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MODELING THE GASIFICATION/COMBUSTION OF WOOD AND CHAR PARTICLES

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Abstract

The state of the art is discussed of transport models for combustion and/or gasification of single biomass/wood particles. A first group of models assumes that the stages of particle devolatilization and char conversion are sequential, where the description of the first stage is usually highly simplified. Models are also available for the sequential occurrence of the various processes of the solid phase which, in some cases, are coupled with detailed descriptions of the external gas-phase phenomena.

1. Introduction

Conversion of biomass in combustors and gasifiers results from a strong interaction between chemical and physical processes. In wood combustion the external gas phase reactions, the surface char reactions and the internal pyrolysis reactions at the surface of the virgin wood are coupled together (Bryden and Ragland (1997)). Thus models should be formulated which take into account both transport phenomena and intrinsic kinetics of the relevant chemical reactions. Compared with pyrolysis, a very few transport models for the combustion/gasification of lignocellulosic particles are available. In the simplest cases, wood/biomass devolatilization and char conversion have been considered as two separate processes. Based on this approximation, in principle, all the transport models of solid pyrolysis recently reviewed (Di Blasi (2006)) could be applied, followed by a model for the conversion of char. The models, specifically proposed for the two stages or for the sole combustion of char, are reviewed here. In a few cases, for biomass particles or black liquor drops, the simultaneous presence of both devolatilization and combustion/gasification reactions is considered. From the conceptual point of view, models for smoldering combustion of wood, generally developed for applications in fire safety science, may also be of a certain interest.

2.1 Models for the sequential occurrence of solid devolatilization and char conversion

Models, where the description of the devolatilization stage is followed by the combustion of char, are proposed by Mukunda et al. (1984), Saastamoinen and Richard (1985), Winter et al. (1996), Leckner et al. (1999), Gera et al. (2002). Models for the processes of char conversion only are presented by (Dasappa et al. (1994a, 1994b, 1998), Mermoud et al. (2006)).

Mukunda et al. (1984) investigated the combustion of wood (teak) spheres (10-25mm diameter). After forced ignition, burning in air takes place. It is observed that the process consists of separate stages. During flaming pyrolysis the sphere diameter and weight are reduced by about 10% and 70-80%, respectively, and the volatile species produced burn in the gas phase with a flame enveloping the sphere. During heterogeneous combustion of char the density decreases by about 10-25%, indicating the possibility of internal reactions with oxygen diffusing from the outer surface through the porous structure. For the largest sizes (diameters of 25mm), some overlap between the two phases is observed. The conversion process is described by one-dimensional models for the two separate stages (Mukunda et al. (1984)). Flaming pyrolysis is described by unsteady heat conduction through the virgin wood, coupled with a quasi-steady formulation of the convection/diffusion (conduction) equations for the char layer and the flame zones. Exothermic pyrolysis takes place at an infinitely thin front at a constant temperature. An infinitely thin reaction zone is also assumed for the flame. For the second stage, in addition to the gas phase equations where no reaction occurs, unsteady species and energy equations are written for the char, where the porosity is used to describe the changes caused by the heterogeneous combustion reaction. Simulation results reproduce the experimental trends. In particular, the particle diameter variation during the char combustion stage follows a d^2 law. Comparison with measurements shows char conversion times shorter than those measured associated with higher temperatures at the particle center (about 1600-1650K against 1000K).

The model proposed by Winter et al. (1996), for particle conversion in a fluidized-bed reactor, assumes that the processes of drying and devolatilization are separated from that of char combustion. The boiling temperature of water is a function of the capillary pressure which, in turn, depends on the pore distribution (values of the boiling temperature comprised between 373-473K). The unsteady heat conduction equation for a cylindrical particle is coupled with the assumptions of constant density and an Arrhenius reaction rate taking place across an isothermal shell. External heat transfer consists of a convective and a radiative contribution. Combustion is described by the unreacted-shrinking core approximation (the density is constant and the size of the particle varies) or by a constant particle size approximation (the particle size remains constant and the density/porosity varies). Simulations and experiments, made by varying several parameters indicate that the fuel properties (beech wood versus coal) are the most important factors affecting combustion, that is, the high volatile content of wood causes much shorter burnout times. Furthermore, the main parameters affecting particle temperature and char combustion times are the bed temperatures and oxygen concentrations.

