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MODELLING OF FLUIDIZED BED BIOMASS GASIFICATION IN THE QUASI-EQUILIBRIUM REGIME FOR PRELIMINARY PERFORMANCE STUDIES OF ENERGY CONVERSION PLANTS

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Thermodynamic equilibrium-based models of gasification process are relatively simple and widely used to predict producer gas characteristics in performance studies of energy conversion plants. However, if an unconstrained calculation of equilibrium is performed, the estimations of product gas yield and heating value are too optimistic. Therefore, reasonable assumptions have to be made in order to correct the results. This paper proposes a model of the process that can be used in case of deficiency of information and unavailability of experimental data. The model is based on free energy minimization, material and energy balances of a single zone reactor. The constraint quasi-equilibrium calculations are made using approximated amounts of non-equilibrium products, i.e. solid char, tar, CH_4 and C_2H_4. The yields of these products are attributed to fuel characteristics and estimated using experimental results published in the literature. A genetic algorithm optimization technique is applied to find unknown parameters of the model that lead to the best match between modelled and experimental characteristics of the product gas. Finally, generic correlations are proposed and quality of modelling results is assessed in the aspect of its usefulness for performance studies of power generation plants.

Keywords: biomass gasification, fluidized beds, thermodynamic equilibrium

1. INTRODUCTION

Thermal conversion of biomass into a gaseous fuel by means of gasification is considered nowadays as one of the most attractive technologies for CO_2 emission reduction and fossil fuel savings. This is mainly due to a high level of power generation efficiency of potential power plants. Nevertheless, a conceptual design, optimization and feasibility studies are required to demonstrate saving potential and economic profitability of biomass to energy conversion projects.

Nowadays the most documented, matured and commercialized biomass gasification technology is the fluidized bed process. In practice this technology dominates the market within the power range above 10 MW (chemical fuel energy input). For research and development purposes fluidized bed reactors are built starting from the power of about 100 kW (Bolhàr-Nordenkampf et al., 2002b). It has already been demonstrated in projects ARBRE (UK), Värnamo (Sweden) and Güssing (Austria) that a variety of technological schemes of power plants can be designed and successfully operated with the medium scale reactors. Due to a relatively high power output fluidized bed gasifiers are more suitable for plants with gas turbines. Nevertheless, examples of plants with gas engines also exist (Bolhàr-Nordenkampf et al., 2002b; Wu et al., 2008).

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There is a number of studies of integrated biomass gasification energy conversion systems available nowadays in the literature. Most of them are based on the thermodynamic equilibrium approach towards modelling gasification. Some examples are given by Brown et al. (2009), Buragohain et al. (2010), Kalina (2010), Klimantos et al. (2009) and Schuster et al. (2001). The equilibrium modelling of gasification reactors is also utilised by software available for computational analysis and simulation of energy plants, that is e.g. the Cycle Tempo (Delft University of Technology, 1980–2006). In general, the equilibrium approach is regarded as satisfactory for this problem, assuming that one is aware of its limitations (Gòmez-Barea et al., 2010). Simple mathematics and independence (to a certain extend) from the reactor design parameters are the advantages. However, if the equilibrium approach is applied for an analysis of a fluidized bed reactor, the results typically show an overestimated mass conversion efficiency, heating value of the producer gas and generation of hydrogen and carbon oxide. The yields of methane and higher hydrocarbons are usually notably underestimated. Mevissen et al. (2009) presented that the differences between predicted and experimental data can be significant. Prins et al. (2007) claim that the equilibrium model gives the highest gasification efficiency that can be possibly attained for a given fuel. In practice, it is difficult to reach equilibrium conditions for solid carbon conversion at gasification temperatures below 1000ºC, which is the case with fluidized bed reactors. A composition of a producer gas that is closer to equilibrium one can be obtained using either in-bed catalyst or a secondary catalytic bed reactor (Asadullah et al., 2003; Corella et al., 1998; Kurkela et al., 2009).

Gil et al. (1999) examined the results of different experimental studies of pine chips gasification in bubbling fluidized silica sand bed using air, pure steam and steam-O_2 mixtures as gasification agents. Similar experimental conditions were taken into account. The reported H_2 content in the dry producer gas was within the range of 5.0 - 16.3%, 38.0 - 56.0% and 13.8 - 31.7% for the three agents respectively. The concentration of CO was 9.9 - 22.4%, 17.0 - 22.0% and 42.5 - 52.0% and the concentration of CH_4 was 2.2 - 6.2%, 7.0 - 12.0% and 6.0 - 7.5%. It can be easily concluded that these concentrations are far from equilibrium ones calculated for the same values of excess air ratio λ.

Gòmez-Barea et al. (2010) conducted an extensive review of modelling of biomass gasification in bubbling and circulating fluidized bed reactors. According to the authors fluidized bed gasifiers have to be modelled using revised pseudo-equilibrium models, or, in some extreme cases, by detailed flow models.

Good assumptions for calculations are required to obtain an agreement between modelling and experimental results. Typically, some empirical parameters, such as concentration of methane in the product gas and/or carbon conversion efficiency, are included in a model. The constrained equilibrium models that nowadays can be found in the literature are usually tuned using a reactor specific experimental data. Consequently, the models have limited predictive capabilities. Another modelling approach is based on the so called quasi-equilibrium temperatures. An example has been given by Brown et al. (2009). They estimated yield and composition of a producer gas using a parametric stoichiometric model where the equilibrium of gasification reactions was calculated at a temperature lower than the real process temperature. The model was calibrated using experimental data from a pilot circulating fluidized bed reactor. According to Prins et al. (2007) such an approach is impractical as the temperature used for calculation appears to be independent from the real process temperature and hardly predictable without experimental data.

In this paper an attempt is made to develop a general constrained quasi-equilibrium model of the fluidized bed gasification process. It is expected that the model would be able to predict a producer gas yield, composition and heating value with an acceptable accuracy. These parameters have significant influence on mass and energy balance as well as on the economic evaluation of a project. They also determine performance of machinery such as gas engines and gas turbines installed downstream of a gasifier. An extensive study of the literature has been made to make reasonable assumptions. Then, the model has been formulated and verified against the published experimental results.

