



Original Research Article

Modelling and Simulation of Biomass Pyrolysis in Thermally Thin Regime

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ABSTRACT

Economic and environmental considerations regarding the use of fossil fuels have brought about the need for renewable energy sources like biomass through pyrolysis. However, thermal regimes (thin, thick and thermal wave) have been lumped up while studying biomass pyrolysis, causing ambiguity in results interpretation and process optimization. This work simulated biomass pyrolysis in thermally thin regime and investigated the effect of process parameters on products evolution and total yields over a wide range of reactor temperatures and heating rates. A chemical kinetic model was adopted, resulting in the development of five ordinary differential equations. Arrhenius rate equation of the first order was used to estimate the reaction rate constants for all the species. All equations were coupled and solved simultaneously using Euler numerical method with a time step of 0.001 s to simulate products evolution. Total yields of product species were obtained by trapezoidal rule. Tar evolution increased with heating rate but declined after reaching its peak until the end of the process. Gas evolution at low and high reactor temperatures increased with heating rate while increase in reactor temperature did not significantly influence gas evolution from 5 to 20 K/s. Char yield decreased with increase in both reactor temperature and heating rate. Maximum tar, gas and char yields were 70.536 (at 1 K/s and 673 K), 31.944 (at 20 K/s and 773 K) and 19.508% (at 1 K/s and 373 K), respectively. The conditions that will optimize the yields of product species in this thermal regime are presented.

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1. INTRODUCTION

An emerging challenge of the 21st century is to seek alternative fuels to address concerns over depletion of fossil fuels resources and the increasing amount of greenhouse gases released to the atmosphere (Mohan *et al.*, 2006). The environmental awareness related to carbon dioxide and other greenhouse gases emissions as

well as concern over the ultimate availability of fossil fuels have increased researchers' interest in using biomass as a renewable resource for chemical feedstock and energy production (Grønli and Melaaen, 2000).

Research and development in thermochemical conversion of biomass have been continuously attracting researchers across the globe, due to the global scare which is evident in the continuous instability of petroleum sector worldwide (Fapetu, 2000; Goyal *et al.*, 2006; Lam *et al.*, 2012; Okekunle *et al.*, 2013). In recent years, these issues have motivated both industrial and academic sectors to examine the feasibility of renewable energy utilization through large scale national and international projects (Painuly and Fenhann, 2002; Park, 2008; Kouei industries, 2011). Biomass, among various alternatives, has become an attractive renewable energy source. This can be credited to its versatility in storage and easy transformation into electricity and heat (Bridgwater, 2003). Supply of energy from biomass plays an increasing role in the debate on renewable energy (Yaman, 2004). The relatively large amount of biomass already used for energy generation reflects mainly the use of wood in the developing countries. This is because their economies are largely based on agriculture and forestry (Vigouroux, 2001). Many works have been done on pyrolysis process (Di Blasi, 2002; Branca and Di Blasi, 2003; Shen *et al.*, 2007; Dupont *et al.*, 2009) but little attention has been paid to distinguishing between the various thermal regimes under which such works were done. This no doubt has led to confusion in proper interpretation of results and understanding of pyrolysis in various thermal regimes. Previous works on thermally thin regime pyrolysis introduced ambiguous orders of reactions in chemical kinetics and failed to clearly depict the effect of heating rates (Srivastava and Jalan, 1994; Srivastava *et al.*, 1996).

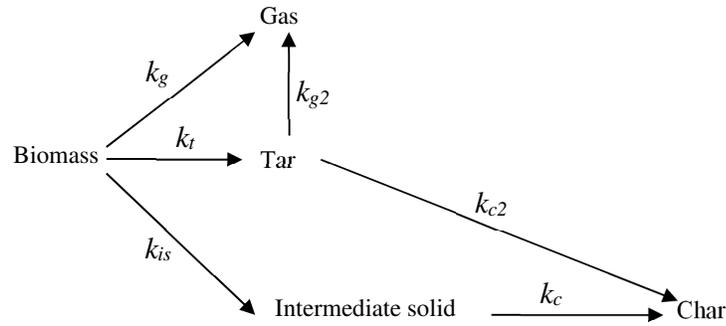
To fully understand the characteristics of biomass conversion at operational conditions relevant to those found in real furnaces, numerical simulations were carried out (Okekunle *et al.*, 2011; Okekunle, 2013; Okekunle and Adeniranye, 2015; Okekunle and Adeoye, 2016). The primary idea is to get a deeper insight of the effects of some basic process parameters in the pyrolysis of biomass but results were not discussed vis-à-vis the thermal regime under consideration. In this study therefore, thermally thin regime pyrolysis of maple wood was modelled and simulated, and the effects of process parameters on products evolution and yields were also investigated.

2. METHODOLOGY

2.1. Pyrolysis Mechanism

Over the years, various kinetic models have been used for numerical investigation of the characteristics of biomass pyrolysis (Di Blasi, 2002; Mohan *et al.*, 2006; Park *et al.*, 2010). The pyrolysis mechanism adopted in this work was developed by Park *et al.* (2010). This is because it accounts for the observed endothermic and exothermic thermal behavior and the solid mass loss measured in their experiments. The model proposed that when a virgin biomass (in this case, maple wood) undergoes thermochemical reactions, it primarily decomposes to yield gas, tar and intermediate solid. The tar generated from primary pyrolysis participates in secondary reactions yielding more gas and char. The intermediate solid also further decomposes through an exothermic secondary reaction into char. The mechanism is illustrated in Figure 1.

