Modeling of Dense Reactive Granular Flows

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Introduction

Many practical multiphase flow systems involve the flow of a dense, reactive, gas-particle mixture. Examples include coal combustion, catalyst regeneration and pyrolysis applications, among others. Because of the lack of fundamental knowledge of both the fluid mechanical and thermal behavior of these systems, the design of these processes has been necessarily based on empirical correlations and experiments performed in laboratory or pilot scale units. Detailed models and simulations of these systems would allow for the optimization of the processes and their scaling, without the need for expensive and time consuming testing. This work is the first step towards providing such a model. In this paper, we present the proposed hydrodynamic model in detail. To illustrate the applicability and level of realism obtained by the model, it is applied to the simulation of biomass particle pyrolysis in a fluidized bed reactor where the focus is mainly on a study of the effects of various operating parameters involved in such a process and the optimization of the process with respect to maximizing the yield of desirable reaction products. The effect of scaling from pilot-plant scale to commercial size will also be discussed.

Brief description of the mathematical model

In the present study, we have derived a comprehensive model for the flow of a multicomponent, dense, reactive, particle mixture (see also Lathouwers and Bellan, 2000). To this end, separate dynamic equations are derived for each particle class describing the evolution of its mean velocity, temperature, etc. These transport equations are coupled through source terms that describe the various non-equilibrium processes, such as mass, momentum and energy transfer, both between particles and the gas, and between the respective particle classes. One of the distinctive characteristics of dense gas-solid flows is that momentum and kinetic energy are primarily exchanged through collisons between particles rather than through the gaseous phase. The particles then interact similarly to the molecules of a dense gas. The present model exploits this analogy through use of the kinetic theory of granular flows. The kinetic theory is based on a Gaussian aproximation for the velocity distribution, assuming the spatial gradients of the mean variables to be small and the particles to be nearly elastic. The model includes thermal balances for the gaseous phase and for all particle classes, which may in general have different temperatures. Both gas-phase and solid phase reactions are allowed as are reactions with phase change.

Application to biomass pyrolysis simulation in fluidized beds

Among the pyrolysis reactor designs investigated for commercial production of condensable tars from biomass, the fluidized bed reactor is potentially efficient due to the high particle heating rates that can be achieved and its excellent mixing properties, assuring a reasonably uniform product quality. A further interesting feature of this type of reactor is that char does not accumulate in the bed, but is rapidly elutriated with the gas flow instead, after which it may be filtered out, making the reactor very suitable for continuous operation.

The fluidized bed reactor contains a large amount of sand that is used as inert bed material which acts as heat capacitor for the biomass that is injected into the bed. Fluidization of the sand-biomass mixture is generally achieved by using either hot steam or nitrogen, some of which is also injected with the biomass, therefore preheating it. Once the particles enter the reactor, the temperature of the biomass rises rapidly and pyrolysis rates increase causing solid biomass species to be converted into tar and gas which are subsequently ejected from the particle, while char which is also formed in the process maintains the particle matrix. Biomass particle pyrolysis is an endothermic process and heat has to be supplied continuously by the surrounding gas, which in turn is heated by the sand. The gaseous reaction products are convected out of the reactor together with the fluidization gas to locations where it is subsequently cooled (quenched) to prevent product degradation.

Although the process has received considerable attention experimentally (Scott and Piskorz, 1982, 1984), currently there are no thorough theoretical analyses available, adressing simultaneously all physico-chemical processes in the reactor.

In the present work, the previously discussed hydroynamic model has been coupled with a realistic pyrolysis chemistry model that is able to differentiate between various biomass feedstock. Results of a series of simulations wil be presented that focus on the determination of the effects of various operating parameters on the efficiency of the pyrolysis process. Scale-up results will also be presented.

Simulated geometry

A sketch of the simulated geometry and the boundary conditions employed is given in Figure 1. The geometry has been chosen to resemble that used in experiments by Scott and Piskorz (1982, 1984), among others. Although the real fluidized bed is a cylindrical vessel, for computational simplicity, the present computational domain is approximated to be rectangular. Some care must therefore be taken in translating/comparing experimental data to simulations. The biomass is fed through an inlet section in one of the side walls, together with an amount of gas, which preheats the biomass during the feeding process. The center of the feedpoint is located 4.6 cm from the bottom of the bed and has a height (area) of 1.86 cm. In the present simulations, the temperature of the gas used for fluidization is equal to that fed through the biomass feed section (T_g). Bagasse, olive husk, maple, and oak have been used as feedstock in the simulations. Furthermore, the fluidization velocity, gas temperature, biomass particle diameter, and scale of the system have been varied, among others. The biomass particles are assumed to have an initial porosity of 0.7 (cf. Miller and Bellan, 1997).