Due to complexity and kinetic limitations of the process it is almost impossible to develop a precise general model of a fluidized bed gasifier. According to Gil et al. (1999) the final composition of a producer gas is influenced by at least 20 operational parameters concerning the reactor and feedstock. In this work the parameters that are assumed to have a key impact on modelling results are: carbon conversion efficiency, yields of methane and higher hydrocarbons (including tar), char composition, yield of NH_3 and heat losses. These are the parameters that reduce the amount of carbon and hydrogen available to equilibrium calculation and influence the process temperature.

Li et al. (2004) proposed to correct deviations between model-predicted and experimentally measured product gas yield and composition by using certain amounts of solid carbon and methane that do not take part in calculating the equilibrium. They defined carbon and hydrogen availability functions as simple correlations of the air ratio. The functions were established using a reactor specific experimental data.

According to Kurkela et al. (2009) carbon conversion efficiency of modern air/O_2 fluidized bed gasification reactors is between 96 - 99%. In allothermal gasifiers, where pure steam is used as the gasification agent and heat is supplied from an external source, the conversion efficiency is between 85 - 92 %. Van der Meijden et al. (2010) modelled an autothermal fluidized bed gasification of biomass using the approximated single value of carbon conversion efficiency of 90%. In the case of allothermal steam gasification they calculated conversion efficiency using a linear function of temperature that gave the values from 71% to 86% within the range from 800 to 900ºC. Van der Drift et al. (2001) presented experimental values of carbon conversion efficiency of gasification of different woody biomass fuels using air as the gasification agent in the range of 85 - 97%, and the average value was 92%. De Souza-Santos (2004) claims that due to fluidization process requirements it is very difficult to achieve the efficiency in excess of 95%. A high carbon conversion requires a long particle residence time and a special design of the reactor. The problem is partly solved in pressurized systems and in oxygen blown reactors. Conversion efficiency is also influenced by the type of in-bed catalyst (Asadullah et al., 2003).

A comprehensive study of gasification in pressurized fluidized bed reactors was presented by De Jong (2005). It can be concluded from the presented results that the higher is the value of the excess air ratio λ the higher is the carbon conversion efficiency. Gasification of miscanthus with λ in the range of 0.31 - 0.45 resulted in carbon conversion between 82 and 92%. The carbon conversion observed during gasification of wood with λ in the range of 0.32 - 0.46 varied very little from 96.6 to 98.7%. Higher carbon conversion efficiency of wood was attributed mainly to smaller particle size and higher reactivity. It can be observed that the contents of methane and higher hydrocarbons in a producer gas (excluding tar) depend linearly on the values of λ. The total content of hydrocarbons in the product gas varied from 2.48 to 5.03 for both analysed fuels. Measurements of the composition of the product gas at different heights of the reactor column show that variations of the gas composition are relatively small along the freeboard. An analysis of the experimental results also shows that 50 – 70% of the fuel nitrogen forms NH3 and 4.5 – 14% forms HCN.

Miccio et al. (1999) presented that carbon conversion efficiency increases both with air to fuel ratio and process temperature. The efficiency asymptotically approaches its maximum value with increasing reactor height. At the bottom of the freeboard carbon conversion sharply approached the value measured at the exit of the freeboard. At the most favourable conditions (i.e. $T = 900$ºC and $\lambda = 0.35$), carbon conversion at the reactor exit reached a maximum value of 97%. The minimum carbon conversion efficiency was 70% (at $\lambda = 0.15$ and $T = 700$ ºC). The presented results showed concentration of methane in the producer gas at the level above 5.7%.

A general estimation of tar content in the dry gas obtained from a fluidized bed reactor is 10 g/Nm^3 Milne et al. (1998). Most of the reported results from gasification tests show tar content below this figure. However, in some unfavorable conditions as much as 100 g/Nm^3 can be generated. Van der

Meijden et al. (2010) assumed generic tar concentration in a dry raw producer gas from fluidized reactors of 30 g/Nm^3. Gil et al. (1999) established the so-called "representative" values for tar contents in a dry producer gas. These values are 2 - 20 g/Nm^3 for gasification with air, 4 - 30 g/Nm^3 for gasification with steam-O_2 mixtures and 30 - 80 g/Nm^3 for gasification with steam. The yield of tar from FICFB gasifier is between 1.5 and 4.5 g/Nm^3 (Bolhàr-Nordenkampf et al. 2002a; Hofbauer et al., 1997).

Corella et al. (2006) presented that the amount of tar from air gasification of biomass decreases with process temperature and air to fuel ratio. On the other hand it increases with heat loss and biomass moisture content. The paper presents that only a relatively dry biomass (12% wt.) gasified with the air equivalence ratio $ER = 0.35$ results in tar content below 2 g/Nm^3. The authors concluded that the two most critical operational conditions are relatively high equivalence ratio and good in-bed material which determine the kinetic constants of the process.

2. MODEL FORMULATION

Within this work a single compartment model of a gasification process is being formulated. It is assumed that a wet solid biomass undergoes thermal conversion, that can be described using the following global stoichiometric reaction:

$$\begin{aligned} & n'_C CH_{b1}O_{b2}N_{b3}S_{b4} + n'_{H2O}H_2O(l) + n_{H_2O}H_2O(g) + \lambda n_{O_2\,\min} + n_{N_2}N_2 = n''_{H_2}H_2 + n''_{CO}CO \\ & + n''_{CH_4}CH_4 + n''_{C_2H_4}C_2H_4 + n''_{CO_2}CO_2 + n''_{H_2O}H_2O(g) + n''_{N_2}N_2 + n''_{SO_2}SO_2 \\ & + n''_C CH_{c1}O_{c2}N_{c3} + n''_{tar}C_{t1}H_{t2}O_{t3} + n''_{NH_3}NH_3 \end{aligned} \tag{1}$$

The process also involves ash and argon from the atmospheric air which are regarded as inert substances.

