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Kinetic Modelling for Tar Evolution and Formation in a Downdraft Gasifier

A. M. Salem, U. Kumar, and M. C. Paul*

Abstract— Biomass gasification modeling is a powerful tool used to optimize the design of a gasifier. A detailed kinetic model was built by the current authors [1] to predict the behavior of air blown downdraft gasifier for a wide range of materials within the range of ($38 \leq C \leq 52$) %, ($5.2 \leq H \leq 7$) %, and ($21.7 \leq O \leq 45$) %. The model was verified and showed a good stability for a wide range of working parameters like equivalence ratio and moisture content. In the current research, 4 main tar species are added to the model to represent tar formation using detailed kinetic reactions. The yield of tar species is discussed for different zones of a gasifier based on temperature of each zone. Mass and energy balance are calculated. 18 different kinetic reactions are implemented in the kinetic code to predict the optimum working conditions that leads to the production of higher value producer gas. Results conclude that using ER of 0.3 with moisture content levels lower than 10% will lead to the production of higher yields of syngas with lower amounts of tar.

Index Terms— Downdraft Gasifier, Kinetic modeling, Tar formation

I. INTRODUCTION

Biomass is one of the promising alternatives to fossil fuels. It is also considered to be a clean and renewable source of energy, and environmentally friendly. Biomass gasification is one of the forms to convert biomass materials to useful gases (e.g. CO, H₂, and CH₄), but the process is affected by the production of undesired gases (N₂, and CO₂), and also with a considerable amount of tar [2], [3], [4]. Tar particularly causes serious problems in any direct downstream application of producer gases from gasification. For example, it can cause fouling and erosion for equipment, and the tar formation wastes about 5-15% of the effective energy from biomass gasification [5].

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Kinetic and equilibrium models were built to simulate gasifier behavior at different working conditions. Several researchers [6], [7], and [8] used equilibrium models based on one global reaction mechanism and succeed to predict product gas composition and gasification temperature at some extend. While other researchers who used multi-step equilibrium models [9], where the output of each zone is fed to the next zone of a downdraft gasifier. However, the equilibrium models are less effective and give an over prediction for the higher heating value and H₂ output with lower amounts of CO [10]. Thermodynamic equilibrium models also fail to take into account the physical and multistep chemical phenomena inside a gasifier, thus originate error in some species estimation [11].

Kinetic models, on the other hand, were built to overcome those problems and proved to be able to simulate a wider range of working parameters of a gasifier e.g. (producer gas composition, temperature profile, heating value and gasifier dimensions) ([1], and [12]). Some other models were used to predict the tar formation during biomass gasification (e.g. see Ref. [13-20]), and tar can be defined as all hydrocarbons that have a molecular weight higher than benzene C₆H₆ [13]. However, tars could form in hundreds of different chemical compounds, but in most cases, about 20 species are considered having significant amounts [14].

While some previous works focused on tar evolution only, other kinetic models presented gas composition and the effect of working parameters on the change of gasifier behavior and gasification efficiency.

The current work is an extension of an existing model developed recently by the authors [1] – a four-zone integrated kinetic model allowing prediction of the optimum working parameters of a downdraft gasifier. The model was tested and verified over a wide range of biomass materials. Tar was assumed to be having one compound represented by formula (C₆H_{6.62}O_{0.2}). This paper presents an extension of the model through implementation of the four main tar species instead of one general formula. Tar species evolution will be tracked through from pyrolysis to oxidation and reduction zones. A well understanding of the evolution of different gas species and tar, and their relationship to temperature at each zone and

other working parameters will be of a high importance when designing a gasifier and also to reduce tar content in producer gas. The results will discuss the optimum working parameters that lead to the production of higher value syngas. To the best of our knowledge, there is no other kinetic model that includes tar formation tracked from each zone, and how they influence on the production of different gas species along the gasifier height at different zones.

II. MODELING PROCEDURE

The kinetic model was described in details at [1]. The current work will discuss how the tar formation model is implemented in it. Primary tars formed during pyrolysis are composed of more than one hundred species, and their formation yields are not well known. The kinetics thus will try to simplify the tar species to be formed during pyrolysis using four main tar species representing major species.

During thermal conversion of biomass, tar is released and can be classified as primary, secondary, and tertiary tars [15]. Primary tars are generally oxygenated, primary organic and condensable molecules. They come mostly from the breakdown of cellulose, hemicellulose, and lignin under 500°C, such as Phenol, acetol (having biggest portion [16]), acetic acid, Guaiacol, and Furfural. As the temperatures rise above 500 °C, primary tars rearrange to form gases and secondary tars (phenol, cresol, indene, and olefins). While tertiary tars like aromatics, toluene and indene are formed at higher temperatures. Condensed tertiary tars make up the PAHs (Benzene, naphthalene (most important [19]), acenaphthylene and pyrene). All primary tars are converted to secondary and tertiary tars as the temperature of gasifiers rises.