Numerical procedure

Spatial discretization of the governing equations is based on a finite volume technique using a staggered grid. All convective fluxes are approximated with a second-order accurate bounded TVD-scheme avoiding the excessive numerical diffusion associated with the first-order accurate upwind scheme. The time discretization is based on a backward Euler scheme in combination with a pressure-correction technique with a timestep adjustment procedure. The momentum equations of all phases are solved in a coupled manner, though separately for each velocity direction. Compared with the well-known Partial Elimination Algorithm (PEA) the present approach is more general (see Lathouwers, 1999 for more details on full-field coupling and multiphase pressure correction algorithms). The species and energy equations constitute a strongly coupled, stiff system of equations. To avoid very large linear systems arising from (the necessarily) implicit discretization, a time splitting is used (Strang, 1968) for the combined species and energy system consisting of three steps: (i) performance of a half convection-diffusion timestep, (ii) time integration of the equations over a full timestep with only the source terms present (reaction terms, radiation, etc.), (iii) performance of another half convection-diffusion timestep. The advantage of this split scheme is that during steps (i) and (iii), the equations are decoupled into standard convection-diffusion systems which are easily handled, whereas in step (ii) there is no spatial coupling. The stiff integration in step (ii) is performed by using either the well-known stiff integrator VODE (Hindmarsh et al., 1989). All sparse linear systems arising from the discretization of convection-diffusion systems are solved with preconditioned Krylov methods (Conjugate Gradient (CG) for the pressure Poisson equation and Generalized Minimum RESidual (GMRES) for the other transport equations; see e.g. Barrett et al., 1994).

Pyrolysis simulations

Among the pyrolysis reactor designs investigated for commercial production of condensable tars from biomass, the fluidized bed reactor is potentially efficient due to the high particle heating rates that can be achieved (Scott et al., 1999) and its excellent mixing properties, assuring a reasonably uniform product quality. A further interesting feature of this type of reactor is that char does not accumulate in the bed, but is rapidly elutriated with the gas flow instead, after which it may be filtered out, making the reactor very suitable for continuous operation.

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Although the process has received considerable attention experimentally (Scott and Piskorz, 1982, 1984), currently there are no thorough theoretical analyses available, adressing simultaneously all physico-chemical processes in the reactor. Most of the work to date has focused on single-particle pyrolysis in a gas stream which requires *a priori* knowledge of ambient gas flow parameters, its temperature in particular (Di Felice et al., 1999).

Below, we present quantitative results from numerical simulations based on a detailed mathematical model, including a realistic chemistry model that is able to differentiate between various feedstock.

Kinetic model for biomass pyrolysis

The main advantage of the present kinetics scheme is the ability to differentiate between feedstock through knowledge of the initial mass composition with respect to the primary components, cellulose, hemicellulose and lignin. All of the reaction rates in the model, K_j, are considered irreversible, first order Arrhenius

$$K_j = A_j exp(-E_j/R^0T), \tag{1}$$

with A denoting the rate constant, and E the reaction's activation energy, were tabulated by Miller and Bellan, 1997, and are dependent of the source component. Due to a lack of more detailed knowledge, the heats of reaction are taken as independent of the source component. The polymerization reaction K_1 has $\Delta h_1 = 0kJ/kg$, reaction K_2 is endothermic with $\Delta h_2 = 255kJ/kg$, and both the char formation and the secondary tar reactions are exothermic with $\Delta h_2 = -20kJ/kg$ and $\Delta h_2 = -42kJ/kg$.

Simulation details

A sketch of the simulated geometry and the boundary conditions employed is given in Figure 1. The geometry has been chosen to resemble that used in experiments by Scott and Piskorz (1982, 1984), among others. Although the real fluidized bed is a cylindrical vessel, for computational simplicity, the present computational domain is approximated to be rectangular. Some care must therefore be taken in translating/comparing experimental data to simulations. The biomass is fed through an inlet section in one of the side walls, together with an amount of gas, which preheats the biomass during the feeding process. The center of the feedpoint is located 4.6 cm from the bottom of the bed and has a height (area) of 1.86 cm. In the present simulations, the temperature of the gas used for fluidization is equal to that fed through the biomass feed section (T_g). To vary the cellulose/hemicellulose/lignin proportions of the feedstock, bagasse, olive husk, maple, and oak are used in the simulations. The initial biomass composition of these biomass types were given by Miller and Bellan, 1997. The diameter of the sand and biomass particles is assumed 0.5 mm, which is a common value in practical operation. The biomass particles are assumed to have an initial porosity of 0.7 (cf. Miller and Bellan, 1997). The biomass feed flux is ramped from 0 at t=0 to a value of 0.5 kg/m^2s at t=0.5 s, after which it is kept constant (when two feedpoints are used, the flow is equally split between feedpoints). A constant value of 1 kg/m^2s is specified for the gas flux through the feeder. The gas flow used for fluidization of the mixture is varied from 0.3 to 0.4 kg/m^2s and is uniform over the bottom of the domain.

