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RESEARCH PAPER

From rice husk waste to useful energy: Influence of pyrolysis on steam gasification

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Abstract

Rice husk gasification has been increasingly paid attention in rice-producing countries. Pyrolysis temperature and heating rate are the most important factors influencing char production and gasification process. However, these parameters have not yet been fully investigated. Moreover, the few existing studies were conducted at the microscopic scale, making it difficult to extrapolate results to the behavior of real particles in a gasifier. In this study, their effects on gasification with steam at a particle scale, in operating conditions relevant to those that exist in industrial gasifiers was conducted using a Macro-thermogravimetric reactor. Regarding pyrolysis, an increase in temperature from 600°C to 900°C slightly lowers rice husk char yield, but did not have much effect on char morphology. Effect of a small change in heating rate from 5°Cmin^{-1} to 20°Cmin^{-1} has not been observed, while a significant increase from 20°C min^{-1} to 1800°C min^{-1} decreases considerably char yield and had an effect on rice husk char characteristics and morphology. Regarding gasification, a char produced at a heating rate of 1800°C min^{-1} increased gasification kinetics by 1.4 times compared to that of 20°C min^{-1}. Results could be useful to set up industrial or academic codes, necessary for the conception of new rice husk gasifiers.

Key words: biomass, gasification, kinetics, rice husk.

Introduction

Rice is grown in more than 75 countries worldwide. In 2016, paddy production was estimated at 747 million tons. This results in approximately 149 million tons of rice husks, i.e. 0.2 tons of husks for each ton of paddy (Bhattacharyya 2014). While rice husks have traditionally been used in low-value applications such as direct burning for cooking, building material and animal husbandry, its potential as a fuel for efficient energy processes, such as gasification, has been attracting increasing attention in recent years. Biomass gasification is a thermochemical conversion process that converts biomass into a CO and H_2 rich-gas called syngas. Syngas can be used to produce heat, electricity or transport fuel. Different types of gasifiers including fixed bed, fluidized, entrained flow and multi-stage gasifiers are used (Balat 2009). Currently, wood gasifiers are the most common type of biomass gasification. Diversifying the feedstock is one of the biggest challenges for this technology, but also involves technical and economic problems. In some rice-producing countries such as China, India, Cambodia, and Thailand, the number of rice husk gasifiers has increased rapidly. However, the current systems show a poor efficiency and high rates of liquid waste emissions, and lacks of scientific knowledge on rice husk gasification are hindering the development of this technology (Nguyen, Ha-Duong, and Van de Steene 2015; Shackley et al. 2012; Laohalidanond, Chaiyawong, and Kerdsuwan 2015; Verbong et al. 2010; Zhou et al. 2012).

Gasification involves a series of processes: drying, pyrolysis, volatile oxidation/cracking, and char gasification. Char gasification is particularly important as it controls the production of syngas, complete carbon conversion and hence the efficiency of the whole process. Detailed knowledge of char gasification kinetics is thus essential for the selection of optimal gasification conditions, as well as for the design of new reactors or the improvement of existing ones. The main reactions involved during char gasification are the following:

$C + H_2O \rightarrow CO + H_2$ (Steam gasification) (1)
$C + CO_2 \rightarrow 2CO$ (Boudouard gasification) (2)

The kinetics of these reactions are known to be highly influenced by the pyrolysis temperature and pyrolysis heating rate, as these parameters define characteristics of the char that will undergo the gasification (Van de steene et al. 2011; Zhai et al. 2016; Xu et al. 2013). Only few studies have focused on kinetics of rice husk gasification with steam as the experimental setup requirements are much harder than Boudouard gasification. Rice husk char gasification in an H_2O atmosphere was investigated by Bhat et al. (Bhat, Ram Bheemarasetti, and Rajeswara Rao 2001) using particles or fine powder, but their results regarding reaction kinetics differed from other published results in the literature (Di Blasi 2009). Zhai et al. (Zhai et al. 2016) conducted pyrolysis experiments using rice husks in a tubular reactor and showed that an increase in the final pyrolysis temperature from 600°C to 1400°C significantly influenced porosity and surface area: the latter presenting a maximum value at 1000°C. A char produced at high temperature was shown to be less reactive to H_2O during gasification. A low pyrolysis heating rate ranging from 5°Cmin^{-1} to 20°Cmin^{-1} has been shown to have no effect on rice husk char gasification kinetics (Alvarez et al. 2015). Meanwhile, no reference for effect of high pyrolysis heating rate on gasification kinetics has been reported in the literature.