Biomass fuel is defined by a proximate and ultimate analysis. The total mass composition from the ultimate analysis of biomass is:

$$c + h + o + s + n + w + ash = 1 \tag{2}$$

Using this data the number of moles of each substrate entering the system with the fuel can be determined:

$$n'_C = \frac{c}{M_C};\ n'_{H_2} = \frac{h}{2M_H};\ n'_{O_2} = \frac{o}{2M_O};\ n'_S = \frac{s}{M_S};\ n'_{N_2} = \frac{n}{2M_N};\ n'_{H_2O} = \frac{w}{M_{H_2O}} \tag{3}$$

The stoichiometric formula of biomass in Equation (1) can be calculated:

$$b_1 = \frac{2n'_{H_2}}{n'_C};\ b_2 = \frac{2n'_{O_2}}{n'_C};\ b_3 = \frac{2n'_{N_2}}{n'_C};\ b_4 = \frac{2n'_S}{n'_C} \tag{4}$$

A bottom feeding of biomass into the reactor is taken into account. The operating temperature is considered to be within the range of 800 – 900°C. The reactor consists of two zones, the so-called bed zone and freeboard. Within the bed zone flash pyrolysis oxidation and char gasification take place. In the freeboard zone there are only homogenous reactions in the gas phase. Any solid particle of char present in the freeboard is considered to be chemically inactive. Gaseous products passing from the bed to freeboard are CO, CO_2, H_2, CH_4, C_2H_4, H_2O, N_2, tar and SO_2 (if sulphur is present in the fuel). Solid products leaving the reactor are ash and unconverted char. The bed material is assumed to have some catalytic properties and the residence time of char particle in the bubble bed is high. Therefore, it can be assumed that carbon conversion efficiency is high and gas composition is close to the equilibrium.

The composition of the product gas is determined assuming the state of thermodynamic equilibrium in gaseous phase. Using the approach based on Gibbs free energy minimization, the objective function can be formulated:

$$G = \sum_{i=1}^{ls} n_i \mu_i \rightarrow \min \tag{5}$$

Given that the chemical potential is equivalent to the partial free enthalpy of components in the mixture and the mixture is a perfect solution of ideal gases at a specified pressure, *p*, and temperature, *T*, it is possible to write the objective function in the following form:

$$\sum_{i=1}^{ls} n_i \left[g_i^0 (T, p) + RT \ln z_i \right] \rightarrow \min \tag{6}$$

Where the partial Gibbs free energy of a pure component is:

$$g_i^0 (T, p) = h_i^0 (T) - T s_i^0 (T, p) \tag{7}$$

H_2, CO, CH_4, CO_2 and H_2O are considered to be the only products i of the gas phase reactions at the equilibrium. Tar, char, NH_3, C_2H_4 and a portion of CH_4 yield are considered to be non-equilibrium products of the process. These components are just withdrawn from the substrates and bypass the calculation of equilibrium.

The constraints for the minimization of the objective function (6) are substance balances of carbon, hydrogen and oxygen:

$$b_1 = n'_C - n''_C - n''_{CO} - n''_{CO_2} - n''_{CH_4} - 2n''_{C_2H_4} - t_1 n''_{tar} = 0 \tag{8}$$

$$b_2 = n'_{H_2} + n'_{H_2O} + n_{H_2O} - n''_{H_2} - n''_{H_2O} - 2n''_{CH_4} - 2n''_{C_2H_4} - \frac{1}{2}\left(c_1 n''_C + t_2 n''_{tar} + 3n''_{NH_3}\right) = 0 \tag{9}$$

$$b_3 = n'_{O_2} + \lambda n'_{O_2\,\mathrm{min}} + \frac{1}{2}\left(n'_{H_2O} + n_{H_2O}\right) - n''_{CO_2} - n''_{SO_2} - \frac{1}{2}\left(n''_{CO} + n''_{H_2O} + c_2 n''_C + t_3 n''_{tar}\right) = 0 \tag{10}$$

The total number of moles of the raw producer gas at the output of the gasifier is:

$$n''_{pg} = n''_{H_2} + n''_{CO} + n''_{CH_4} + n''_{C_2H_4} + n''_{CO_2} + n''_{H_2O} + n''_{N_2} + n''_{SO_2} + n''_{Ar} + n''_{tar} + n''_{NH_3} \tag{11}$$

It is assumed that remaining solids consist of ash and unconverted char $CH_{c1}O_{c2}N_{c3}$. The mass of carbon in the char is determined using the assumed value of carbon conversion efficiency, which can be given by:

$$\varepsilon_C = \frac{n'_C - n''_C}{n'_C} \tag{12}$$

To calculate the amounts of hydrogen and oxygen in the char the empirical correlations developed by Richard et al. (2002) are used:

$$\frac{1}{c_1} = 0.55 \exp\left[0.0032 (T - 273)\right] \tag{13}$$

$$\frac{1}{c_2} = 1.7 \exp\left[0.0035 (T - 273)\right] \tag{14}$$

The amount of nitrogen in the char is assumed to be equal to 40% of fuel nitrogen. The remaining 60% of nitrogen present in biomass forms ammonia NH_3 (Van der Drift, 2001).

The tar present in the products is modelled as a mixture of benzene C_6H_6, naphthalene $C_{10}H_8$ and phenol C_6H_5OH. The molar composition of the tar is assumed as follows: C_6H_6 - 30%; $C_{10}H_8$ - 50%; C_6H_5OH - 20%. The yield of the tar is calculated in grams per kg of dry biomass fuel (g/kg$_{db}$) using the following formula:

$$m_{tar} = \delta_1 \left(90 f_{H_2O} + 30\right)\left(1 + w_{db}\right) \tag{15}$$

To estimate the yield of methane it is assumed that a portion of methane from pyrolysis remains unconverted during the process and goes into the product gas. This amount of CH_4 is associated with fuel hydrogen content. Therefore, it can be written as:

$$n''_{CH_4} = \delta_2 n'_{H_2} + \left(n''_{CH_4}\right)_{eq} \tag{16}$$

The yields of hydrocarbons C_2H_2, C_2H_4 and C_2H_6 are represented by equivalent yield of C_2H_4 that is assumed to be equal to:

$$n''_{C_2H_4} = \delta_3 n'_{H_2} \tag{17}$$

The variables ε_C, δ_1, δ_2 and δ_3 are four undetermined parameters of the model. In the process of model verification these parameters are varied to obtain the best agreement of modelled results with experimental data.

Finally, there are 5 unknowns to be determined by the minimization of the objective function (6). These are n''_{H2}, n''_{CO}, n''_{CO2}, n''_{H2O} and $(n''_{CH4})_{eq}$. The task can be solved by the method of undetermined Lagrange multipliers. In this case the modified objective function to be minimized takes the form:

$$F = \sum_{i=1}^{ls} n''_i \left[\frac{g_i^0(T,p)}{RT} + ln\, n''_i - ln\, n''_{pg} \right] + \sum_{k=1}^{3} \lambda_k b_k \rightarrow min \tag{18}$$

Minimization procedure now requires a solution of eight equations with eight unknowns (including λ_1, λ_2, λ_3). Five equations result from the condition of zeroing the partial derivatives of function F with respect to the unknowns:

$$\frac{\partial F}{\partial n''_i} = 0 \tag{19}$$

Equations (18) for gas components take the final form:

$$n''_i = n''_{pg}\, exp\left[-\frac{g_i^0(T,p)}{RT} - \sum_{k=1}^{3} \sum_{j=1}^{ls} \lambda_k a_{kj} \right] \tag{20}$$

The remaining three equations are substance balances (8), (9) and (10).

