang D, Wang J, Zhou Y. Kinetics study of gas pollutant adsorption and thermal desorption on silica gel. *Appl Sci* 2017;7:609. doi: 10.3390/app7060609.
- Thu K, Chakraborty A, Saha BB, Ng KC. Thermo-physical properties of silica gel for adsorption desalination cycle. *Appl Therm Eng* 2013;50:1596–602. doi: 10.1016/j.applthermaleng.2011.09.038.
- Ma X, Zhou B, Gao W, Qu Y, Wang L, Wang Z, et al. A recyclable method for production of pure silica from rice hull ash. *Powder Technol* 2012;217:497–501. doi: 10.1016/j.powtec.2011.11.009.
- Bakar RA, Yahya R, Gan SN. Production of high purity amorphous silica from rice husk. *Procedia Chem* 2016;19:189–95. doi: 10.1016/j.proche.2016.03.092.
- Umeda J, Kondoh K. High-purity amorphous silica originated in rice husks via carboxylic acid leaching process. *J Mater Sci* 2008;43:7084–90. doi: 10.1007/s10853-008-3060-9.
- Sapei L, Pramudita A, Widjaja LB. Pure amorphous silica derived from calcined acid-leached rice husk. *Pittcon Conf Expo* 2014:1.
- Mahmud A, Megat-Yusoff PSM, Ahmad F, Farezzuan AA. Acid leaching as efficient chemical treatment for rice husk in production of amorphous silica nanoparticles. *ARPN J Eng Appl Sci* 2016;11:13384–8.
- Kalpathy U, Proctor A, Shultz J. A simple method for production of silica from rice hull ash. *Bioresour Technol* 2002;85:285–9. doi: 10.1016/S0960-8524(02)00116-5.
- Zulfiqar U, Subhani T, Wilayat Husain S. Towards tunable size of silica particles from rice husk. *J Non Cryst Solids* 2015;429:61–9. doi: 10.1016/j.jnoncrysol.2015.08.037.

- [21] Pa FC, Kein WK. Removal of iron in rice husk via oxalic acid leaching process. *IOP Conf Ser Mater Sci Eng* 2019;701:012021. doi: 10.1088/1757-899x/701/1/012021.
- [22] Brunauer S, Emmett PH. The use of low temperature van der Waals adsorption isotherms in determining the surface areas of various adsorbents. *J Am Chem Soc* 1937;59:2682–9.
- [23] Barrett EP, Joyner LG, Halenda PP. The determination of pore volume and area distributions in porous substances: I. Computations from nitrogen isotherms. *J Am Chem Soc* 1951;73:373–80.
- [24] Tchobanoglous G, Burton FL, Stensel HD. Metcalf & eddy, inc. Wastewater Engineering Treatment and Reuse. 2003.
- [25] Martínez JD, Pineda T, López JP, Betancur M. Assessment of the rice husk lean-combustion in a bubbling fluidized bed for the production of amorphous silica-rich ash. *Energy* 2011;36:3846–54. doi: 10.1016/j.energy.2010.07.031.
- [26] Geankoplis CJ. Agitation and mixing of fluids and power requirements. *Transp. Process. Unit Oper. Third Ed.*, 1993:141–51.
- [27] McCabe W, Smith JC, Harriot P. Agitation and mixing of liquids. *Unit Oper. Chem. Eng.* 1993:236. 264–5.
- [28] Sarikaya M, Depci T, Aydogmus R, Yucel A, Kizilkaya N. Production of nano-amorphous SiO<sub>2</sub> from Malatya pyrophyllite. *IOP Conf Ser Earth Environ Sci* 2016;44. doi: 10.1088/1755-1315/44/5/052004.
- [29] Carvalho C. Thermal and chemical treatments to rice husk valorization. 2015.
- [30] Ul-Haq I, Akhtar K, Malik A. Effect of experimental variables on the extraction of silica from the rice husk ash. *J Chem Soc Pakistan* 2014;36:382–7.
- [31] Setyawan N, Hoerudin, Wulanawati A. Simple extraction of silica nanoparticles from rice husk using technical grade solvent: effect of volume and concentration. *IOP Conf Ser Earth Environ Sci* 2019;309. doi: 10.1088/1755-1315/309/1/012032.