To sum up, the influence of pyrolysis temperature and pyrolysis heating rate on steam gasification of rice husk char have not yet been fully investigated. Moreover, the few existing studies were conducted at the microscopic scale, i.e. a few milligrams of powders were used in a thermogravimetric analyzer, making it difficult to extrapolate results to the behavior of real particles in a gasifier. In addition, the experimental results were validated without mentioning the applicability of the findings to other types of rice husks.

The objective of this study was thus to study the effects of pyrolysis temperature and pyrolysis heating rate on gasification with steam at a particle scale, in operating conditions relevant to those that exist in industrial gasifiers, and to verify if results from a specific rice husk could be applied for another types. Results obtained could help researchers or engineers aiming to optimize rice husk gasification using computational fluid dynamics models or in the design of new prototypes.

Material and Methods

Rice husk feedstock

Rice husk used for experiments was the popular "Te do", taken from the Red River Delta, Vietnam. Proximate and ultimate analysis were performed and results were given in Table 1.

Table 1: Proximate and ultimate analysis of "Te do" rice husk

V_{db}%	A_{db}%	FC_{db}%	C_{daf}%	H_{daf}%	O_{daf}%	N_{daf}%	HHV (MJ/kg)
64.6	16.4	19.0	41.1	5.9	52.6	0.4	15.1

V: Volatile, A: Ash, FC: Fixed carbon, HHV: higher heating value, db: dry basis, daf: dry ash-free basis.

Rice husk char preparation

For the purpose of the study, five different chars were produced by varying the heating rate (5°Cmin^{-1}, 20°Cmin^{-1}, and 1800°Cmin^{-1}) and the final temperature (600°C, 750°C and 900°C). About 400 mg of rice husk were placed in an air-tight refractory steel box of 25 cm diameter and 20 cm height. The box was swept with N_2 to avoid oxidation and placed in the muffle furnace. Note that this system allowed us to produce a big quantity of char that will be used for the char gasification study. The final temperature and heating rate of the furnace were controlled. For the char produced at a high heating rate, i.e. 1800°Cmin^{-1}, we used the macro-thermogravimetric reactor described below. Whatever the pyrolysis reactor used, the final temperature was maintained for one hour.

Experimental setup

A new macro-thermogravimetric reactor (Macro-TG) was designed and set up at University of Science and Technology of Hanoi to experiment char gasification kinetics (Figure 1). The reactor consisted of a ceramic tube, 111 cm in length, with an internal diameter of 7.5 cm (1), placed in an electrical furnace (2). Heating was ensured by three independently controlled heating zones, ensuring the temperature was uniform throughout the reactor. The reaction atmosphere was generated by a mixture of N_2 and H_2O in selected proportions. Each gas was controlled by mass flowmeters (M1, M2, and M3). The gas mixture was preheated in a 2 m long coiled tube (3) located in the upper heated part of the reactor.

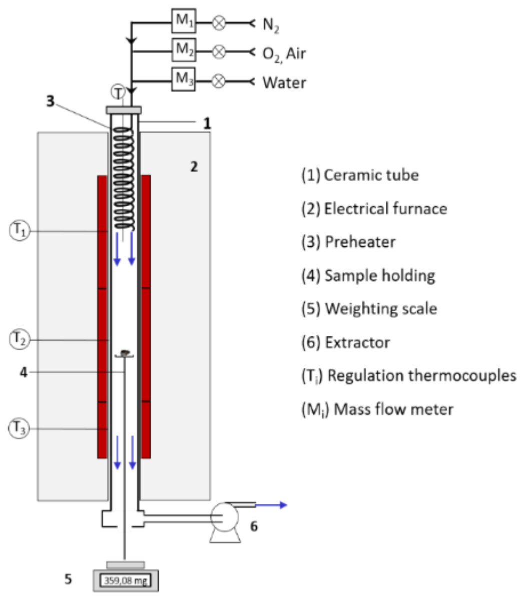


Figure 1: Description of the Macro-TG

The experiment consisted of gasifying rice husk char particles at atmospheric pressure, in a well-controlled atmosphere in terms of temperature and partial pressure of reacting gas. Sample mass was measured and recorded continuously (Figure 2). For each experiment, the reactor was first heated to the desired operating temperature. Then the sample holding (4) containing char particles was lifted from the bottom of the reactor to the desired position and maintained under N_2 to release all remaining volatile matter. When a constant mass was achieved, gas flows were established. As gasification took place, the mass of the char progressively decreased until constant mass - that corresponded to the ash content - was obtained.

