

MODELLING UPDRAFT GASIFICATION OF BIOMASS WITH A HIGH MOISTURE CONTENT: INFLUENCE OF TAR CONDENSATION AND SUBSEQUENT REACTIONS

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ABSTRACT: Updraft gasification of high moisture biomass is investigated in this work with a reactor model considering a tar condensation model and secondary tar charring/cracking reactions. The reactor model is a one dimensional model which solves mass and energy balance equations along the reactor length. It incorporates a comprehensive pyrolysis scheme which is able to predict a detailed product composition. The tar condensation model is an equilibrium model based on the application of the Raoult law. It allows calculating the amount of tar that condenses at a defined temperature.

Tar secondary charring/cracking reactions, occurring after condensation of the tar, play a main role in updraft gasification for high moisture fuels. These reactions lead to the movement of the pyrolysis reaction front to the top of the reactor avoiding extinction of the process. This self-regulating behaviour is produced because tar charring/cracking is exothermic and char that is generated in this reaction is then exothermally combusted at the bottom of the reactor.

Keywords: updraft gasification, fixed bed, tar, condensation, model.

1 INTRODUCTION

Gasification of ligno-cellulosic biomass is a technique of major importance because of its highly flexible application. In an updraft gasification process, biomass is fed at the top of the reactor and moves downwards as a result of the conversion and the removal of the ash. Primary air intake takes place from the bottom and the producer gas leaves the reactor at the top. It is a staged process and the sub-processes drying, pyrolysis, char gasification and oxidation are taking place sequentially, as shown in Fig. 1.

The major advantages of updraft gasification are the geometric simplicity, high char conversion and gasification efficiency and low producer gas exit temperature. In comparison to downdraft gasification, updraft gasification allows considerably higher fuel flexibility in terms of particle size and especially moisture content [1]. Moisture contents between 5 and 50% (in mass % w.b.) are possible. Updraft gasification of high moisture biomass is investigated in this work with a reactor model considering a tar condensation model and secondary tar charring/cracking reactions. The reactor model is presented in Section 2, the condensation model in Section 3 and both models are applied in Section 4. Finally, the conclusions are exposed.

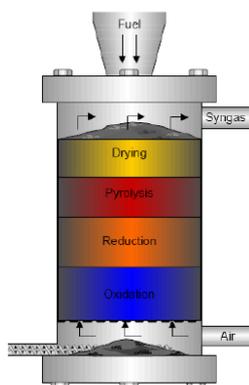


Figure 1: Updraft gasifier. Figure from [1].

2 REACTOR MODEL

The reactor model is a one dimensional fixed-bed updraft gasifier model. The solid/liquid (wet biomass) and the gas phase are considered in the reactor bed. It is a quasi-continuous model for both phases and the mass and energy balance equations are solved along the reactor length. The assumptions of the model are similar than the ones of the reference work of Di Blasi [2]. Due to the relevance in the investigated topic, the models for drying, pyrolysis and secondary tar reactions are explained in detail in the next subsections.

2.1 Drying model

The drying process is modelled with an equilibrium model, where the reaction rate is calculated so that the partial water vapour pressure is at equilibrium. The reciprocal of the equilibrium time constant is set to 100 1/s in this work. The partial vapour pressure of equilibrium is a function of the vapour saturation pressure and sorption isotherm. The sorption isotherm is taken from Gronli [3].

2.2 Pyrolysis model

The pyrolysis scheme is based on the mechanistic scheme developed by Ranzi et al. [4] and adapted by this group [5] to consider secondary char formation reactions, which are relevant for particles of a certain thickness. The pyrolysis scheme is applied in this work for spruce. The initial dry ash-free composition in percentage of mass is:

- Cellulose (CELL): 41.4%.
- Hemicellulose (HC): 28.2%.
- Lignin (LIG): 6.1% lignin-C, 18.3% lignin-H and 6.0% lignin-O.

