

# 1. Penulis Anggota

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# A Simple Material and Energy Input–Output Performance in Evaluating Silica Production from Conventional, Fume, and Biomass Thermochemical Conversion Routes

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## Abstract

Rice husk valorization to produce silica is believed to involve an eco-friendly process rather than silica production from conventional and fume routes. Nevertheless, the quantitative point of view regarding this is still not widely disclosed. In this study, a simple material and energy input–output analysis (M&E I/O) is employed to compare the environmental impact of the production of 1 tonne of silica from conventional, fume, and biomass thermochemical conversion routes. The scope consideration includes raw material, transportation, utility systems, main production process, and output streams as environmental impact. Results show that conventional and fume routes need 3.86 tonnes of sandstone and biomass thermochemical conversion route needs 6.56 tonnes of rice husk. For conventional and fume routes, energy is supplied from 1194.08 and 1954.99 kg of coal combustion, whereas the biomass thermochemical conversion route uses rice husk as fuel and additional coal of only 238.38 kg. Further, the lowest CO<sub>2</sub>-equivalent emission of 0.85 tonnes is nominated to the biomass thermochemical conversion route, while conventional and fume routes are 10.09 and 18.62 tonnes, respectively. The produced wastewater from conventional, fume, and biomass thermochemical conversion routes is 27.27, 27.13, and 24.76 tonnes, successively. This study concludes and proves that silica production from rice husk is more eco-friendly and has low environmental impact, but wastewater treatment to meet the effluent standard should be applied.

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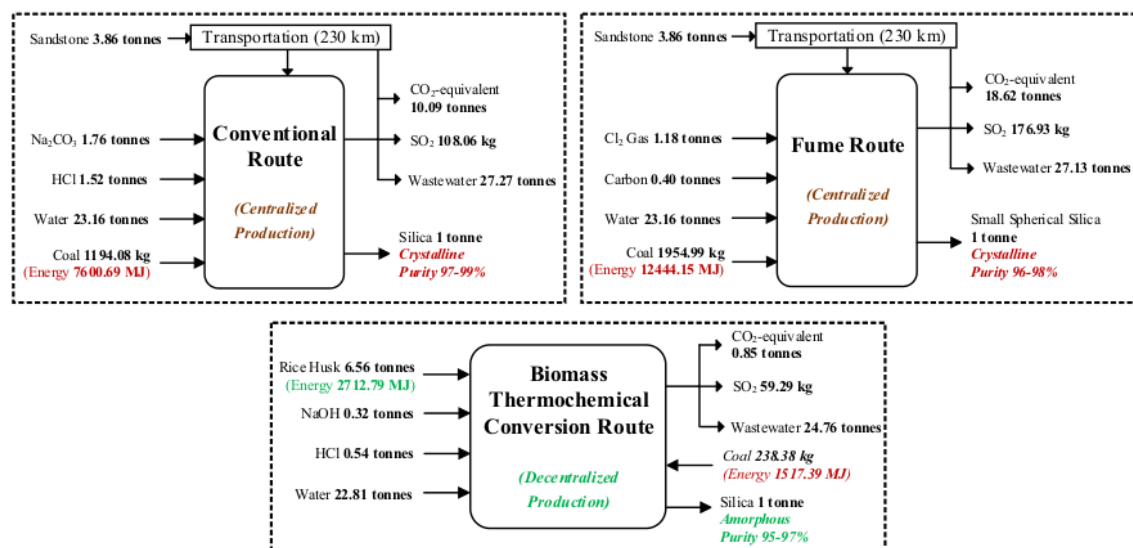
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## Graphical Abstract



**Keywords** Rice husk · Silica · Environmental impact · Sustainable · Electricity · Quartz

### Statement of Novelty

Rice husk is an attractive agricultural biomass because it is rich in silica and has a significant heating value. There have been many studies that dealt with rice husk valorization and concluded its eco-friendly production. However, there are still no studies that quantitatively prove using material and energy input-output (M&E I/O) analysis that the silica production from rice husk is more eco-friendly than conventional and fume routes that employ sandstone as raw material. According to the results, this study convinces the sustainability of rice husk processing as potential silica and energy from a more renewable resource.

### Introduction

Silica is an abundant mineral that has many applications in diverse fields, e.g., medical field (for drug delivery vector) [1], construction field (for mixture agent in cement portland) [2, 3], chemicals field (for chemical adsorbent, moisture adsorber, catalyst support) [4–6], electronics field (for semiconductor, transistor, or printed circuit board) [7]. The production of high-purity silica requires a significant amount of fossil resources which leads to severe environmental

impacts, such as acid rain, ozone depletion, and global warming [8–11].

Mainly, silica is synthetically produced from sandstone through conventional or fume route. Conventional route using sandstone which contains K-feldspar and quartz. The process involves quartz heating with high temperatures of 1200–1800 °C [12–16]. Meanwhile, the quartz heating in fume route is carried out under plasma electric arc furnaces under extreme temperatures, surpassing 3000 °C at the arc zone. The high temperatures cause the material to vaporize and the resulting fumes are collected and condensed to obtain silica [17–19]. Besides the huge energy supplied, high process temperatures lead to enormous  $\text{NO}_x$  formation [20, 21]. For conventional and fume routes, crushing and washing of sandstone are employed as pretreatment.

To overcome the environmental problems and to meet the goal of sustainability, silica production is proposed using biomass thermochemical conversion route [22, 23]. Biomass from agricultural residues represents abundant and renewable resources in agrarian nations which is extensively exploited to produce chemicals and fossil fuel substitutes [24, 25]. Rice husk, as a residue in the rice milling house, stands out among the various types of agricultural biomass. It occupies approximately 20w/w% from straw-free rice [26, 27]. Rice husk also has heating value of 14–17 MJ/kg and silica content in ash up to 95w/w% [28–32].

On the basis of these interesting facts, rice husk valorization has become promising [12, 33], particularly in agricultural countries like Indonesia. However, rice husk is only piled up or open-burned on the landfill which becomes a burden to the environment [34] whereas it can play a role as potential and sustainable resource in terms of silica and energy producer. Through biomass thermochemical conversion route, rice husk is combusted to produce energy and ash. The ash then undergoes the sol-gel process to synthesize silica that employs alkaline as an extracting solution and acid or CO<sub>2</sub> gas as a precipitating agent [35–37]. The process holds the potential to reduce dependency on fossil resources while mitigating greenhouse gas emissions.

Many studies state the sustainability of rice husk utilization, but there is no study that directly compares the material and energy input–output (M&E I/O) performance of silica production through conventional, fume, and biomass thermochemical conversion routes. Quispe et al. calculated that rice husk utilization in Peru can decrease 97% of global warming potential. Their study uses life cycle assessment to reveal the environmental impact of coal and rice husk for electricity purposes [9]. However, this study has not dealt with silica production from rice husk yet. In addition, Joglekar et al. performed a cradle-to-gate impact analysis on silica extraction from rice husk ash. They conducted a comprehensive input–output calculation and reported a recommendation that biomass utilization should be accompanied by renewable electricity [12]. Nevertheless, their study still does not compare with other routes in producing silica.