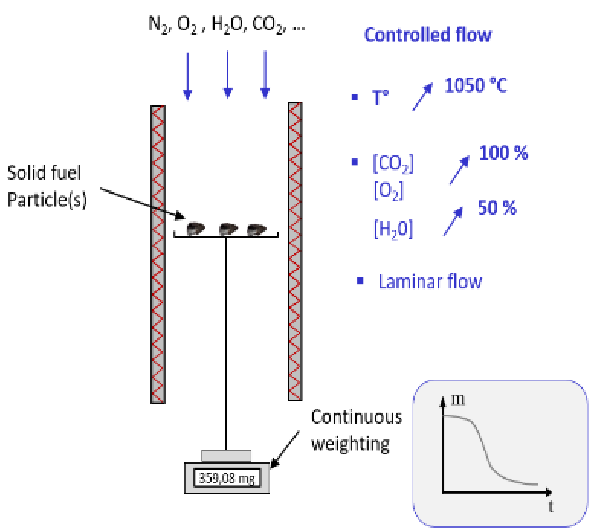


Figure 2: Principe of the Macro-TG

Rice husk char yield was determined as follow:

$$Char\ yield\ (\%) = \frac{m_{char}}{m_{rice\ husk}}\ x\ 100\% \qquad (3)$$

where $m_{rice\ husk}$ (g) and m_{char} (g) are respectively the initial mass of rice husk and the mass of char obtained after pyrolysis.

Energy transferred in the char (E) was calculated as follows:

$$E(\%) = \frac{HHV_{char}\ x\ Char\ yield}{HHV_{rice\ husk}}\ x\ 100\% \qquad (4)$$

where HHV_{char} (MJ/kg) and $HHV_{rice\ husk}$ (MJ/kg) are respectively the higher heating value of the char and the higher heating value of the rice husk.

Conversion X during gasification was calculated as follows:

$$X = \frac{m_i - m}{m_i - m_{ash}} \quad (5)$$

where m_i, m, and m_{ash} are respectively the initial mass, the mass at time t and the mass of ash.

Gas flow rate of 5Nlmin^{-1} and samples of 100 mg were chosen for all experiments. We previously checked that these values allowed gasification to take place in a chemical regime. A deviation of less than 10% in measurements was observed, which is acceptable considering the heterogeneity of rice husk and equipment accuracy (Mermoud et al. 2006). All data presented below were an average of at least 2 repeatability experiments.

Results and Discussions

Pyrolysis of rice husk

Influence of pyrolysis temperature

Pyrolysis experiments were performed (Table 2) considering 3 types of chars produced at different final temperatures of 600°C, 750°C and 900°C for this study. Results showed that rice husk char yield slightly decreased with an increase in final pyrolysis temperature, from 39.5% at 600°C to 36.6% at 900°C. Volatile matter from char also decreased from 7.3% to 2.9% when final pyrolysis temperature raised from 600°C to 900°C.

Table 2: Influence of final temperature (T_{final}) (heating rate of 20°Cmin^{-1})

T_{final} (°C)	V_{db}%	A_{db}%	FC_{db}%	HHV (MJ/kg)	Yield (%)	E (%)
600	7.3	37.8	54.9	19.0	39.5	49.7
750	5.5	38.3	56.2	19.2	38.1	48.4
900	2.9	39.9	57.2	19.3	36.6	46.8

In contrarily, final pyrolysis temperature did not affect much higher heating value (HHV) of the char. Besides, from the measurement of char yield and HHV, the ratio of energy transferred from the rice husk to the char has been calculated. It can be seen that about 47% of energy was transferred in the char after pyrolysis at 900°C. This value slightly increased at lower final temperature, i.e. 48.4% at 750°C and 49.7% at 600°C, as char yield increased accordingly. Results above showed that characteristics of rice husk char after pyrolysis stays relatively stable for high temperatures (above 500°C), which is consistent with the previous studies (Zhang, Liu, and Liu 2015; Al-Wabel et al. 2013).

