



Research Article

Modelling of integrated processes for the pyrolysis and steam reforming of rice husk (*Oryza sativa*)

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Abstract

Thermochemical processes can be used to harness the energetic content of agricultural residues. This study utilises ASPEN Plus v8.8 to develop thermodynamic models for pyrolysis and in-line steam reforming of rice husk (*Oryza sativa*). The pyrolysis simulation at 500 °C and 1 atm gave a product yield of 36.3% oil, 49.6% char and 14.1% gas. Whilst the gas was composed of lighter hydrocarbons, the char was primarily elemental carbon and SiO₂. The pyrolysis oil was composed of higher hydrocarbons, an assortment aromatic compounds and pyrolytic water. Optimum parameters for the steam reforming process were 700 °C, 1 atm and a steam-to-gas molar ratio of 7. The product composition at optimal conditions was 67% hydrogen gas, 19% carbon dioxide, 12% carbon monoxide and 2% methane. For a theoretical biomass feed of 200 kg/h and steam feed of 1400 kg/h, the synthesis gas flowrate obtained from the process was 204 kg/h and the rest were char and condensate water. This has gone a long way to reinforce our idea that the energy content of locally sourced rice (*O. sativa*) husk can be harnessed via different thermochemical techniques to give good yields of very useful products.

Keywords ASPEN Plus · Modelling · Pyrolysis · Steam reforming · Rice husk · Thermodynamics

1 Introduction

Rice is a monocotyledonous plant of the genus *Oryza*. The genus consists of two cultivated species and 21 wild species [1]. The cultivated species *Oryza sativa* and *Oryza glaberrima* originated from Asia and Africa, respectively. *O. sativa* has superior yield and milling quality and is commercially grown in 112 countries from all continents. In contrast, *O. glaberrima* is a semi-aquatic plant which is only grown in the West Africa region [1]. Rice (*Oryza sativa*) is one of the major agricultural products of tropical West Africa in general and Nigeria in particular. The by-product from the milling of rice is the husk. The husk of the plant is currently incinerated by both commercial and local farmers.

Rice husk of course is a very rich cellulosic source for energy recovery processes [2], and it is readily available, especially in West African countries [3]. Rice (*Oryza sativa*)

husk can be converted into useful forms of energy via different novel thermochemical and biochemical conversion techniques. Thermochemical methods include direct combustion, pyrolysis and gasification, whilst biochemical methods include the anaerobic digestion and fermentation processes [1]. Amongst the various biomass thermochemical technologies, pyrolysis presents an interesting opportunity to local stakeholders in harnessing the energetic content of rice (*Oryza sativa*) husk. Biomass consists of mainly three components: hemicellulose, cellulose and lignin [4]. Each of these three components possesses very different thermal behaviours [5]. The nature and yield of the pyrolysis products are also dependent on the distribution of hemicellulose, cellulose and lignin in the biomass.

The experimental thermochemical conversion of rice (*Oryza sativa*) husk into useful products has been investigated over the years [6–12], reactor designs inclusive [13, 14]. Alvarez et al. [15] utilised a conical spouted bed reactor

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for bio-oil production from rice husk pyrolysis and obtained a maximum bio-oil yield of 70 wt% at 450 °C. Tsai et al. [7] investigated the product yield and composition from the pyrolysis of rice husk in a fixed-bed tubular reactor. They observed an oil yield of just above 40% at the optimal temperature of about 500 °C. Natarajan and Ganapathy [16] also studied rice husk pyrolysis in a fixed-bed reactor and obtained a maximum of about 31% oil yield at 500 °C. Williams and Nugranad [17] compared products from catalysed and uncatalysed pyrolysis of rice husk and observed that the oils from an uncatalysed process were more homogeneous, of low viscosity and highly oxygenated. In-line steam reforming is unreported yet for rice husk.

Numerous chemical reactions occur during rice husk pyrolysis; therefore, simple stoichiometric reaction equations represent the entire process cannot be practically elucidated. Studies have been conducted on reaction sequences and overall reaction kinetics for biomass pyrolysis in general [18] and rice husk in particular [6]. For numerous biomass samples, kinetics and reaction sequence has been exhaustively examined [19]. Several theoretical in silico models for energy recovery from rice husk have also been developed and validated [20–23] over the years using a variety of approaches.