The temperature T is determined from the energy balance of the process. For 1 kg of a wet biomass at the input temperature 298 K the balance of the process takes the form:

$$\begin{aligned} LHV_{db}(1-w) - wr + \sum_i n_i \Delta h_i \Big|_{298K}^{T_i} + Q_{in} = \sum_j n''_j \left(MLHV_j + \Delta h_j \Big|_{298K}^{T} \right) \\ + n''_C \left(MLHV_C + \frac{c_1}{2} MLHV_{H_2} \right) + \left(m_{char} c_{char} + m_{ash} c_{ash}\right)(T - 298) + Q_{out} \end{aligned} \tag{21}$$

The heat capacity of the char is calculated using the following formula (Thunman et al., 2001)

$$c_{char} = -334 + 0.441T - 3160 \cdot 10^{-6} T^2 + 1010 \cdot 10^{-9} T^3 - 119 \cdot 10^{-12} T^4 \text{, J/kgK} \tag{22}$$

The heat capacity of the ash is modelled using a correlation developed by Kirov (1965), that for ash takes the following form:

$$c_{ash} = 0.18 + 1.4 \cdot 10^{-4} T \text{, kJ/kgK} \tag{23}$$

A code has been written using Engineering Equation Solver (EES) to solve the reactor model. Properties of substances are calculated using JANAF and NASA tables attached to the software package as the external routines.

3. RESULTS AND DISCUSSION

In order to verify the model the results of calculations were compared with experimental data published in the literature. The first set of experimental data was taken from Miccio et al. (1999). The gasified feedstock was beech wood of the following characteristics (dry basis): volatiles – 84.90%, fixed carbon – 14.07%, ash – 1.03%; mass composition: C – 42.18%, H – 6.03%, N – 0.22%, O – 50.54%; *LHV* – 18380 kJ/kg. Detailed results of gasification are reported for the temperature T = 800ºC and equivalence ratio ER = 0.15. The composition of the dry product gas was: H_2 - 15.02%, CO - 19.20%, CH_4 - 6.13%, C_2H_x - 2.30%, CO_2 - 18.20%, N_2 - 39.55%. The carbon conversion efficiency of the experiment was 78% (read from the chart). In the simulation the assumed values of ε_C, T and ER were kept the same as the reported experimental ones. To find values of the undetermined parameters δ_1, δ_2 and δ_3 the EES embedded genetic algorithm optimization procedure was applied. The minimization of the sum of squares of relative deviations between the measured and calculated concentrations of the product gas components the objective was:

$$\Gamma = \sqrt{\sum_i \left(\frac{(z_i)_e - (z_i)_m}{(z_i)_e} \right)^2} \rightarrow min \tag{24}$$

The number of individuals was set to 64 and the number of generations was set to 256. The optimized undetermined parameters were: $\delta_1 = 0.0024$, $\delta_2 = 0.1231$ and $\delta_3 = 0.0445$. The calculated composition of the dry product gas (tar and argon free) was: H_2 - 19.44%, CO - 15.71%, CH_4 - 6.56%, C_2H_x - 2.37%, CO_2 - 21.74%, N_2 - 34.19%. The calculated lower heating value of the gas was 175389 kJ/kmol versus 170254 kJ/kmol calculated for the experimental composition. The influence of undetermined parameters on the objective function Γ (24) is shown in Fig. 1. The relative deviations of gas heating value are shown in Fig. 2.

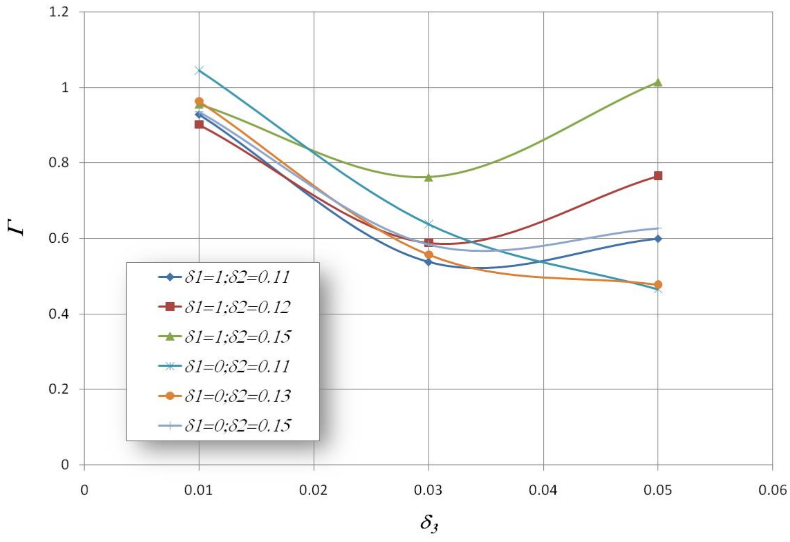

Fig. 1. Objective function Γ (24) as a function of undetermined parameters δ_1, δ_2 and δ_3

The second set of experimental data was taken from the work presented by Narváez et al. (1996). The data is presented in Table 1. The gasified feedstock was pine sawdust of the following characteristics (dry basis): volatiles – 81 to 83%, fixed carbon – 16 to 17%, ash – 0.5 to 1.2%; mass composition: C - 50.0%, H – 5.7%, N – 0.1 to 0.3%, O – 44.1%, S – 0.03%; *LHV* – 18000 to 18400 kJ/kg. Again the optimization procedure was applied to find the values of undetermined parameters. In this case, however, the value of ε_C was not known so it was added to the set of decision variables.