Toluene was reported as the best representative for secondary tars while naphthalene as a PAH representing tertiary class tars and benzene as a model for primary tars although it is no longer considered as a tar [17]. The four compounds, (benzene, naphthalene, toluene, and phenol) were used for the model of [18] who simplified 10 tar species to those four lumped tar species.

The current model aims to address the formation and tar destruction for tar species along a downdraft gasifier having different zones based on the equations and kinetic relations stated in Table 1 and 2). The reactions start based on the given initial conditions of every species and the temperature of the gasifier zones. Initially the model assumes tar release with volatiles at the pyrolysis zone based on the pyrolysis temperature.

III. PYROLYSIS TAR FORMATION

Biomass first decomposes to volatiles, char and tar. Volatiles decomposition was shown in details in

[1]. Tar decomposition based on pyrolysis temperature will be addressed and discussed in the present work. Ref. [18] reported parameters for the empirical correlations of pyrolysis products as shown in Table 1 based on experimental data taken from [19] which gives the mass yield of tar evolution during pyrolysis process in (g tar/ kg biomass).

TABLE 1
CORRELATIONS FOR PYROLYSIS PRODUCTS [18]

	<i>a</i>	<i>b</i>	<i>c</i>
C ₇ H ₈	-6E-5	0.10701	-48
C ₁₀ H ₈	-0.0001	0.218	-115.32
C ₆ H ₆	-0.0003	0.7017	-387.6
C ₆ H ₆ O	2E-5	-0.068	46.42

The mass yield of different tar species *Y*, in (g/kg biomass), can be derived by using equation (1)

$$Y = aT^2 + bT + c \quad (1)$$

After calculating the mass yield of the four main tar species at the pyrolysis zone, they are added to the pyrolysis products and an energy balance is made to calculate the pyrolysis temperature through equation (2).

$$\sum X_i \cdot (h_f + C_p \cdot \Delta T)_{pyrolysis\ products} = \sum X_i \cdot (h_f + C_p \cdot \Delta T)_{combustion\ products} + Q_{loss} \quad (2)$$

The heat loss is mentioned in the oxidation zone only as it is higher in temperature than other zones, and the overall heat loss is assumed to be 10% of the product of the equivalence ratio (ER) and HHV [20]. The same energy balance principle is made for every zone to get the corresponding temperature.

After calculating the temperature, a backward calculation is made to predict the gas composition for the pyrolysis products including the tar species.

IV. TAR SPECIES IN COMBUSTION AND REDUCTION ZONES

The products of pyrolysis are used as feed to the oxidation zone. The reactions stated in Table 2 are implemented in the kinetic model for both the combustion and reduction zones. Those reactions are taken from the references mentioned in the table. Other reactions for the gasification and combustion were already discussed in [1] and will not be repeated here. Again, after calculating the gas composition, energy balance is made to get the oxidation and gasification temperature and backward calculations are made to get the correct gas composition coming out of combustion. The model uses 18 different kinetic reactions in the combustion and reduction zone for the calculation of different gas and tar species.

V. MODEL VALIDATION

The initial kinetic code was validated in [1] over a wide range of biomass materials for the gas composition, temperature profile and gasifier dimensions. The validation in this paper will only focus on the tar species formation before discussing other results.

TABLE 2
REACTIONS OF TAR SPECIES IMPLEMENTED IN THE MODEL.