Researchers have been exploiting ASPEN Plus in modelling different aspects of renewable energy processes. Key research domains have come in the areas of process design, modelling and integration [24–28], feasibility studies [29], thermodynamic analysis [30–32], life cycle assessment [33–35], energy and exergy analysis [36], greenhouse gas assessment [37] and cost analysis [38] amongst other industrial applications and research application [39–42]. Thermodynamic modelling of biomass pyrolysis has previously been conducted for banana residues [43] and sugarcane bagasse [44]. There has not been any report for rice husks. The aim of this study is to utilise ASPEN Plus v8.8 to develop models for the pyrolysis and in-line steam reforming of rice (*Oryza sativa*) husk to more useful products based on a thermodynamic approach. The models are used to evaluate the product yield and feedstock potential of rice husk for the thermochemical processes. A key advantage of this approach is that it eliminates other extraneous factors present in experimental studies. This helps to paint a true picture of how this process factor affects product yield and portray the true potential of the biomass for thermochemical processing.

2 Methodology

The commercial software package ASPEN Plus v8.8 was utilised for the modelling and simulation of the process. ASPEN Plus is a complete integrated solution for

process systems engineering [26]. It does not possess any preloaded models but consists of several unit operation blocks. ASPEN Plus can be used to model fast pyrolysis and predict the output of bio-oil, gas and char [43, 44]. It can also be used for steam reforming simulations [30, 45, 46]. The simulations are based on a minimisation of Gibbs free energy calculation technique. Details of the technique have been explained elsewhere [47, 48]. The feedstock to be pyrolysed in the simulation is rice (*Oryza sativa*) husk. Proximate, ultimate and chemical analyses of rice (*Oryza sativa*) husk were obtained from reported values for West African agricultural residues [49].

2.1 Model component specification

The global stream class of the simulation was set as MIXCINC. This informs that there are conventional and non-conventional solids apart from the conventional components. The particle size distribution of the solid component is not required under this selection. Non-conventional components are not present in ASPEN Plus database as they do not have a molecular formula. In the simulation environment, they are specified by the information from their proximate and ultimate analyses. Enthalpy and density are the only properties calculated for non-conventional components, and they are computed by empirical correlations. The specific property methods for enthalpy and density for rice husk were chosen as HCOAL-GEN method and DGOALIGT method, respectively. Rice (*Oryza sativa*) husk is a non-conventional material which can be easily modelled in ASPEN Plus v8.8 by specifying proximate and ultimate analyses. Table 1 shows the necessary information required in modelling the biomass in the software.

The conventional components added to the simulation includes saturated aliphatic hydrocarbons C_1 – C_{25} , nitrogen gas, hydrogen sulphide, hydrogen chloride, water, some aromatic compounds and elemental carbon. Water was added to account for the moisture content of the feed. The decomposition products were cellulose, hemicellulose and lignin. Hemicellulose and cellulose are represented in the simulation by their monomers: $C_5H_8O_4$ (xylan) and $C_6H_{10}O_5$ (xylose-like cellulose monomer), respectively [50]. Lignin was represented in the simulation by a phenyl propane monomer. The nitrogen content of the biomass is taken into account by including pyrrole in the simulation components. The hydrogen sulphide gas accounts for all sulphur contents, whilst hydrogen chloride gas accounts for the chlorine content of the feedstock. Ash was considered to be SiO_2 as studies show that above 90% of the rice husk ash is SiO_2 [2]. For the estimation of the physical properties of the conventional components in the simulation, the Peng–Robinson with Boston–Mathias alpha

Table 1 Proximate, ultimate and chemical analysis of rice (*Oryza sativa*) husk [49]

<i>Proximate analysis (wt% wet basis)</i>	
Moisture	8.59
Fixed carbon	8.48
Volatile matter	58.22
Ash	24.71
<i>Ultimate/elemental analysis (wt% moisture free)</i>	
Carbon	34.9
Hydrogen	5.15
Sulphur	0.64
Oxygen	59
Nitrogen	0.31
Chlorine	<0.01
<i>Chemical analysis (wt%)</i>	
Cellulose	37.34
Hemicelluloses	10.07
Lignin	41.08
Extractives	11.51

function equation of state (PR–BM) was used. Alpha is a temperature-dependent parameter that improves the pure component vapour pressure correlation at very high temperatures [33]. For this reason, PR–BM was suitable for the pyrolysis process since relatively high temperatures are involved [43].

