Investigation of the Reaction Behavior of *Albizia gummifera* Wood under Steam Gasification with Varied Partial Pressures and Gasification Temperatures

Kehinde Oluoti* and Tobias Richards

The astronomical increase in global energy demand makes locating energy sources other than fossil fuels worthwhile. The use of tropical biomass wood waste as a renewable energy source was investigated in this study. The thermal conversion analysis of *Albizia gummifera* (ayinre) was carried out in a thermobalance reactor *via* steam gasification under varying temperature (700 to 1000 °C) and steam partial pressure (0.020 to 0.050 MPa). The experimental data was evaluated using three gas-solid reaction models. The modified volume reaction model (mVRM) gave the overall highest coefficient of determination (0.9993) and thereby the best conversion prediction. The observed char activation constant rates (from paired reaction conditions) indicated, on average, an increase in reactivity as the parameters increased. The results showed that the activation energy of the mVRM gave the lowest value (32.54 kJ/mol) compared with those of the shrinking core model (SCM) and the volume reaction model (VRM) (49.29 and 49.89 kJ/mol, respectively).

Keywords: Steam gasification; Tropical biomass; Kinetic parameters; Albizia gummifera; Gas-solid reaction models

*Contact information: Swedish Centre for Resource Recovery (SCRR), University of Borås, SE-501 90, Borås, Sweden; *Corresponding author: kehinde.oluoti@hb.se; oluotikehinde@yahoo.com*

INTRODUCTION

The unprecedented increase in global population has correspondingly increased the total energy demand. Fossil fuel, the world's largest energy source (Loáiciga 2011), is non-renewable and causes environmental pollution problems resulting from its exploration and processing (Shafiee and Topal 2009). Ongoing problems include climate change, ozone depletion, drought, flooding, and melting glaciers. Many of these problems are man-made and attributed to carbon dioxide emissions resulting from fossil fuel combustion.

In addition to the climatic and health concerns of fossil fuels, the increasing cost of processing and product security are two major factors that make downstream products very expensive. The economic downturn and the rise in energy demand have promoted the search for other affordable sources of energy. Biomass is an abundant source of renewable energy and an alternative to fossil fuels. The overall carbon emission during energy recovery processes is low, and it is much easier to source (Oluoti *et al*. 2014). Biomass is the leading sustainable source of biofuels (Huber *et al*. 2006), and it is potentially the largest sustainable energy source with an annual global availability of 220 billion dry tons (De Lasa *et al*. 2011).

Thermo-chemical conversion is a well-known method for deriving energy products from biomass (Ahmed and Gupta 2009). As the name implies, gasification converts biomass into a gaseous intermediate prior to its final use, *e.g.*, to produce electricity or liquid fuels *via* the Fischer-Tropsch process. The operating variables of gasification include gasifying agents and temperature. The most common gasifying agents are air, steam, O_2, CO_2, steam-oxygen, air-steam, and O_2-enriched air.

According to Sun *et al.* (2007), there are many applications of gasifying agents, with varying outcomes. Air gasification in food waste treatment produced a fuel gas with a low heating value of 4 to 6 MJ/m^3 and 8 to 14% by volume hydrogen (H_2) content. When cotton wood and Douglas fir chars were gasified under carbon dioxide or steam atmospheres, the latter had a lower activation energy for the same conversion. Fluidized-bed steam gasification of biomass produced a medium heating value (MHV) (10 to 16 MJ/m^3) gas with a 30 to 60% H_2 content. The fluidized-bed air-gasification system that uses sawdust produced a fuel gas with a heating value of 5.0 MJ/Nm^3 and an H_2 content of 9.27% (Cao *et al*. 2006).

Steam gasification of recycling waste in a bubbling fluidized-bed reactor produced a syngas with a heating value of 8.6 MJ/m^3 (Slapak *et al*. 2000). Carbonization products from brown coal gave increased gasification activity as a result of increased carbon compound interaction with water vapor (Kuznetsov *et al.* 2015). Steam gasification of sewage sludge increased the hydrogen yield three-fold compared with air gasification (Nipattummakul *et al*. 2010). Finally, Ahmed and Gupta (2011) gasified woodchip chars with steam and CO_2. The reactor temperature was maintained at 900 °C, and the partial pressures of the gasifying agents ranged from 1.5 to 0.6 bar. The average reaction rate for steam was almost twice that of CO_2.

Temperature is an important variable during gasification processes. Air gasification of mixed fuels (biomass and coal-coke) produced a syngas with increased H_2 concentration as the temperature increased (Lapuerta *et al*. 2008). Syngas (H_2 and CO) yield, in terms of mass fraction, was favored by high-oxidation zone temperature during air gasification of dry refinery sludge using an updraft gasifier (Ahmed and Sinnathambi 2013). At sufficient residence time, increasing the gasification temperature led to higher H_2 yields, CO efficiency, and higher heating value (HHV) of the product gas (Aljbour and Kawamoto 2013).