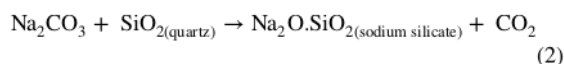
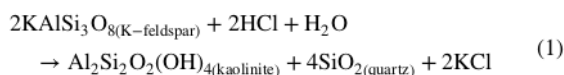
Besides, a review study by Prasara-A and Grant states rice husk utilization in Thailand can help to reduce impacts on fossil fuel depletion and climate change [24]. However, they focus on electricity and ethanol production from rice husk, not yet on silica production. Apart from that, Kubota et al. conducted a technical study related to environmental impact analysis of rice husk combustion. They found that the process has a low environmental impact [38]. The study did not specifically talk about silica production and again has not comprehensively compared the results with other processes through input–output calculations.

In order to quantitatively prove the sustainability of rice husk conversion to produce silica, this study aims to communicate an overall M&E I/O of silica production from three different aforementioned routes. M&E I/O is a simple tool used to analyze raw material and energy requirements, utility involved, and output streams as environmental impact potential [39]. It is also applied for environmental performance comparison between the existing and newly developed processes [22, 38]. The aspects of raw material, transportation, main production process, utility systems such as energy, chemicals, and water involved in supporting process production, and environmental potential analysis are

considered. All the results in this study are based on 1 tonne of silica produced.

### Silica Production from Conventional, Fume, and Biomass Thermochemical Conversion Routes

Prior to the main processing of silica production from conventional route, sandstone is first washed and ground. The washed and ground sandstone is reacted under aqueous acidic conditions to convert K-feldspar into quartz according to reaction 1. The quartz is then heated at high temperatures, up to 1800 °C, and undergoes a reaction with Na<sub>2</sub>CO<sub>3</sub> to produce Na<sub>2</sub>O.SiO<sub>2</sub> and CO<sub>2</sub> following reaction 2. The heating is conducted in the conventional electric furnace in which the heat source is produced from wire-coiled elements. Silica is finally obtained from the acid precipitation of Na<sub>2</sub>O.SiO<sub>2</sub> through reaction 3 [12, 13]. The obtained product from quartz heating process can achieve a purity of 97–99% SiO<sub>2</sub> [40] but it has high crystalline phase due to extreme heating temperature. The M&E I/O stream for silica production from conventional route is depicted in Fig. 1.



In fume route, sandstone is initially pretreated by washing and grinding. Subsequently, pretreated sandstone is reacted with acid following reaction 1 to obtain quartz. Quartz is then reacted with Cl<sub>2</sub> gas and carbon in the form of char at 1100 °C to produce SiCl<sub>4</sub> through reaction 4. This is followed by a reaction at up to 2000 °C in plasma electric arc furnaces (the temperature at the arc zone can exceed 3000 °C) until silica vapor is formed, reaction 5 [17–19, 41]. This extreme temperature is generated by applying high voltage of electricity of about 1400 kV [12]. The silica vapor is then condensed to produce small spherical silica particles [17–19, 42] with the phase of crystalline. The composition of this product reaches 96–98% SiO<sub>2</sub>, 0.1–1.5% Al<sub>2</sub>O<sub>3</sub>, 0.5–0.6% Fe<sub>2</sub>O<sub>3</sub>, P<sub>2</sub>O<sub>5</sub> 0.1%, CaO 0.1%, MgO 0.2%, K<sub>2</sub>O 0.4%, and Na<sub>2</sub>O 0.1% [17, 18]. Figure 2 presents the silica production from fume route accompanied by its M&E I/O stream.



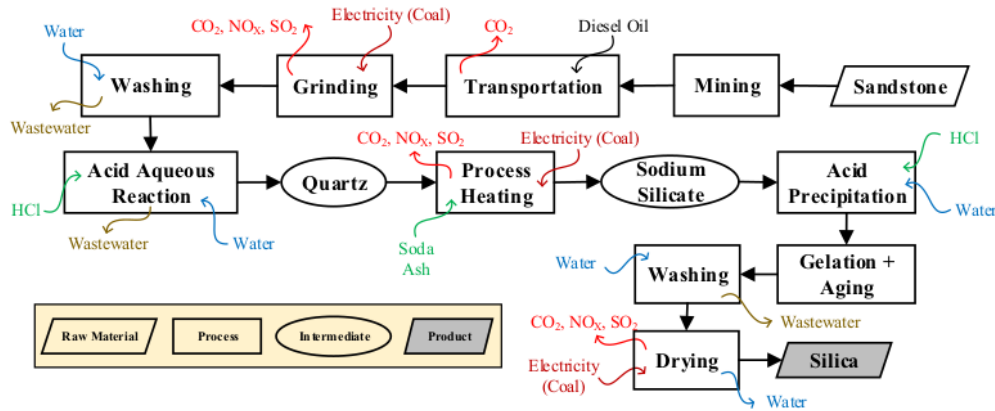


Fig. 1 M&E I/O stream for silica production from conventional route

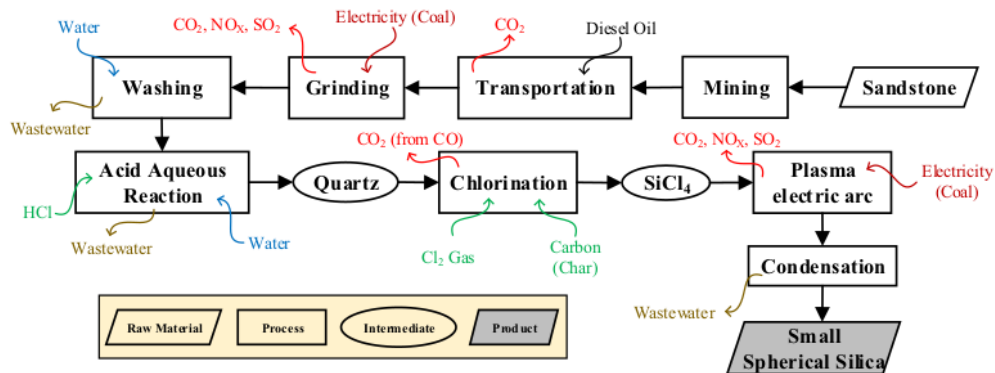
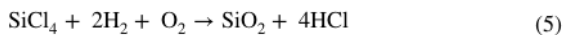
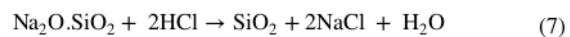
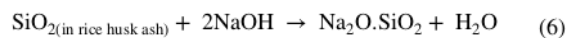