	Reaction and rate expression	A, s ⁻¹	E, kJ/mol	Ref
1	$C_7H_8 \rightarrow 0.17C_{10}H_8 + 0.89C_6H_6 + 0.67H_2$ $r = k_1 [C_7H_8]$	2.23E13	315	[21]
2	$C_{10}H_8 \rightarrow 10C + 4H_2$ $r = k_2 [C_{10}H_8]^2 [H_2]^{-0.7}$	5.56E15	360	[22]
3	$C_{10}H_8 + 4H_2O \rightarrow C_6H_6 + 4CO + 5H_2$ $r = k_3 [C_{10}H_8] [H_2]^{0.4}$	1.58E12	324	[22]
4	$C_7H_8 + H_2 \rightarrow C_6H_6 + CH_4$ $r = k_4 [C_7H_8] [H_2]^{0.5}$	1.04E12	247	[22]
5	$C_6H_6 + 5H_2O \rightarrow 5CO + CH_4 + 6H_2$ $r = k_5 [C_6H_6]$	4.4E8	220	[22]
6	$C_6H_6 + 7.5O_2 \rightarrow 6CO_2 + 4H_2O$ $r = k_6 [C_6H_6]^{-0.1} [O_2]^{1.25}$	17.83	125.5	[22]
7	$C_6H_6 + 3O_2 \rightarrow 6CO + 3H_2$ $r = k_7 [C_6H_6] [O_2]$	1.58E15	202.6	[22]
8	$C_7H_8 + 9O_2 \rightarrow 7CO_2 + 4H_2O$ $r = k_8 [C_7H_8]^{-0.1} [O_2]^{1.25}$	14.26	125.5	[22]
9	$C_6H_6O \rightarrow CO + 0.4C_{10}H_8 + 0.15C_6H_6 + 0.1CH_4 + 0.75H_2$ $r = k_9 [C_6H_6O]$	1.0E7	100	[18], [16]

The comparison shown in Fig. 1 between the results obtained by the present model and the other experimental data [23], shows a good agreement for the total tar amount. Maximum tar produced by the model shows values around 5 g/Nm³, the fact that also has an agreement with [24], where they stated that tar produced in downdraft gasifiers is in the range of 0.01-6 g/Nm³. In the experimental work of [23], they used a non-woody biomass material (corn stalks) with moisture content level of 6.17%, and the comparison is made for the different values of the air equivalence ratio to measure the stability of the current model for a normal range of working conditions.

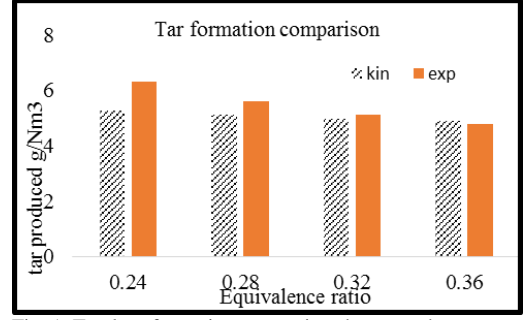


Fig. 1. Total tar formation comparison between the present model and experimental results [23].

VI. TAR SPECIES EVOLUTION AND FORMATION ALONG THE GASIFIER

Tar evolution during pyrolysis and its relation with temperature is illustrated and first implemented in the current kinetic code. Tar destruction, formation and converting to other species based on the reactions stated in Table 2 is shown and was first implemented in the kinetic code. Tar formation results are illustrated in Fig. 2, and Fig. 4). Different tar species used in the model are traced from its formation in pyrolysis then the combustion and reduction zones along the gasifier height, and they depend on the temperature of each zone. The effect of the varying moisture content and equivalence ratio will be discussed and presented. Initial investigations are carried out at a fixed ER of 0.326, and with varying moisture content to study its effect on the tar formation and subsequently, predict the optimum conditions that lead to less amount of tar in the producer gases. Other simulations are carried out at a fixed moisture content of 10% but with a varying equivalence ratio.

Fig. 2 shows the evolution of different tar species along the gasifier height during the rubber wood (ultimate analysis from [25]) gasification with varying moisture content. Phenol formation starts in pyrolysis, then decreases in oxidation and tends to disappear or exist in a very small amount in the producer gas, that is because it is a primary tar compound. Primary tars start forming at temperatures 673-973 K [26], and at temperatures above 773 K they start reforming [27] and convert to secondary then tertiary tars. Temperature profile along the gasifier within the different moisture content or equivalence ratio is shown in Fig. 3. Temperatures of the oxidation and reduction zones that are higher than 1000 K are enough to destroy the primary tar species to reform them into other compounds.