Varying the partial pressure of the gasifying agent has a considerable influence on the overall gasification process and the outcome (Slapak *et al*. 2000). Sharma *et al*. (2009) investigated the effect of steam partial pressure on gasification rate and gas composition of product gas from catalytic steam gasification of HyperCoal. The results showed that partial pressure changed with the rate constant and the latter was also identified as a means of controlling the H_2/CO ratio of the synthetic gas. In the steam gasification of coke from an Indian high-ash coal (Aranda et al., 2016), it was established that both reaction rate and the conversion increased with increasing steam partial pressure.

These examples show that using steam as a gasifier agent together with temperature as a reaction variable produces a syngas with high energy content. Steam gasification was therefore chosen in this study for investigating the gasification reaction behavior of *Albizia gummifera* (ayinre).

With the ultimate ambition of converting waste material into electricity, tropical wood waste from industries, companies, and other facilities are gasified to produce fuel gas. Gasification utilizes the appropriate gasifier agents and operating conditions. Temperature is an important operating condition that is varied to obtain the optimal conversion. This study aimed to find a renewable solution to meet national energy needs.

Currently in Nigeria, biomass wood waste is being used only for cooking and other domestic purposes. Biomass wood waste is of particular interest as a potential fuel to address the inadequate power supply produced by the national grid. On an industrial scale, a fixed-bed gasifier could be used to convert fuel materials and study the process kinetics. In this study, a lab-scale version of the gasifier—the thermogravimetric analyzer (TGA)—was employed. The results from this study potentially could be extrapolated to represent those obtained at the industrial scale.

The aim of this study was to investigate the behavior of ayinre under varied partial pressures of steam and temperatures during steam gasification. The kinetic parameters were determined. Specifically, the reaction constant K, was obtained from the most suitable model applied in this study, and the activation energy E_a, was used to evaluate the behavior of the sample during the thermal/steam gasification process.

EXPERIMENTAL

Sample Preparation and Properties

Ayinre samples were collected from sawmills in Nigeria. The dried pieces were further dried in the sun for approximately three weeks to ensure low moisture content. The wood pieces were ground until they had an average final fineness size of 0.25 to 20.00 mm (Fig. 1). The chemical composition of a sample determines its energy content. Table 1 shows the composition of the ayinre sample as evaluated by Bränsle & Energilaboratoriet (BELAB) AB, Norrköping, Sweden.

Fig. 1. A photograph of the ayinre fuel sample

Table 1. Composition of Dry Ayinre

Local Name	Botanical Name	Moisture % (105 °C)	Ash %ts (550 °C)	C %ts (dry)	H %ts (dry)	N %ts (dry)	S %ts (dry)	HHV (kJ/kg) (dry)
Ayinre	*Albizia gummifera*	5.3	1.5	49.4	6.1	0.36	0.027	19.42

ts, total solid; C, carbon; H, hydrogen; N, nitrogen; S, sulphur

Equipment

Cutting mill

The size of the material was reduced with a Retsch SM 100 model cutting mill (Retsch Technology GmbH, Haan, Germany) using cutting and shearing forces. The sample came into contact with the rotor and was then comminuted between the blades and the stationary standard cutting bars inserted in the housing. The dwelling time of the sample in the chamber was short; the articles were discharged as soon as they were small enough to pass through openings in the bottom sieve, and then they were collected in the receptacle. The rotor speed of 1,500 $\min^{-1}$ ensured gentle and rapid size reduction.

Bomb calorimetry

An IKA C 200 bomb calorimeter (M2 Scientifics, Hudsonville, MI, USA) was used to determine the HHV of the sample, using the ASTM D 5865 (2010) standard.

Thermogravimetric analysis (TGA)

Laboratory-scale gasification was performed using a DynTHERM HP TGA device (Rubotherm GmbH, Bochum, Germany). This device is a combination of a magnetic suspension balance and a gas/vapor dosing system into which a sample was loaded by a motor-assisted movement of the balance into a convenient loading position. In all runs, the sample was deposited in a small cylindrical ceramic crucible of 10 mm in diameter and 10 mm in height. The average mass of the sample used in each run was approximately 0.160 g. The change in mass of the reacting sample was recorded as the reaction proceeded. The sensitivity of the balance was 10^{-5} g.

Sample Conversion Procedure

Devolatilization process: Pyrolysis

Prior to the pyrolysis reaction, the material was dried further in the TGA device at 105 °C to a constant mass. The resulting dried material was subjected, under a nitrogen atmosphere and a heating rate of 10 °C/min, to 900 °C and then held at this temperature for 2 h to complete the pyrolysis process. The char (residual material left in the crucible) was then subjected to in-situ steam gasification.