Fig. 2 M&E I/O stream for silica production from fume route

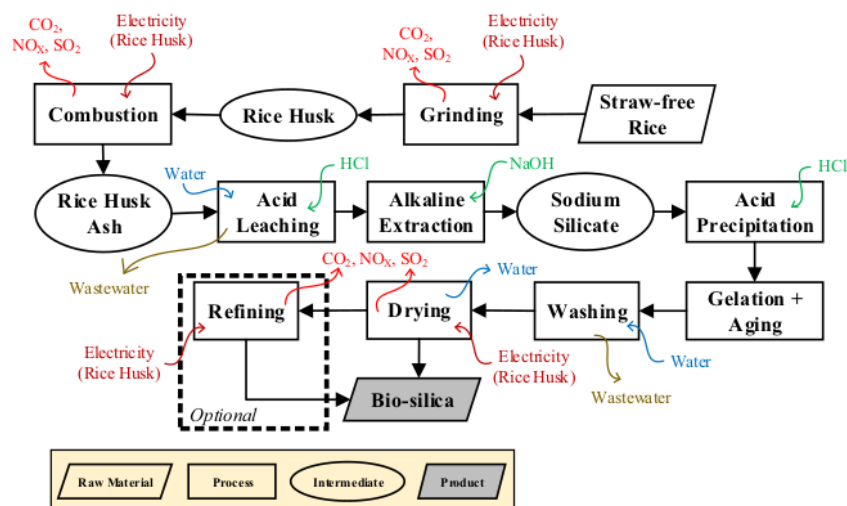


For biomass thermochemical conversion route, rice husk combustion should be performed at a maintained temperature of 700 °C because higher temperature has a high potential to transform silica phase in ash into crystalline [43] which can be detrimental to human health if exposed frequently [44, 45]. Combustion can be held in an electric furnace, but temperature control needs to pay attention, remembering the combustion should not exceed 700 °C. After combustion, rice husk ash is initially leached by 1 mol/L HCl at 100 °C to remove impurities. Leached rice husk ash is then reacted with 1 mol/L NaOH at 120 °C for 2 h through reaction 6. Afterward, the sodium silicate is precipitated by 1 mol/L HCl at 25 °C until gel is formed which follows reaction 7. Gel is aged and washed

to remove the remaining acid and base. Aged gel is finally dried to obtain amorphous silica [46]. Silica refining under box furnace is optionally carried out if the product is still blackish which is usually caused by biomass ash feed in the sol-gel process still containing unburned carbon content [47]. Following this route, the product can be acquired in fully amorphous phase with a composition of 95–97% SiO<sub>2</sub>, 0.1–0.2% Al<sub>2</sub>O<sub>3</sub>, 0.01% Fe<sub>2</sub>O<sub>3</sub>, 0.32% P<sub>2</sub>O<sub>5</sub>, 0.1% CaO, 1.5% K<sub>2</sub>O, 1.1–1.2% Na<sub>2</sub>O [35, 47]. M&E I/O stream for silica production from biomass thermochemical conversion route is served in Fig. 3.



**Fig. 3** M&E I/O stream for silica production from biomass thermochemical conversion route



## Calculation Methods

The calculation method depends on factors such as the availability of raw material, raw material characteristics, transportation, process efficiency, utility systems, fuel amount and properties, and output stream impact.

## Raw Material and Transportation

Silica production through conventional and fume routes utilized sandstone which was presumed to consist of 60w/w% K-feldspar [14, 48–50]. It was employed from four different local quarry sites in Subang, West Java, Indonesia due to the rich resources in sandstone [51]. The scope in conventional and fume routes started from transported sandstone and ended in silica product, so the energy of human power involved in the sandstone quarrying process was excluded.

All of the quarried sandstone was then transported to the silica production plant in Cilegon, Banten. This location was chosen because Cilegon is dubbed as the area with the highest concentration of petrochemical industries [52]. Based on the calculation which was assisted by Google Maps, the average distance from 1 sandstone quarry site and silica production plant was approximately 230 km.

The transportation of sandstone used dump truck with a 5-tonne capacity. It was fueled with diesel oil which contains  $C_{16}H_{34}$  with a density of 0.85 kg/L [53]. According to the field experience, the specific diesel oil consumption for full load dump truck was 0.362 L/km. This value is in accordance with the common fuel consumption for dump trucks found in the literature, which is 0.3–0.4 and 0.38 L/km for large truck sizes [54]. The diesel oil combustion inside the engine of dump truck was completely undergone to form  $CO_2$  and  $H_2O$ .

On the other hand, silica production from biomass thermochemical conversion route utilizing rice husk. It did not need to be transported to the centralized silica production plant but the production was encouraged and proposed to be held at the rice milling house itself, taking into account the combustion temperature of less than 700 °C. This concept is known as decentralized production where rice milling house is combined with on-site silica production. In accordance with the previous experimental studies by Steven et al., rice husk ash occupies 20.44w/w% from rice husk [47], silica content in rice husk ash was 87.69w/w% [35], and the yield of silica extraction was approximately 85% [35, 47]. Similar to conventional and fume routes, the scope of biomass thermochemical conversion route started from harvested straw-free rice and ended in silica product, so the energy of human power involved in rice harvesting was nullified.

## Utility Systems Requirements

Utility systems for silica production include water, chemicals, and energy (in the form of electricity).

## Water and Chemicals

Conventional route required chemicals in the form of  $Na_2CO_3$  for quartz heating process (Reaction 2). It also used HCl for precipitating silica from  $Na_2O \cdot SiO_2$  (Reaction 3). Fume route employed  $Cl_2$  gas and carbon (char) as chemicals to produce  $SiCl_4$ , which was then heated in plasma electric arc furnace. In silica production from rice husk, NaOH is applied for extraction with the amount of 6 L/kg rice husk ash [35, 46, 47].

In addition, HCl was also employed for acid leaching and precipitation. The HCl amount for acid leaching was 5 L/kg

of rice husk ash [46, 55] while HCl amount for precipitation was determined from reactions 6–7 by stoichiometric calculation. Water was subjected to dissolve acid and base. For conventional and fume routes, water was employed for sandstone washing with a total amount of 6 L/kg of sandstone.

### Energy Demand During Process

The energy demand for quartz heating follows Eq. 8.

$$E_{Hq} = P_F \cdot m_S \cdot C_{p_q} \cdot (T_f - T_0) \quad (8)$$

where  $E_{Hq}$  is energy demand for quartz heating [kJ],  $P_F$  is proportion of K-feldspar in sandstone [%],  $m_S$  is sandstone amount [kg],  $C_{p_q}$  is specific heat capacity of sandstone [0.74 kJ/kg/°C] [56],  $T_f$  is final heating temperature [1800 °C for conventional route and 3000 °C for fume route], and  $T_0$  is initial sandstone temperature [25 °C].