2.2 Reactor model description

The pyrolysis reaction was modelled in the simulation by a combination of three blocks: the RSTOIC, RYIELD and RGIBBS reactors. The RSTOIC block is a stoichiometric reactor

that utilises stoichiometric equations. This was used to model the drying (evaporation of moisture) of rice (*Oryza sativa*) husk at the commencement of heating. The RYIELD (yield) reactor carries out the conversion of the dry non-conventional feedstock to conventional simulation components (cellulose, hemicellulose and lignin). The RGIBBS (Gibbs) reactor does the calculation of the final component distribution and phase equilibrium through the minimisation of Gibbs free energy. Both the yield reactor and the Gibbs reactor do not require the specification of stoichiometric equations. Table 2 shows the description of each unit involved in both simulations.

In the development of the simulation model, several assumptions were implemented. Both models prepared with ASPEN Plus v8.8 are steady-state isothermal sequential modular models. In the model for pyrolysis, it was considered that about 95% of the moisture is evaporated as water vapour. All the sulphur in the biomass is considered as organic sulphur, and it leaves the processes as hydrogen sulphide gas. All chlorine in the processes leaves as hydrogen chloride gas. The ash content of the feedstock was represented as SiO₂ alone. All elements take part in the chemical reaction except ash which is considered as inert.

2.3 Process model description (pyrolysis)

A steady-state sequential modular model was developed using ASPEN Plus v8.8. In ASPEN Plus, sequencing connotes a block-by-block calculation method where the results of one module serve as the basis for the next. The simulation model is similar to previously developed and validated models albeit with an improvement to account for the initial pre-drying stage [43, 44]. The simulation ambient temperature and pressure were specified as 25 °C

Table 2 ASPEN Plus unit operations model description

ASPEN Plus ID	Block ID	Description
RSTOIC	DRYER	Models the moisture content reduction/drying of the biomass at the initiation of heating
RYIELD	DECOMP	Models the conversion of non-conventional materials (rice husk) to conventional simulation components (cellulose, hemicellulose and lignin)
RGIBBS	PYRO	Calculates the pyrolysis products distribution via the minimisation of Gibbs free energy method
FLASH2	SEP1	Removal of moisture from the pyrolysis vapour stream
SSPLIT	SEP2	Separates the char from the product vapour by specified split ratio
HEATER	COOLER	Induces condensation of pyrolysis liquid products by reducing the vapour stream temperature
FLASH2	SEP3	Separates the pyrolysis oil from non-condensable gases
CALCULATOR	WATER	Specifies 90% moisture removal to the RSTOIC block
CALCULATOR	PYROLYSE	Specifies the RYIELD mass fractions of cellulose, hemicelluloses and lignin based on their proportions in the chemical analysis
HEATER	HEATER	Raises the temperature of the water feed and converts it to steam
RGIBBS	REFORMER	Predicts the steam reforming reactions via the minimisation of Gibbs free energy method
FLASH2	SEP4	Removes moisture from the reformer vapour stream via condensation

and 1 atm, respectively. The rice (*Oryza sativa*) husk feedstock (200 kg/h) and nitrogen purge gas (0.1 kmol/hr) were fed into the RSTOIC reactor block at ambient conditions, and drying was stipulated to occur at a temperature of 130 °C. The nitrogen helps in the removal of evolving water vapour. In fact, all nitrogen fed into system leaves with the water vapour. About 95% of the moisture was stipulated to be removed by the dryer by the 'water calculator' block. This was effected by the use of Fortran declarations in the simulation. The RYIELD reactor block carried out the conversion of the non-conventional components to cellulose, hemicellulose and lignin. The split fractions were specified by the 'pyrolyse' calculator block, using the values in the chemical analysis earlier stated. The equations in both calculator blocks were stated using Fortran statements. The process flow diagram (PFD) of the simulation is presented in Fig. 1.

The RGIBBS reactor predicts the final product distribution via the minimisation of Gibbs free energy. The nitrogen gas and SiO₂ were specified as inert in the Gibbs reactor. The temperature and pressure of the reactor system are taken as the temperature and pressure of the final reactor block, and they were specified at 500 °C and atmospheric pressure. The choice of pyrolysis temperature was based on the results of Ji-Lu [8] who observed that for rice husk, optimum pyrolysis temperature was between 420 and 540 °C. The cyclone is used to model the separation of the char from the vapour products. The vapour is then condensed to ambient temperature before the final separation of the non-condensable gases from the oil.