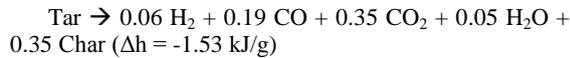
Secondary charring reactions produced during pyrolysis are considered by introducing an additional parameter “x”. This parameter represents the amount of the initial primary products that will further react to form the secondary charring products during pyrolysis. “x” is set to 0.35 for all components. A value in this range was employed for pyrolysis in a fixed bed producing good

results [5]. The product composition is calculated at a heating rate of 100 K/min. The obtained products in percentage of mass are:

- Solid: 25.1% char with composition $\text{CH}_{0.193}\text{O}_{0.071}$. Char consists on pure carbon and the char intermediate species $\text{G}\{\text{COH}_2\}$ and $\text{G}\{\text{H}_2\}$.
- Permanent gases: 6.1% CO , 11.5% CO_2 , 0.5% H_2 , 2.2% CH_4 and 1.8% C_2H_4 .
- Condensable species: 16.3% H_2O and 36.5% organic condensable species (tars) with composition $\text{CH}_{1.706}\text{O}_{0.627}$. The latter includes 12 compounds with phenolic, carbonyl, alcohol or furan structures, which are detailed in Tab. 1.

2.3 Secondary charring/cracking reactions of tar

Tars that are produced in the pyrolysis reaction may afterwards condense on biomass, before leaving the bed, and move downwards together with biomass. The condensed tars could then suffer evaporation or charring/cracking reactions when they get in contact with the char, which acts as a catalyst. The product composition of these possible charring/cracking reactions is based on the experiments of Boroson et al. [6], where volatiles from pyrolysis flow through a char bed. The product composition of this reaction is approximated to resemble these experiments, closing the elemental mass balances and considering the tar composition described in Section 2.2:



The possible tar charring/cracking reaction, produced after condensation of the tar, is included in the pyrolysis reaction. The tar fraction that suffers the tar charring/cracking reaction after condensation, related to the total tar produced in pyrolysis, is denoted by “y”. The condensed amount of tar can be roughly estimated with the tar condensation model that will be presented in the next section. Moreover, it should be considered that just a fraction of the total tar that condenses will suffer charring/cracking reactions. The other fraction will evaporate.

3 TAR CONDENSATION MODEL

An equilibrium model is presented, where the amount of tar that condenses in equilibrium at a defined temperature is calculated. The condensation model is based on the application of the Raoult law and considers an ideal gas phase.

The tar compounds predicted by the previously presented pyrolysis scheme (see Tab. 1) are classified in different groups based on their structure and main functional groups:

- Phenolic compounds: aromatic compounds with hydroxyl (-OH) and other O-containing groups linked. Their main origin is lignin.
- (Hetero)cyclic compounds: furans such as 5-hydroxymethyl-furfural.
- Not cyclic compounds: compounds with carbonyl and/or alcoholic groups.

Table 1: Tar composition.

Name	Symbol	% mass
Hydroxyacetaldehyde / Acetic acid	HAA / AA	26.0
Glyoxal	GLYOX	6.6
Propanal (Acetone)	$\text{C}_3\text{H}_6\text{O}$	12.2
Propanedial	$\text{C}_3\text{H}_4\text{O}_2$	0.4
Formaldehyde	CH_2O	7.2
Methanol	CH_3OH	8.3
Acetaldehyde (Ethanal)	CH_3CHO	4.2
Ethanol	ETOH	2.2
Total not cyclic		67.1
5-hydroxymethyl-furfural	HMFU	14.3
Total (hetero)cyclic		14.3
Paracoumaryl alcohol	pCOUMARYL	1.6
Phenol	PHENOL	0.8
Sinapaldehyde	FE2MACR	16.2
Total phenolics		18.6

The tar groups phenolic and not cyclic are subdivided into two groups. The molecular weight and the percentage of mass of each group are calculated from the tar composition predicted by the pyrolysis scheme and shown in Tab. 1. The boiling points (B_p , in °C) and saturation pressures at 25°C (P_s , in Pa) at atmospheric pressure are then estimated based on correlations obtained with data from species from these groups and shown in Eq. (1) and (2). The enthalpy of vaporization (L) is obtained from the Clausius–Clapeyron equation, shown in Eq. (3). The condensation properties of the groups are summarized in Tab. 2 and the predicted condensation of tars over temperature by the model is shown in Fig. 2.