Reactor efficiency parameters

The true measure of the reactor efficiency for the tar production can be quantitatively determined by comparing the actual mass of tar exiting the reactor (Ω_{tar}) and the amount still present in its interior (M_{tar}) to the biomass feed flow (M_{feed}):

$$\Omega_{tar} = \int_0^t \int_{outlet} (\alpha_g \rho_g \mathbf{u}_g Y_{tar}) \cdot \mathbf{dA} dt' \tag{2}$$

$$M_{tar} = \int_{reactor} (\alpha_g \rho_g Y_{tar}) dV \tag{3}$$

$$M_{feed} = \int_0^t \int_{inlet} (\alpha_b \rho_b \mathbf{u}_b) \cdot \mathbf{dA} dt' \tag{4}$$

where the subscript b denotes biomass. With these definitions, two measures for reactor performance are the yield (η) and the differential reactor efficiency (DRE), ϑ, cf. Miller and Bellan (1998):

$$\eta_{tar}(t) = \frac{\Omega_{tar} + M_{tar}}{M_{feed}} \tag{5}$$

$$\vartheta_{tar}(t) = \frac{\Omega_{tar} + M_{tar}}{\sum_\xi (\Omega_\xi + M_\xi)} \tag{6}$$

where the summation is over the species tar, gas and char. These measures quantify the relative efficiency of reactor tar production relative to the biomass feed flow, and the relative proportion of tar produced compared to gas and char. Similar definitions are used for the reactor performance with respect to gas and char production.

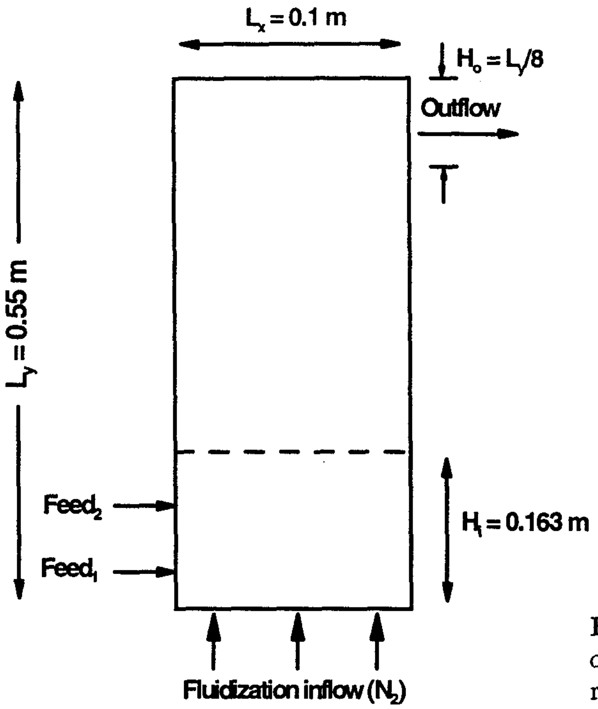
Qualitative behavior

In Figure 2, the y-coordinate of the center of mass of the sand and biomass are plotted as a function of time. It shows the particular transient structure due to the bubbling of the bed (both sand and biomass fluctuate in phase). In general, the biomass is situated higher up in the bed than is the sand. This is caused by two effects: (i) the biomass density is much smaller than that of the sand due to its high porosity, and (ii) the general motion of the biomass is directed upwards right after feeding due to bubbling, while on downward motion the particles have already partially pyrolysed, before reaching the bottom of the bed (an exception being formed by the unreactive char which remains). This particle segregation may be detrimental to tar production, indicating the need to optimize this yield as a functions of reactor and feedstock parameters.

Parametric study

A parametric study has been carried out in order to find the optimum conditions for maximizing tar yield as function of the relevant operating parameters. The parameters investigated include the temperature of the inlet gas, the feed temperature of the biomass, the fluidization velocity (or flux), the location of the feedpoint(s), and the type of feedstock used for pyrolysis.

