

Chapter 12

Computational Fluid Dynamics Modeling of Biomass Gasification and Pyrolysis

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Biomass thermochemical conversion holds great promise for producing biofuels and will play a determining role in displacing petroleum-based fuel consumption toward renewable sources. Empirical approaches have shown severe limitations in their capability to understand and control the conversion processes. However, without the ability to accurately predict and optimize thermochemical conversion performance, large-scale commercialization of these systems is severely compromised. In this context, Computational Fluid Dynamics (CFD) appears as an essential tool to better comprehend the complex physical and chemical processes involved, paving the way toward efficient control and design strategies. After a brief description of the numerical models needed to simulate biomass gasification and pyrolysis, the contributions of CFD to process design and optimization are detailed. Finally, the state of the art in terms of numerical models for the dense, reactive particulate flows typically found in conversion processes are reviewed. Shortcomings of existing CFD simulations, especially in terms of validation and predictability, are examined; and directions for future research based on the progress of CFD in other fields are suggested.

Introduction

Currently, crude oil is almost exclusively used for producing transportation fuels worldwide. In fact, more than 97% of transportation fuel needs are met with crude oil (1). To reduce our sole dependence on crude oil to meet transportation

needs, and to limit the environmental impact of crude oil usage such as greenhouse gas emissions, many countries and regions are rapidly developing and deploying biofuels and have set some very aggressive goals for near-term deployment. For example, the EU has mandated that biofuels account for 10% of transportation fuel use by 2020 (2). Furthermore, the United States has set both a near-term goal of a 20% reduction in 2007 gasoline usage by 2017, to be met predominantly with increased biofuels production (3), as well as a long term “30x30” goal to displace 30% of the 2004 gasoline demand with biofuels by 2030 (4).

Although many countries are rapidly deploying biofuels, this first wave of development focuses almost exclusively on first-generation biofuels technologies that utilize food- or feed-based feedstocks. Brazil and the United States are rapidly moving forward with developing and deploying ethanol technology, with Brazil using sugarcane as the feedstock and the United States using corn. Brazilian sugar cane ethanol is generally regarded as having little to no impact on primary food supplies and prices, because Brazil has increased its sugar cane production to more than offset the amount of sugar diverted to ethanol production. However, food supply and price concerns have been raised about corn ethanol production in the United States (5), because corn grain is an important food and animal feed commodity. The EU, the largest biodiesel producer, uses rapeseed oil as its main feedstock and again concerns about fats and oils supplies and prices have been raised over the diversion of rapeseed oil to biodiesel production.

Because of these concerns and the overall limitations of first-generation biofuels technology primarily due to feedstock restrictions, advanced or second-generation biofuels technologies, based on sustainable, non-food sources of feedstocks, will be required to meet aggressive volume goals for biofuels deployment (6). Several different technologies exist (6–8) for converting cellulosic biomass to biofuels. The predominant differentiation between the conversion options is the primary catalysis system (9). Biochemical conversion routes rely on biocatalysts, such as enzymes and microbial cells, in addition to heat and chemicals, to convert biomass first to an intermediate mixed sugar stream and then to ethanol or other fermentation-produced biofuel. Conversely, thermochemical conversion technologies rely on heat and/or physical catalysts to convert biomass to an intermediate gas or liquid, followed by an additional conversion step to transform that intermediate to a biofuel. Thermochemical conversion processes will play a determining role in the development and deployment of second generation biofuels, because they offer significant advantages, such as the ability to robustly handle a wide range of feedstocks, and they are capable of producing various types of transportation fuels. Biofuels production via thermochemical approaches shows great promise for being economically competitive with conventional petroleum derived gasoline and diesel (4) in both the near and long term. Additionally, the economics of biofuels production via thermochemical approaches compare favorably with the economics of other biomass-to-biofuels conversion routes, such as biochemical approaches (10).