Influence of pyrolysis heating rate

Pyrolysis experiments of rice husk were performed at heating rates of 5°Cmin^{-1}, 20°Cmin^{-1} and 1800°Cmin^{-1} to study the influence of pyrolysis heating rate to the rice husk char. A heating rate of 5°Cmin^{-1} can be found in the case that rice husk particles have little contacts with heat sources in a gasifier, while a heating rate of 20°Cmin^{-1} is close to typical values in an industrial process. High values of heating rate can be found in a fluidized bed, or in the case that a particle enters into contact with a heated wall (Mermoud et al. 2006).

Results showed that a small change in heating rate, i.e. from 5°Cmin^{-1} to 20°Cmin^{-1} mostly did not impact char yield, as well as other physical property of rice husk char.

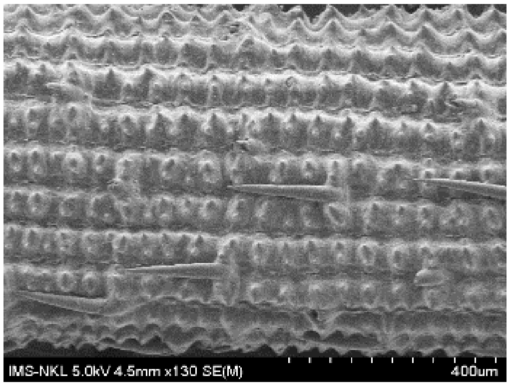
Table 3: Influence of heating rate (Final temperature: 900°C)

HR (°Cmin^{-1})	V_{db}%	A_{db}%	FC_{db}%	HHV (MJ/kg)	Yield (%)	E (%)
5	2.6	39.2	58.2	19.6	36.9	47.9
20	2.9	39.9	57.2	19.5	36.6	47.2
1800	1.9	46.5	51.6	16.3	29.7	32.0

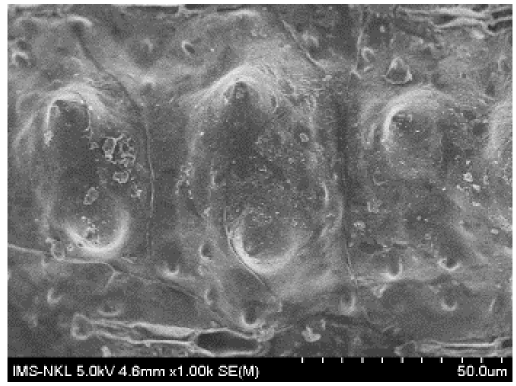
In contrarily, when heating rate was highly increased, i.e. from 20°Cmin^{-1} to 1800°Cmin^{-1}, char yield and volatile matter significantly decreased from 47.9% to 32.0%, and 2.9% to 1.9%, respectively. Higher heating value also significantly decreased, as a consequence of a decrease in fixed carbon and volatile matter of the char.

Morphology of rice husk and rice husk chars

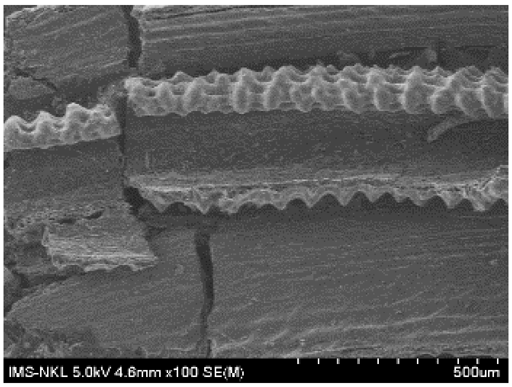
The scanning electron microscopy (SEM) of the surface of rice husk particle and its chars has been performed to observe the morphology of rice husk particle, and the change in morphology of its chars under different pyrolysis conditions. Figure 3a showed a high degree of roughness at the outer surface of rice husk. A large number of serrated peaks (Figure 3b) are placed in lines and are parallel to each other. Some pointed thorns are found on the surface and these are easy to be broken by thermal effects. The inner surface of the rice husk, in contrarily, is almost completely flat as shown in Figure 3c. A cross-section of a rice husk particle is presented in Figure 3d.