As an example, figures 3 and 4 show the yield and differential reactor efficiency with respect to tar production, respectively. The strong dpendence of both yield and DRE are noteworthy. Comparing the yield with the DRE, it can



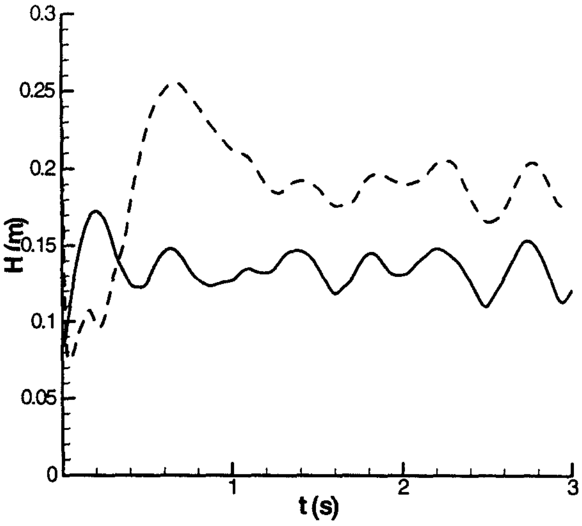


Figure 2: Averaged vertical coordinate of center of mass of sand (———) and biomass (- - - - -) particles in the reactor. Data reflect case no 4.

Figure 1: Schematic of the fluidized bed reactor.

be concluded that the reactor efficiency is a more practical way of examining efficiency than is the yield which is, however, the quantity of ultimate industrial interest. Both figures indicate that the efficiency for tar production reaches a maximum at either 750 or 800 K; an extended simulation time is required to draw a stricter conclusion with respect to the optimum temperature. The abrupt decrease in tar DRE at small times corresponds to the large char DRE (see Fig. 8) resulting from the low biomass temperature. As the biomass temperature increases, so does the tar DRE concomitantly with the reduction in char DRE. The tar DRE is reduced by the large char DRE at small gas temperatures, and by the large gas DRE at high gas temperatures (see Fig. 6). Figures 5 and 6 show similar results for the gas production. The yield indicates a monotonic increase as temperature is increased, however, the gas DRE seems to be minimized for $T_g = 750K$, indicating that this is the most likely optimal temperature for maximizing tar yield. Similarly, for the production of char, Figures 7 and 8 show that the of char production increases with T_g, as pyrolysis is more vigorous. However, the char DRE decreases with increasing T_g, in agreement with the known hindrance of char production at larger reactor temperatures.

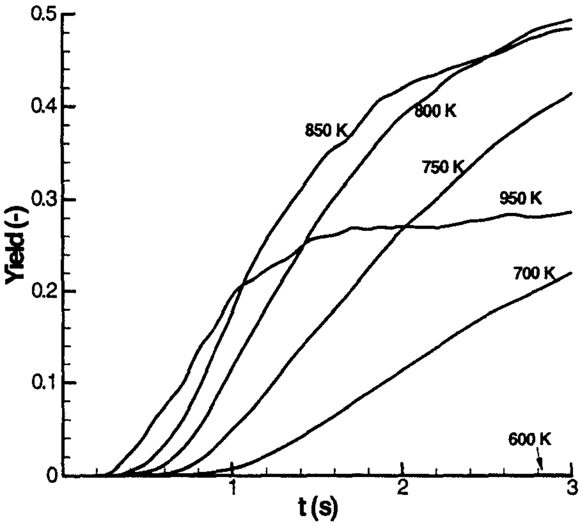


Figure 3: Tar yield as function of time for various gas temperatures.

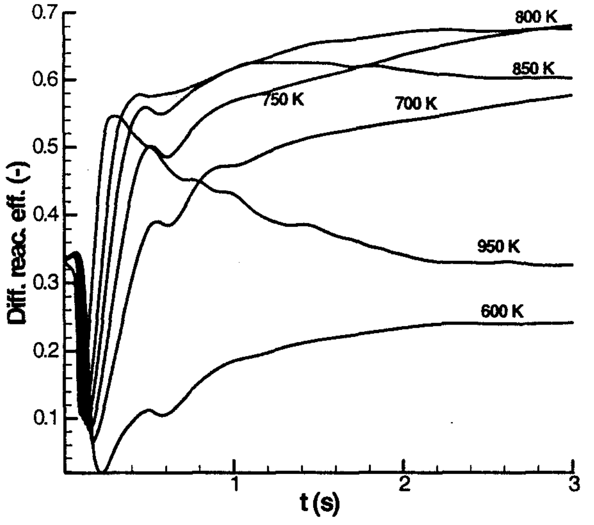


Figure 4: Tar differential reactor efficiency as function of time for various gas temperatures.