Thermochemical conversion technologies for producing transportation fuels can be categorized as either gasification or pyrolysis (11). Gasification is a complete depolymerization of biomass with limited oxygen at high temperatures,

typically $> 850^{\circ}\text{C}$, to a gaseous intermediate synthesis gas (syngas) consisting of H_2 and CO . A review of the existing types of gasifiers and their relative advantages and disadvantages for transportation fuel production is provided by Spath and Dayton (12). Pyrolysis, on the other hand, is the milder depolymerization of biomass producing a liquid intermediate (pyrolysis oil or “bio-oil”) in the absence of added oxygen at lower temperatures, typically in the range of 400°C to 650°C . Detailed reviews of pyrolysis techniques and their current technical status are provided by Bridgwater and Peacocke (12) and Czernik and Bridgwater (13). Although there are a number of gasification and pyrolysis processes under development (14), fluidized bed processes are attractive for converting biomass (15) because they are easily scalable, very robust, and do not require significant size reduction of the feedstock, which can be problematic for biomass.

Current State of Development and Research Needs

Driven by a worldwide desire to develop second-generation biofuels and the high potential offered by thermochemical conversion technologies, considerable progress has been achieved for both gasification and pyrolysis routes for biofuels production (16). Despite these advances, some technical challenges still need to be solved to enable large-scale industrialization of these processes (17).

One of the big challenges associated with either fluidized-bed gasification or pyrolysis is the high variability in reactor performances, noticeably increasing the risks associated with the development of industrial-scale facilities. For example, Figure 1 shows the hydrogen-to-carbon monoxide ratio as a function of the operating temperature reported in the literature for several existing biomass gasification fluidized-bed reactors. Hydrogen-to-carbon monoxide ratio is a critical output parameter for liquid fuel synthesis, and still, variations of nearly an order of magnitude are observed, which cannot be explained by the current state of understanding of these systems.

Another challenge is the issue of undesirable tar production. In this context, the term “tar” refers to the complex mixture of organic compounds that are produced by either biomass gasification or pyrolysis. In fuel synthesis operations, tars are especially problematic, because if they are not fully reacted to product gases or removed, they can condense and rapidly foul downstream equipment, significantly decreasing the overall efficiency of the conversion process. The ultimate nature of the tar produced from gasification or pyrolysis is a combination of primary tar formation and secondary tar reactions that alter both the total amount and composition of tars produced (17). Although a considerable amount of work has focused on addressing the tar production problem, the overwhelming majority of this previous work tends to fall in two distinct categories: the use of catalysts to prevent or reduce tar formation in the fluidized bed (24, 25), or the use of downstream tar-reforming catalysts to reform the tars into additional syngas (26, 27). However, a fundamental understanding of the chemical and physical processes responsible for tar formation is essential to develop efficient and viable control strategies.

Computational fluid dynamics (CFD) has been recognized as a powerful design and development tool in many industrial areas. For example, spectacular

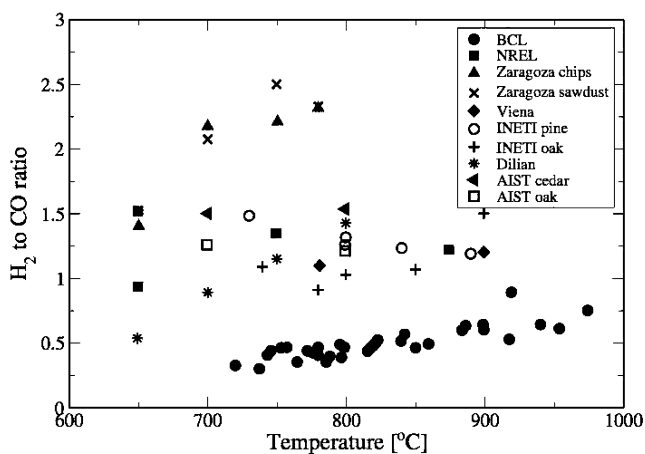


Figure 1. Reported hydrogen-to-carbon monoxide ratios from existing fluidized-bed gasifiers. (sources: (18–23))

progress has been made in aircraft design that enabled a deep understanding of crucial processes such as laminar to turbulent transitions, and fluid/structure interactions for external flows, or turbulence/chemistry interactions, flame stability and pollutant formation for engines, leading to the computational exploration of novel, more efficient designs (28–30). These advances in numerical techniques, applied to biomass thermochemical conversion systems, provide a unique opportunity to improve CFD predictive capabilities and move beyond the strictly empirical strategies that have shown severe limitations in terms of cost, flexibility, reactor scale-up, and optimization of reactor design and operating conditions.