Leckner et al. (1999) propose a model for the devolatilization of wood pellets and the subsequent combustion of char in a fluidized-bed reactor. The devolatilization model consists of the unsteady heat conduction equation and three devolatilization reactions for the three main components of wood with kinetic parameters as estimated by Gronli (1996). External heat transfer takes into account the convective term, following correlations specifically developed by the authors for fluidized-bed reactors and corrected by a factor of 0.85 in the case of very large particles. An analytical solution is proposed valid for infinite cylinders or slabs. The devolatilization times are predicted using constant property values and a zero value of the reaction enthalpy obtaining acceptable agreement with measurements (bed temperatures between 873-1173K, initial particle diameters of 6-10mm, moisture contents of about 8%db). Char particles are assumed to present the same size as the original wood pellet and to burn in accordance with the unreacted-shrinking-core treatment at a constant density in the absence of spatial gradients of temperature. The application of the model is limited to the evaluation of the parameters of the apparent combustion kinetics (activation energies of 28-48kJ/mol).

The biomass particle is treated as a coupled system for devolatilization and combustion in the model proposed by Gera et al. (2000) for co-firing applications in pulverized coal boilers. To take into account the large length/diameter ratio on the temperature gradients, the unsteady two-dimensional version of the heat conduction equation for cylindrical particles is used for the first stage. The one-step devolatilization reaction is first order in the amount of volatile released, as determined from proximate analysis, with guessed values of the kinetic parameters. The particle diameter (and density) is related, by means of a shrinkage factor, to the current amount of volatiles still retained by the particle. Once the devolatilization process is terminated, combustion occurs at the external particle surface and is controlled by oxidant diffusion and corrected by an enhancement factor to take into account the effects of non spherical shape. The char density is constant, while variations in the particle mass cause a continuous reduction in the diameter. The particle model is coupled with the gas phase processes in a pulverized coal burner, modeled by a commercial CFD code. A Lagrangian formulation is used for the particle phase and the coupling with the gas phase is made via production terms. Together with the cylindrical particle model, described above, a simplified model based on the assumption of perfectly stirred system (absence of spatial gradients) is also examined. It is found that, due to a large surface area, the transport model for the particle gives rise to shorter burn-out times. Shape effects are important for the particle trajectories and residence times. For a given volume, the sphere presents the minimum surface area. The cylindrical or ellipsoidal shape, typical of biomass particles, results in a larger surface (when compared with a sphere of equivalent volume). This enhances oxygen diffusion towards the char combustion zone, thus affecting both ignition and burn-out times. Compared with coal, it appears that much larger sizes of biomass particles can be used without increase in unburned

carbon in fly ash, owing to the higher content of volatile matter and aerodynamic (shape) factors which allow for longer residence times inside the boiler.

As for the char particle, the homogeneous and the shrinking core models represent extreme conditions of char gasification/combustion, corresponding to very rapid and very slow diffusion of reacting gases. Un example of a comprehensive model for coal char is proposed by Morel et al. (1990). It summarizes the state of the art for this fuel and also lists the chief literature. Global one-step kinetics are used for the gasification (CO_2 , H_2O and H_2) and oxidation reactions. Gas-phase oxidation of carbon monoxide and hydrogen and water-gas shift take place in the surrounding gas layer and in the void space of the char particles. A few models have also been proposed for wood char.

Dasappa et al. (1994a) couple the spherically symmetric one-dimensional equations for diffusion and convection of chemical species and energy in a porous medium with a volumetric rate expression for a multi-step mechanism of char gasification in the absence of oxidant. Effective values are introduced for the diffusion coefficients and the thermal conductivity. The equations for the gas layer adjacent the particle are assumed to be steady. This assumption coupled with that of a unit Lewis number allows a closed-form solution to be obtained for the profiles of the main variables. The conditions at the particle surface are based on the continuity of heat and mass fluxes. The most important assumption is that the particle volume remains constant during gasification while the porosity varies. Physical properties are selected in accordance with experimental measurements but the activation energy is used as an adjustable parameter to get quantitative predictions of the conversion curves. The agreement between measured and predicted conversion curves is good for low conversions (below 60%). The disagreement at high conversions is attributed to ash fusion which makes the carbon inaccessible to further gasification (Standish and Tanjung (1988)), a feature not described by the model. However, the validity of the model and the criticism about the shrinking-core regime evidenced by the measurements of Standish and Tanjung (1988) are highly questionable. Indeed, the low ash content of wood chars does not justify the constance of the particle size assumed by this model.