The auxiliary process for silica production encompassed material size reduction (grinding), drying, acid leaching, extraction, and refining [35, 47]. Quarried sandstone should be ground to ease the further process whereas straw-free rice was held to separate rice husk from rice grain. The specific grinding energy for straw-free rice was 107.7–112.7 kJ/kg [57] and the other stated at 160.6 kJ/kg [58], so the average value was 127.0 kJ/kg. The energy demand for straw-free rice grinding can be expressed in Eq. 9a.

$$E_{GR} = SE_{GR} \times \left( \frac{m_{RH}}{P_{RH}} \right) \quad (9a)$$

In order to calculate the energy for sandstone grinding, a variable named working index should be addressed. It is gross energy involved to comminute materials until 80% of the material amount can pass to 100 microns of sieve [59]. The sandstone working index is 11.53 while straw-free rice has working index which was similar to calcined clay (1.43) due to its softness and brittleness [59]. The ground sandstone particle size was similar to the ground straw-free rice particle, so the required energy for sandstone grinding is written in Eq. 9b.

$$E_{GS} = \left( \frac{WIS}{WIR} \right) \times SE_{GR} \times m_S \quad (9b)$$

where  $E_{GR}$  is grinding energy of straw-free rice [kJ],  $SE_{GR}$  is average specific grinding of straw-free rice [kJ/kg],  $m_{RH}$  is rice husk amount [kg],  $P_{RH}$  is average proportion of rice husk from straw-free rice [20%],  $E_{GS}$  is grinding energy of sandstone [kJ],  $WIS$  is sandstone working index [kJ/kg],  $WIR$  is straw-free rice working index [kJ/kg], and  $m_S$  is sandstone amount [kg].

Gel drying was conducted at 105 °C for 6 h in a Heratherm Thermo-scientific oven with 0.3 kW of power

consumption and 70% process efficiency [57]. Several processes such as 1 h of acid leaching and 2 h of extraction were held using a Thermo-scientific Cimarec hotplate stirrer which has 0.6 kW of power consumption. The process efficiency of this hotplate was stated at 50% due to significant heat dissipation to the environment. Silica refining (optional process) was held for 2 h in a box-type resistance 5-12D model of furnace which has a dimension of 30 × 20 × 12 cm, power consumption of 5 kW, and process efficiency of 90% as its better insulation [60, 61]. The energy in the auxiliary process was stated in Eqs. 10a–d.

$$E_{Aux,c} = E_{GS} + E_D \quad (10a)$$

$$E_{Aux,f} = E_{GS} \quad (10b)$$

$$E_{Aux,b} = E_{GR} + E_k = E_{GR} + E_D + E_{AL} + E_E + E_R \quad (10c)$$

$$E_k = \sum_{k=D}^{AL,E,R} \left( \frac{PC_k \cdot t_k}{\eta_k} \right) \quad (10d)$$

where  $E_{Aux,i}$  is energy involved in auxiliary processes in conventional (c) / fume (f) / biomass thermochemical conversion route (b) [kJ],  $E_D$  is energy for gel drying [kJ],  $E_{AL}$  is energy for acid leaching [kJ],  $E_E$  is energy for extraction [kJ], and  $E_R$  is energy for refining [kJ],  $PC$  is power consumption of process  $k$  [W],  $t$  is duration of process  $k$  [s],  $\eta$  is efficiency of process  $k$  [%], and  $k$  is dummy variable for process  $D, AL, E$ , and  $R$ .

### Fuel Amount and Properties

The required energy for conventional and fume routes was supplied from sub-bituminous coal due to wide and abroad abundance in Indonesia [62, 63]. It was converted into electricity with combustion efficiency of 85%, Rankine cycle efficiency of 30%, and generator mechanical efficiency of 90% [29, 56, 64]. During handling and preparation, coal is presumed to have mass loss percentage of 10% [65]. The required coal is accordingly determined as in Eqs. 11a–c.

$$m_{coal} = \frac{E}{(1 - m_L) \times \eta_c \times \eta_{RC} \times \eta_g \times HV_{coal}} \quad (11a)$$

$$E = E_{Hq} + E_{Aux,c}, \text{ for conventional route} \quad (11b)$$

$$E = E_{Hq} + E_{Aux,f}, \text{ for fume route} \quad (11c)$$

In the meantime, the required energy for biomass thermochemical conversion route was supplied by rice husk itself. Rice husk was combusted using 100% of excess air and the generated energy was calculated following the procedure

**Table 1** Rice husk and sub-bituminous coal properties on dry-ash-free basis

Properties	Units	Coal [62]	Rice husk [29]
C	w/w%	80.78	42.33
H	w/w%	5.85	6.90
O	w/w%	8.08	50.08
N	w/w%	1.67	0.46
S	w/w%	3.62	0.23
Heating value	kJ/kg	30,820	13,030

from Steven et al. [29]. The thermal energy was then converted to small-scale electricity by organic Rankine cycle apparatus with thermal efficiency of 10% and generator mechanical efficiency of 90% [55, 66–68]. If there is a deficit, the remained electricity needs are then covered by coal (Eq. 11d). The properties of rice husk and sub-bituminous coal are summarized in Table 1.

$$E = (E_{CR} \times \eta_{ORC} \times \eta_g) - E_{Aux,b}, \text{ for biomass route} \quad (11d)$$

where  $m_{coal}$  is required coal amount [kg],  $E$  is required energy which was fulfilled from coal [kJ],  $m_L$  is mass loss percentage of coal during handling and preparation [%],  $\eta_c$  is coal combustion efficiency [%],  $\eta_{RC}$  is Rankine cycle efficiency [%],  $\eta_g$  is generator mechanical efficiency [%],  $HV_{coal}$  is coal heating value [kJ/kg],  $E_{CR}$  is generated energy from rice husk combustion using 100% excess air [kJ], and  $\eta_{ORC}$  is organic Rankine cycle efficiency [%].

**Output Stream Impact**

All silica production from three routes releases output streams that contain wastewater and gas emissions. Wastewater had a composition of water, liquid chemicals in output reaction side, and impurities from sandstone washing. Gas emission from three routes was dominated by CO<sub>2</sub>, NO<sub>x</sub> (a mixture of NO, NO<sub>2</sub>, and N<sub>2</sub>O), and SO<sub>2</sub> [29, 69]. The flue gas amount was preliminary calculated by mass balance where C, N, and S elements from each fuel ultimate analysis undergo complete combustion, thus no CO emission was detected. The calculation from mass balance is then corrected by the thermodynamics conversion ( $X_{eq}$ ) which is derived from the equilibrium constant ( $K_{eq}$ ).

The  $K_{eq}$  value is obtained from Eq. 12 where  $X_{eq}$  is determined from Eq. 13 [56].

$$\prod (y_i)^{v_i} = K_{eq} = \exp \left[ \frac{-\Delta G_0^o}{RT_0} + \frac{-\Delta H_0^o}{RT_0} \left( 1 - \frac{T_0}{T} \right) + \Delta A \left( \ln \frac{T}{T_0} - \frac{T - T_0}{T} \right) + \frac{1}{2} \Delta B \frac{(T - T_0)^2}{T} + \frac{1}{2} \Delta C \frac{(T - T_0)^2}{T^2 T_0^2} \right] \quad (12)$$