2.4 Process model description (in-line steam reforming)

The simulation was adjusted for the steam reforming process. The addition of an in-line steam reforming unit is similar to that previously utilised in a previous work [44]. Conventional components: hydrogen gas, carbon dioxide and carbon monoxide, were added to the simulation. The pre-drying section was removed as water in the pyrolysis oil will favour the reforming reaction. Consequently, the pyrolysis oil obtained in this process will have a high moisture content. The vapour condensation section of the previous simulation was removed, and a steam reformer was introduced instead. Water was pre-heated to a temperature of 120 °C to obtain a steam feed and then sent into the reformer (Fig. 2).

The vapour products stream was sent directly into the reformer with an inflow of steam going in at a specific steam-to-gas molar ratio (STGR). The reformer is also a Gibbs (RGIBBS) reactor which requires no reaction

stoichiometry. Factors such as STGR, reformer temperature and reformer pressure were optimised to give maximum yield of hydrogen from the process. The reformed products are then condensed back to room temperature to separate the excess water from the gaseous products. Response surface methodology (RSM) was used to study the factor interactions and for the determination of optimum parameters for the steam reforming of glycerol. In order for more convenient report, notations in Table 3 are given to the factors/independent variables. For this study, the temperature range considered was 300 and 1100 °C, pressure was between 0.1 and 5 atm, whilst steam-to-gas ratio was between 1 and 12 mol/mol.

3 Results and discussion

3.1 Pyrolysis results

After running the simulation, the results obtained at feed rate and pyrolysis temperature of 200 kg/h and 500 °C, respectively, are presented in Table 4. The results demonstrate a decent yield of pyrolysis oil with respect to the other products. From the simulation results, the synthesis gas is composed mainly of C₁–C₄ hydrocarbons with traces of other gases in the simulation as impurities. The char was composed primarily of elemental carbon and SiO₂. The pyrolysis oil was composed of the higher hydrocarbons, an assortment aromatic compounds and some pyrolytic water. The moisture content of the oil obtained was 2.02%. This value is quite fantastic because the feedstock is first pre-dried to remove moisture content by 90%. The difference in moisture content is a reason for variations in product composition and yield in experimental works. Ji-Lu [8] summarily observed this also after oil characterisation. It can be seen from Table 5 that simulation results were fairly consistent with experimental works, albeit at different levels. The investigation by Tsai et al. [7] shows a better consistency than that by Ji-Lu [8]. It can be surmised that locally sourced rice (*Oryza sativa*) husk can be pyrolysed to give decent oil yields. The biomass has been shown to be a promising feedstock for bio-oil production via the pyrolysis process. The results of the thermodynamic predictions reveal that rice husk is inherently more suitable for the production of char via char optimised processes.

3.2 In-line steam reforming results

For the stipulated ranges of STGR, temperature and pressure, a numerical optimisation was carried out to