In Figure 1, k_g , k_t , k_{is} , k_c , k_{g2} and k_{c2} are formation reaction rate constant for gas, tar, intermediate solid, char, secondary gas and secondary char, respectively.

Figure 1: Schematic illustration of pyrolysis mechanism (Park *et al.*, 2010)

2.2. Numerical Simulation

Based on Figure 1, the mass conservation equations for various species were developed and presented as follows:

2.2.1. Mass conservation equation for solid species

The virgin biomass instantaneous mass balance equation contains three consumption terms, each for the reaction yielding gas, tar and intermediate solid, and is given as:

$$\frac{\partial \rho_w}{\partial t} = -(k_g + k_t + k_{is})\rho_w \quad (1)$$

Where ρ_w is the instantaneous density of the wood

The intermediate solid instantaneous mass balance equation (Equation 2) contains two terms, one for the decomposition of wood to yield intermediate solid and the other for the conversion of intermediate solid to yield char, and is given as:

$$\frac{\partial \rho_{is}}{\partial t} = k_{is}\rho_w - k_c\rho_{is} \quad (2)$$

Where ρ_{is} is the instantaneous density of the intermediate solid

Similarly, the mass balance equation for char yield (Equation 3) contains two terms, one for the conversion of intermediate solid to char and the other for the transformation of tar to char, and is given as:

$$\frac{\partial \rho_c}{\partial t} = k_c\rho_{is} + k_{c2}\rho_t \quad (3)$$

Where ρ_t is the instantaneous density of tar

2.2.2. Mass balance equation for gas

The gas instantaneous mass balance equation (Equation 4) has two terms, one for the conversion of the virgin biomass solid (wood) to gas and the other from tar secondary reaction to yield gas.

$$\frac{\partial \rho_g}{\partial t} = k_g \rho_w + k_{g2} \rho_t \quad (4)$$

ρ_g is the instantaneous density of the gas.

2.2.3. Mass balance equation for tar

The tar instantaneous mass balance equation (Equation 5) contains three terms, the first for the conversion of virgin biomass to tar, the others for the conversion of tar to gas and char.

$$\frac{\partial \rho_t}{\partial t} = k_t \rho_w - k_{g2} \rho_t - k_{c2} \rho_t \quad (5)$$

Equations (1) to (5) are the ordinary differential equations describing the mass change of biomass and its products based on the pyrolysis mechanism shown in Figure 1. The reaction rates were assumed to follow Arrhenius rate expression of the form:

$$k_i = A_i \exp\left(-\frac{E_i}{RT}\right) \quad (6)$$

Where A_i , E_i , R and T are the activation pre-exponential factor, activation energy, universal gas constant and temperature, respectively and $i = g, t, is, c, c2, g2$.

The kinetic parameters used were obtained from Shen *et al.* (2007) and are presented in Table 1. In Table 1, t, g, is, c, c2 and g2 are the formation reactions for the tar, gas, intermediate solid, char, secondary char and secondary gas, respectively.

Table 1: Kinetic parameters

Reaction (<i>i</i>)	A_i (s^{-1})	E_i (J/mol)
t	1.08×10^{10}	148,000
g	4.38×10^9	152,700
is	3.75×10^6	111,700
c	1.38×10^{10}	161,000
c2	1.0×10^5	108,000
g2	4.28×10^6	108,000

2.2.4. Numerical solution

The ordinary differential equations (Equations 1-5) were solved simultaneously by applying Euler explicit method to simulate biomass disintegration and products evolution. Simulation time step was 0.001 s. The process was terminated when biomass apparent density was reduced to 0.0003 kg/m³. Products yields were then estimated by trapezoidal rule.

3. RESULTS AND DISCUSSION

3.1. Temperature and Heating Rate

Figure 2(a-e) shows the conversion profiles of the pyrolyzed solid (maple wood) at different heating rates (1, 5, 10 and 20 K/s) and reactor temperatures (373, 473, 573, 673 and 773 K). As seen from the figure, the biomass sample at all reactor temperatures and heating rates was almost 100 % converted. This was because

the conversion process was terminated when biomass apparent density became very small (0.0003 kg/m^3). It can also be clearly seen that increase in heating rate enhanced the decomposition rate of biomass at all the reactor temperatures considered. However, commencement of significant biomass decomposition took a longer time from 373 to 573 K at 1 K/s. This was because a longer time was required for particle heat-up before the initiation of primary decomposition reactions. Moreover, the figures also show that the biomass conversion profiles for all heating rates at 373 and 473 K were similar. This implies that at these temperatures, there may be no significant difference in the rate of biomass primary decomposition reactions. Increase in reactor temperature to 573 K caused the biomass decomposition to commence earlier than at 373 and 473 K. Meanwhile, at reactor temperatures of 673 and 773 K, the decomposition process commenced immediately. This is evident in Figures 2(d) and 2(e) where high temperatures caused the biomass sample to heat-up rapidly. This accelerated the initiation of sample decomposition. Increasing the heating rate at 773 K has a little effect on the process (Srivastava *et al.*, 1996).