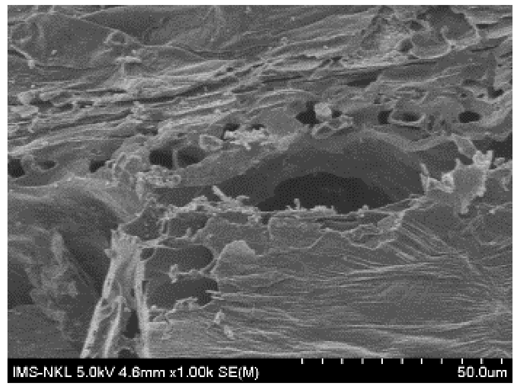
Outer surface



Serrated peaks



Inner surface



A cross-section

Figure 3: SEM of a rice husk particle

After the pyrolysis process, all volatiles have been released, however the surface structure remains similar compared to the original rice husk (Figure 4). The SEM of chars produced at different pyrolysis temperatures (Figure 4a and Figure 4c) showed that pyrolysis temperature has virtually no effect on the outer surface of rice husk. It is noteworthy that rice husk contains 15% to 22% of ash, most of which is SiO_2. Silicate systems have a marked effect on the stability of a rice husk when it undergoes thermal decomposition.

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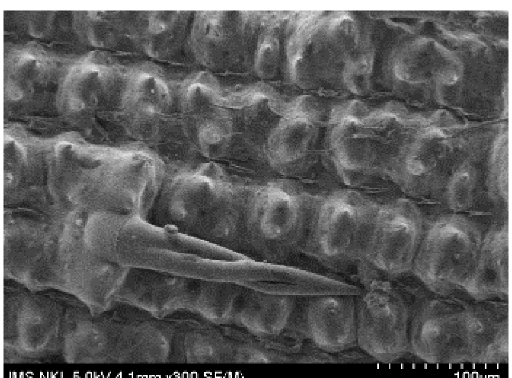
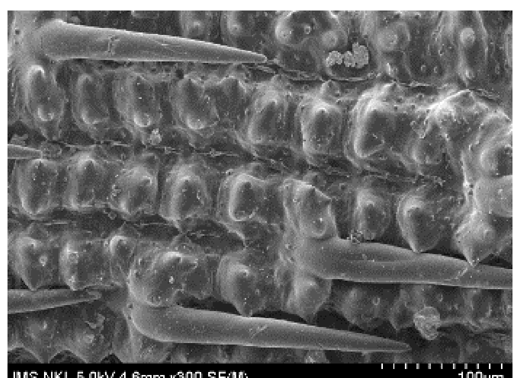
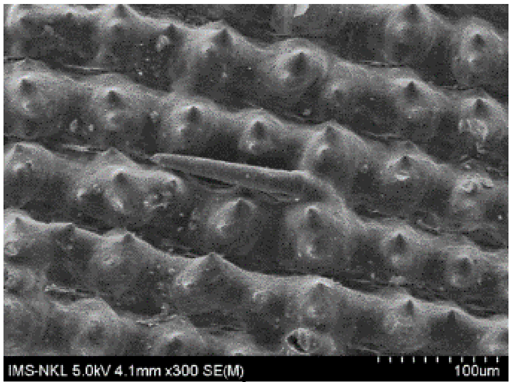


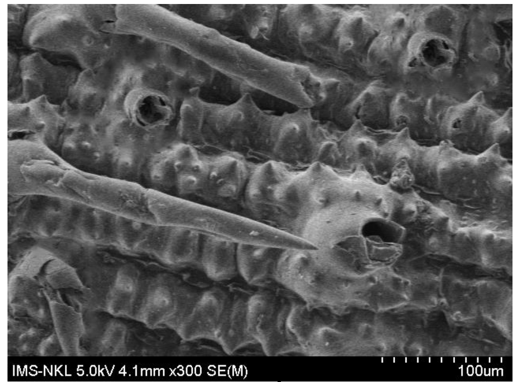
(a) 600°C, 20min^{-1} char



(b) 900°C, 5°Cmin^{-1} char



(c) 900°C, 20°Cmin^{-1} char



(d) 900°C, 1800°Cmin^{-1} char

Figure 4: SEM of chars produced under different pyrolysis conditions

A small change in the pyrolysis heating rate has no effect on char morphology as shown in Figure 4b and Figure 4c. In contrarily, the surface of rice husk char produced at 1800°Cmin^{-1} seems to be more damaged, expressed through the presence of some cracks on the outer surface of rice husk. This may presumably due to the sudden change of temperature during pyrolysis.