The gasification model (Dasappa et al. (1994a)) was properly modified to take into account char combustion (Dasappa et al. (1994b)). The main scope of the work was to produce a better agreement with measured conversion times and temperature profiles with respect to the previous combustion models (Dasappa et al. (1984)). The combustion reaction takes into account the adsorption and desorption processes. The model is used to interpret the results of an experimental activity focused on the effects of oxygen concentration and flow velocity on the weight loss characteristics and surface/core temperatures of wood char (spheres of 2-15mm diameters obtained from Ficus wood) burning in air after forced ignition. The burning time in air can be expressed by a power law ($n=1.87-2$) dependence on the particle diameter. Extinction occurs for reduced oxygen mass fraction (below 0.14 for air temperatures of 300K). Furthermore, only partial combustion is observed at ambient conditions when the Reynolds number exceeds 500. Parametric and sensitivity analyses are also carried out. The core temperature is significantly affected by the endothermicity of the gasification reaction and mainly by the thermal conductivity of char (namely its dependence on temperature). The former parameter also strongly affects the conversion time. Extinction reproduces results already well known for the idealized perfectly stirred reactor. Thus they are dominated by the ratio between the rates of heat generation and heat loss. Although the key variable for the description of the transformation of the char particle during conversion is the porosity (Dasappa et al. (1994a,b)), a comparison is proposed between two different approaches based on the assumptions that 1) the ash layer is not removed (constant particle size) or 2) ash layer is stripped away by the gas flow (continuously shrinking particle). Experimental measurements of the conversion times as functions of the Reynolds number are comprised between these two limits. In particular the assumptions 1) and 2) are best suited for the low and high Reynolds number, respectively.

The model presented by Dasappa et al. (1998) extends, with the inclusion of the effects of steam gasification, the work done by the same research group on wood char conversion by carbon dioxide gasification (Dasappa et al. (1994a)) and combustion (Dasappa et al. (1994b)). Experiments are carried out on single char spheres (diameters of 4-15mm) obtained from Ficus wood and exposed to temperatures in the range 1250-1390K with atmospheres typical of fixed-bed gasification (mixtures of steam, carbon dioxide, carbon monoxide and nitrogen). The steam gasification of char is modeled by a multi-step mechanism as in Blackwood and McGrory (1958) with equilibrium conditions for the water gas shift reaction. The activation energy, evaluated by examining the predictions of two conversion curves (temperatures of 1250 and 1388K) for a 8mm thick particle, is 217kJ/mol. The simulation results are highly affected by the char thermal conductivity, whose expression takes into account variations in both temperature and gas composition. The exponent of the power-law dependence of the conversion time on the particle diameter is comprised in the range 1-1.3 for carbon dioxide and steam environment (against 2 for air/oxygen). The differences with combustion are a consequence of different temperature and chemical species dynamics, in particular reaction energetics and reactant diffusivities, which also result in different limit values of the particle size for a transition from a chemical to a diffusional control. It is also found that the conversion time for carbon dioxide gasification is about 3.5 times longer than that for a steam atmosphere.

Recently a model for steam gasification of a thick wood char particle has been proposed (Mermoud et al. (2006)) to simulate experiments carried out for different particle sizes (diameters from 10 to 20mm), external temperatures (1100- 1300K), steam partial pressures (0.1-0.4 atm of steam), and external gas velocities (0.09-0.30m/s). The model comprises the conservation equations for mass (solid and gaseous species) and enthalpy (thermal equilibrium between the solid and the gas phase) with the Darcy Law for a spherically symmetric, one-dimensional particle. The following main assumptions are made: uniform conditions at the external surface and spherical shape during conversion, diffusion by the Fick law (effective diffusion coefficients), no tar formation, absence of chemical reaction in the pores of the particle, conversion described in term of the overall reaction of carbon gasification according to Blackwood and McGrory (1958) with adjustable parameters to fit the experimental results, the product of the surface area and reactivity is constant through gasification until the entire structure suddenly collapses. Also, the evolution of the char particle during gasification is described in terms of porosity and the boundary conditions are expressed in term of global heat and mass transfer coefficients. The fitting procedure allows the determination of kinetic parameters valid for every conversion for thin particles and for values up to 60% for thick particles. Apart from some critical assumptions of the model, such as that of spherical symmetry, the authors point out that mechanism phenomena may become controlling at this stage (large fracture formation, peripheral fragmentation), clearly indicating a need of further research effort in this sector.