Char-ash conversion tests: Gasification

In order to observe the gasification behavior under varied steam partial pressures and temperatures, temperatures in the range of 700 to 1000 °C were tested, with steam partial pressures from 0.020 to 0.050 MPa. The statistics used for choosing the parameters of each run were two-factor—temperature and steam—experiments with temperature and steam pressure ranges. A full factorial experimental matrix was adopted. For every partial pressure of steam used, *i.e.*, 0.020, 0.030, 0.040, and 0.050 MPa, the complementary partial pressure of nitrogen used was 0.080, 0.070, 0.060, and 0.050 MPa, respectively, to keep the same volume flow of 100 mL/min. Each sample of char had a residence time of 3 h to ensure complete conversion.

Selection of the Kinetic Model for Char Gasification

The overall reaction rate for gasification can be expressed as follows (Zhang *et al.* 2014),

$$\frac{d\alpha}{dt} = K(P_g, T).f(\alpha) \tag{1}$$

where K is the apparent reaction rate of the gasification process, when K is a function of the reaction temperature T, and P is the pressure of the steam used as the gasifying agent. Changes in the properties of the sample under investigation are denoted by $f(\alpha)$.

With the reaction solely dependent upon the reaction temperature as a result of using constant pressure, concentration, flow rate, or other properties of the gasifying agent, the Arrhenius equation representation of the apparent reaction rate constant K can be expressed as,

$$K = A\, exp\, \left(\frac{-E_a}{RT}\right) \tag{2}$$

where A and E_a are the pre-exponential (or frequency) factor and the activation energy (kJ/mol), respectively, and R (kJ mol^{-1} K^{-1}) is the universal gas constant (Xu *et al*. 2014).

The extent of conversion (α) of the char was calculated according to the following expression,

$$\alpha = (m_o - m_t)/(m_o - m_{ash}) \tag{3}$$

where m_o denotes the initial mass of the sample, m_t is its mass at a particular time of reaction, and m_{ash} is the remaining mass (*i.e.*, ash) after complete gasification.

The mathematical representation (rate equation) of a reaction model (conceptual picture) describes the manner and progress of a reaction, *i.e.*, the kinetics of a reaction is described satisfactorily if the model closely represents how the reaction proceeds. However, according to Wu *et al.* (2009), no mathematical model can describe exactly and completely the kinetics of a conversion reaction, and the choice of model for the reactivity study of char is influenced greatly by the morphology of the fuel material. The heterogeneous structure of char means that different kinetic models are chosen for different reaction conditions for different types of char (Zhang *et al*. 2014).

In this study, the conversion data from the steam gasification reaction was interpreted using three selected gas-solid gasification models: the shrinking-core model (SCM), the volumetric reaction model (VRM), and the modified volume reaction model (mVRM). The term $f(\alpha)$ in Eq. 1 had different interpretations and expressions when each of the above-listed models were considered (Zhang *et al*. 2014). Regardless of the form of $f(\alpha)$, the general algorithmic kinetic expression for all of the models is given as,

$$K = Ae^{-(E/RT)}P_{steam}^{n} \tag{4}$$

where P_{steam} is the partial pressure of steam and n is the reaction order.

According to Seo *et al.* (2010), Eq. 4 expresses the dependence of the reaction rate constant on the reaction order n. The value of n is determined by constructing a log-log plot of K *vs.* the partial pressure of the steam; the slope is then equal to n. However, in order to determine the partial pressures (P_{steam}) of steam from the flow rates used during the experimental runs, the equation of state comes into use, as follows,

$$\mathrm{P}_{steam} = \frac{\dot{\mathrm{V}}_{steam}}{\dot{\mathrm{V}}_{N_2}+(\dot{\mathrm{V}}_{steam})} \cdot (P_0) \tag{5}$$

where $\dot{\mathrm{V}}_{steam}$ and $\dot{\mathrm{V}}_{N_2}$ are the volumetric flow rates (mL/min) of the steam and nitrogen used during the gasification reactions, respectively, and P_0 is the atmospheric pressure.

Shrinking-Core Model (SCM)

Known also as the grain model, this model stipulates that reactions must occur at the surface of the particle (Zou *et al*. 2007; Nowicki *et al*. 2011). Taking into account the effect of particle size and mass transfer during char-gas reactions, SCM is an appropriate candidate for studying the kinetics of such reactions (Zheng and Morey 2014). The gas fluid does not diffuse into the core section because the grains, which are solidly packed, are either non-porous or the reaction is so fast that it reacts and forms the products as soon as the gas reaches the surface of the core. However, the high porosity of the ash shell means that the gas can diffuse through it. Heat and mass transfers are affected considerably by shrinkage (Peters *et al.* 2012). Smaller dimensions mean that heat and mass reach the center of a shrinking particle at a faster rate. As the reaction proceeds, each of these grains (with assumed spherical shape) exhibits continuous reduction in core diameter (Fermoso *et al*. 2008). The overall reaction rate following from Eq. 1 (Tangsathitkulchai *et al*. 2013) is given as,

$$\frac{d\alpha}{dt} = K_{SCM}(1-\alpha)^{2/3} \tag{6}$$

where k_{SCM} denotes the reaction rate constant.