This chapter will focus on the modeling of the fluidized-bed reactor itself, disregarding the subsequent tar-reforming and fuel-synthesis processes. Performance during this first stage is crucial to the overall process, as it determines the extent and cost of the treatments required to clean and condition the synthesis gas prior to power generation or fuel synthesis. The fluidized-bed reactor combines most of the possible phenomena involved in thermal conversion, including hydrodynamics, chemical processes and heat release, in a single unit operation. Therefore, progress in this area will help develop comprehensive models that encompass the additional units necessary for the gasification or pyrolysis of biomass. For instance, in the case of indirect gasification, modeling heat production through char combustion in an annex reactor can directly take advantage of the multiphase reactive models developed for the gasifier. Accurate, validated modeling of biomass behavior in fluidized-bed reactors is the crucial next step for advancing computational modeling in biomass gasification and pyrolysis.

The sections in this review include a description of biomass gasification processes and existing numerical approaches that may be used to model them, the role of CFD in reactor and process design and optimization, and the steps necessary to translate descriptive CFD into predictive models.

Biomass Thermochemical Conversion Processes and Modeling

Physical and Chemical Characteristics of Biomass Gasification

The first step of biomass gasification occurs when pelletized biomass enters a fluidized-bed reactor (Figure 2). A fluidized-bed reactor consists of a bed of inert material such as olivine sand, which is fluidized by injecting a gaseous medium from the bottom. The bubbling or turbulent motion of the bed ensures good mixing properties and uniform heat distribution. Superheated steam is usually used to create fluidization when gasification is desired, as it actively contributes to conversion to syngas. Biomass is injected into the reactor, either at the bottom or top of the bed, and is quickly heated by the hot flowing gas and through collisions with the hot sand.

The first stages of gasification are similar to those of pyrolysis. First, water contained in the biomass evaporates. Then, volatiles are released, producing permanent gases and primary tar, leaving behind fixed carbon in the form of char. Primary products undergo further decomposition both inside the biomass particle and in the gas phase. The composition of the gas released from the biomass as well as the amount of carbon left in the char is highly dependent on both the intra-particle thermal and chemical processes and the coupling with the external flow. Gas flow in the reactor and particle collisions provide heat and carry away devolatilization products, driving the biomass conversion to syngas. To emphasize the coupling between all these processes, Figure 3 shows the structural changes occurring in the plant cell walls when subjected to intense external heat as encountered in fluidized beds. While the heat-exposed side (left) suffers extensive deformation, the interior walls (right) remain nearly intact. Clearly, heat and mass transfer inside the particle plays a crucial role in biomass conversion, and need to be taken carefully into account in CFD to correctly describe gasification and pyrolysis.

Some phenomena, similar to those briefly outlined above, can be found in other engineering applications in which CFD might be at a more advanced stage and, therefore, have been already studied extensively. Others are specific to biomass conversion, and the available work mostly concentrates on local, detailed modeling that has been rarely ported into CFD codes. Therefore, as illustrated in Figure 4, capturing the complex dynamics of the multiphase reacting flows encountered in a fluidized-bed gasifier requires combining a significant number of models from various backgrounds and levels of accuracy and identifying and handling potential interactions that may exist between these models. Next, we give a short description of each of the processes and existing models that can be applied in the context of fluidized beds.

Hydrodynamics of Particle-Laden Flows

Solid Phase Description

Gas-solid fluidized beds, characterized by their excellent mixing and heat transfer properties, are used in a wide variety of chemical and engineering industrial processes (31). The overall dynamic of these systems is dominated by