2.2 Simultaneous occurrence of solid devolatilization and char conversion

Modeling the gasification/combustion of char particles after drying and pyrolysis of the starting material is a high simplification of the conversion process. Indeed, an overlap may be established of the different processes, as also observed for coal combustion (Saastamoinen et al. (1993)), Gurgel Veras et al. (1999)). Models accounting for this feature in wood conversion include the studies by Ragland et al. (1988), Saastamoinen and Richard (1989), Bryden and Ragland (1996, 1997), Ouedraogo et al. (1998), Galgano and Di Blasi (2006), Galgano et al. (2006). The models by Jarvinen et al. (2002, 2003) for black liquor gasification, and some models of smoldering combustion of wood (Souza Costa and Sandberg (2004), Boonmee and Quintiere (2005)) are also of interest given some similarities with biomass particles.

The work presented by Ragland et al. (1988) and Bryden and Ragland (1996, 1997) is focused on the conversion of chunk wood or whole-tree sized fuels for conditions typical of large boilers. In (Ragland et al. (1988)) experiments are carried out with samples of yellow poplar and spruce (15 cm diameter) inserted onto the grate of a spreader stoker boiler with characteristic temperatures above the bed of about 1473K. It is observed that chunk wood burns as a shrinking sphere (rate of about 1.8mm/min) consisting of a relatively thin layer of char (5-15mm) surrounding the core of unburned wood. The presentation of the mathematical model is not detailed, but it can be understood that devolatilization is described by an unsteady one-dimensional heat conduction equation including temperature-dependent physical properties (thermal conductivity, density, specific heats). Temperatures of 373 and 773K are associated with the occurrence of moisture evaporation and wood pyrolysis, respectively. Combustion occurs at the surface and is controlled by oxygen diffusion described by the Ranz-Marshall correlation with a correction factor for turbulence and non-spherical shape. The oxygen mass fraction is assumed to remain constant and equal to 0.049kg/m^3 . Acceptable predictions of the mass loss curves are reported.

Successive experiments (Bryden and Ragland (1997)) carried out with thick wood (thickness above 10cm) under condition typical of packed-bed reactors confirm that the processes of drying, pyrolysis and combustion/gasification are coupled. The measurements are used to get an empirical correlation for the growth of the char layer as a function of the initial and average diameters and the initial moisture content. This description of the pyrolysis process is coupled with a model for the heterogeneous conversion of char by means of one-step reactions for oxidation, steam gasification and carbon dioxide gasification with kinetic constants derived for coal and modifications in the surface area to account for cracks and fissures. Energy and mass balances are written for the char surface. In particular, mass balances are written by equating the reactant consumed by the reaction with char with the reactant flow through the surface and the diffusion of the reactant through the boundary layer. The predictions of the combustion rates and solid mass fractions agree within 30% of the measured values. The model of wood combustion presented by Ouedraogo et al. (1998) is still based on the shrinking core treatment of the reaction fronts. Compared with the work developed by Bryden and Ragland (1997), the heterogeneous combustion of char is assumed to be controlled by diffusion. The overall mass transfer coefficient incorporates the blowing correction but also an empirical geometry factor. The temperature of the unreacted core is uniform and equal to the ambient value, whereas a quasi-steady version of the energy conservation equations is written for the char and the core surface. However, as observed from the application of more complete pyrolysis models using the same treatment (Galgano and Di Blasi (2003)), this assumption highly affect the validity of the simulation results even from the qualitative point of view and introduces a degree of arbitrariness about the duration of the transients of the initial formation of the char layer and the related thickness. Therefore, the agreement between model predictions and the measurements by Bryden and Ragland (1997) is most likely the result of a specific selection for these parameters and those of the global mass transfer coefficient.