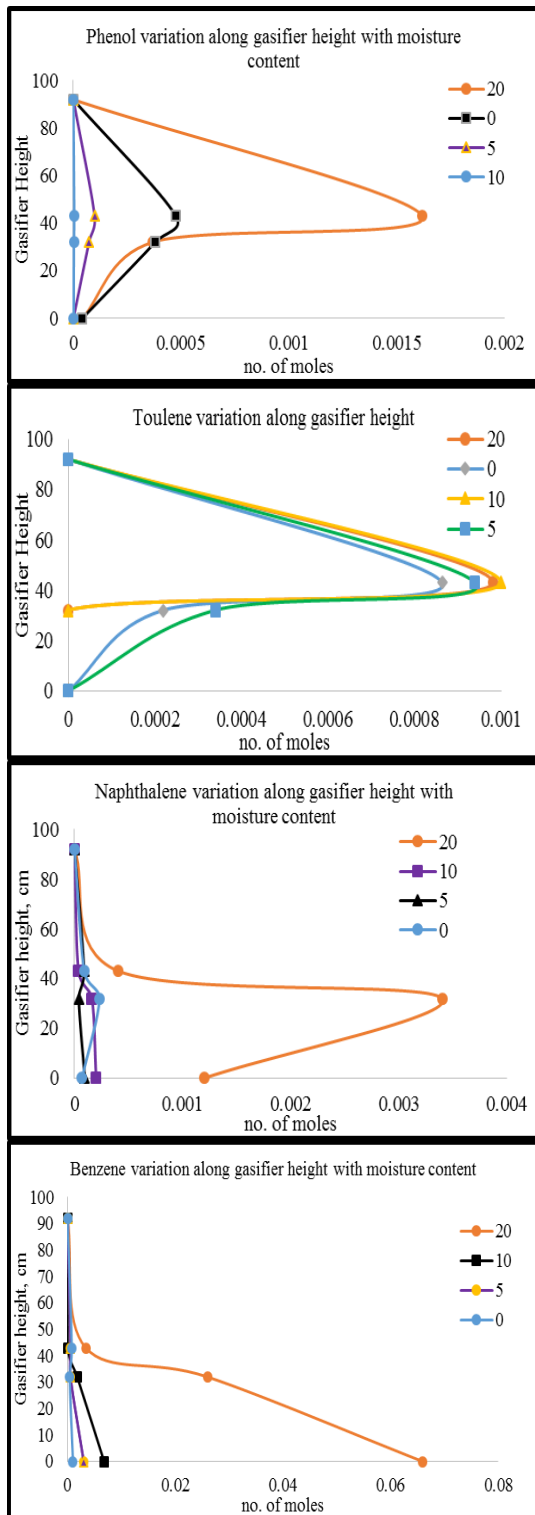


Fig. 2. Tar evolution and formation along the gasifier height at different moisture content levels.

As seen in Fig. 2, toluene formation along the gasifier has the same trend of phenol: higher concentration in the pyrolysis zone, followed by destruction in the oxidation and reduction zone. Temperatures above 1173 K are enough to crack all

the phenol and toluene and subsequently, convert them into benzene and other lighter species [14].

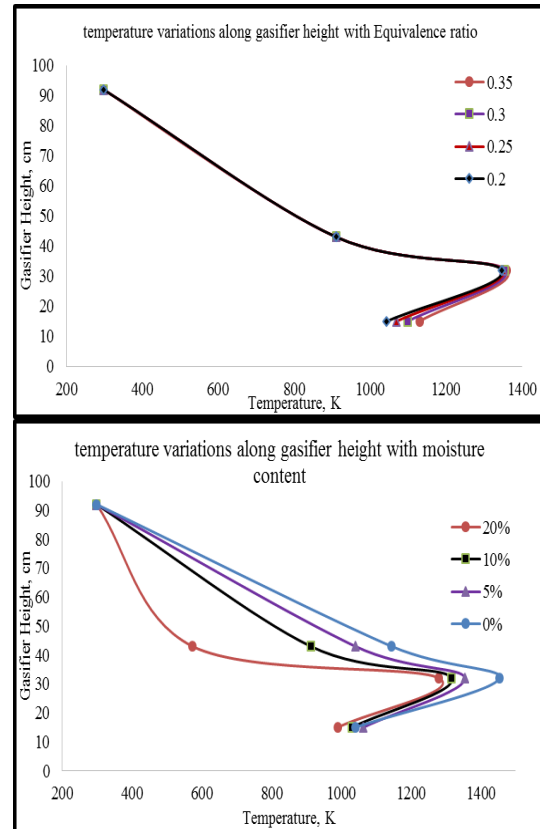


Fig. 3. Temperature variations along gasifier height.

Naphthalene formation, however, takes a different trend than other species. It is formed and present in considerable amounts in producer gas. Small amounts are produced during pyrolysis, because it is a tertiary tar which requires higher temperatures to present and formed. Fig. 3 shows that higher temperatures in the oxidation zone $>1300\text{K}$ are favorable for the naphthalene formation which starts conversion for temperatures greater than 1300K and achieves a total conversion at 1600K [28]. Based on reactions 2 and 3 in Table 2, naphthalene is converted to char, H_2 , CO and benzene. Those reactions are tend to take place in the combustion and reduction zone, however it is more likely to happen in the reduction zone because of the presence of water vapor. Higher concentration of naphthalene in the oxidation zone is mainly due to the conversion of lighter species (phenol and toluene) and also because of the oxidation temperature which is in the ideal range of naphthalene formation and never exceeds this to the destruction temperatures ($>1600\text{K}$).