Using Eqs. 1, 4, and 6, the char activation reaction rate for the SCM is:

$$\frac{d\alpha}{dt} = Ae^{-(E/RT)}P_{steam}^{n}\,(1-\alpha)^{2/3} \tag{7}$$

With the surface area decreasing as the reaction proceeds, the model also predicts a decreasing rate of reaction. A linearized version of Eq. 6 after the separation of variables and integration gives:

$$3[1-(1-\alpha)^{1/3}] = K_{SCM}t \tag{8}$$

Volume Reaction Model (VRM)

With conversion, the VRM assumes that the reaction surface area of the char decreases linearly, with a preceding homogeneous reaction throughout the particle. The implication here is that the gas reacts with the particle in all directions. Considering the assumption that the reactions between the char and the gasifying agents occur at all active sites (distributed uniformly in and around the particle surface), the volumetric model does not consider the structural changes of the char during the gasification process (Zhang *et al*. 2010). Assuming that the gas reacts with the particle in all directions, it follows that the density of the reacting particle will be changed (Zhang *et al.* 2014). The overall rate of reaction is expressed as,

$$\frac{d\alpha}{dt} = K_{VRM}(1-\alpha) \tag{9}$$

where k_{VRM} denotes the reaction rate constant.

The char activation reaction expression for VRM is given as:

$$\frac{d\alpha}{dt} = Ae^{-(E/RT)}P_{steam}^{n}\,(1-\alpha) \tag{10}$$

Integration of the first order reaction rate (Eq. 9) yields the following linear expression:

$$\ln(1-\alpha) = K_{VRM}t \tag{11}$$

Modified Volume Reaction Model (mVRM)

As the gasification reaction proceeds, the modified volume reaction model assumes that the apparent rate constant changes with the solid conversion (α). The reaction rate can be expressed by the following equation,

$$\frac{d\alpha}{dt} = K(\alpha)(1-\alpha) \tag{12}$$

where $k(\alpha)$ is the apparent rate constant at conversion α.

For the purpose of comparison with other models, an average rate constant K_{mVRM} (as an index of reactivity) for this model can be obtained (Murillo *et al*. 2004):

$$K_{\,mVRM} = \int_{0.01}^{0.99} K(\alpha)d\alpha \tag{13}$$

The quantity $k(\alpha)$ can be calculated using the integrated form of the equation,

$$K(\alpha) = a^{1/b}b[-ln(1-\alpha)]^{(b-1/b)} \tag{14}$$

where a and b are modified empirical model parameters with no physical meaning (Tomaszewicz *et al*. 2013).

The time-conversion equation for this model is (Sun *et al.* 2007):

$$at^b = -\ln(1-\alpha) \tag{15}$$

For the purpose of use with experimental data, Eq. 15 can be modified to obtain:

$$ln[-ln(1-\alpha)] = ln\alpha + blnt \tag{16}$$

RESULTS AND DISCUSSION

Impact of Operating Parameters

In this study, steam gasification reactions of char from pyrolysed ayinre wood were performed at different steam pressures and gasification temperatures. A conversion interval of 0.1 to 0.9 was chosen in order to avoid problems associated with reaction start-up and approaching the end point. From Table 2 it is clear that changes in steam partial pressure and gasification temperature had effects on the char conversion rate constants. In essence, the char conversion rate constant increased with steam partial pressure (minimally) and gasification temperature (more pronounced).

Table 2. Evaluated Char Gasification Rate Constants K_{VRM} (min^{-1}) Using Gasification Temperatures and Steam Partial Pressures as Paired Reaction Variables

Temperature (°C)	Pressure			
	0.02 MPa	0.03 MPa	0.04 MPa	0.05 MPa
700	0.0486	0.0559	0.0725	0.0747
800	0.1082	0.0937	0.1987	0.1882
900	0.1652	0.1435	0.3123	0.2389
1000	0.2420	0.2475	0.2942	0.3284

The observed changes in the char conversion rate constant with the steam partial pressure could be related to an alteration in the flux of water molecules (steam) to the char's outer surfaces. Figure 2 shows the effect of applying different temperatures at the same steam partial pressure of 0.04 MPa. At higher temperatures, the conversion was faster. The fastest reaction occurred at the highest temperature of 1000 °C.