Steam gasification of rice husk

For further experiments of rice husk char gasification with steam, operating conditions that are close to those exist in current industrial processes were chosen: partial pressure of H_2O (in N_2): 0.2 atm and temperature of reacting gas: 900°C.

Influence of pyrolysis temperature

Chars produced at different pyrolysis temperatures (600°C, 750°C and 900°C) were gasified and kinetics were shown in Figure 5. Results clearly indicated that the pyrolysis temperature has no effect on the char conversion in an H_2O atmosphere, as conversion curves of 3 types of chars were identical and conversions were all complete after 3500s.

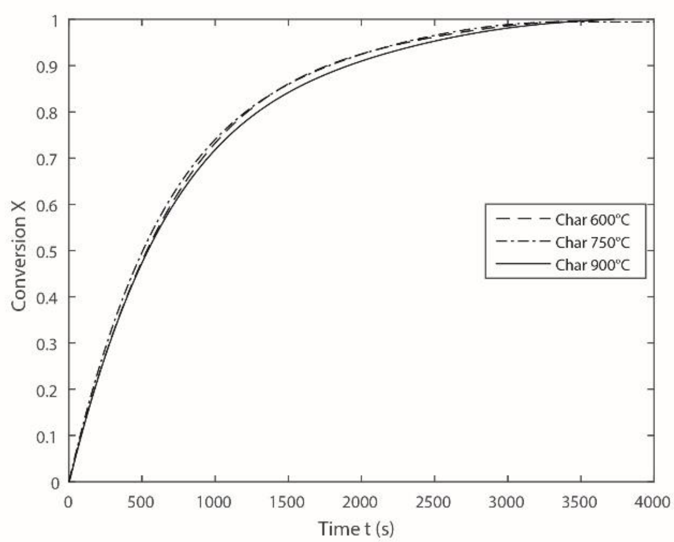


Figure 5: Influence of pyrolysis temperature on char conversion in an H_2O atmosphere (partial pressure: 0.2 atm, temperature: 900°C)

Influence of pyrolysis heating rate

Rice husk chars produced at different heating rate (5°Cmin^{-1}, 20°Cmin^{-1} and 1800°C.min^{-1}) were gasified and kinetics were shown in Figure 6. No difference in kinetics has been reported for chars produced at low heating rates of 5°Cmin^{-1} and 20°Cmin^{-1}, which is relevant to the literature (Alvarez et al. 2015).

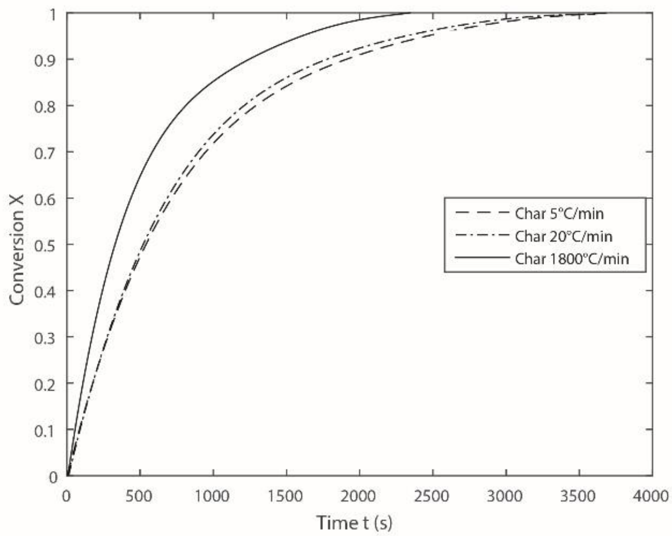


Figure 6: Influence of pyrolysis heating rate on char conversion in an H_2O atmosphere (partial pressure: 0.2 atm, temperature: 900°C)

However, a high pyrolysis heating rate accelerated the conversion rate of rice husk char gasification. For a char produced at 20°Cmin^{-1}, gasification was complete after 3500s, whereas for a char produced at 1800°Cmin^{-1}, gasification was complete after 2300s. Gasification rate was thus about 1.4 times faster with a char produced at 1800°Cmin^{-1} than a char produced at 20°Cmin^{-1}. The effect of heating rate on rice husk char gasification seems to be less important compared to results obtained with wood char gasification, where a much clearer difference in reaction rates was found between chars produced at different heating rates of 2.6°Cmin^{-1}, 12°Cmin^{-1} and 900°Cmin^{-1} (Mermoud et al. 2006).