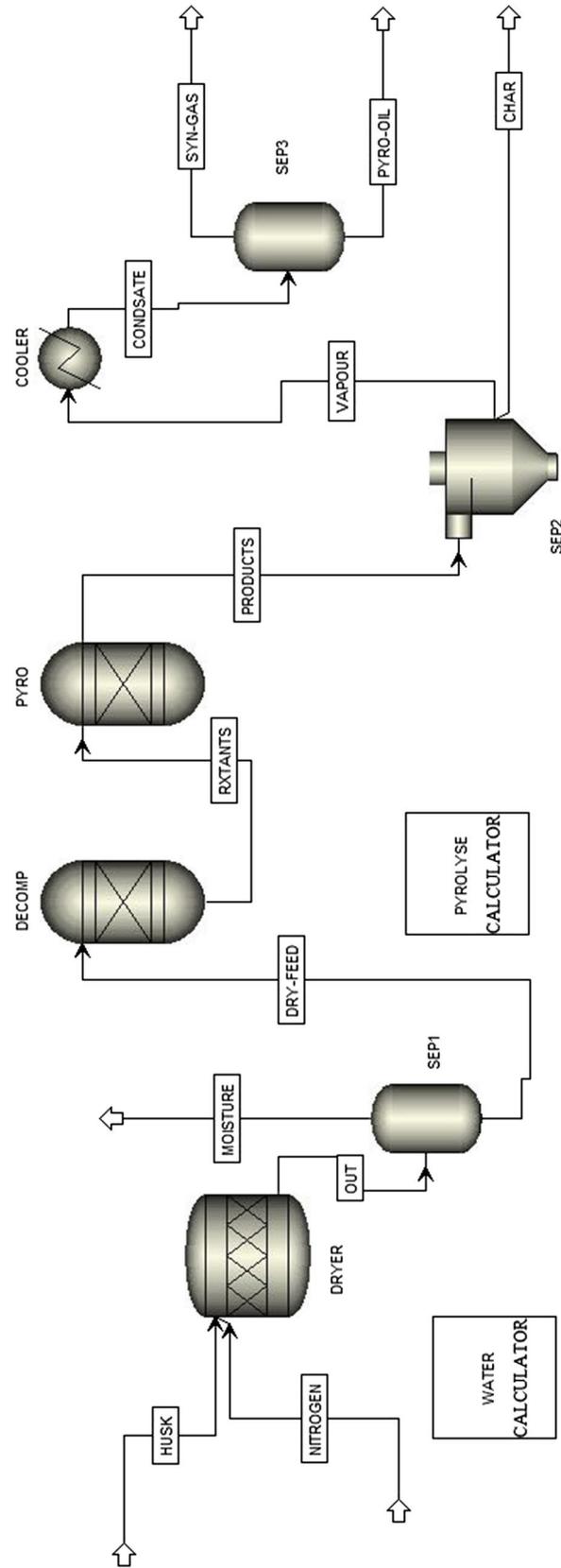


Fig. 1 The process flow diagram of the pyrolysis simulation

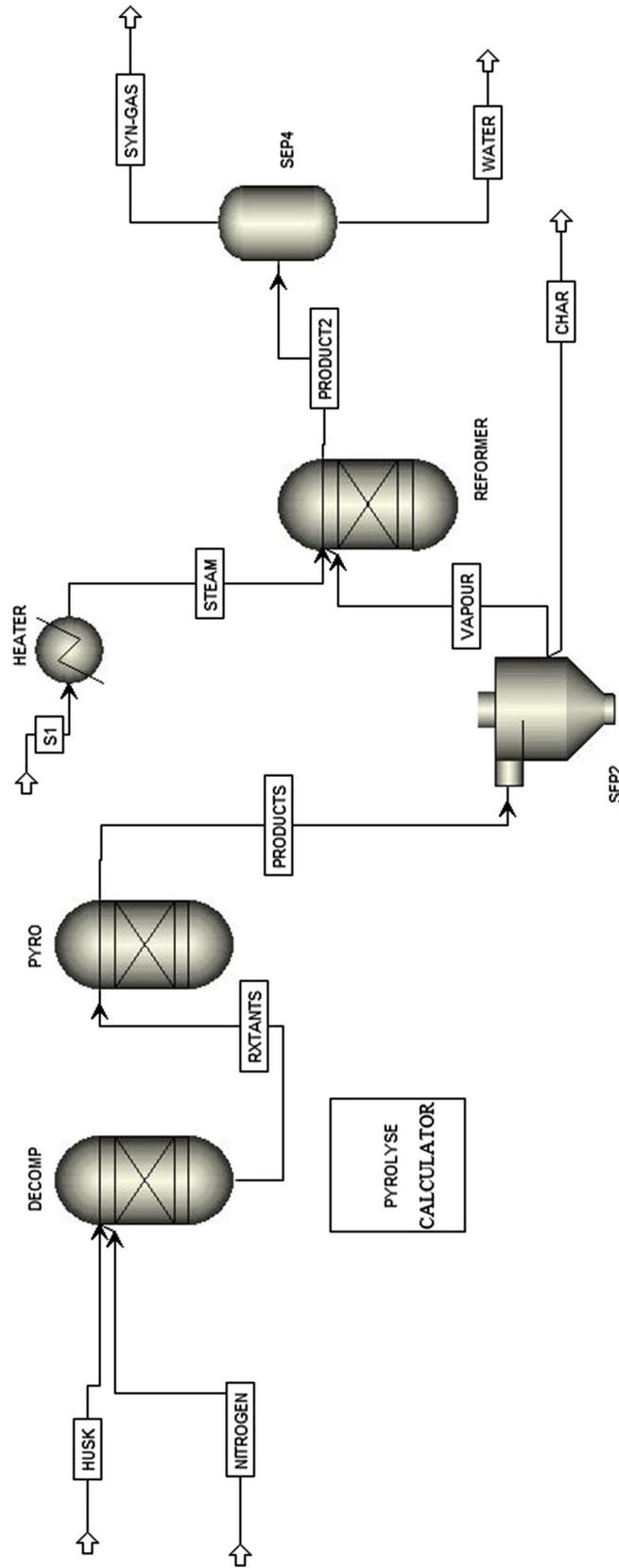


Fig. 2 The process flow diagram of the steam reforming simulation

Table 3 Notations of factors/independent variables

Factor	Unit
Reaction temperature	°C
Reactor pressure	atm
Steam-to-gas ratio (STGR)	mol/mol
H ₂ % Molar composition	%
CO% Molar composition	%
CO ₂ % Molar composition	%
CH ₄ % Molar composition	%