The main limitation of the current model is that in the phenolics group just one ring (monomers) phenolic compounds are considered. Two or more ring phenolic compounds are not considered in the model, which condense at higher temperatures (“heavy tars”).

$$B_p = 1.8413 M_m - 13.459 \quad (1)$$

$$B_p = -17.99 \log_{10}(P_s) + 257.94 \quad (2)$$

$$\ln\left(\frac{P_2}{P_1}\right) = \frac{L}{R} \left(\frac{1}{T_1} - \frac{1}{T_2} \right) \quad (3)$$

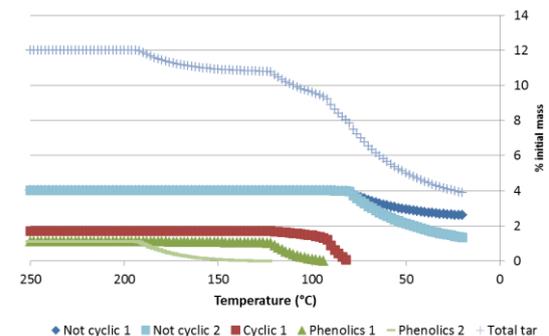


Figure 2: Tar condensation model applied for gas phase composition typical for updraft gasification: 12% tars, 33% H_2O , 55% permanent gases.

Table 2: Percentage of mass and condensation properties of tar groups.

	% mass	Ps (Pa, 25°C, 1 atm)	Bp (°C, 1atm)	L (J/mol)	Mm (g/mol)
Not cyclic 1	33.5	59897.4	60.0	12390	40
Not cyclic 2	33.5	12901.8	87.7	29416	55
(Hetero)cyclic	14.3	9.0	218.4	58769	126
Phenolics 1	9.3	0.278	281.0	68724	160
Phenolics 2	9.3	0.00060	391.5	85185	220

4 MODEL RESULTS

Model simulations are presented for a cylindrical gasifier with an internal diameter of 75 cm and height of 100 cm, which is fuelled with spruce chips. The fuel and primary air inlets are 171.8 and 125.4 kg/h, respectively, and fuel moisture is 45% w.b. The pyrolysis product composition has been shown in Section 2.2. Other reactor and feedstock properties are stated in Tab. 3.

Results are shown in Fig. 3 and Fig. 4 for “y” (tar fraction that suffers the tar charring/cracking reaction after condensation, see Section 2.3) equal to 0.15 and 0.20.

In the oxidation zone, at the bottom of the gasifier, primary air is quickly consumed through oxidation reactions producing the energy needed to run the gasifier. Oxygen is mainly consumed in the char oxidation reaction, reducing the solid velocity, and also in the CO combustion reaction. This oxidation layer is very thin; oxygen is consumed in the first 2 cm in good agreement with other models [2]. The oxidation reactions lead to high temperatures near the grate and provide heat for the endothermic char gasification reactions. These reduction reactions take place approximately in the first 10 cm of the reactor. At the bottom of the reactor higher temperatures are obtained by the model than in the experiments. This is probably due to the presence of hot and cold zones in the reactor, with different residence times, due to local ash melting, which was experimentally detected. The measurement values in this zone probably correspond to a cold zone, where there is locally a lower primary air and solid biomass input rate.

The position of the drying and pyrolysis zones changes with variations in the “y” parameter, representing the tar fraction that suffers the tar charring/cracking reaction after condensation. In the drying zone moisture is released as water vapour. Dry biomass is then pyrolyzed, producing char that moves downwards and volatiles, that leave the reactor through the top together with water from drying and the products of the oxidation/reduction layer. The primary tar produced in primary pyrolysis can also be converted by a secondary charring/cracking reaction after condensation.

With “y” = 0.15 (Fig. 3), the pyrolysis reaction front is close to the bottom of the reactor. With “y” = 0.20 (Fig. 4), the pyrolysis reaction takes place in the middle of the reactor. An increase of “y” leads to the movement of the pyrolysis reaction front as tar charring/cracking is exothermic and char that is generated in this reaction is then exothermally combusted at the bottom of the reactor. The experimental temperatures show that the pyrolysis front is at a reactor height of 0.30 – 0.40 m. Therefore, the appropriate value for “y” in these conditions may be in the range 0.15 – 0.20.