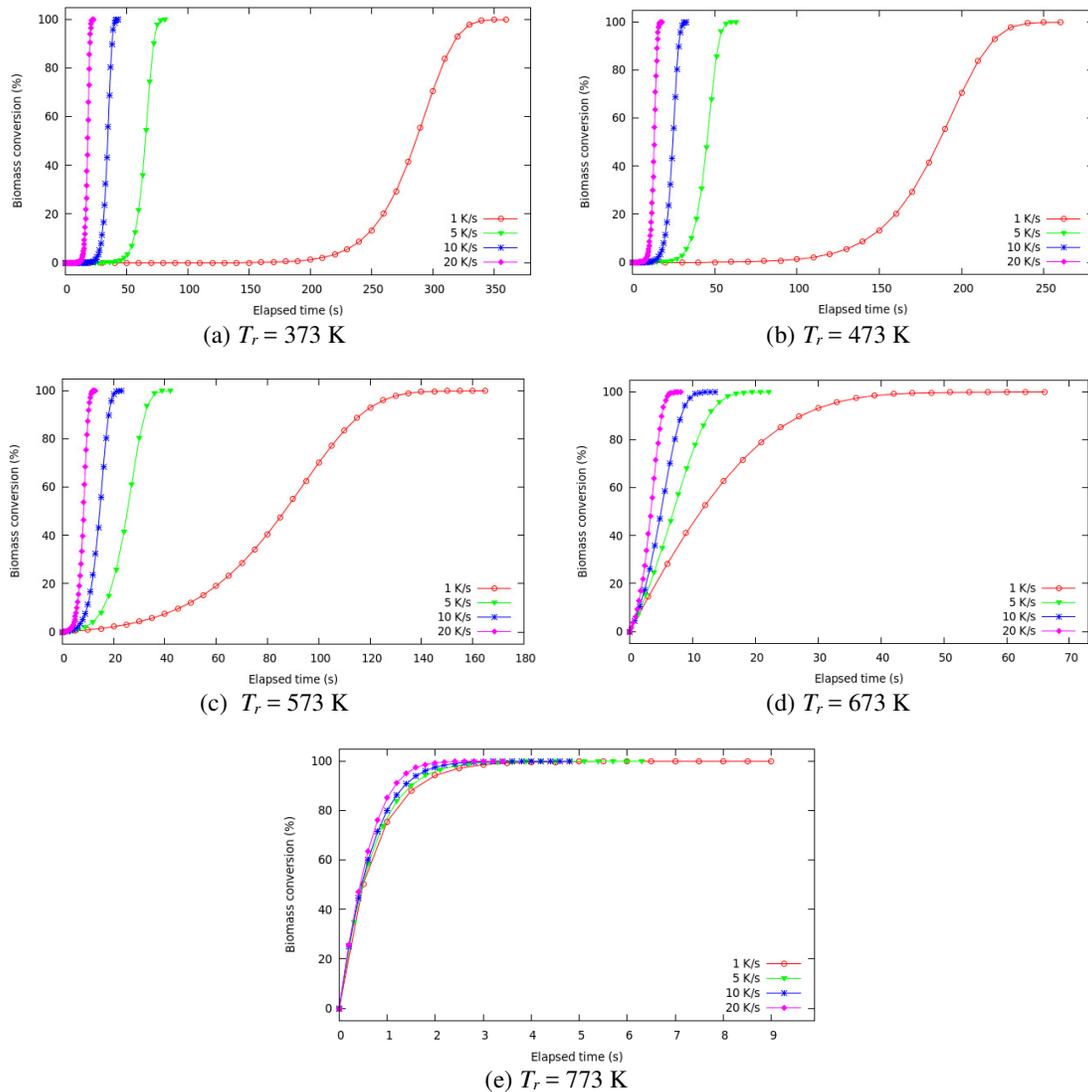


Figure 2: Biomass conversion at different heating rates and reactor temperatures

3.2. Tar Evolution

Figure 3(a-e) shows tar evolution at different heating rates and reactor temperatures. At all reactor temperatures considered, tar evolution increased with increase in heating rate. Furthermore, tar evolution, after reaching its peak in each case declined. The decline could be as a result of reduction in tar production from primary pyrolysis and volatiles secondary reactions to yield more gas (Park *et al.*, 2010; Okekunle *et al.*, 2013).