The most recent contribution in the modeling of wood log combustion is due to Galgano and Di Blasi (2006) and Galgano et al. (2006). The shrinking-unreacted-core of moist wood pyrolysis, previously developed (Galgano and Di Blasi (2003,2004,2005)) has been modified to include the processes of char gasification and combustion. Heterogeneous gasification/combustion is assumed to occur at the external surface of the char layer, consisting of pure carbon. The rates of the steam and carbon dioxide gasification are modeled as in Barrio and Hustad (2001) and Barrio et al. (2001), respectively. The rate of the oxidation reaction is assumed to be diffusion controlled. Moreover, it is assumed that the solid

phase is in thermal equilibrium with the volatile pyrolysis products and steam. The solid-phase model is coupled with a two-dimensional CFD model for the gaseous phase processes, solved by the commercial code CFX4.3. Coupling between the two models at the solid/gas interface is made assuming that the characteristic times of the gas phase processes are much shorter than those of the solid phase. Simulations of the combustion of single wood logs, as obtained with the coupled solid- and gas-phase models and with a solid-phase model alone incorporating global heat and mass transfer coefficients and a constant property gas phase, are compared with experimental measurements. It is found that acceptable agreement is obtained only in the first case, whereas the solid-phase model alone highly underestimates the conversion time and overestimates the mean mass loss rates.

A highly simplified model for the combustion of wood particles is presented by Thunman et al. (2002) with the scope of describing single particle effects in reactor modeling. The model for moisture evaporation and wood pyrolysis, proposed by Saastamoinen and Richard (1996), is extended to include char combustion and shrinkage for a finite cylinder and a parallelepiped. Similar to devolatilization, char combustion occurs at an infinitely thin region. The particle consists of four layers: moist wood, dry wood (where pyrolysis is under way), char and ash, where the size of each layer is determined by the amount of fuel at a certain stage of conversion. Temperature and other physical and thermo-chemical properties are averaged across each layer. Moreover, the ash layer is inert and fixed on the particle surface during the entire conversion process. Release of water and volatile pyrolysis products can be retained to take place at two assigned temperatures, although Arrhenius rate expressions are used to facilitate the numerical solution of the equations. Char is predominantly consumed by the oxidation reaction, but steam, carbon dioxide and hydrogen gasification is also taken into account. The rates of conversion are functions of the diffusion of the reactant from the bulk into the reactive surface and the reaction rate at the reactive surface. Comparison between measured and predicted times for devolatilization and char burn-out present some scatter, owing to the simplifications of the model.

The propagation of infinitely thin fronts of drying (temperature of 373K), pyrolysis (temperatures above 500K) and combustion (temperatures above 650K) across a horizontal semi-infinite log is modeled by Souza Costa et al. (2004). The propagation speed is assumed to be the same for the three fronts. The assumptions of one-dimensional burning, steady state, absence of chemical reactions between char and volatile pyrolysis products, absence of structural failure and thermal equilibrium for the solid and the gaseous phase reduce the mathematical model essentially to the enthalpy conservation equation for the moist wood, dry wood and char zones. Closed-form solutions are provided with and without circumferential heat losses and for the single front of drying and pyrolysis. The application of the model is limited to a parametric analysis about the effects of several parameters (initial moisture content, log diameter, char density) on the smoldering characteristics. The results are only qualitative and the assumptions made, specifically that of semi-infinite system and coincident rates of the thin fronts, are scarcely valid for thermochemical conversion processes.

To investigate the physical and chemical processes controlling the processes of surface oxidation (glowing), the wood pyrolysis model developed by Kung (1972), has been modified by Boonmee and Quintiere (2005). Compared with the original model, the chemistry of pyrolysis uses a first-order global reaction for each of the three main wood components. The oxidation of char is assumed to occur only at the surface (with kinetics derived from previous literature), where oxygen diffusion from the surrounding environment is described by a one-dimensional stagnant layer model. Surface regression, following char combustion, produces a moving boundary problem. It is observed that the char surface oxidation is initially controlled by chemical kinetics and then by oxygen diffusion. A temperature of about 673K is indicated as a boundary for the transition from one regime to another. Acceptable agreement is observed with measurements (Boonmee and Quintiere (2002)) but the kinetic constants

for both the pyrolysis and combustion reactions are significantly different from the values currently accepted.