- [32] Su Y, Liu L, Zhang S, Xu D, Du H, Cheng Y, et al. A green route for pyrolysis poly-generation of typical high ash biomass, rice husk: effects on simultaneous production of carbonic oxide-rich syngas, phenol-abundant bio-oil, high-adsorption porous carbon and amorphous silicon dioxide. *Bioresour Technol* 2020;295:1–9.
- [33] Al-maamori MH, Ahmed JK, Ali HM. Production of nano-silica from water glass production of nano-silica from water glass. *Acad Res Int* 2015;6:89–97.
- [34] Jain A, Ong SP, Hautier G, Chen W, Richards WD, Dacek S, et al. Commentary: the materials project: a materials genome approach to accelerating materials innovation. *APL Mater* 2013;1:1–11.
- [35] Muljani S, Setyawan H, Wibawa G, Altway A. A facile method for the production of high-surface-area mesoporous silica gels from geothermal sludge. *Adv Powder Technol* 2014;25:1593–9. doi: 10.1016/j.apt.2014.05.012.
- [36] Phooinkong W, Kitthawee U. Low-cost and fast production of nano-silica from rice husk ash. *Adv Mater Res* 2014;979:216–9. doi: 10.4028/www.scientific.net/AMR.979.216.
- [37] Chakraverty A, Mishra P, Banerjee HD. Investigation of combustion of raw and acid-leached rice husk for production of pure amorphous white silica. *J Mater Sci* 1988;23:21–4.
- [38] Chun J, Lee JH. Recent progress on the development of engineered silica particles derived from rice husk. *Sustainability* 2020;12:1–19.
- [39] Pantos E, West JB, Dokter WH, Van Garderen HF, Beelen TPM, Van Santen RA. Growth and aging phenomena in silica gels. *J Sol-Gel Sci Technol* 1994;2:273–6. doi: 10.1007/BF00486255.
- [40] van Santen RA, Beelen TPM, van Garderen HF, Dokter WH, Pantos E. Aggregation and aging in silica gel. *Nucl Inst Methods Phys Res B* 1995;97:231–7. doi: 10.1016/0168-583X(95)00194-8.
- [41] Brinker CJ, Scherer GW. *Sol-Gel science: the physics and chemistry of sol-gel processing*. Academic Press, Inc.; 1990.
- [42] Hench LL, West JK. The sol-gel process. *Chem Rev* 1990;90:33–72. doi: 10.1021/cr00099a003.
- [43] Ghosh R, Bhattacharjee S. A review study on precipitated silica and activated carbon from rice husk. *J Chem Eng Process Technol* 2013;04. doi: 10.4172/2157-7048.1000156.
- [44] de Lima SPB, Paiva RP de V e OA, Cordeiro GC, Chaves MR de M, Fairbairn RDTF e E de MR. Production of silica gel from rice husk ash. *Quim Nov* 2011;34:71–5.
- [45] Sing KSW. Reporting physisorption data for gas/solid systems with special reference to the determination of surface area and porosity. *Pure Appl Chem* 1985;57:603–19.
- [46] Brunauer S, Emmett PH, Teller E. Adsorption of gases in multimolecular layers. *J Am Chem Soc* 1938;60:309–19.
- [47] Rouquerol F, Rouquerol J, Sing KSW. Adsorption by powders and porous solids: principles, methodology and applications. 1999.
- [48] Gregg SJ, Sing KSW. Adsorption surface area and porosity, Second Edition. 1982.
- [49] Thommes M, Kaneko K, Neimark AV, Olivier JP, Rodriguez-Reinoso F, Rouquerol J, et al. Physisorption of gases, with special reference to the evaluation of surface area and pore size distribution (IUPAC technical report). *Pure Appl Chem* 2015;87:1051–69.
- [50] Pijarn N, Jaroenworarluck A, Sunsaneeyametha W, Stevens R. Synthesis and characterization of nanosized-silica gels formed under controlled conditions. *Powder Technol* 2010;203:462–8. doi: 10.1016/j.powtec.2010.06.007.
- [51] Zhang Y, Zhang Y, Zhang H. Study on preparation of a novel silica adsorbent and its selective separation applied to genistein. *Brazilian J Chem Eng* 2008;25:201–6. doi: 10.1590/S0104-66322008000100020.
- [52] Kamath SR, Proctor A. Silica gel from rice hull ash: preparation and characterization. *Cereal Chem* 1998;75:484–7. doi: 10.1094/CCHEM.1998.75.4.484.