Table 4 Pyrolysis results

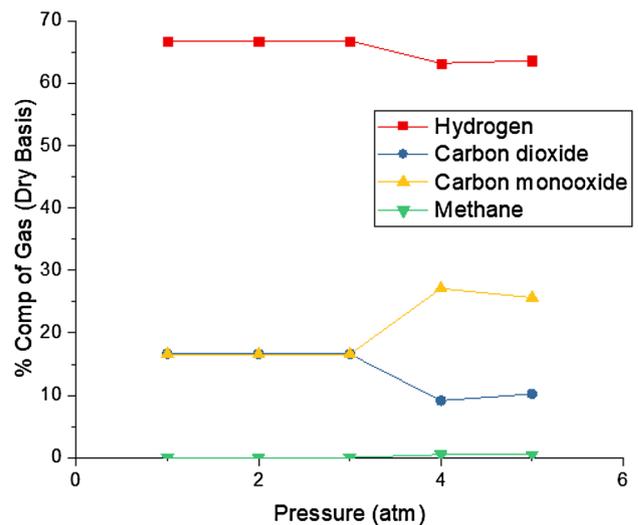
Product	Yield (wt%)
Oil	36.30%
Char	49.60%
Gas	14.10%

Table 5 Validation of results

	Oil	Char	Gas
Current study (500 °C)	36.30%	49.60%	14.10%
Tsai et al. [7] (500 °C)	35.90%	47.10%	17.00%
Ji-Lu [8] (500 °C)	53%	21%	26%

determine the optimum parameters for hydrogen gas production. Optimum parameters were obtained as 897 °C, 4.34 atm and STGR of 7.82 mol/mol. These values are considered to be purely numerical; hence, a secondary optimisation will be carried out with the simulation itself to get the best possible parameters considering not only hydrogen maximisation, but carbon monoxide maximisation and methane and carbon dioxide minimisation. The above numerical optimisation results will provide the initial basis for examining the simulation.

From the figure above, hydrogen gas production is constant from atmospheric pressure to 3 atm and then it falls. Methanation is fairly independent of pressure. Between 3 and 5 atm, carbon monoxide production is favoured against carbon dioxide. CO is usually desired in synthesis gas above CO₂ because it is also combustible. However, in considering the process only on the basis of optimising hydrogen generation, 1 atm remains the optimal pressure for the process. Adeniyi et al. [44] explained that atmospheric pressure is optimal for in-line steam reforming as biomass as lower pressures shifts the chemical equilibrium in favour of the lighter chemical species. Hydrogen is the lightest species in the system; hence, the observed trend in Fig. 3 is validated.

**Fig. 3** Gas composition with pressure at 897 °C and 7.82 mol/mol steam-to-gas ratio

Now utilising the optimal pressure alongside the numerical optimal STGR of 7 mol/mol, we obtain the temperature sensitivity of the synthesis gas produced. Hydrogen production peaks at 700 °C and starts dropping as the threshold is exceeded. Lower temperatures favour methanation and CO₂, whilst higher temperature favours CO production. Optimal temperature for hydrogen production is 700 °C. The rise in hydrogen production with an increase in temperature is due to the endothermic nature of the steam reforming reaction which is favoured at higher temperatures [30]. The rise in methane production at lower temperatures is due to the exothermic nature of methanation reaction which is favoured at those conditions [30]. The drop in hydrogen production beyond 700 °C was explained by Adeniyi and Ighalo [48] as competing reaction such as high-temperature thermal cracking of oxygenates, reverse water–gas-shift and coke deposition reactions (Fig. 4).