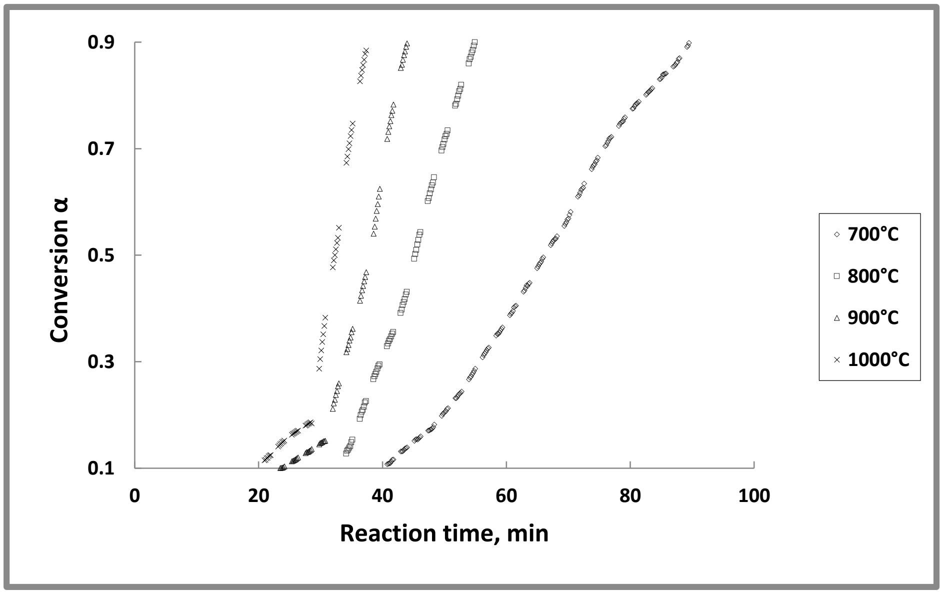

Fig. 2. A typical conversion versus reaction time plot for steam gasification of ayinre char

Gasification Behavior Described by the Models

To examine the steam gasification behavior using the three models, a partial steam pressure of 0.04 MPa was chosen at random. Figures 3, 4, and 5 show how the models fit the conversions.

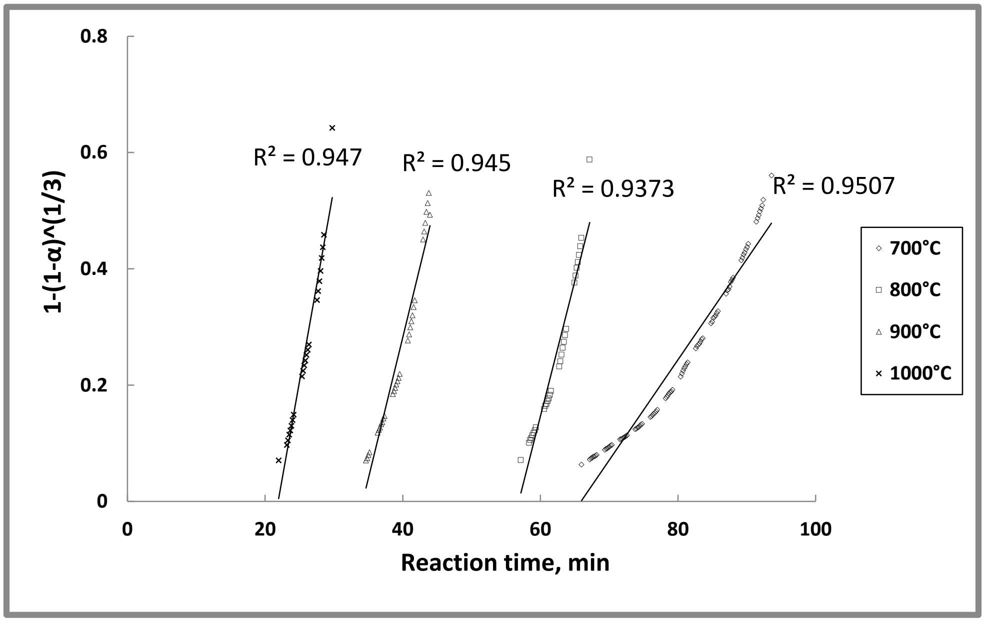

Fig. 3. An SCM conversion *versus* reaction time plot for steam gasification of ayinre char