**Table 2**  $\Delta G_0^o$ ,  $\Delta H_0^o$ ,  $A$ ,  $B$ , and  $C$  values for several elements and compounds [56]

Element/compound	$\Delta G_0^o$ (kJ/mol)	$\Delta H_0^o$ (kJ/mol)	A	B ( $\times 10^3$ )	C ( $\times 10^{-5}$ )
C	0	0	1.771	0.771	-0.867
H <sub>2</sub>	0	0	3.249	0.422	0.083
O <sub>2</sub>	0	0	3.639	0.506	-0.227
N <sub>2</sub>	0	0	3.280	0.629	0.014
S	0	0	4.114	-1.730	-0.783
CO	-137,169	-110,525	3.376	0.557	-0.031
CO <sub>2</sub>	-394,359	-393,509	5.457	1.045	-1.157
SO <sub>2</sub>	-300,194	-296,830	5.699	0.801	-1.015
NO	86,550	90,250	3.387	0.629	0.014
NO <sub>2</sub>	51,310	33,180	4.982	1.195	-0.792
N <sub>2</sub> O	104,200	82,050	5.328	1.214	-0.928

$$X_{eq} = \left( 1 - \frac{n}{n_0} \right) \times 100\% = \left( 1 - \frac{y_i n_t}{n_0} \right) \times 100\% \quad (13)$$

where  $y_i$  is fraction composition of  $i$  compound in equilibrium,  $v_i$  is reaction coefficient of  $i$  compound,  $-\Delta G_0^o$  is standard Gibbs free energy of formation [kJ/mol],  $-\Delta H_0^o$  is standard enthalpy of formation [kJ/mol],  $A$ ,  $B$ , and  $C$  are constants for heat capacity,  $T_0$  is 25 °C,  $T$  is reaction temperature [°C],  $n_0$  is initial amount of  $i$  compound [mol],  $n$  is equilibrium amount of  $i$  compound [mol], and  $n_t$  is total outlet amount of  $i$  compound [mol]. The value of  $\Delta G_0^o$ ,  $\Delta H_0^o$ ,  $A$ ,  $B$ , and  $C$  are listed in Table 2.

Both CO<sub>2</sub> and NO<sub>x</sub> are candidate compounds that cause global warming where NO<sub>x</sub> is 310-fold more risky than CO<sub>2</sub>, while SO<sub>2</sub> in a large amount could cause acid rain [10, 70, 71]. The CO<sub>2</sub> and NO<sub>x</sub> emissions are determined as CO<sub>2</sub>-equivalent as written in Eq. 14. It is calculated from element balance but for biomass, there is a correction factor where CO<sub>2</sub> emission biomass combustion is reabsorbed by biomass itself with an efficiency of 98.62% [22]. Other than that, the amount of released SO<sub>2</sub> was served as kg SO<sub>2</sub>/kg fuel combusted, Eq. 15.

$$CO_2 - eq = \frac{(m_{CO_2} \cdot (1 - \xi)) + (310 \cdot X_{eq,NOx} \cdot m_{NOx})}{1000} \quad (14)$$

$$SO_2 \left[ \frac{kg}{kg\text{fuel}} \right] = \frac{m_{SO_2}}{m_{fuel}} \quad (15)$$



where  $CO_2 - eq$  is  $CO_2$ -equivalent amount [tonne],  $m_{CO_2}$  is emitted  $CO_2$  amount [kg],  $\xi$  is efficiency of emitted  $CO_2$  reabsorption [0% for coal; 98.62% for biomass],  $X_{eq,NO_x}$  is thermodynamics conversion of  $NO_x$  formation at a certain process temperature,  $m_{NO_x}$  is emitted  $NO_x$  amount [kg],  $m_{SO_2}$  is emitted  $SO_2$  amount [kg], and  $m_{fuel}$  is fuel amount (coal for conventional and fume routes; rice husk for biomass thermochemical conversion route) [kg].

## Results and Discussion

### Silica Production Concept from Conventional, Fume, and Biomass Thermochemical Conversion Routes

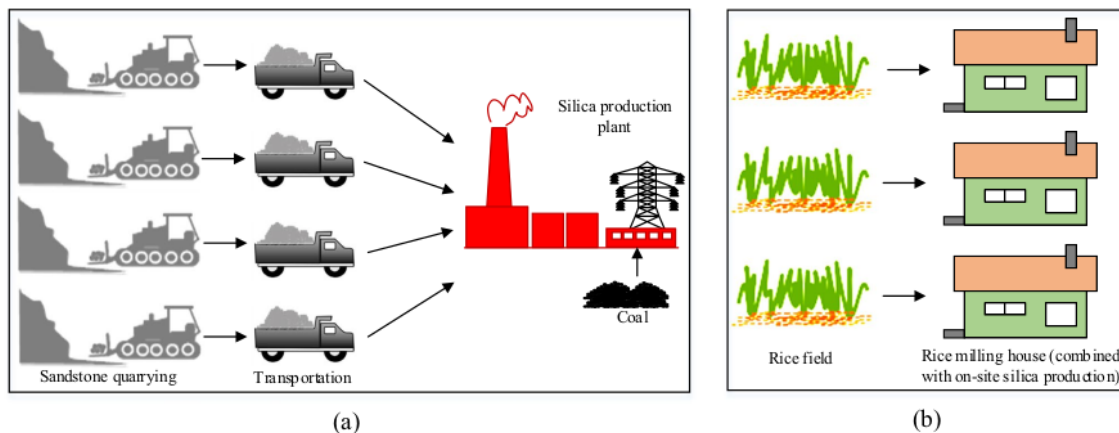
Silica production concept from conventional and fume routes is centralized. Quarried sandstone from 4 different local quarry sites in Subang is transported to the centralized silica production plant which is located in Cilegon. The energy requirement for silica production plant is fully generated from coal combustion. On the other hand, silica from biomass thermochemical conversion route is produced in the rice milling house. It combines the on-site silica production in the rice milling house which is known as decentralized production concept. Decentralized production does not require transportation of raw materials and this becomes an advantage for biomass thermochemical conversion routes. It should be noted that the transportation of agricultural biomass is disadvantageous in terms of its low bulk density [9].

According to Indonesian Central Bureau of Statistics, Indonesia has rice fields with total areas of 10.61 million Ha and total production of straw-free rice of 55.67 million

tonnes in 2022 [72]. It can be said that the annual productivity of Indonesian straw-free rice is equal to 5.25 tonnes/Ha. From the simple calculation, as much as 6.56 tonnes of rice husk is required to produce 1-tonne silica [Recall in Sect. 2.2: ash content in rice husk = 20.44w/w%, silica content in ash = 87.69w/w%, and extraction yield = 85%]. The processing of 6.56 tonnes of rice husk accordingly requires 32.8 tonnes of straw-free rice.

In other words, 1 tonne of on-site silica production can be realized and managed in 6–7 rice milling houses per 1 Ha of rice field area. If rice plants are presumed to have 3 harvesting periods in a year, following decentralized production concept, one rice milling house takes account of 5.25 tonnes of straw-free rice per year or 1.75 tonnes of straw-free rice per harvesting period. This amount does not require the consumption of diesel oil in transportation (does not require transportation using a truck) because the transfer of 1.75 tonnes of straw-free rice can be carried out by all farmers therein considering the rice milling house location is close to the rice fields.

The harvested rice plant from the field is first separated from straw, husk, and bran. In the rice milling house, straw-free rice is ground to acquire rice husk. Rice husk is then directly processed on-site through combustion and silica extraction from rice husk ash. The energy requirement for grinding, drying, acid leaching, extraction, and refining processes is fulfilled from rice husk combustion. Coal is used as additional energy supply only when the electricity generated from rice husk combustion has not been completely covered yet. Figure 4 illustrates the comparison between centralized and decentralized silica production concepts.