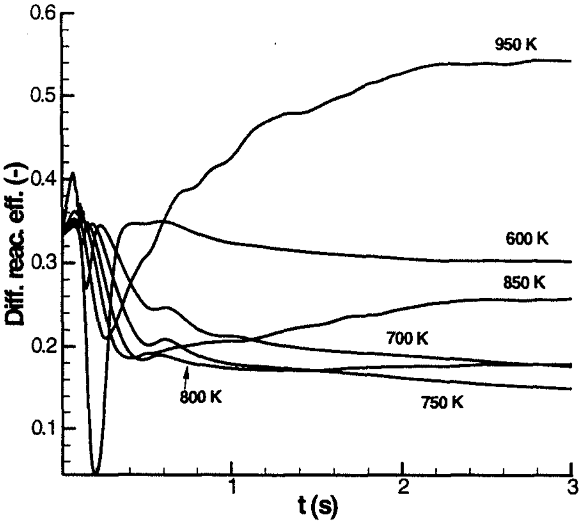


Figure 6: Gas differential reactor efficiency as function of time for various gas temperatures.

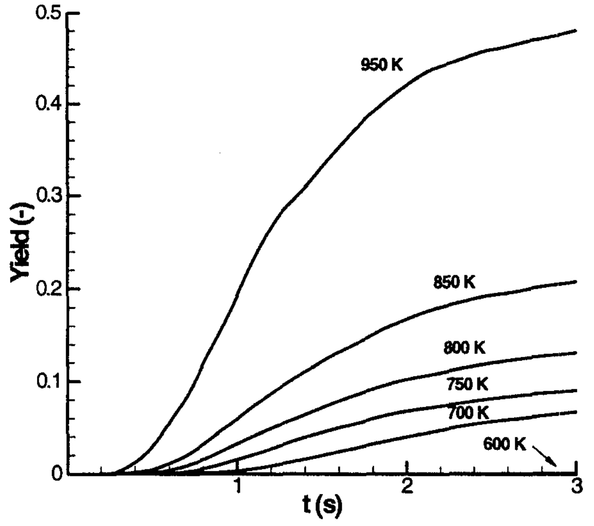


Figure 5: Gas yield as function of time for various gas temperatures.

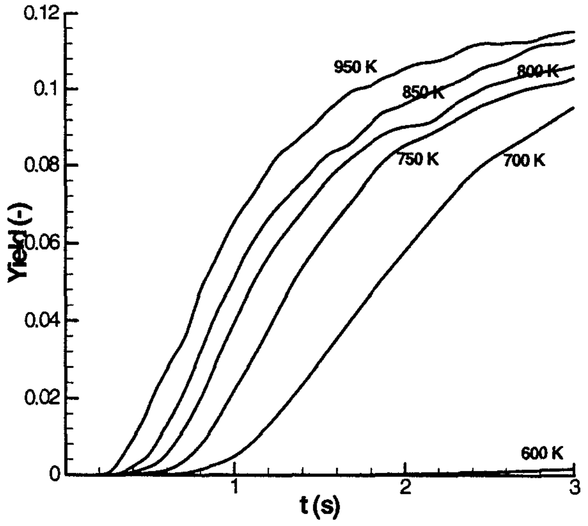


Figure 7: Char yield as function of time for various gas temperatures.

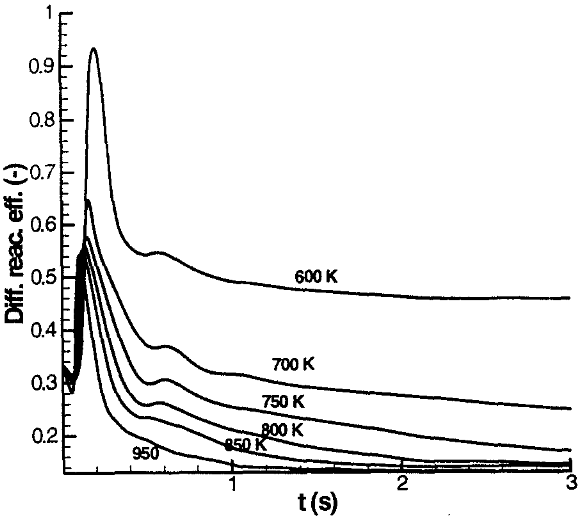


Figure 8: Char differential reactor efficiency as function of time for various gas temperatures.

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