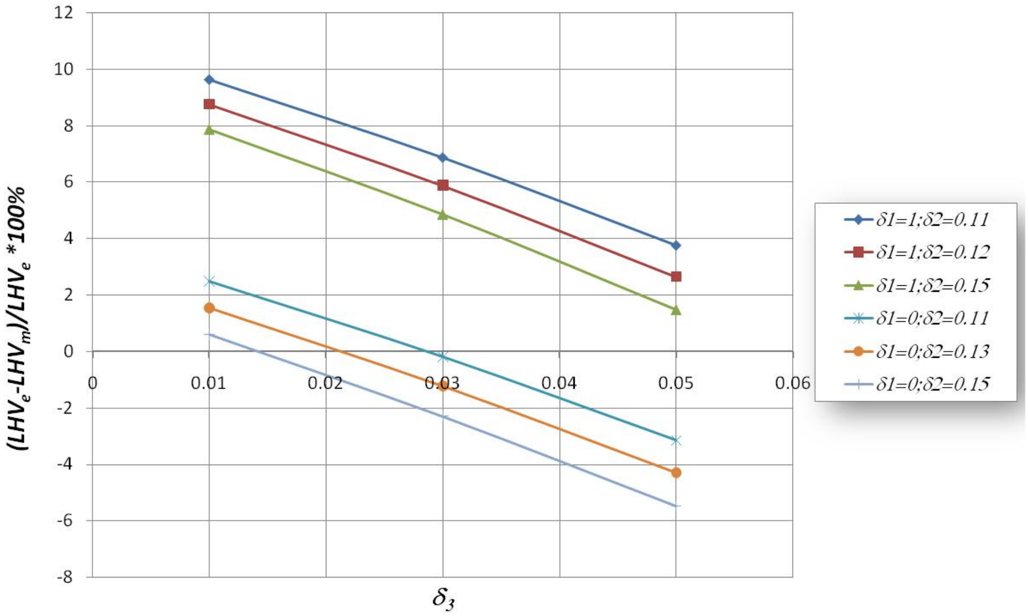

Fig. 2. Relative deviation of heating value of the gas as a function of undetermined parameters δ_1, δ_2 and δ_3

Table 1. Experimental data presented by Narváez et al. (1996)

Biomass moisture, %wt	23.5	21	23	22	25
ER	0.32	0.37	0.47	0.26	0.36
Freeboard temperature, °C	540	550	500	600	560
Bed temperature, °C	800	800	810	800	790
Dry tar free gas composition, %vol					
H_2	7	9.5	8	9.5	9.5
CO	14	13	10	13	13
CO_2	13.5	15	12	15	15
CH_4	3	2.7	2.4	2.7	2.7
C_2H_4	1.2	1.6	1.1	1.6	1.6
N_2	61.3	58.3	66.5	58.3	58.3
Concentration of tar, mg/Nm^3	3733	7163	2987	2011	2011
Gas yield, Nm^3/kg daf	2.3	2.5	2.5	2.1	2.4
VLHV, MJ/Nm^3	4.3	4.6	3.7	4.6	4.6
Stoichiometric oxygen requirement, kmol/kmol of gas	0.2010	0.2145	0.171	0.2145	0.2145

It was found that the minimization of the objective function Γ (24) resulted in relatively high deviations of the heating value of the gas and stoichiometric oxygen requirement for its combustion. In most cases also carbon conversion efficiency was too optimistic. Therefore, the objective function was modified to the following form:

$$\Gamma' = \sqrt{\left(\frac{V_e - V_m}{V_e}\right)^2 + \left(\frac{VLHV_e - VLHV_m}{VLHV_e}\right)^2} \rightarrow min\,, \tag{25}$$

The idea behind this modified approach was to minimize errors of the estimation of the product gas chemical energy flow. In this way the energy balance of a plant is close to the realistic one. The results of the calculation for a fixed value of freeboard temperature are given in Table 2.

Table 2. Predicted product gas characteristics

1. Dry tar free gas composition, %vol					
H_2	15.56	11.21	5.55	22.22	11.22
CO	5.15	5.25	1.34	9.32	4.19
CO_2	24.08	23.99	24.96	21.46	24.63
CH_4	5.03	6.69	5.18	2.44	5.37
C_2H_4	0.29	0.56	1.30	0.31	1.59
N_2	49.89	52.30	61.67	44.25	53.00
Concentration of tar, mg/Nm^3	12680	1029	3668	18630	6993
Gas yield, Nm^3/kg daf	2.27	2.50	2.69	2.08	2.40
VLHV, MJ/Nm^3	4.30	4.60	3.39	4.63	4.60
Heat losses, % of fuel energy input	13	19	30	5	16
Stoichiometric oxygen requirement	0.213	0.233	0.177	0.216	0.232
2. Relative deviations from data presented by Narváez et al. (1996)					
H_2	-122.29	-18.00	30.60	-133.89	99.29
CO	63.22	59.62	86.59	28.28	67.77
CO_2	-78.37	-59.93	-108.00	-43.07	-64.20
CH_4	-67.73	-147.93	-115.79	9.74	-98.93
C_2H_4	75.65	65.19	-18.09	80.58	0.50
N_2	18.61	10.29	7.26	24.10	9.09
Concentration of tar, mg/Nm^3	-239.67	85.63	-22.80	-826.40	-247.74
Gas yield, Nm^3/kg daf	1.48	0.00	-7.72	1.14	0.00
VLHV, MJ/Nm^3	-0.07	0.04	8.41	-0.67	-0.04
Stoichiometric oxygen requirement	-5.95	-8.57	-3.52	-0.60	-8.27
3. Calculated values (EES genetic algorithm)					
δ_1	0.7252	0.0671	0.2510	0.9952	0.4154
δ_2	0.0157	0.1747	0.1992	0.0117	0.1727
δ_3	0.0104	0.0219	0.0550	0.0101	0.0600
ε_C	0.8938	0.9925	0.9966	0.8188	0.9868

In another test also the temperature of the process was added to the set of decision variables for optimization procedure. It was found that in both cases the relative deviations from the experimental values of *VLHV*, gas yield and stoichiometric oxygen requirement were reduced. It was also concluded from the results that a correlation between the calculated carbon conversion efficiency ε_C and equivalence ratio *ER* can be established. This observation is in agreement with some published data Li et al. (2004). Therefore, the generated results were put together with other values of carbon conversion efficiency that were collected from the literature for wood gasification (De Jong, 2005; Miccio, 1999; Van der Drift, 2001). A single variable function has been established and it is shown in Fig. 3. The lack of experimental data does not allow for better estimation of influence of process parameters such as pressure, temperature, presence of in-bed catalyst and steam delivered on conversion efficiency.