**Fig. 4** Illustration of silica production through centralized (a) and decentralized (b) concepts

### Raw Material, Transportation, and Utility Systems Analysis

The basis of material requirements for conventional and fume routes are determined by mass balance involving Reactions 1–5. Based on the calculation,  $\text{Na}_2\text{CO}_3$ , HCl, and sandstone amounts to produce 1 tonne of silica from conventional route are 1.76, 1.52, and 3.86 tonnes, respectively. This process also needs 23.16 tonnes of water and 7600.69 MJ of energy from 1194.08 kg of coal combustion. This amount of energy has already included sandstone grinding and gel drying, which occupies 3951.89 and 0.93 MJ. In conventional route, the overall yield of silica from sandstone is 25.91%.

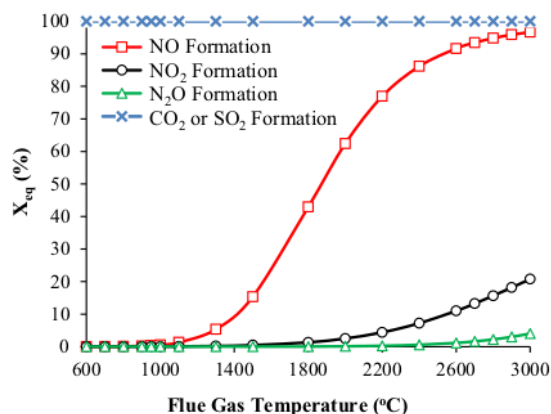
Similar to conventional route, silica production from fume route requires 3.86 tonnes of sandstone (overall yield of silica 25.91%). The chemicals involved are 1.18 tonnes of  $\text{Cl}_2$  gas and 0.40 tonnes of char as a carbon source. The water utilization in fume route is 23.16 tonnes. The calculation also informs that fume route is energy-consuming. Approximately 12,444.15 MJ of energy should be prepared, including 3951.89 MJ for sandstone grinding. It is generated from 1954.99 kg of coal combustion. The amount of sandstone grinding in conventional or fume route is equal to 1-h grinding in jaw crusher with power consumption of 1.1 MW. This value is in the range of industrial small jaw crusher equipment with power consumption that ranges from 0.75–7.46 MW [59].

Silica production from rice husk utilizes 6.56 tonnes of rice husk, 0.32 tonnes of NaOH, 0.54 tonnes of HCl, and 22.81 tonnes of water to obtain 1 tonne of silica. The overall yield of silica from rice husk following this route is 15.24%. The required energy includes 4167.96 MJ for grinding and 62.22 MJ for the auxiliary process. In the meantime, the combustion of 6.56 tonnes of rice husk only produces 2712.79 MJ of energy. Hence, biomass thermochemical conversion route needs coal to cover the deficit but with a far lower amount from conventional and fume routes, 238.38 kg.

Conventional and fume routes both still adopt the centralized production concept. The transportation from quarry sites to the silica production plant, in consequence, occurs. The carbon footprint from transportation through conventional and fume routes is recorded in the same amount, 0.88 tonnes of  $\text{CO}_2$ -equivalent, which is produced from the internal combustion of 283.33 kg of diesel oil. In biomass thermochemical conversion route, raw material is not needed for transportation as conventional and fume routes do because silica is produced on-site in the rice milling house.

**Table 3** The equilibrium constant ( $K_{eq}$ ) for emitted gas at various flue gas temperatures

Gas	$K_{eq}$				
	700 °C	1100 °C	1500 °C	2000 °C	3000 °C
$\text{CO}_2$	$4.25 \times 10^{21}$	$5.30 \times 10^{15}$	$3.76 \times 10^{12}$	$1.93 \times 10^{10}$	$1.87 \times 10^9$
NO	$3.88 \times 10^{-4}$	$2.67 \times 10^{-2}$	$3.62 \times 10^{-1}$	$3.32 \times 10^0$	$5.85 \times 10^1$
$\text{NO}_2$	$1.35 \times 10^{-4}$	$1.87 \times 10^{-3}$	$1.23 \times 10^{-2}$	$7.51 \times 10^{-2}$	$1.13 \times 10^0$
$\text{N}_2\text{O}$	$7.27 \times 10^{-8}$	$6.36 \times 10^{-6}$	$1.17 \times 10^{-4}$	$1.60 \times 10^{-3}$	$5.97 \times 10^{-2}$
$\text{SO}_2$	$4.96 \times 10^{17}$	$5.22 \times 10^{13}$	$5.37 \times 10^{11}$	$2.51 \times 10^{10}$	$1.89 \times 10^9$



**Fig. 5** Thermodynamics conversion ( $X_{eq}$ ) for gas emission at various flue gas temperatures

### Gas Emission Amount and Environmental Impact Analysis

The output streams from this study contain wastewater and gas emissions that impact the environment. The calculation of gas emission amount is first obtained from the mass balance which is corrected with thermodynamics conversion at the temperature at which the gas is emitted. The coal combustion is targeted until 1800 °C for conventional route or 3000 °C for fume route to supply energy for quartz heating. Under this temperature, the equilibrium constant for  $\text{CO}_2$ , NO,  $\text{NO}_2$ ,  $\text{N}_2\text{O}$ , and  $\text{SO}_2$  are tabulated in Table 3. The  $K_{eq}$  results show that  $\text{CO}_2$ , as well as  $\text{SO}_2$ , is formed very spontaneously at all emission temperatures. Contrary,  $\text{NO}_x$  might form at extreme temperatures through a reversible reaction.

The tremendous  $K_{eq}$  values of  $\text{CO}_2$  and  $\text{SO}_2$  lead to nearly 100% of their thermodynamics conversion ( $X_{eq} \approx 100\%$ ).

It can be said that C and S elements from the fuel ultimate analysis produce  $\text{CO}_2$  and  $\text{SO}_2$  spontaneously. In contrast, the N element from fuel partially produces  $\text{NO}_x$  with  $X_{eq}$  lower than 100%. Figure 5 plots the  $X_{eq}$  result for  $\text{CO}_2$ ,  $\text{SO}_2$ ,  $\text{NO}$ ,  $\text{NO}_2$ , and  $\text{N}_2\text{O}$  respectively. From Table 3 and Fig. 5, the  $K_{eq}$  and  $X_{eq}$  for  $\text{NO}_2$  and  $\text{N}_2\text{O}$  are found in a lower amount compared to  $\text{NO}$ , therefore  $\text{NO}_x$  can be represented by only  $\text{NO}$  compound. Figure 5 also informs that 42.93% of N element from coal is converted to  $\text{NO}$  under combustion at  $1800^\circ\text{C}$  (conventional route) and the value escalates to 96.69% when emitted at  $3000^\circ\text{C}$  (fume route). In the meantime, the thermodynamics conversion of N element from rice husk to form  $\text{NO}$  is only 0.069% at  $700^\circ\text{C}$ . This calculation proves that rice husk combustion temperature at  $700^\circ\text{C}$  is able to suppress  $\text{NO}_x$  emission as well as prevent silica crystalline formation in rice husk ash [73, 74].