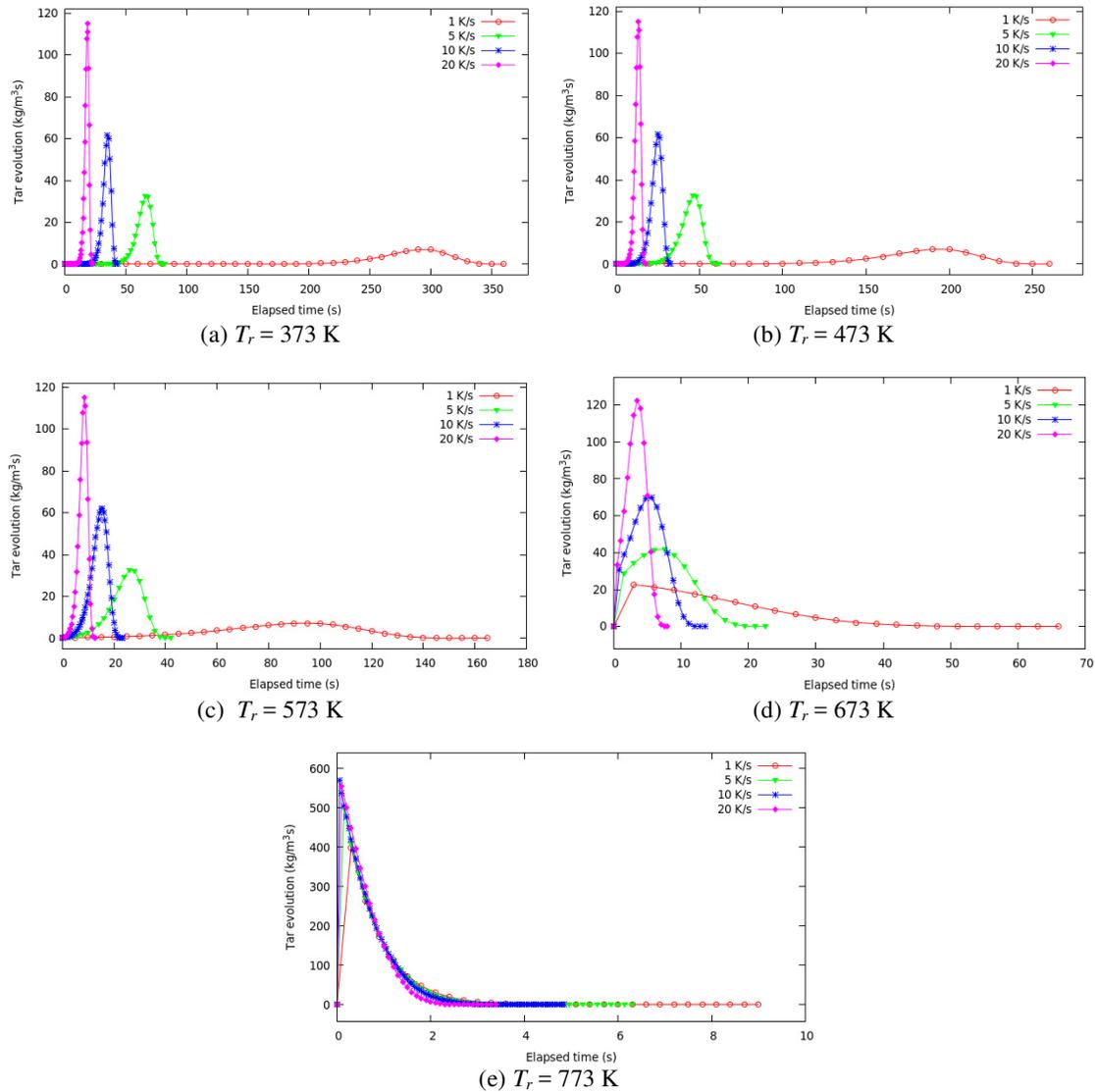


Figure 3: Tar evolution at different heating rates and reactor temperatures

It could also be seen from the figures that the tar evolution period decreased with increase in heating rate. Tar evolution took a longer time before commencement especially at 1 K/s for 373 and 473 K reactor temperatures but a slight decrease in time of evolution start-up was observed at 573 K. At a reactor temperature of 673 K, increase in heating rate further enhanced the rate of biomass decomposition, thereby

accelerating the rate of tar evolution. Tar production commenced with a rapid shoot within 1 s before a steady increase and declination on reaching its peak was noticed at all heating rates. The same trend was noticed at 773 K. In fact, tar evolution has already reached its peak before 1 s. It can be deduced that heating rate did not significantly affect the commencement of tar evolution process at 773 K. This is because the temperature was high enough to initiate the primary pyrolysis almost immediately. This suggests that at high temperatures, regardless of the heating rate, tar evolution peaks do not increase significantly with increase in heating rate. This result agrees with the findings of Okekunle *et al.* (2015).

3.3. Gas Evolution

Figure 4(a-e) shows gas evolution at different heating rates and reactor temperatures.