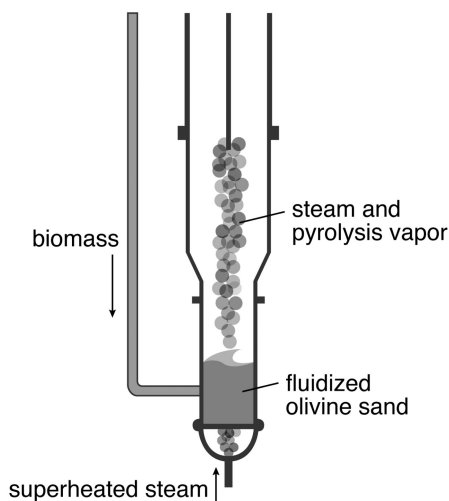


Figure 2. Schematic of a fluidized bed reactor

large-scale structures such as bubbles and recirculation regions along the walls, which, in turn, are controlled by interactions at the particle scale. Although robust and accurate numerical methods have been developed over the years for single-phase flows involving a gas or a liquid, such methods are still in the development stage for multiphase flows or flows involving a particulate phase. In a recent review of existing numerical models for gas-solid fluidized beds, Van der Hoef et al. (32) identified five major classes of methods based on the strategy, either Eulerian or Lagrangian, used to describe the gas phase and the solid phase, respectively. Among them, the two-fluid model (TFM) and the unresolved discrete particle model (DPM) have been the topics of a very large number of studies. Both approaches usually make the assumption of constant size, spherical particles.

TFM (33) follows a Euler-Euler approach that considers both phases as continuous interpenetrating media described using Navier-Stokes-type conservation equations. However, although easily implemented in pre-existing CFD codes and relatively computationally inexpensive, this method requires all processes at the particle scale, such as drag, collision and friction forces, and heterogeneous chemistry to be included as phase-interaction terms into the governing equations. Closures are obtained using the kinetic theory of granular flows (34) or experimental correlation, for example for the fluid-solid drag force (33, 35).

Two-fluid methods can be extended for polydisperse, particle-laden flows involving particle size distributions and evolving particle sizes by using the more general and rigorous framework of the methods of moments. Equations are derived for a particle number density and its moments, conditioned on size, density or velocity (36). Although mostly applied to dilute solid-phase systems such as sprays (37), recent developments focused on the dense granular flows found in fluidized beds (38).

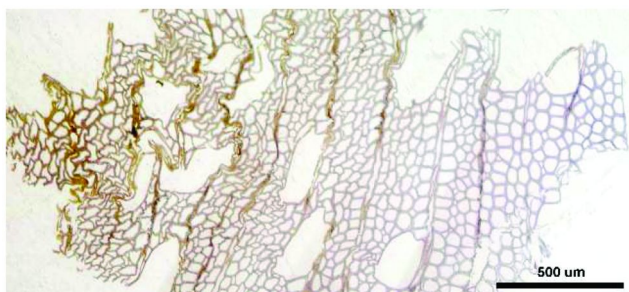


Figure 3. Structural changes in a heated biomass particle. (courtesy of Drs. T. Haas and B. Donohoe, National Renewable Energy Laboratory, Golden, CO)

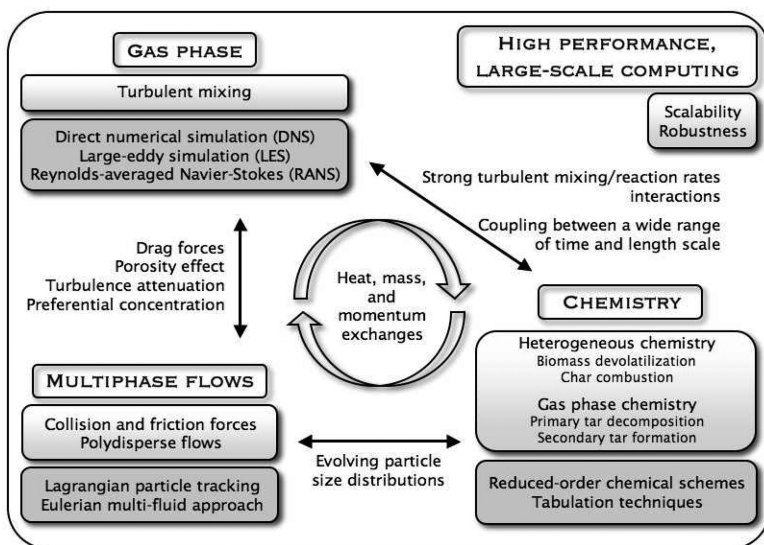


Figure 4. Modeling of physical and chemical processes interactions in biomass thermochemical conversion.