Overall, benzene has the highest portion of tar species, which is usually greater than 37% by weight of total tars produced [29]. Also the data collected from [19], [17], and [15] shows that the highest portion of tar compounds is for benzene which meets a good agreement with the model.

VII. EFFECTS OF EQUIVALENCE RATIO ON THE TAR SPECIES

The results shown in Fig. 4 illustrate the effects of the equivalence ratio on the tar evolution at the different zones of the downdraft gasifier. Rubber wood was used as feedstock with moisture content of 10%. The same trend shown with varying moisture content is also noticed with the equivalence ratio. All the tar species evolution starts from pyrolysis to oxidation and reduction. More attention will be focused on the benzene formation with varying equivalence ratio as it is the major of tar species formed during gasification and also because of the similar trend of all species with varying equivalence ratio.

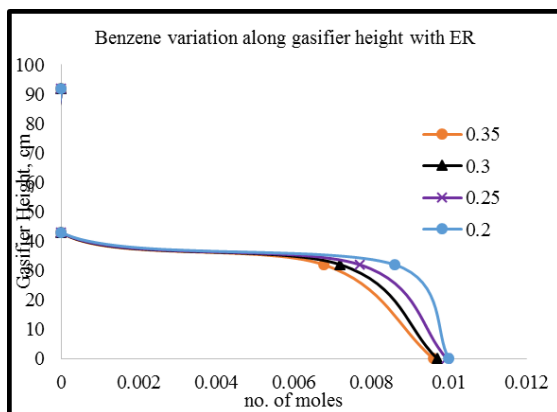


Fig. 4. Benzene evolution and formation along gasifier height at different equivalence ratios.

Furthermore, as illustrated and discussed before, benzene has the biggest portion of tar produced during biomass gasification. The results of changing moisture content or equivalence ratio show the same trend and find good agreement with the other previous works like [28], [17], [18] and [21]. Generally, a higher equivalence ratio tends to increase the oxidation and reduction temperatures, Fig. 3. This is because it increases the air amount and circulation inside the gasifier and hence, increasing the oxidation process reactions. Oxidation reactions are normally exothermic which release heat and thus increase temperature inside the gasifier.

VIII. OPTIMUM WORKING PARAMETERS

After validating the current model, it is used to address the optimum working parameters that lead to higher quality syngas (high heating value with lower moisture content). Lower amount of water content in biomass leads to a significant increase in the heating value which has a good agreement with ([30], [31], and [32]). On the other hand higher levels of moisture content requires more heat for removal, this heat is not recovered again and reduces producer gas heating value. In contrast, lower moisture content is favorable for lower tar amount

levels in the producer gas. The results reveal a decrease in tar amount produced from the gasifier by more than 40% when decreasing moisture content from 10 to 0%. On the other hand, increasing equivalence ratio from 0.2-0.35 leads to a decrease of tar produced by 6%.

Highest heating value of 5.96 MJ/Nm³ is found for the rubber wood gasification under ER=0.2, and MC of 10%. Higher tar levels (5.3 g/mol of biomass = 22.5% by weight) are also found for using rubber wood at 20% moisture content. While lower tar amounts is found at using 0% MC at ER= 0.326, which gives 0.1 g tar/mol biomass. In conclusion, biomass materials with moisture content levels lower than 10 %, and using equivalence ratio of 0.3 will increase the yield of syngas, leading to an increase of heating value with a reasonable amount of tar content in producer gas.

IX. CONCLUSION

The current model is a four-zone kinetic model for air-blown downdraft gasifiers. The model is verified and found good agreement with other experimental data. The model can predict producer gas composition, four main tar species formation, and gasifier dimensions design. The model is used to address the evolution of different gas species, char and tar species along gasifier, starting from devolatilization process to combustion and reduction. The results show the evolution and variation of different tar species with different working conditions of moisture content and equivalence ratio. Finally, the model is used to find the optimum working parameters for a downdraft gasifier that leads to the production of higher yield of syngas with lower tar amounts. Using equivalence ratio of 0.3, with lower values of moisture content < 10% will increase the yield of syngas, leading to an increase of heating value with a reasonable amounts of tar content in producer gas.

Future work on this model will try to address new ideas regarding tar destruction and converting it to useful compounds.

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