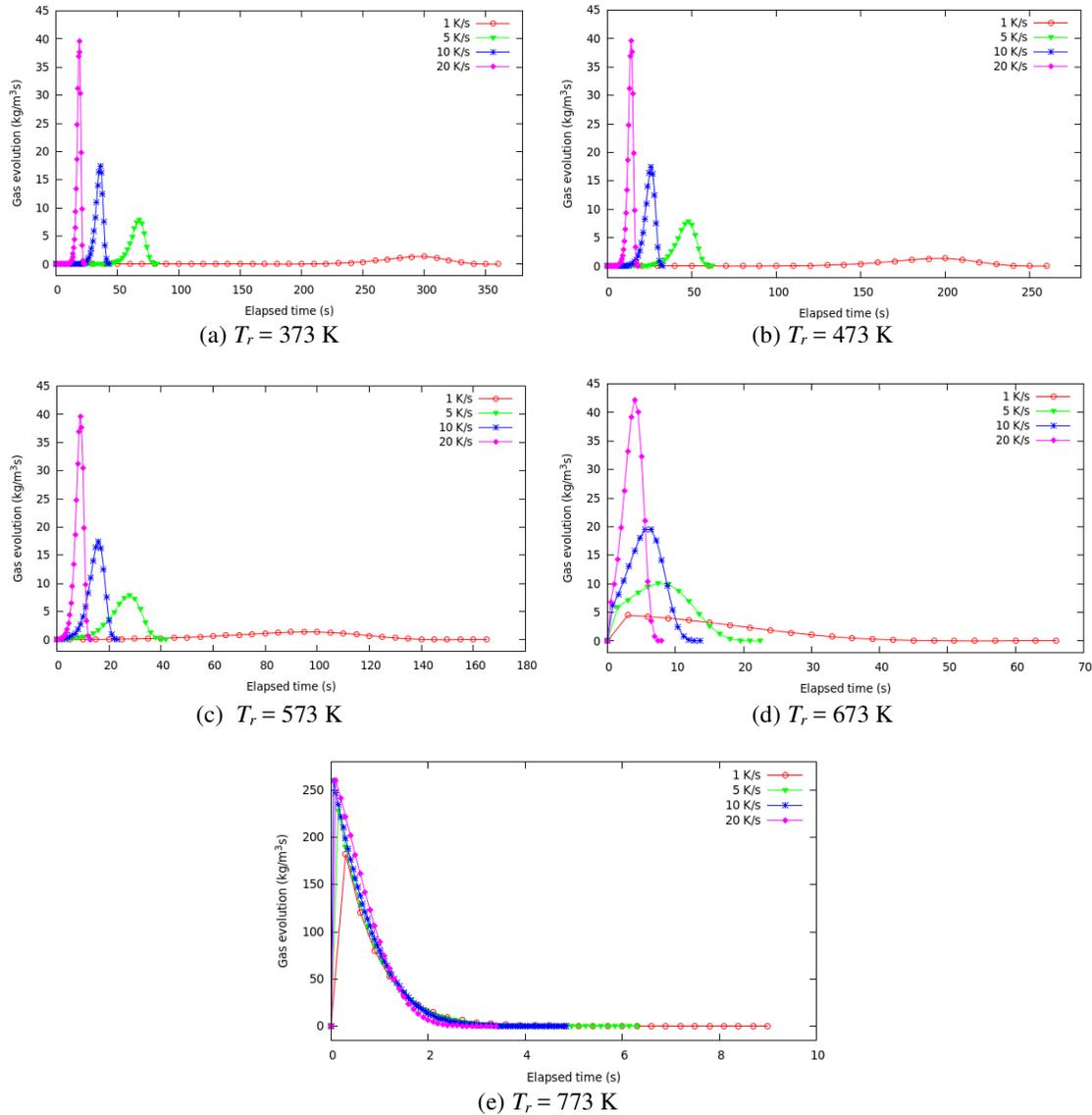


Figure 4: Gas evolution at different heating rates and reactor temperatures

At all reactor temperatures considered, gas evolution increased with increase in heating rate while evolution period decreased with the latter. As the reactor temperature increased, the time lag between gas evolution at different heating rates became shorter. Gas evolution commencement took a longer time at 1 K/s for 373, 473 and 573 K reactor temperatures. At a reactor temperature of 673 K, gas production commenced with a brisk shoot within 1 s before a steady inclination and declination on reaching its peak were noticed at all heating rates. This is because temperature increase hastened gas evolution from biomass primary pyrolysis by accelerating sample conversion and favouring gas evolution kinetics. The same trend was noticed at 773 K. In fact, gas evolution has already reached its peak before 1 s. It can be deduced that increase in heating rate did not significantly affect the commencement of gas evolution at 773 K. This is because the temperature was high enough to initiate biomass conversion to gas almost immediately. This suggests that at high temperatures, regardless of the heating rate, gas evolution occurs rapidly. This is in agreement with the work of Okekunle *et al.* (2015).

3.4. Intermediate Solid Formation

Figure 5(a-e) shows the rate of intermediate solid formation at different heating rates and reactor temperatures. At all reactor temperatures considered, the formation rate increased with increase in heating rate. Intermediate solid evolution, after reaching its peak declined. The declination was as a result of its secondary reactions to yield char. Furthermore, the evolution period of this species decreased with increase in heating rate because biomass disintegration was accelerated. Intermediate solid formation initiation took a longer time at 1 K/s for 373 and 473 K reactor temperatures because more time was needed to heat the sample up to decomposition temperature. At reactor temperatures of 573 and 673 K, increase in heating rate further enhanced the rate of biomass decomposition, thereby accelerating the rate of intermediate solid formation which commenced with an invigorating shoot within 1 s before a steady increase and declination on reaching its peak at all heating rates. The same trend was noticed at 773 K. It can be inferred that heating rate did not significantly affect the commencement of the intermediate solid formation process at 773 K. This suggests that at high temperatures, regardless of the heating rate, intermediate solid formation occurs rapidly. According to the mechanism shown in Figure 1 and Equation 2, the initial rise in intermediate solid evolution with respect to time was due to its formation from biomass degradation while the subsequent decrease was due to its conversion to char. Besides, as biomass disintegration continued, the rate of formation of products would also decrease.

3.5. Char Formation

Figure 6(a-e) shows the rate of char formation at different heating rates and reactor temperatures. At all reactor temperatures considered, char formation rate increased with increase in heating rate. Furthermore, char formation, after reaching its peak declined. However, as the temperature increased, the time lag between char evolution at different heating rates became shorter. A longer time was required for commencement of char evolution at 1 K/s for 373 and 473 K reactor temperatures. At reactor temperatures of 573 and 673 K, temperature increase enhanced primary pyrolysis and secondary reactions. Hence, char formation commenced within 1 s and progressed steadily before declining after reaching its peak at all the heating rates considered. The same trend was noticed at 773 K. It can be deduced that heating rate does not significantly affect the commencement of char formation process at 773 K. At the initial stage, when biomass concentration is high, the rate of char formation from biomass will also be high. This is the reason for a steady increase in char formation at the initial stage. However, as biomass disintegration continues, its concentration drops and so also the rate of char formation, hence, the subsequent decrease.