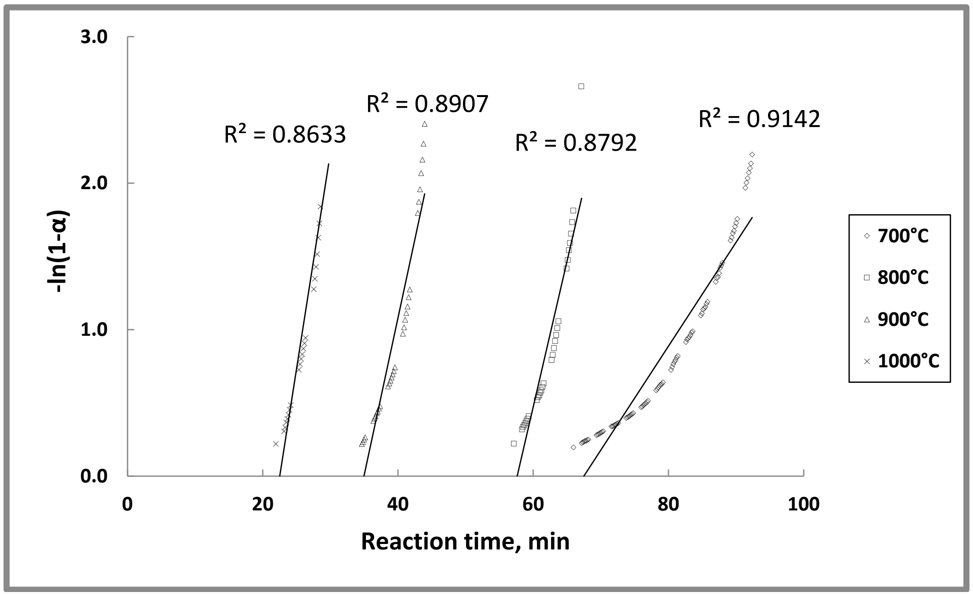

Fig. 4. A VRM conversion *versus* reaction time plot for steam gasification of ayinre char

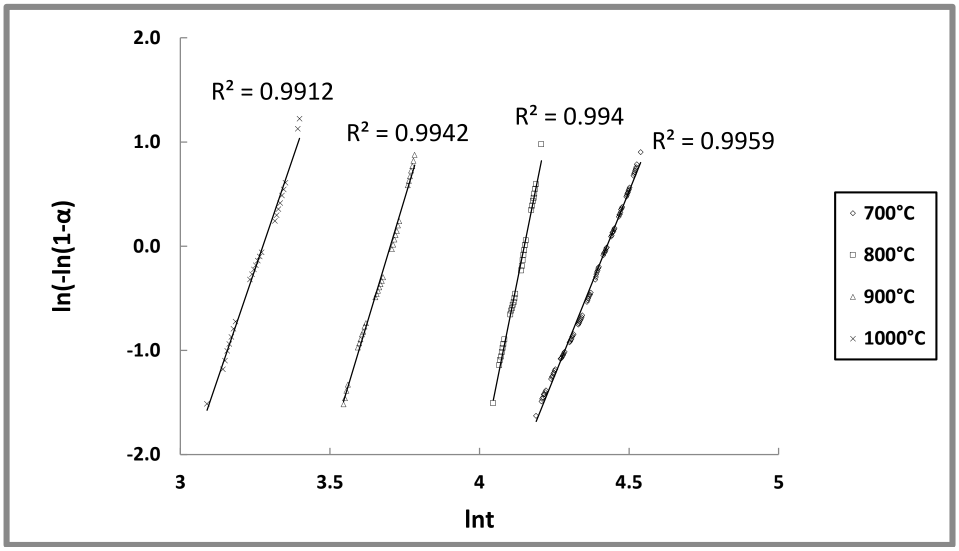

Fig. 5. An mVRM conversion *versus* reaction time plot for steam gasification of ayinre char

Of the three models, mVRM had the most perfect fit, with an R^2 value (from the trend lines) of 0.9959. With some exceptions from the VRM (shown in Fig. 4) with R^2 values less than 0.9 (which was insufficient), all others showed a good fit to the experimental data. Various corresponding reaction constant rates were determined from the plots, as well as from the values calculated from the plots involving models. Figure 4 shows an example of the array of the reaction rate constant K (min^{-1}) from the VRM-treated experimental data.

Determination of Reaction Kinetic Parameters

The kinetic parameters determined included the activation energy E_a, the kinetic rate constant K, and the pre-exponential factor A. For example, an Arrhenius plot using a steam partial pressure of 0.03 MPa during gasification section is shown in Fig. 6. The activation energy was calculated from the slope of the straight line, and the pre-exponential factor was determined by the intercept. This calculation was valid for all models used in this study; a complete compilation of the values is shown in Table 3. A combined Arrhenius plot of the linear and the calculated (model-fitting) data using a steam partial pressure of 0.030 MPa is shown in Fig. 7.