DPM methods, on the other hand, use Lagrangian particle tracking to describe the solid phase, in which each particle is transported following Newton's laws of motion (Figure 5). Particles are typically very small compared to the gas flow computational cells. Therefore, details of the flow around the particle are not resolved, and drag forces need to be modeled in a similar way as in TFM. However, collisions between particles are considered explicitly using, for example, hard-sphere (39) or soft-sphere (40, 41) collision models. Porosity is taken into account by introducing the void fraction in the gas-phase governing equations. Due to the very large number of particles involved in fluidized beds, DPM approaches may become prohibitively expensive and are applied most often in two-dimensional configurations.

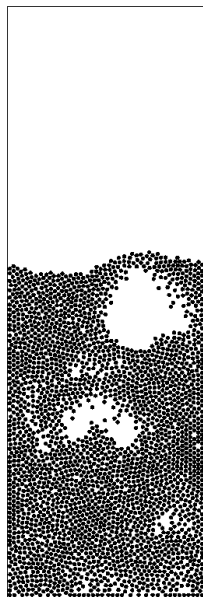


Figure 5. DPM simulation of bubble formation in a fluidized bed.

Gas Phase Coupling

In most practical applications, the gas flow is fully turbulent and must be treated adequately to obtain meaningful results. Mature techniques developed for aeronautics or internal engine flows may be extended to chemical reactors. Depending on the required resolution of the solution and the computational resources, Reynolds average equations (42) (RANS) or Large Eddy Simulation (43) (LES) may be used. The former solves for the mean quantities of the flow while modeling the fluctuations; whereas the latter can be seen as a filter operation, resolving the large, energy-containing scales and modeling the smallest scales. RANS methods are computationally affordable, as they usually require only a few additional evolution equations, compared to laminar configurations, for the kinetic energy and eddy dissipation rate. LES simulations are more expensive because they demand a higher grid resolution, but provide a much more accurate description of the unsteady, large-scale processes.

The interactions between gas and particles can have a significant impact on the turbulence intensity of the flow through, and increase the apparent gas viscosity (44). Precisely modeling the gas-particle couplings is important to accurately capture the mixing properties of the bed, which are expected to have a predominant impact on the overall reactor performances. A detailed review of the numerical aspects associated with gas-solid coupling in particle-laden flows is given in Curtis *et al.* (45).

To simulate biomass conversion, a description of the complex chemical processes needs to be combined with the above models for the hydrodynamics of fluidized-bed reactors. The chemistry of biomass conversion has been the topic of many experimental and modeling studies reviewed by Di Blasi (46) among others. Most studies focus on the heterogeneous reactions associated with biomass devolatilization. They often rely on kinetically controlled experiments providing weight mass loss as a function of time or temperature (47–50). The resulting data are used to derive kinetic rates for global schemes involving only a few representative compounds and reactive steps. Kinetic control, however, is not representative of the conditions found in realistic reactors and might even be difficult to ensure in the experiments (46). Instead, heat transfer to and inside the reactive particle by conduction, convection, and radiation is often the limiting process. However, the details of heat transfer inside the reactor are much more difficult to quantify experimentally. To understand heat transfer better, kinetic data can be coupled to one-dimensional partial differential equations describing the temperature evolution inside the particle. Most of these simulations are done on one single particle in an infinite domain, and the results largely depend on the kind of assumptions introduced (51–54). When more realistic configurations are considered, heat transfer is often modeled using correlations based on a few characteristic dimensionless numbers such as the Nusselt (ratio of conductive to convective heat transfer) and Sherwood (ratio of convective to diffusive mass transfer) numbers (55, 56). Although very convenient to use, these models have been shown to be inadequate, especially for dense granular flows (57).

Comparatively few systematic studies into the nature or kinetics of gas-phase tar formation from biomass devolatilization products have been conducted. Taralas and Kontiominas (58) looked at secondary pyrolysis of vaporized unsaturated hydrocarbons in the presence of water vapor and oxygen using toluene and benzene as model compounds. Morf *et al.* (59) performed a mechanistic study of primary and secondary tar reactions and concluded that secondary tar reactions become important at temperatures higher than 650°C. These studies have just begun to address the important issue of tar formation, and more detailed work needs to be performed in this important area.