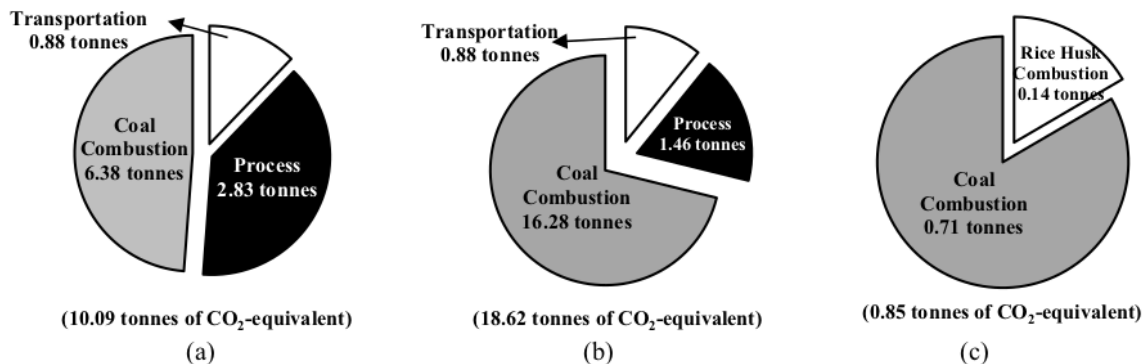
It must be admitted that the process involving fossil fuels should be reduced [75–77]. The emission of  $\text{SO}_2$  is particularly important for Indonesia since an extensive utilization of coal, which has high sulfur content [63]. Likewise,  $\text{NO}_x$  emission from fossil fuel combustion occurs because of its high process temperature [21]. Both could harm the ecosystem.  $\text{SO}_2$  formed in the atmosphere provokes corrosive acid rain which can damage the environment.  $\text{NO}_x$  also has a global warming potential of 310 times greater than  $\text{CO}_2$  [10].

Coal and other fossil fuels contribute to tremendous  $\text{CO}_2$  emissions. Unlike them, emitted  $\text{CO}_2$  from biomass combustion is reabsorbed by itself for photosynthesis [78]. However, it cannot be simply stated as zero carbon emission because several researchers report that biomass utilization is not fully carbon neutral as long as it is not combined with carbon capture process [23, 78, 79]. Chungsangunsit et al. found that  $\text{CO}_2$  reabsorption efficiency is 98.62% [22] so the rest amount becomes emission. Rice husk combustion temperature, heating value, and C, N, and S contents are also all lower than coal [45, 80]. All those facts cause rice

husk combustion to release lower emissions of  $\text{CO}_2$ ,  $\text{NO}_x$ , and  $\text{SO}_2$ .

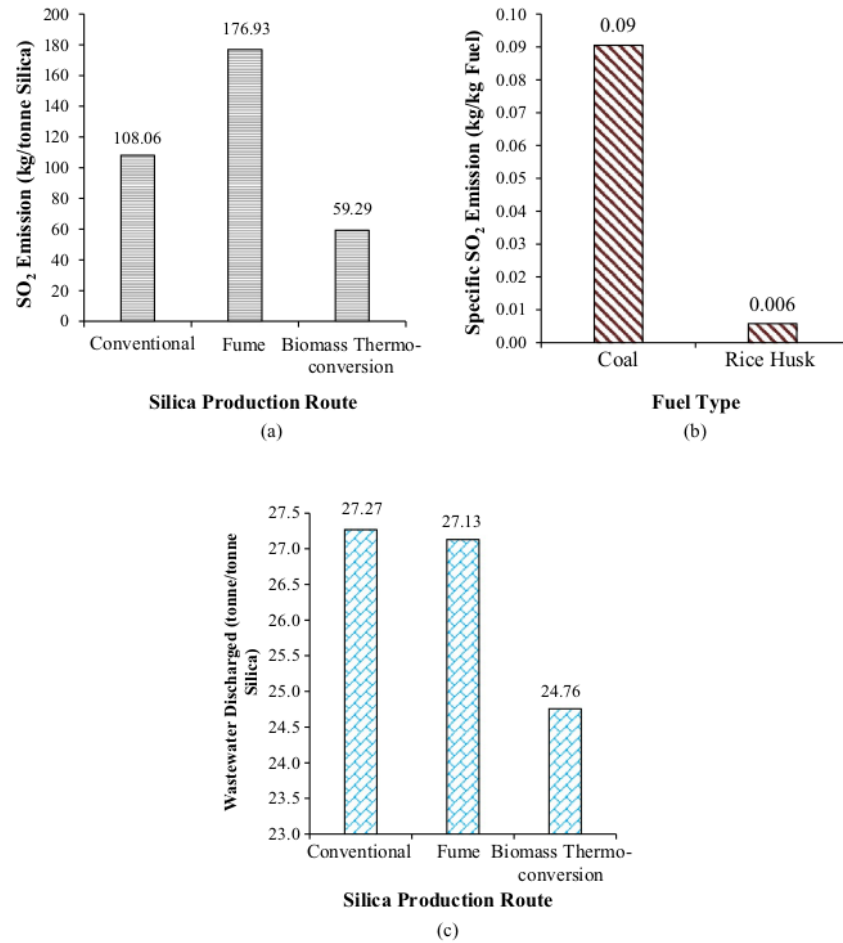
According to the environmental impact analysis, gas emission is produced from the main production process, coal and/or rice husk combustion, and transportation which is sourced from diesel oil combustion. The emission in terms of  $\text{CO}_2$ -equivalent for conventional, fume, and biomass thermochemical conversion routes is 10.09, 18.62, and 0.85 tonnes. The amount of 10.09 and 18.62 tonnes  $\text{CO}_2$ -equivalent in conventional and fume routes entirely come from coal combustion. From 0.85 tonnes of  $\text{CO}_2$ -equivalent in biomass thermochemical route, as much as 0.71 tonnes is from coal combustion and only 0.14 tonnes is from rice husk combustion. Figure 6 is the pie chart for equivalent  $\text{CO}_2$  emission amount from three routes. Transportation occupies 0.88 tonnes of  $\text{CO}_2$  for both conventional and fume routes.

Sulfur content that exists in both coal and rice husk will lead to  $\text{SO}_2$  formation. Figure 7a describes that to produce 1 tonne of silica, conventional route produces 108.06 kg of  $\text{SO}_2$  to the atmosphere (from 1194.08 kg of coal combustion). Fume route emits 176.93 kg of  $\text{SO}_2$  (from 1954.99 kg of coal combustion). As much as 59.29 kg of  $\text{SO}_2$  is also emitted from biomass thermochemical conversion route where 6.56 tonnes of rice husk combustion contributes to 37.72 kg of  $\text{SO}_2$  emission and 238.38 kg of coal combustion contributes to 21.57 kg of  $\text{SO}_2$  emission. In other words, it can be recalculated that the specific  $\text{SO}_2$  emission amount is 0.09 kg/kg for coal combustion and only 0.006 kg/kg for rice husk combustion as communicated in Fig. 7b. Furthermore, as much as 24.76 tonnes of wastewater is discharged from biomass thermochemical conversion route which consists of impurities leachate and rinsed chemicals. Conventional and fume routes discharge 27.27 and 27.13 tonnes of wastewater, Fig. 7c. Before being discharged into the environment, wastewater is desired to be treated through demineralization until reaches the effluent standard [81, 82].