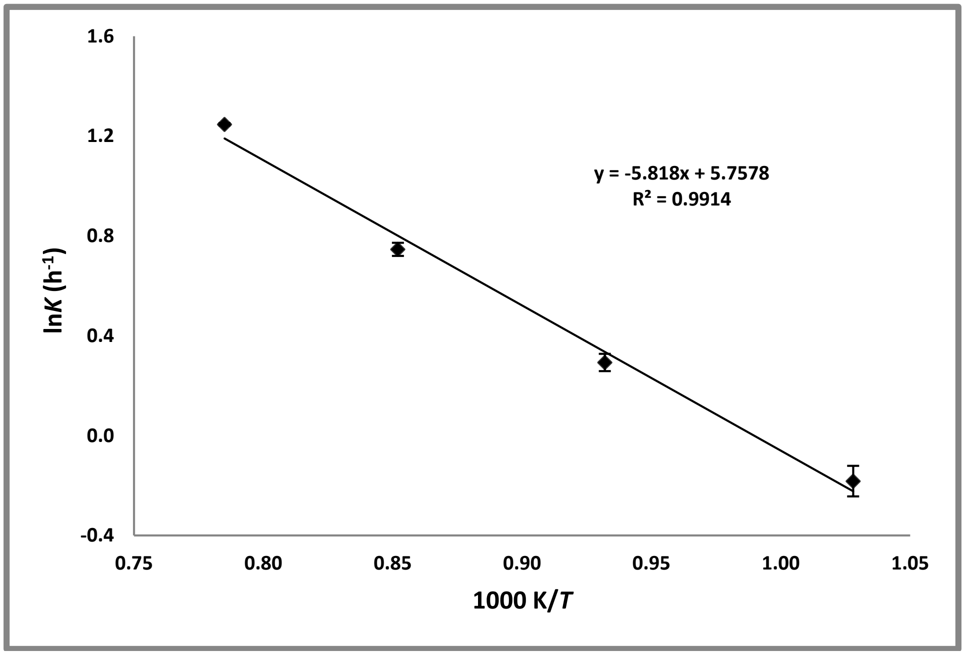

Fig. 6. An Arrhenius plot of experimental data for the steam gasification reaction of ayinre char at a partial pressure of steam of 0.03 MPa using the SCM

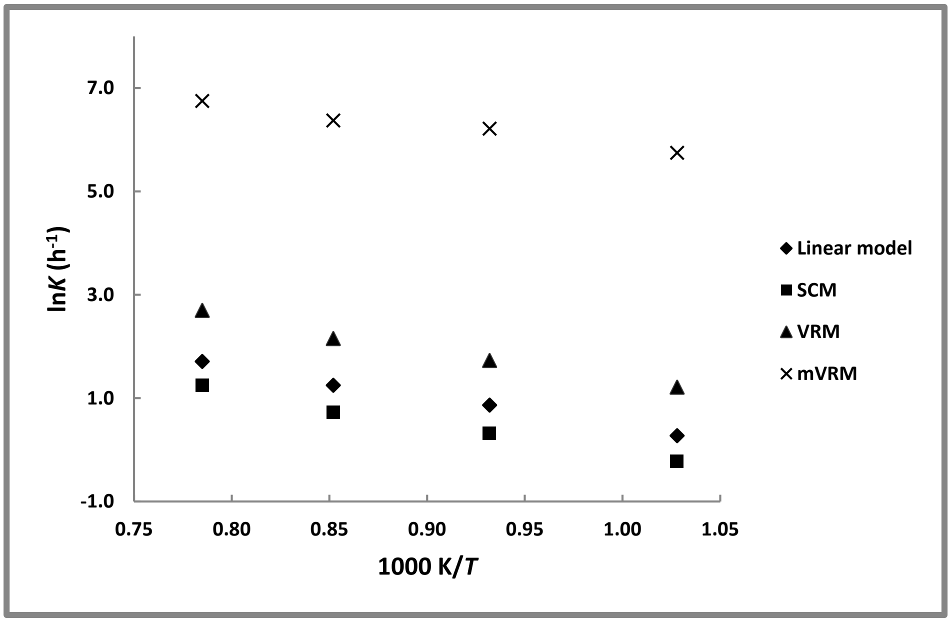

Fig. 7. A combined Arrhenius plots for both linear and calculated (model-fitting) data for ayinre char conversion at a partial pressure of 0.030 MPa

Table 3. The Activation Energy E_a (kJ/mol) and Pre-exponential Factors *A* (h^{-1}) for Steam Gasification of Wood Char Using Experimental and Calculated (Model-fitting) Data from Different Models

Data Type	E_a (kJ/mol)	*A* (h^{-1})
Experimental	48.13	5.09E + 02
SCM	49.29	3.45E + 02
VRM	49.89	1.55E + 03
mVRM	32.54	1.79E + 04

Following from Eq. 5, the relationship between the average reaction rate and the partial pressure of steam was as shown in Fig. 8 for experimental data at 700 °C. The reaction order n was 0.58, and the R^2 was 0.97. Considering calculated (model-fitting) data with an example in the mVRM at 700 °C, the value of n was estimated at 0.54, and the R^2 value a low 0.73, indicating a bad fit for the model.