The above plot in Fig. 5 elucidates gas composition responses to STGR changes at optimal temperature and pressure. Appreciable increase in the hydrogen composition stops at an STGR of about 8 mol/mol. However, in the simulation a steady but minor increase in hydrogen composition is observed even up to an STGR of 15 mol/mol. However, in determining the optimal STGR, several points are put in consideration. The combustible methane and CO are favoured at lower STGR compared to the non-combustible CO₂. Steam economy is also crucial if the process is to possess any financial appeal to investors. Considering these, optimal STGR is 7 mol/mol because beyond that point hydrogen composition does not rise significantly enough considering the fall in the composition of

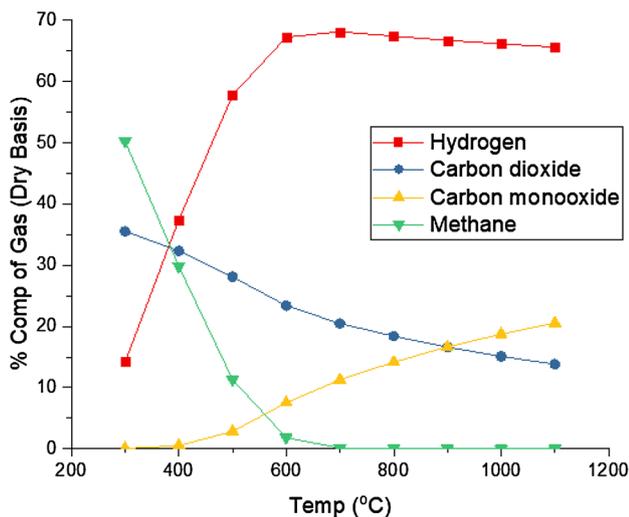


Fig. 4 Gas composition with temperature at 1 atm and 7.82 mol/mol steam-to-gas ratio

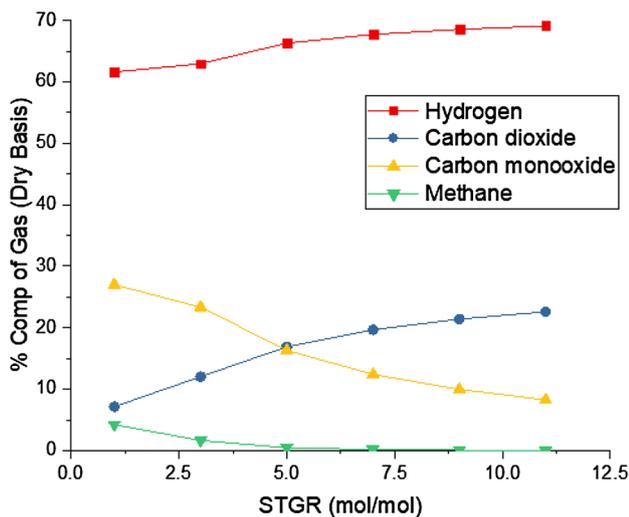


Fig. 5 Gas composition with steam-to-gas ratio at 1 atm and 700 °C

the other combustible products. In summary, optimum parameters are 700 °C, 1 atm and an STGR of 7 mol/mol and these will give a product composition of 67% hydrogen gas, 19% carbon dioxide, 12% carbon monoxide and 2% methane. The synthesis gas flowrate obtained from the process is 204 kg/h (for a theoretical 200 kg/h biomass and 1400 kg/h steam feed rate, respectively) and the rest of the materials are char and condensate water (Fig. 6).

From the above surface plots, we observe that temperature is the most significant factor in the steam reforming of rice (*Oryza sativa*) husk for hydrogen production. The reforming pressure and STGR demonstrate a far less