**Fig. 6** Equivalent  $\text{CO}_2$  emission amount of conventional (a), fume (b), and biomass thermochemical conversion (c) routes

**Fig. 7** SO<sub>2</sub> emission amount from three routes (a); specific SO<sub>2</sub> emission from coal or rice husk (b); and wastewater discharged amount (b) from three routes



### Overall M&E I/O Summary

Actually, the silica production from conventional, fume, and biomass thermochemical conversion routes provides various options with their own characteristics in terms of raw material requirement, energy consumption, and environmental impact. The detailed results of M&E I/O above inform the overall and component balances as well as energy balance for both input and output streams in all routes. In summary, the production of 1-tonne silica through conventional and fume routes has energy demands of 1.80 and 2.94 times higher than biomass thermochemical conversion route.

Still, conventional and fume routes have a greater global warming potency than biomass thermochemical conversion route, with a factor of 11.81 and 21.80, successively. Furthermore, SO<sub>2</sub> specific emission from coal combustion is 15.75-fold more significant than rice husk. Because all routes produce a significant amount of wastewater, chemical

treatment is desired to be performed by demineralization to fulfill the effluent quality regulation as well as save the chemicals cost.

Despite the enormous energy requirement and highest negative environmental effects in fume route, it can directly produce silica into a small spherical particle, whereas other routes do not have this added value. In both conventional and biomass thermochemical conversion routes, silica is produced by precipitating sodium silicate with acid until it forms silica gel. The common morphology of this silica particle is prism shape with irregular, non-uniform, non-homogeneous, and unlaminated characteristics [35, 47]. The small spherical shape of particle is difficult to achieve since the particle formation is not easy to be controlled [83]. To obtain small and spherical shape, further treatment such as redispersing the particle in ethanol can be an attractive option [84].

**Table 4** Summary of M&E I/O performance of conventional, fume, and biomass thermochemical conversion routes

Parameters	Units	Conventional route	Fume route	Biomass thermochemical conversion route
Silica produced amount	Tonne	1	1	1
Product characteristics:				
- Silica phase	–	Crystalline	Crystalline	Amorphous
- Particle morphology	–	Irregular prism	Spherical	Irregular prism
- Purity	% SiO <sub>2</sub>	97–99	96–98	95–97
Production concept	–	Centralized	Centralized	Decentralized
Raw material	–	Sandstone	Sandstone	Rice husk
Process temperature	°C	1800	3000	700
Raw material amount	Tonnes	3.86	3.86	6.56
Overall yield of silica	%	25.91	25.91	15.24
Chemicals amount	Tonnes	3.28	1.58	0.86
- Na <sub>2</sub> CO <sub>3</sub>	Tonnes	1.76	–	–
- HCl	Tonnes	1.52	–	0.54
- Cl <sub>2</sub> gas	Tonnes	–	1.18	–
- Char (carbon)	Tonnes	–	0.40	–
- NaOH	Tonnes	–	–	0.32
Required water amount	Tonnes	23.16	23.16	22.81
Total energy involved	MJ	7600.69	12,444.15	4230.18
$E_{Hq}$	MJ	3639.54	8492.26	–
$E_{GS}$	MJ	3951.89	3951.89	–
$E_{GR}$	MJ	–	–	4167.96
$E_D$	MJ	9.26	–	9.26
$E_{At}$	MJ	–	–	4.32
$E_E$	MJ	–	–	8.64
$E_R$	MJ	–	–	40.00
Fuel amount	kg	1194.08 (Coal)	1954.99 (Coal)	6560 (Rice husk) 238.38 (Coal)
Energy released from fuel	MJ	7600.69	12,444.15	2712.79 (Rice husk) 1517.39 (Coal)
Equivalent CO <sub>2</sub> emission	Tonnes	10.09	18.62	0.85
- CO <sub>2</sub> emission from transportation	Tonnes	0.88	0.88	–
- CO <sub>2</sub> emission from process reaction	Tonnes	2.83	1.46	–
- CO <sub>2</sub> emission from coal combustion	Tonnes	3.54	5.79	0.71
- CO <sub>2</sub> emission from rice husk combustion	Tonnes	–	–	0.14
- CO <sub>2</sub> from NO <sub>x</sub> emission from coal combustion	Tonnes	2.84	10.84	0.0009
- CO <sub>2</sub> from NO <sub>x</sub> emission from rice husk combustion	Tonnes	–	–	0.007
SO <sub>2</sub> emission	kg	108.06	176.93	59.29
- SO <sub>2</sub> emission from coal combustion	kg	108.06	176.93	21.57
- SO <sub>2</sub> emission from rice husk combustion	kg	–	–	37.72
- Specific SO <sub>2</sub> emission	kg/kg fuel	0.09	0.09	0.006
Wastewater discharge	Tonnes	27.27	27.13	24.76
- Water	Tonnes	23.16	23.16	22.81
- Impurities	Tonnes	1.54	1.54	–
- NaCl	Tonnes	1.95	–	1.95
- HCl	Tonnes	–	2.43	–
- KCl	Tonnes	0.62	–	–

Yet, biomass thermochemical conversion route has also a superior point where it is believed to be able to produce amorphous silica due to its maximum process temperature not exceeding 700 °C [43]. Moreover, the product purity is no less different from conventional and fume routes. In contrast, conventional and fume routes have high possibility of transforming the silica phase to crystalline since the exposure at extreme temperatures (> 1200 °C) in their processing [55]. Although amorphous and crystalline silica has their own special uses, frequent exposure to crystalline silica can threaten human health [85]. The overall results comparison is outlined in Table 4.

## Conclusions and Recommendations

The M&E I/O performance for silica production from three different routes by considering transportation, utility requirement, process production, and environmental impact has been studied. The least environmental impacts of biomass thermochemical conversion route are also discussed. It is observed that CO<sub>2</sub> is the most influential flue gas produced from all routes, but the lowest amount is found in biomass thermochemical conversion route. NO<sub>x</sub> and SO<sub>2</sub> are also produced for rice husk combustion, but a greater amount is found in other routes that utilize full coal combustion for energy supply. Wastewater is also found in all three routes, which consists of water, raw material impurities, and rinsed chemicals. This study concluded and clarified that silica production from biomass thermochemical conversion route is the most sustainable. Moreover, there is no transportation needed and less coal requirement. Although the study performs M&E I/O of silica production, the results could be different between the Indonesian case and others in nature. The results depend on the different presumptions and diverse variability of internal country databases related to the raw material and detailed process. Nevertheless, this study helps to convince and highlight the sustainability of rice husk as a potential silica source.

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## Declarations

**Competing interest** There is no conflict of competing of interest to declare.

**Ethical Approval** This declaration is not applicable.

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