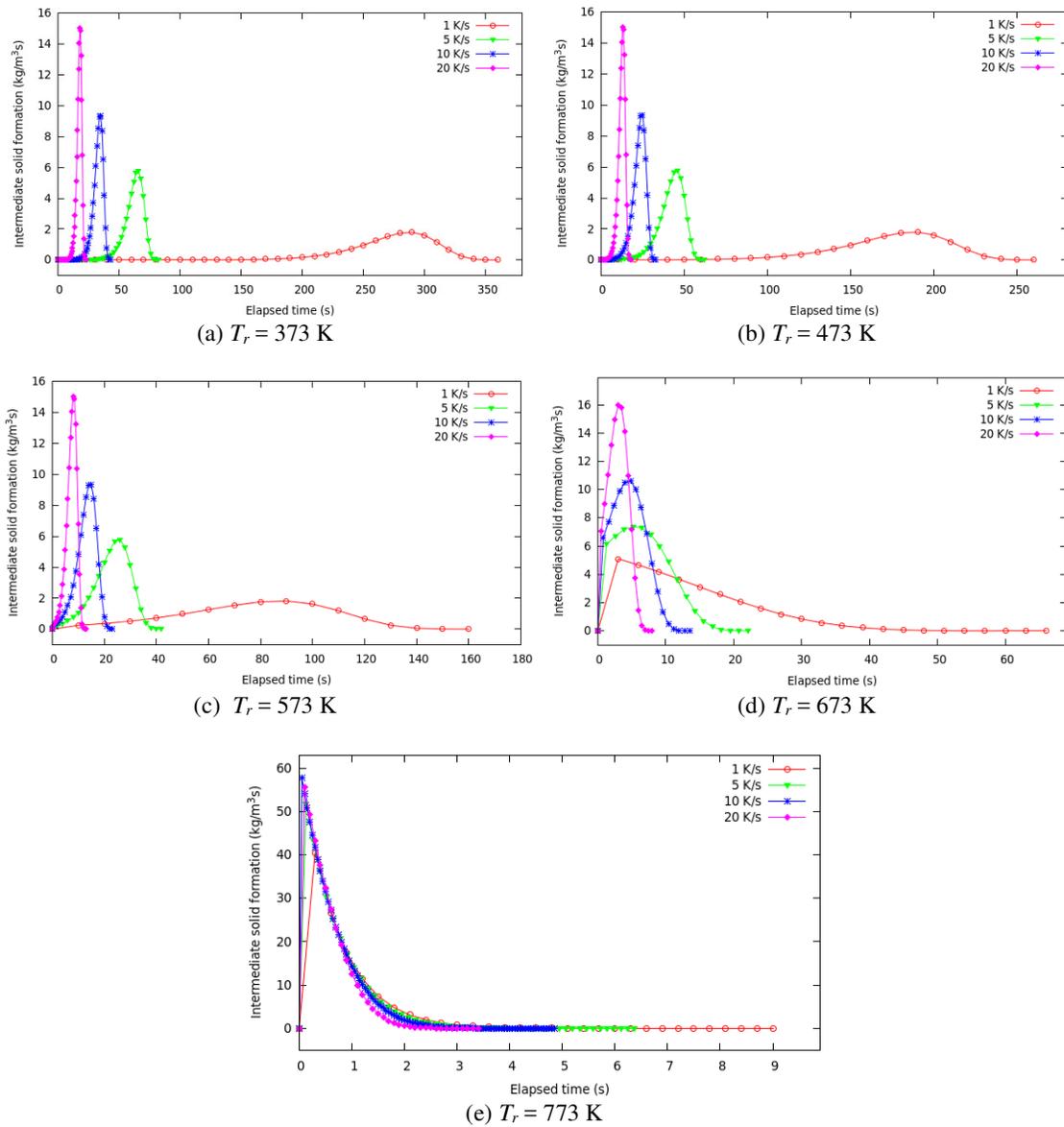


Figure 5: Intermediate solid formation at different heating rates and reactor temperatures