The gasification of black liquor drops modeled by Jarvinen et al. (2002, 2003) is of importance from the conceptual point of view for the topic examined here. The authors of this work observe that thick and wet particles present the simultaneous presence of drying, devolatilization and combustion/gasification zones. Furthermore, CO₂ and H₂O produced from the more internal zones of the particle may cause the conversion of char into volatile species, while crossing the more external particle layer given temperatures sufficiently high. This process, indicated as "autogasification", can be very important for wet, thick fuel particles and for fuels with a very reactive char, such as in the case of black liquor.

The unsteady model developed by Jarvinen et al. (2002, 2003) takes into account the chief processes of the solid phase, including autogasification, for a one-dimensional spherical particle, but extra-particle processes are neglected. The different phases are in local thermal equilibrium. The fluxes of gas in a multi-component gas mixture in a porous medium are given by the Stefan-Maxwell diffusion equations combined with the viscous contribution and Knudsen flow. Drying is assumed to be heat transfer controlled with the local boiling temperature function of the solid mass fraction. More precisely, the droplet is assumed to be isothermal (a perfectly mixed reactor) until a critical water content (10%) is reached. The pyrolysis rate is assumed to be linearly dependent on the heating rate, whereas the ratio between volatiles and char yields is an empirical function of temperature. One-step global kinetics are used for CO₂ and H₂O gasification together with finite-rate kinetics for the water-gas shift reaction. Swelling is modeled by using a uniformly expanding grid and an empirical swelling coefficient. Char conversion, with reference to each single cell of the discretized domain, occurs with constant volume until a conversion of 80% is reached. Then the cell is eliminated and the remaining mass is transferred into the other cells in proportion to their volumes. Simulations indicate that autogasification becomes important when the surface temperature exceeds 1173K, the Biot number 0.5 and pyrolysis and drying are still under way. These results are peculiar of black liquor. For other fuels (wood, peat) where the gasification rates of char are much slower, much longer pyrolysis times (thicker particles) are required. In other words autogasification is important when the ratio of the time for chemical conversion of the char and pyrolysis is small.

3. Conclusions

Transport models for the dynamics of single wood/biomass particles belong to three main categories:

- a) models for the sequential occurrence of pyrolysis followed by the gasification and combustion of char;
- b) models for the combustion/gasification of char;
- c) models for the simultaneous occurrence of both pyrolysis and gasification/combustion.

Models of type a) consider isolated particles burning in air, after forced ignition, in fluidized beds and in co-firing applications by means of pulverized coal boilers. Simplified description of the pyrolysis stage are followed by the unreacted shrinking core (the density is constant and the size of the particle varies) or the constant particle size (the particle size remains constant and the density/porosity varies) approximation for char conversion. These treatments are generally used also by the models of type b). In this case, experimental validation has been carried out for char particles in different atmospheres (air, carbon dioxide, steam). It is observed that, for large particles typical of fixed-bed gasifiers, predictions are accurate only up to 60% conversion. The difficulty in the correct simulation of high conversions is attributed to either anisotropy and peripheral fragmentation or ash fusion, which makes carbon inaccessible to further gasification.

The most comprehensive models of type c) include descriptions limited to the porous solid only and both to the porous solid and the adjacent gas phase. The most advanced treatment describes the conversion of chunk wood or whole-tree size fuels for conditions typical of large boilers, using the unreacted-core-shrinking treatment for the heterogeneous processes of char conversion. The main difference among models lies in the different degree of detail for the pyrolysis models. As for the experimental validation, particularly important is the description of the processes occurring in the external gas phase for the quantitative prediction of the conversion time and the average rate of volatile release.

Future research should consider the development of more accurate models, including both chemical kinetics and structural modifications during char conversion. In fact, the phenomena of reactive surface evolution and particle fracturing are scarcely understood. More accurate descriptions of char conversion should be coupled with accurate models for the pyrolysis stage already available. Furthermore, experimental data, for model validation, should be produced under well defined conditions. The development of simplified models is also of interest for both design and scaling rule and for incorporating single particle effects in reactor models.

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