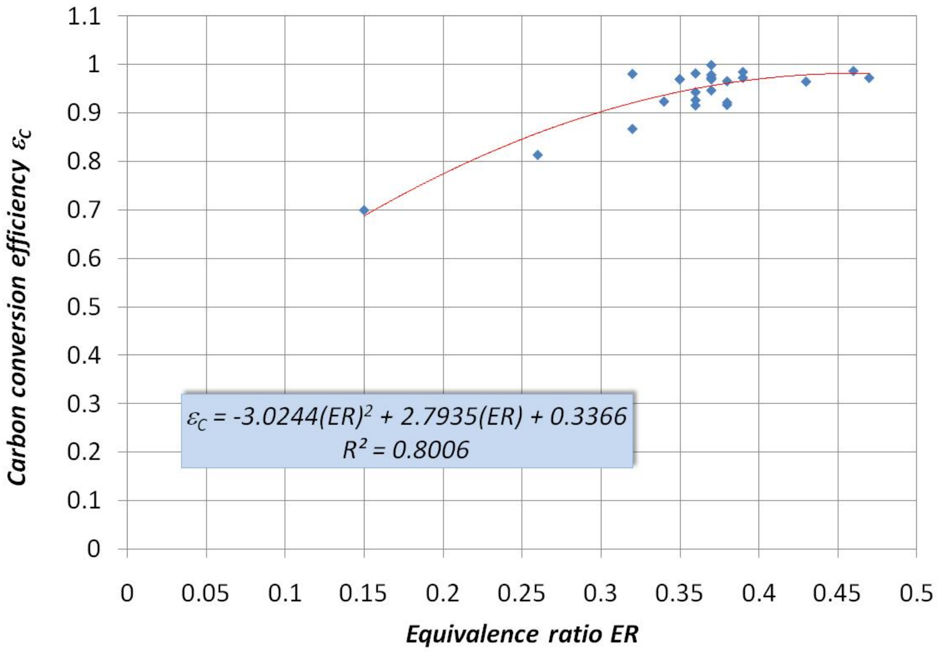

Fig. 3. Carbon conversion efficiency of authothermal air gasification of wood in fluidized bed gasifier as a function of equivalence ratio

A similar correlation of ε_C as a function of *ER* was presented by Li et al. (2004) for an experimental CFB gasifier. However, the correlation developed in this work results in higher values of carbon conversion efficiency.

The experimental data presented by De Jong (2005) were used in another analysis. The gasified feedstock consisted of wood pellets of the following characteristics (as received basis): volatiles – 74.9%, fixed carbon – 16.5%, ash – 0.13%, moisture – 8.4%; mass composition: C – 47.0%, H – 6.5%, N – 0.15%, O – 46.1%, S – 0.1%; *HHV* – 18600 kJ/kg.

After the optimization using the objective function (25) it was found that the variations of the optimal values of undetermined parameters were: $\delta_1 = 0.1$ to 0.96, $\delta_2 = 0.015$ to 0.20 and $\delta_3 = 0.016$ to 0.06. It was not possible to establish any reasonable correlation between the observed values and excess air ratio λ. Therefore, it was finally decided to use the average values from all the test runs of the model. These values are $\delta_1 = 0.40$, $\delta_2 = 0.12$ and $\delta_3 = 0.025$. A comparison between the experimental and modelling results, that were obtained using carbon conversion efficiency given in Fig. 3 and the average values of δ_1, δ_2 and δ_3, is presented in Table 3. The calculations were made using a given experimental freeboard temperature.

Table 3. Comparison of model results with experimental data presented by De Jong (2005)

run	011030	011127	020111	020129	020205	020212	020220	020226
ER	0.36	0.32	0.39	0.38	0.37	0.39	0.37	0.46
Freeboard temperature, K	1057	1004	1037	1092	1090	1103	1033	1078
Bed temperature, K	1160	1131	1175	1214	1207	1167	1087	1117
Gasification pressure, kPa	350	350	350	500	500	500	350	350
Steam to air mass ratio	0	0	0	0	0	0.1	0.099	0.1
Dolomite to fuel ratio	0	0	0.036	0.036	0.036	0.036	0	0
Experimental characteristics of the product gas								
CO, %vol	9.74	11.3	9.13	10.67	9.62	7.16	7.42	5.78
H_2, %vol	6.82	7.27	5.24	6.39	6.37	6.09	6.18	5.38
CH_4, %vol	3.91	3.97	3.26	3.91	3.87	3.26	3.01	2.48
C_2H_4, %vol	0.8	0.86	0.57	0.33	0.30	0.34	0.63	0.45
C_2H_6, %vol	0	0.2	0.09	0.07	0.09	0.11	0.13	0.07
CO_2, %vol	14.89	14.82	14.81	14.90	15.37	15.29	14.87	15.12
H_2O, %vol	11.09	11.77	19.12	12.21	13.86	18.54	18.70	22.20
N_2, %vol	52.1	49.12	47.12	50.90	49.83	48.64	48.49	47.91
Ar, %vol	0.5	0.53	0.60	0.57	0.57	0.54	0.54	0.56
HHV, MJ/Nm^3	4.17	4.64	3.55	3.98	3.82	3.27	3.41	2.73
ε_C	0.982	0.981	0.973	0.966	0.973	0.99	0.979	0.987
Stoich. oxygen requirement	0.185	0.205	0.157	0.176	0.169	0.146	0.152	0.121
Calculated characteristics of the product gas								
CO, %vol	10.15	10.07	9.02	9.82	10.08	6.71	6.41	4.99
H_2, %vol	10.22	11.61	9.38	9.12	9.44	9.18	10.51	7.16
CH_4, %vol	3.14	3.52	3.00	2.99	3.03	2.65	2.76	2.48
C_2H_4, %vol	0.65	0.68	0.61	0.62	0.63	0.55	0.57	0.52
CO_2, %vol	14.41	14.84	14.79	14.00	13.93	14.36	15.07	15.22
H_2O, %vol	12.66	12.13	12.45	12.84	12.80	20.13	19.69	20.55
N_2, %vol	48.04	46.42	50.01	49.89	49.38	45.71	44.29	48.34
Ar, %vol	0.56	0.53	0.58	0.57	0.56	0.52	0.51	0.58
HHV, MJ/Nm^3 (tar free wet gas)	4.15	4.44	3.837	3.916	4.004	3.512	3.659	3.004
ε_C	0.9503	0.92	0.966	0.961	0.9562	0.966	0.9562	0.9816
Stoich. oxygen requirement	0.184	0.199	0.170	0.173	0.177	0.149	0.157	0.126
Relative deviations between calculated and experimental values								
HHV, MJ/Nm^3	0.60	4.42	-8.08	1.61	-4.82	-7.40	-7.30	-10.04
ε_C	3.23	6.14	0.72	0.52	1.73	1.93	2.33	0.55
Stoich. oxygen requirement	0.56	2.79	-8.27	1.67	-4.52	-2.35	-3.40	-3.82