The comparison between the experimental producer

gas composition and model predictions is detailed in Tab. 4. There is a good prediction of H₂O, CO, CH₄ and N₂ yields, under-prediction of CO₂ and H₂ yields and over-prediction of tar and light hydrocarbons yields. The inclusion of the tar charring/cracking reaction improves the predictions of the model, regarding the tar, H₂ and CO₂ yields. The CO₂ yield remains however under-predicted, probably due to the presence of cold zones at the bottom of the reactor, where the temperatures are not high enough to consume the CO₂ through the Boudouard char gasification reaction.

Table 3: Reactor and feedstock properties.

Property	Value	Unit
Internal diameter	0.75	m
Primary air input	125.4	kg/h
Biomass input	171.8	kg/h
Moisture content	45	% w.b.
Bed height	1	m
Representative input biomass	0.78	m
Initial particle diameter	0.008	m
Dry particle density	430	kg/m ³
Dry bed density	177	kg/m ³
Energy input	444.6	kW
Bottom heat release	21.3	kW
Heat radiation to the top	2.2	kW
Lateral thermal resistance	2.2	(m ² *K)/W
Temperature pre-heated primary air	250	°C
Ambient temperature	25	°C

Table 4: Producer gas composition.

	Exp.	Model		
		y = 0.00	y = 0.15	y = 0.20
CO	19.6	18.6	19.2	20.7
CO ₂	10.1	4.5	5.4	4.7
H ₂	0.6	0.3	0.4	0.5
CH ₄	0.7	0.7	0.7	0.7
C ₂ H ₄ + C ₃ H ₆	0.1	0.6	0.6	0.6
Tar	7.0	12.4	10.2	9.5
H ₂ O	29.6	33.2	32.5	32.0
N ₂	32.2	29.6	31.0	31.3

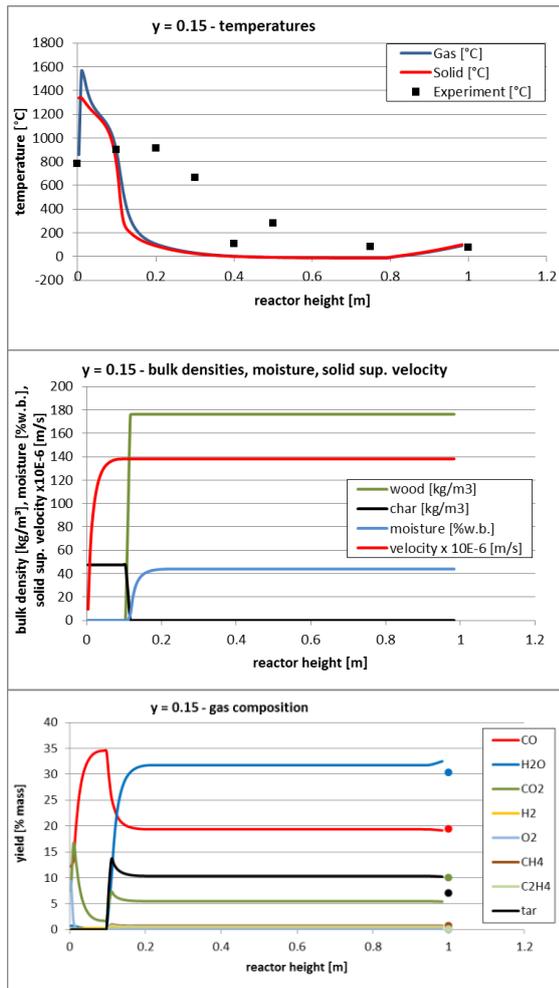


Figure 3: Model results for the $y = 0.15$ case (98.5% solid d.a.f. conversion): temperatures (top), solid bulk densities, moisture content and solid superficial velocity (middle) and gas composition in mass yields (bottom) with experimental data in points.

The tar condensation model described in the previous section was applied to the product composition predicted by the model with “ y ” = 0 (see Fig. 2). Condensation of phenolics starts at temperatures of 190°C and condensation accelerates at temperatures lower than 100°C. The model predicts condensation of $\approx 1/3$ of the tar at the minimum experimental temperature in the reactor ($\approx 80^\circ\text{C}$). Taking into account that, according to the reactor model, around 15-20% (“ y ” = 0.15 – 0.20) of the total tar would react in the cracking/charring reaction, it can be considered as a first approximation that around half of the condensed tar would suffer the charring/cracking reaction while the other half may evaporate again.