Influence of nature of rice husks

The nature of rice husks can differ considerably from one variety to another. Dupont et al. (Dupont et al. 2016) reported that the silicon content of a biomass can influence the conversion rate in a gasification process. In the case of rice husks, this element can be tracked by measuring ash content, as it contains from 90% to 97% of SiO2 (Issagulov et al. 2014). This is of importance thus to investigate how the nature of the rice husk char influences the conversion. For this purpose, we chose eight varieties of rice which are popular in Vietnam. The composition of the eight types of rice husks was determined to enable us to select rice husks with significantly different ash contents (T). Based on our results, Te do, Nep Ha Noi and Khang dan rice husks were chosen, and rice husk chars were produced for experiments using the muffle furnace following the same method described above.

Table 4: Composition of the selected Vietnamese rice husks

No	Rice husk type	V_{db}%	A_{db}%	FC_{db}%
1	Te do	64.6	16.4	19.0
2	Khang dan	66.1	14.5	19.4
3	Thien Uu	65.0	15.8	19.2
4	Can Tho	66.5	13.7	19.8
5	Nep Ha Noi	69.3	12.8	17.8
6	Bac Thom 7	65.1	15.5	19.4
7	B.C	67.6	13.3	19.1
8	Viet Huong	66.3	13.3	20.4

Figure 6 revealed that there were no significant differences in the conversion rates of the three different rice husk chars, suggesting that findings obtained with a specific type of rice husk could be applicable to other types.

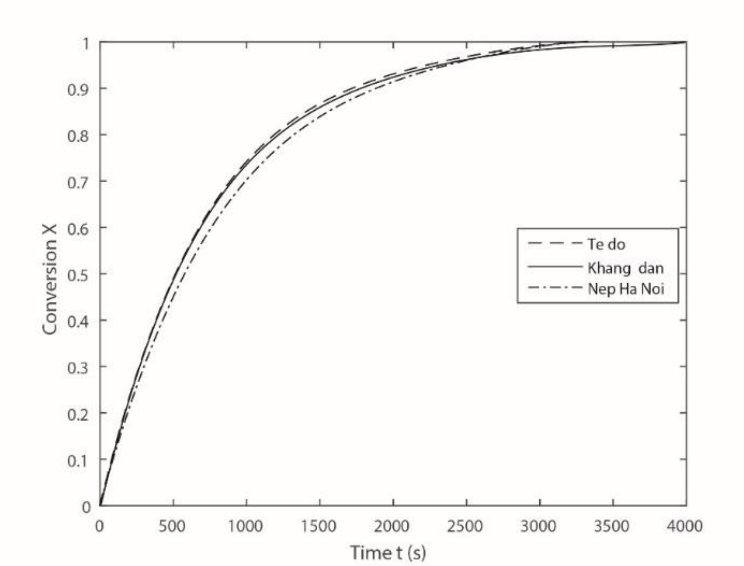


Figure 7: Influence of the nature of rice husk on char conversion in an H_2O atmosphere (partial pressure: 0.2 atm, temperature of reacting gas: 900°C).

Conclusion

The influence of pyrolysis temperature and pyrolysis heating rate on steam gasification of rice husk char at a particle scale was investigated using a macro-thermogravimetric reactor.

Regarding pyrolysis, an increase in pyrolysis temperature from 600°C to 900°C slightly decreased rice husk char yield, but didn't have much effect on rice husk char characteristics and morphology. A significant increase of pyrolysis heating rate from 20°Cmin^{-1} to 1800°Cmin^{-1} decreased considerably char yield and had an effect on rice husk char characteristics and morphology. Regarding gasification, a char produced at high heating rate (1800°Cmin^{-1}) increased gasification kinetics by about 1.4 times compared to a char produced at low heating rate (20°Cmin^{-1}). Comparison of gasification kinetics of different types of rice husks also suggested that findings obtained with a specific type of rice husk could be applicable to other types. Results and data produced could be useful to set up industrial or academic codes, necessary for the conception of new rice husk gasifiers.

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