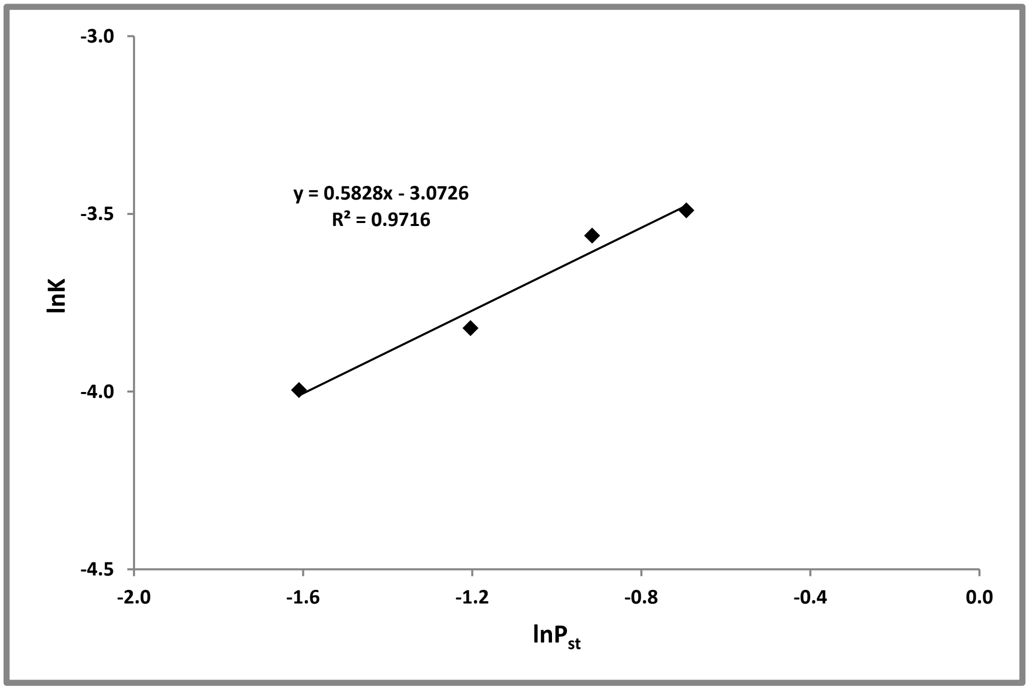

Fig. 8. A natural logarithm plot of the average reaction rate *versus* partial pressure of steam using experimental data at 700 °C

Several n values have been reported for gasification reactions of different materials. An n value of 0.58 was recorded for filter carbon at 850 °C (Sun *et al*. 2007). Gasification of Japanese cedar char in an entrained-flow gasifier with steam and oxygen between 900 and 1000 °C gave an n value of 0.22 (Matsumoto *et al*. 2009). Steam gasification of grapefruit char that was pyrolysed for 1 to 2 h between 900 and 1000 °C gave exponent numbers between 0.5 and 0.6 (Marquez-Montesinos *et al*. 2002). Gasification of birch char in a steam atmosphere between 1023 and 1223 °C gave an n value of 0.57, while beech wood gave 0.51 as the exponential number under the same conditions (Barrio *et al*. 2008). However, according to Matsumoto *et al*. (2009), the result from the Japanese cedar indicating low reactivity with an n of 0.22 was possibly due to the low concentration of alkali metals in the feedstock ash and also to high O/C ratio in the biomass.

Using Eq. 4, the kinetic expression of the reaction rate of steam gasification of ayinre char is given as:

$$\frac{d\alpha}{dt} = 1.79 \times 10^4 e^{-32540/8.314T} P_{steam}^{0.58} \quad (17)$$

The modeled kinetic expression (Eq. 17) will ultimately assist in the design and configuration of boilers most suitable for the optimal conversion of energy from tropical wood biomass.

CONCLUSIONS

1. The use of steam gasification as an alternative for thermal conversion was investigated using a thermo-gravimetric balance. By varying the temperature of the gasification and the partial pressure of the steam, it was observed that the conversion reaction of the char was enhanced by either an increase in the partial pressure of steam and/or the gasification temperature.
2. Widely-used gas-solid reaction models, including SCM, VRM, and mVRM, were employed to determine the best conversion possible during the course of a reaction. The mVRM showed to have the best fit, with the lowest activation energy E_a of 32.54 kJ/mol and a pre-exponential factor A of 1.79E + 04 h^{-1}.
3. The reaction order with respect to steam was 0.58, which was close to published results.
4. Finally, the kinetic expression of the reaction rate of steam gasification of ayinre char using mVRM was derived.

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