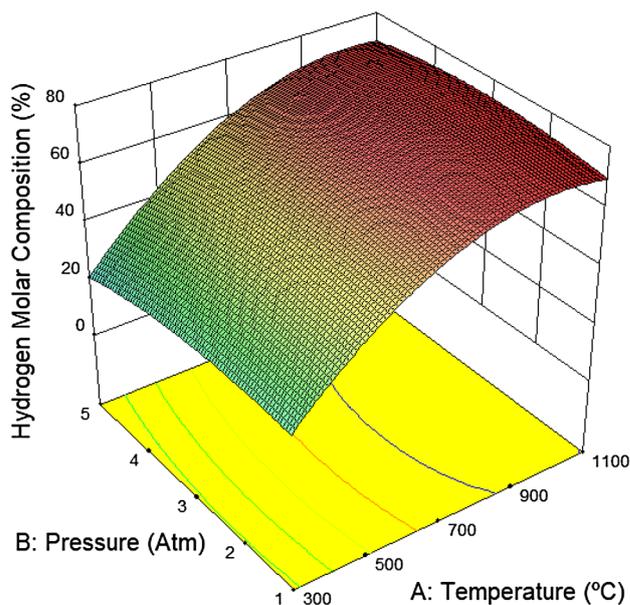


Fig. 6 Surface plot of hydrogen composition response to pressure and temperature

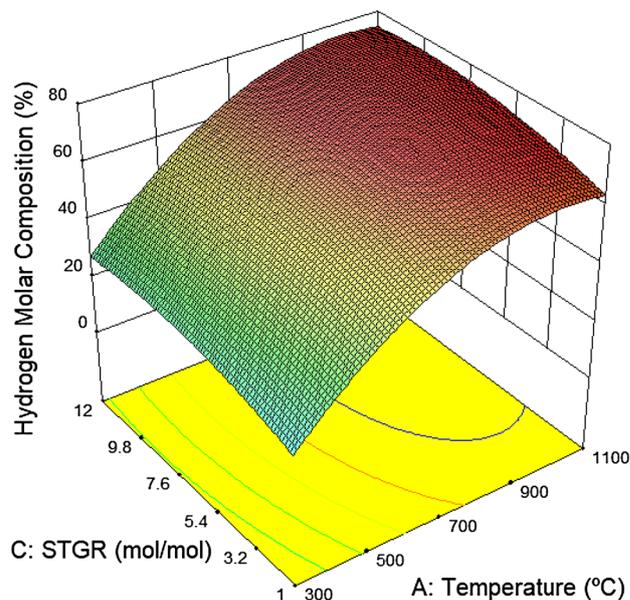


Fig. 7 Surface plot of hydrogen composition response to steam-to-gas ratio and temperature

significant effect on the composition of hydrogen in the synthesis gas stream. The results of the steam reforming study reveal that rice husk can be harnessed to give an excellent yield of hydrogen via the steam reforming process (Figs. 7, 8).

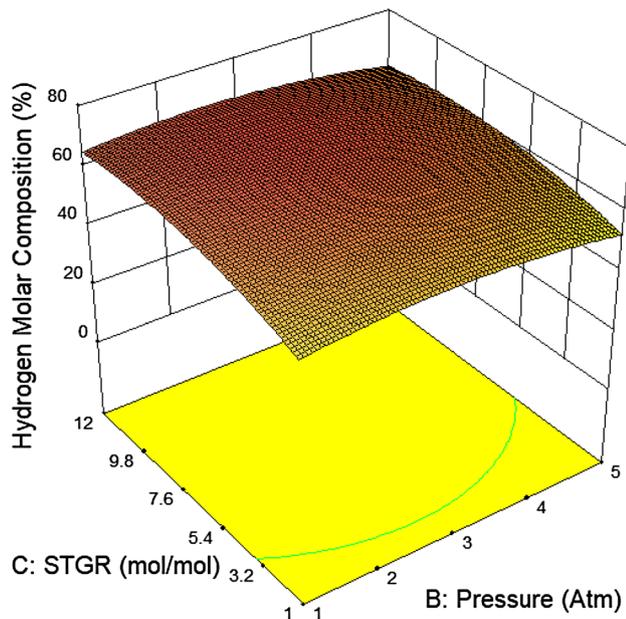


Fig. 8 Surface plot of hydrogen composition response to steam-to-gas ratio and pressure

4 Conclusion

ASPEN Plus v8.8 was used to develop a steady-state model for the pyrolysis of rice (*Oryza sativa*) husk based on a thermodynamic approach. The simulation pyrolysis conditions were at 500 °C and atmospheric pressure. After the simulation was run, a yield of 36.3% oil, 49.6% char and 14.1% gas was obtained. The pyrolysis yield was shown to be consistent with experimental studies at similar temperatures. The gas is composed mainly of light hydrocarbons, whilst the char was composed primarily of elemental carbon and as SiO₂. The pyrolysis oil was composed of the higher hydrocarbons, an assortment aromatic compounds and 2.02% pyrolytic water. The results of the thermodynamic predictions reveal that rice husk is inherently more suitable for the production of char via char optimised processes. Optimum parameters for the steam reforming process were 700 °C, 1 atm and an STGR of 7 mol/mol, and these gave a product composition of 67% hydrogen gas, 19% carbon dioxide, 12% carbon monoxide and 2% methane. The synthesis gas flowrate obtained from the process was 204 kg/h. The results of the steam reforming study reveal that rice husk can be harnessed to give an excellent yield of hydrogen via the steam reforming process.

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Compliance with ethical standards

Conflict of interest No potential conflict of interest was reported by the authors.

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