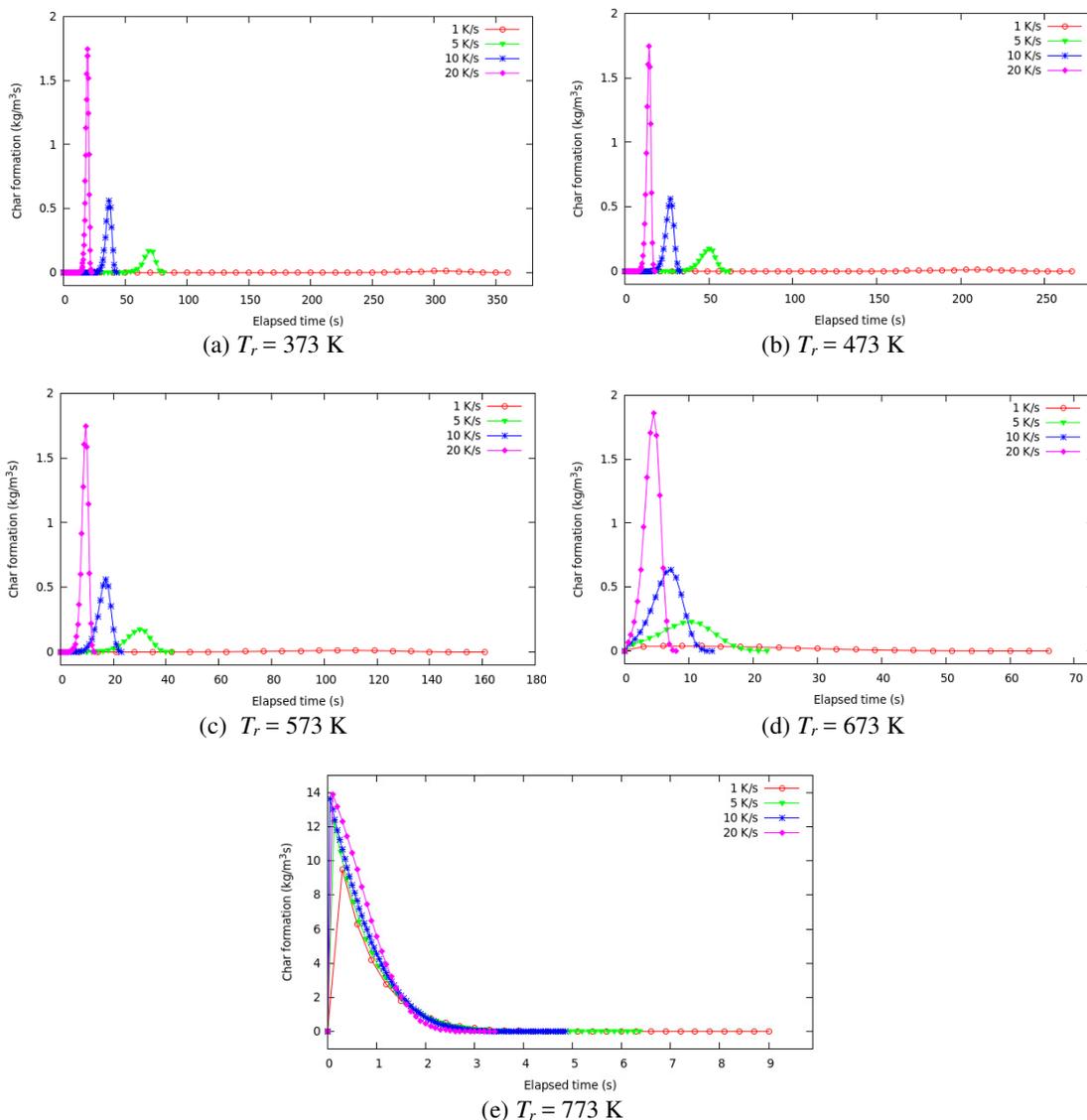


Figure 6: Char formation rate at different heating rates and reactor temperatures

3.6. Total Tar Yield

Figure 7 shows the total tar yields at different reactor temperatures and heating rates. From the figure, tar yield at reactor temperatures of 373, 473 and 573 K increased with increase in heating rate from 1 to 5 K/s. It reached its peak at 5 K/s and took a downward toll from 5 - 20 K/s. This is because tarry compounds participated in secondary reactions to yield non-condensable gas and possibly char through repolymerisation (Okekunle *et al.*, 2011). At 673 K, the tar yield decreased with increase in heating rate. This is also because volatile secondary reactions were facilitated (Okekunle *et al.*, 2011). It is expected that this decrease in tar yield will be accompanied with corresponding increase in gas yield. The same trend was observed when the reactor temperature increased to 773 K. Furthermore, as shown in the figure, temperature increase from 373 to 473 K has no significant effect on total tar yields. This result suggests that despite the temperature increase, tar production chemical kinetics do not change significantly. A slight increase in the yield was however

observed at 573 K for 1 K/s heating rate. At 673 K, the temperature was high enough to accelerate volatiles secondary reactions. Reactor temperature of 773 K was seen to favour tar molecules secondary reactions. Hence, tar yield dropped significantly and continuously from 1 to 20 K/s at 773 K. Maximum tar yield of 70.536 % was obtained at reactor temperature of 673 K and 1 K/s, while the minimum was 60.577 % at a temperature of 773 K and 20 K/s. These results agree with the findings of Dupont *et al.* (2009).