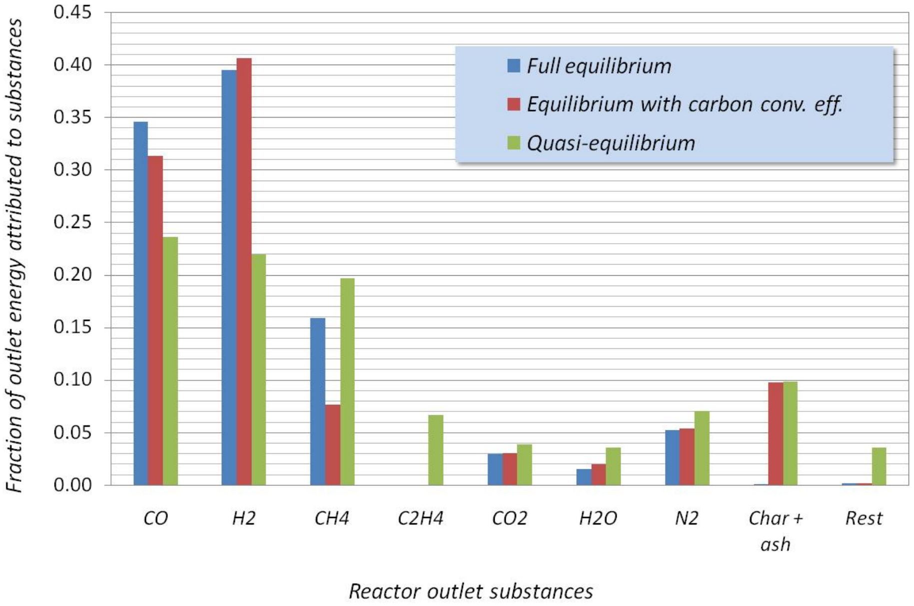

Fig. 4. Distribution of enthalpies of substances leaving the reactor calculated using various approaches towards equilibrium modelling

Table 4. Comparison of results from different thermodynamic equilibrium based models

Approach	Full equilibrium	Equilibrium with ε_C	Quasi-equilibrium
δ_1	0	0	0.4
δ_2	0	0	0.12
δ_3	0	0	0.025
ER	0.3	0.3	0.3
Product gas composition (tar free), %vol			
CO	15.63	14.2	11.29
H_2	20.71	21.4	12.20
CH_4	2.6	1.262	3.42
C_2H_4	0	0	0.71
C_2H_6	0	0	0.00
CO_2	13.76	13.57	14.14
H_2O	9.259	11.29	16.99
N_2	37.48	37.7	40.53
Ar	0.4798	0.4826	0.52
Tar free gas *LHV*, kJ/Nm^3	5146	4562	4395
ε_C	1	0.9025	0.9025
Dry gas yield, Nm^3 /kg$_{db}$	2.487	2.417	2.101
Process temperature, K	929.9	947.4	1128
Tar yield g/Nm^3 dry gas	0	0	6.719
$\dfrac{\left(V_{dg}VLHV_{dg}\right)_F-\left(V_{dg}VLHV_{dg}\right)_Q}{\left(V_{dg}VLHV_{dg}\right)_Q}$	0	0.187	0.276

Eventually a comparison was made between different equilibrium approaches towards modelling of a gasification process. The reactor is authothermal and atmospheric air at 298 K is used as the gasification agent. The calculations were performed for wood of the heating value LHV_{db} = 17680 kJ/kg and moisture content of 15%. According to Van der Drift et al. (2001) and Corella et al. (2006) heat losses from the gasifier were assumed to be 3% of the total heat released. The results are presented in Table 4. The share of enthalpy of particular outlet product in the total reactor outlet energy is presented in Fig. 4. It can be observed that differences between the results are quite considerable. It is important that the deviation in dry gas total enthalpy is at the level of 20 - 30 %, depending on the modelling approach. It may have a significant influence on the results of an the examined energy system study.

Table 5. Comparison of modelling results with experimental data presented by Hofbauer et al. (1997) and Pfeifer et al. (2004)

1. Experimental results			
Gasification temperature, ºC	745	850	838
In-bed catalyst	none	olivine	Ni-olivine
H_2, %vol	31.5	38.9	43.9
N_2, %vol	2.79	n.a.	n.a.
CO, %vol	22.66	29.1	27.2
CH_4, %vol	11.21	11.4	8.3
CO_2, %vol	27.46	17.5	18.8
C_2H_4, %vol	3.52	2	1.3
C_2H_6, %vol	0.55	n.a.	n.a.
higher C_xH_y, %vol	0.31	n.a.	n.a.
LHV, kJ/Nm^3	13015	13800	12400
V_g, Nm^3 dry gas/kg of dry biomass	n.a.	0.95	0.99
2. Results of calculation			
δ_1	0.0105	0.0019	0.0156
δ_2	0.2258	0.2025	0.1605
δ_3	0.0668	0.0367	0.0254
H_2, %vol	42.36	45.56	48.89
N_2, %vol	0.03	0	0
CO, %vol	18.06	18.02	22.99
CH_4, %vol	12.26	11.3	8.1
CO_2, %vol	23.38	23.15	18.74
C_2H_4, %vol	3.486	1.96	1.26
C_2H_6, %vol	0	0	0
higher C_xH_y, %vol	0	0	0
LHV, kJ/Nm^3	13286	12392	11824
V_g, Nm^3 dry gas/kg of dry biomass	1.34	1.36	1.39
V_g estimated using corelation given by Fercher et al. (1998)	1.02	1.23	1.20

The model was also used to simulate an FICFB process. The results presented by Hofbauer et al. (1997) and Pfeifer et al. (2004) were used for tuning and validation of the model. In the work presented by Hofbauer et al. (1997) the gasified feedstock were wood chips of the following characteristics (wet basis): volatiles – 73.3%, fixed carbon – 14.0%, moisture – 12.1%, ash – 0.6%, *LHV* – 15600 kJ/kg; mass composition (dry basis): C – 51.5%, H – 6.3%, N – 0.22%, O – 44.3%, S – 0.05%. Pfeifer et al. (2004) used wood chips of the characteristics as follows (dry basis): C – 49.0%, H – 6.52%, N – 0.12%, O – 44.31%, S – 0.05%; *LHV* – 17120 kJ/kg.