It should be considered that neither the tar condensation model nor the model of the tar charring/cracking reaction were yet thoroughly validated; there is a great uncertainty in these points. However, it is clearly shown that tar secondary charring/cracking reactions, occurring after condensation of the tar, play a main role in updraft gasification for high moisture fuels and these phenomena should be considered to correctly describe this process.

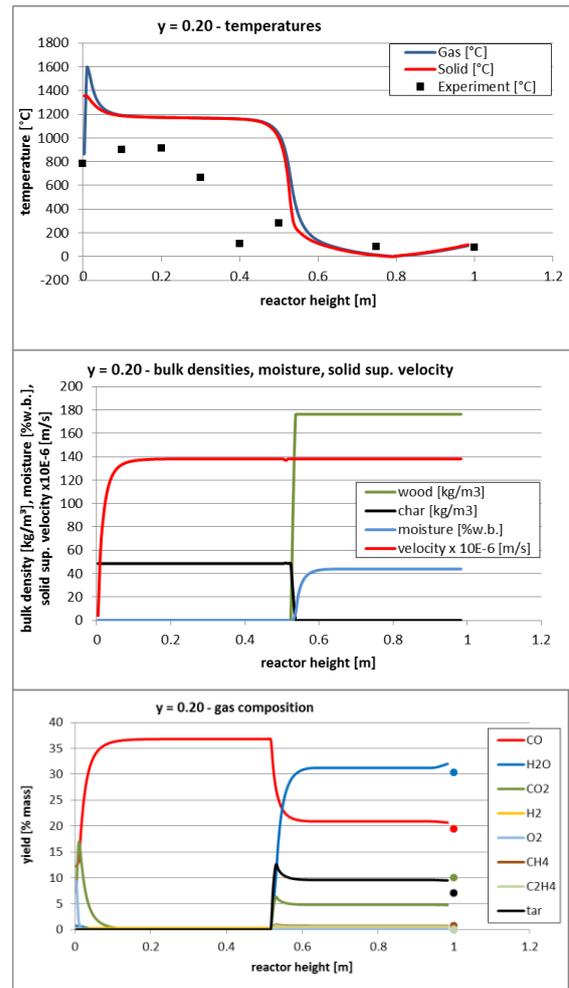


Figure 4: Model results for the $y = 0.20$ case (99.5% solid d.a.f. conversion): temperatures (top), solid bulk densities, moisture content and solid superficial velocity (middle) and gas composition in mass yields (bottom) with experimental data in points.

5 CONCLUSIONS

Updraft gasification of high moisture biomass was investigated in this work with a reactor model considering a tar condensation model and secondary tar charring/cracking reactions. The reactor model is a one dimensional model which solves mass and energy balance equations along the reactor length. It incorporates a comprehensive pyrolysis scheme which is able to predict a detailed product composition, including 12 tar compounds with phenolic, carbonyl, alcohol or furan structures, and tar charring/cracking reactions. The product composition of these reactions is taken from literature. The tar condensation model is an equilibrium model based on the application of the Raoult law. It allows calculating the amount of tar that condenses at a defined temperature. To develop the model the tar compounds predicted by the detailed pyrolysis scheme are classified in different groups based on their structure and relevant properties are calculated for each group.

Tar secondary charring/cracking reactions, produced after condensation of the tar, play a main role for high moisture fuels. These reactions lead to the movement of

the pyrolysis reaction front to the top of the reactor avoiding extinction of the process. This self-regulating behaviour is produced because tar charring/cracking is exothermic and char that is generated in this reaction is then exothermally combusted at the bottom of the reactor. It was predicted that, during gasification of spruce chips with a moisture content of 45% w.b., around one third of the tar would condense and the half of this condensed fraction would further react in a secondary tar charring/cracking reaction.

It was concluded that the presented reactor and tar condensation models are useful to understand the phenomena taking place in fixed-bed updraft gasification of fuels with high moisture contents. This is very helpful for designing updraft gasifiers which can exploit the limits of maximum moisture content of the fuels.

Future work could include a validation of the tar condensation model and product composition of the tar charring/cracking reaction, as well as a deeper global validation of the reactor model with more experimental results, including product composition, at different conditions.

6 REFERENCES

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