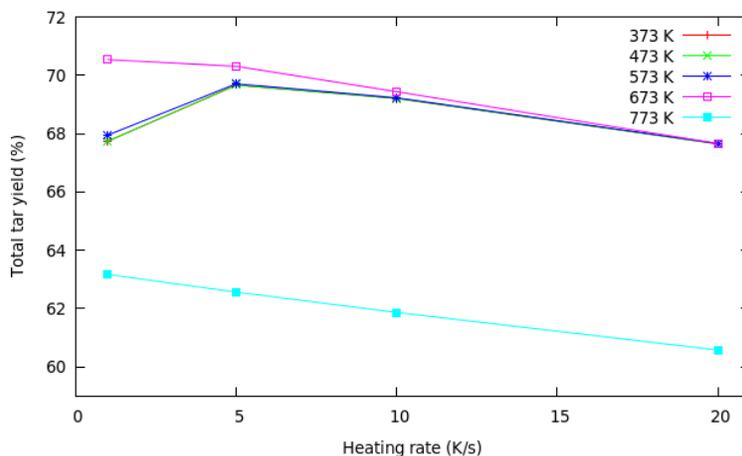


Figure 7: Effects of reactor temperature and heating rate on total tar yield

3.7. Total Gas Yield

Figure 8 shows the total gas yield at different reactor temperatures and heating rates. From the figure, total gas yields for all the reactor temperatures increased with increase in heating rate. Furthermore, as shown in the figure, temperature increase between 373 and 573 K has no effect on total gas yields at all heating rates. This result suggests that gas production chemical kinetics do not change significantly, despite the temperature increase. At 673 K, the temperature was high enough to favour gas production from primary pyrolysis and facilitated tar secondary reactions, hence yielding more gas. Reactor temperature of 773 K was significantly high, such that gas yield has maximum values for all heating rates considered. This is because the temperature was high enough also to favour biomass conversion to gas and tar secondary reactions to produce more gas. Maximum gas yield of 31.944 % was obtained at reactor temperature of 773 K and 20 K/s, while the minimum was 12.752 % at a temperature of 373 K and 1 K/s. Yaman (2004) and Dupont *et al.* (2009) have reported a similar trend in gas yield. Comparison of Figures 7 and 8 shows that total tar yield and total gas yield are inversely proportional (Okekunle and Adeniranye, 2015).

3.8. Total Char Yield

Figure 9 shows total char yields at different reactor temperatures and heating rates. From the figure, total char yield at all reactor temperatures decreased with increase in heating rate. A rapid decrease was observed from 373 to 573 K at 1 – 5 K/s but a steady reduction in char yield took place between 5 and 20 K/s. Furthermore, as shown in the figure, temperature increase between 373 and 573 K has no significant effect on total char yields. It is plausible that increase in temperature at this region has no significant effect on char formation kinetics. At 673 K, char yield reduced for all heating rates because biomass conversion to gas and tar was more favoured. A significant change was observed at a reactor temperature of 773 K, such that char yield has minimum values for all heating rates considered. This is because the temperature was high enough to convert most of the biomass to gas and tar. Maximum char yield of 19.508 % was obtained at reactor temperature of 373 K and 1 K/s heating rate, while the minimum was 7.479 % at a temperature of 773 K and

20 K/s. This result agrees with Park (2008), who reported that as pyrolysis temperature varies from 632 to 873 K, char yield ranges from 8.1 to 7.4%.

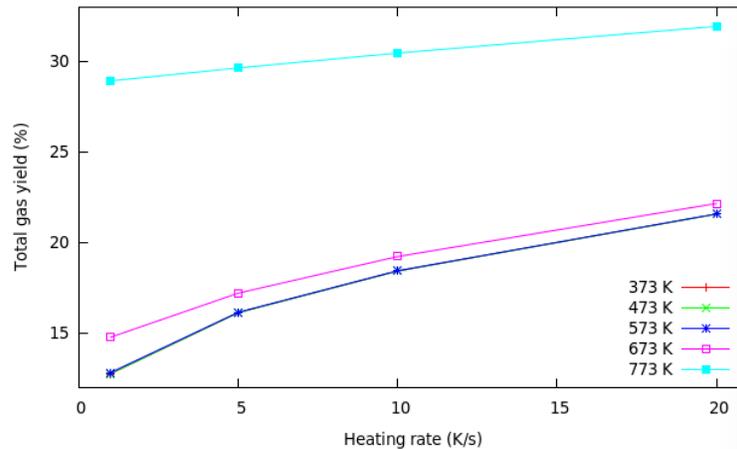


Figure 8: Effects of reactor temperature and heating rate on total gas yield

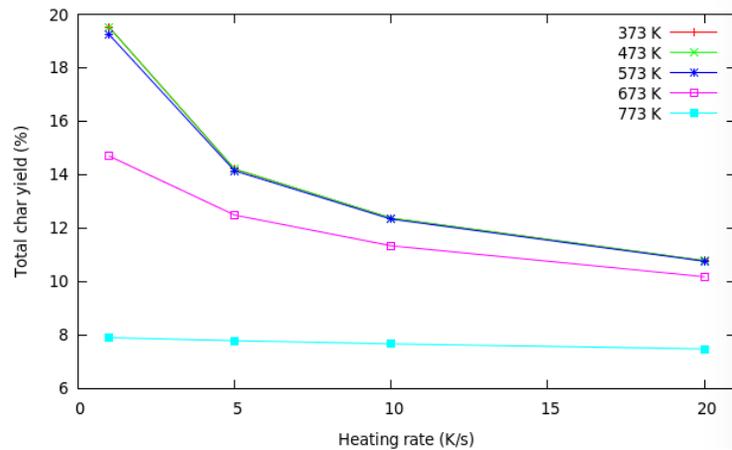


Figure 9: Effects of reactor temperature and heating rate on total char yield

4. CONCLUSION

Effects of reactor temperature and heating rate on the product yields of biomass pyrolysis in thermally thin regime have been investigated. A kinetic model, consisting of five ordinary differential equations was adopted. Results showed that increase in heating rate enhanced the rate of biomass conversion. Tar evolution increased with increase in heating rate at all the reactor temperatures considered. The evolution takes longer time when the heating rate was low compared to higher heating rates. Gas evolution at low and high pyrolysis temperatures increased with heating rate. Char formation kinetics is more sensitive to changes in heating rate between 373 and 573 K than at other temperatures considered. High reactor temperatures resulted (673 and 773 K) in much lower yield of char.

5. ACKNOWLEDGMENTS

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6. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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