During the calculations the temperature of the process was assumed the same as given in the cited publications. Three values of carbon efficiency were tested. The first one was $\varepsilon_C = 0.85$ as suggested by Schuster et al. (2001), the second one was $\varepsilon_C = 0.89$ as suggested by Fercher et al. (1998) and the third one was calculated from the correlation presented by Van der Meijden et al. (2010) that resulted in significantly lower values. It was found that at the highest value of ε_C the highest deviation was observed between the reported and calculated dry tar free gas yield values. At the lowest value of ε_C calculated using the correlation of Van der Meijden et al. (2010) the gas yield was in a relatively good agreement with the experiment but the generation of tar and the composition of the gas were radically different. At the value of $\varepsilon_C = 0.85$ the results of the calculation were in a relatively good agreement with the measurements (for a system using a nickel-olivine catalyst). The only relatively high deviation was observed in the case of gas yield. However, the calculated values were closer to the values of gas yield estimated from the correlation given by Fercher et al. (1998). The results are presented in Table 3.

Finally, for allothermal gasification in a catalytic bed using steam as the gasification agent a single value of carbon conversion efficiency of 0.85 is suggested. There were not enough data to decide the values of δ_1, δ_2, δ_3 for this process. For initial studies the values given in Table 5 can be used.

5. CONCLUSIONS

Thermodynamic equilibrium based models of fluidized bed biomass gasification if not constrained result in too optimistic results. An overestimated product gas yield and heating value can lead to a significant error of financial profitability analysis of a biomass-to-energy plant project. To correct results and forecast economic indices with a better accuracy it is recommended that gasification reactors are modelled using the constraint thermodynamic equilibrium approach. Four parameters can be specified to perform quasi-equilibrium calculations. These are the amounts of the non-equilibrium products, i.e. solid char, tar, CH_4 and C_2H_4. After tuning and validation of the model the average values of the constraints were proposed. It was found that predictions of the producer gas composition using the quasi-equilibrium approach very relatively close to the values reported for wood by several references. It has to be, however, stressed that despite a considerable number of experimental and analytical studies of fluidized bed biomass gasification process only few publications contain data suitable for validation of models. This conclusion is in line with observations made by Gòmez-Barea et al. (2010). Therefore, there is a need for future work aimed at updating the elaborated values of model parameters.

It was observed in different runs of the model that if the calculated content of H_2 in the product gas increases, the content of CO decreases as compared to the experimental values. As these gases have similar heating values and the same stoichiometric oxygen requirement it can be concluded that equilibrium models are good for systems with gas engines and turbines. In a possible analysis of a fuel cell system or gas combustion kinetics the results of equilibrium based calculations can lead to more significant errors.

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SYMBOLS

ash	mass fraction of ash in wet biomass, kg/kg
a_{kj}	the number of particles of element *k* (C, O_2, H_2) in the gas component *j*
b	indicator of constraint equation
c	mass fraction of carbon in wet biomass, kg/kg
c	heat capacity, kJ/kg/K
$CH_{b1}O_{b2}N_{b3}S_{b4}$	stoichiometric formula of biomass
$CH_{c1}O_{c2}N_{c3}$	stoichiometric formula of char
$C_{t1}H_{t2}O_{t3}$	stoichiometric formula of tar
f_{H_2O}	molar fraction of steam in gasification agent
g	gas
G	Gibbs free energy, kJ
g_i^0	standard partial free enthalpy of pure component *i*, kJ/kmol
h	mass fraction of hydrogen in wet biomass, kg/kg
h_i^0	standard enthalpy of pure component *i*, kJ/kmol
$\Delta h_i\big\|_{298K}^{T_i}$	physical enthalpy of component *i* at its temperature T_i, kJ/kmol
l	liquid
LHV	dry basis lower heating value of biomass, kJ/kg
ls	number of gaseous components
m	mass, kg
M	molecular mass, kg/kmol
MLHV	molar lower heating value of gaseous product, kJ/kmol
n	mass fraction of nitrogen in wet biomass, kg/kg_{wb}
n	number of moles of component, kmol
n'	primary number of moles of component in biomass, kmol
n''	number of moles of product, kmol
o	mass fraction of oxygen in wet biomass, kg/kg_{wb}
p	pressure, kPa
r	heat of evaporation of water, kJ/kg H_2O
R	universal gas constant 8.314 kJ/kmolK
s	mass fraction of sulfur in wet biomass, kg/kg_{wb}
s_i^0	standard entropy of pure component *i*, kJ/kmolK
T	temperature, K
Q_{in}	heat delivered to the process, kJ
Q_{out}	heat heat lost from the process, kJ
V	producer gas yield, Nm^3
VLHV	volumetric lower heating value of the gas, kJ/Nm^3
w	mass fraction of liquid water in wet biomass, kg/kg
z_i	molar fraction of component *i* in the mixture

Greek symbols

δ	model undetermined parameters
ε	conversion efficiency
Γ, Γ'	objective functions for optimization
λ	excess oxygen factor; Lagrange multipliers
μ_i	chemical potential of component *i*

subscripts

ash	related to ash
b_1, b_2, b_3,b_4	stoichiometric numbers of mols of components H, O, N in biomass, kmol
c_1, c_2, c_3	stoichiometric numbers of mols of components H, O, N in char, kmol
C	carbon
char	related to unconverted char
db	dry basis
dg	dry gas
e	related to experimental value
eq	resulted from equilibrium calculations
H	hydrogen
H_2O	water
i	element indicator
j	phase or product indicator
k	constraint indicator
m	related to modeled value
N	nitrogen
O	oxygen
O_2min	minimum number of moles of oxygen required for total combustion
pg	product gas
S	sulfur
t_1, t_2, t_3	stoichiometric numbers of mols of components H, O, N in tar, kmol
wb	wet basis

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