CFD Contribution to Process Design and Optimization

CFD appears as a cost-effective option to explore various configurations and operating conditions directly at the industrial scale to find the optimal configuration depending on the project specifications. However, CFD results are only valid if appropriate chemical and physical models are developed and validated using lab-scale experiments and robust numerical methods are used. In the case of indirect gasification, the optimal configuration corresponds to minimum tar content in the syngas; whereas in the case of pyrolysis, the tar output needs to be maximized. This translates into different operating temperatures and gas residence time inside the reactor. However, it must be emphasized

that biomass pyrolysis and gasification in a fluidized-bed involve the same small-scale phenomena. Therefore, both types of conversion can be studied simultaneously and effectively using CFD, providing a versatility that cannot be found in experimental approaches. The advantages of using CFD in conjunction with well-designed validation experiments are given below, namely a global understanding of the large-scale impact of local phenomena, the identification of the most sensitive parameters, and guidelines for the industrial scale-up of new reactors.

Numerical Experiments

As mentioned earlier, a current limitation of biomass gasification in a fluidized-bed reactor is the high tar level in the product gas, causing fouling and hazardous waste. Costly cleaning treatments to control the tar content of the exiting gas are usually applied downstream. An appealing approach would be to directly optimize the gasifier design and operating conditions to the biomass properties for minimal tar production. Tar formation involves a lot of different steps occurring at different time and length scales. Primary tars are formed through biomass devolatilization inside the bed, which is highly sensitive to the local heat transfer between the sand and biomass particles and gas flow around the particles. Primary tar molecules are then transported across the reactor, where their decomposition and further reactions to form secondary tars depend on the temperature profile, residence time in the gas phase, and turbulence impact on reaction rates. Developing efficient control strategies requires understanding in detail each of these steps and how they are coupled with the larger scale flow phenomena happening in the reactor. However, the types of measurement that can be done on a full-scale reactor are considerably restricted, because visual access inside a three-dimensional fluidized-bed reactor is very limited, and the dense gas-solid mixture is difficult to sample without disruption. In most cases, only global time-averaged data are available, such as exit gas composition, flow rate, and temperature. To access internal details, some small-scale processes have been extensively studied in well-controlled environments. They include biomass devolatilization, non-reactive particle collisions, and gas-phase chemical reactions. CFD offers a unique opportunity to incorporate these fundamental results into a larger scale framework, in which process interactions can be studied.

Parameterization of the simulated configurations enables sensitivity analysis studies, normally an expensive and highly time-consuming process when performed experimentally. Sensitivity analysis should be conducted with two objectives in mind: identifying the limiting steps, and providing guidelines for further detailed modeling, as significant progress will be achieved if the most important and sensitive processes are correctly and accurately modeled first. Identifying the limiting processes and focusing on developing accurate models for them are crucial steps toward optimizing biomass gasification, as they will determine the exit gas composition and the amount of char left behind.

The CFD capabilities outlined above require that every component of the whole CFD model be extensively validated, which will be detailed in a later section.

Reactor Scale-Up

CFD models based on first principles should be able to predict accurately the gas-solid hydrodynamics at all scales, provided that a thorough validation of the code has been performed. A fundamental challenge faced when trying to develop new industrial-scale reactors based on laboratory results is the scale-up problem. Scaling laws based on conserving non-dimensionless numbers from one scale to another are often too simplistic or overly constraining to be reliable. The limited success of the scaling approach comes from the fact that the hydrodynamic of fluidized-bed reactors is not scale-similar because they involve scale-related phenomena, e.g., bubble dynamics and wall effects, which promote better mixing in small reactors (60). Instead, scale-up is usually done using one-dimensional models that incorporate simple descriptions of the mixing processes and heat transfer along the reactor. However, a pilot scale is often required between the lab and full-scale reactors, as these simplified models have little predictability (60). Minimizing or skipping pilot-plant validation of scale up through strategic use of CFD models would represent both capital cost and time-to-market savings. Also, the risk associated with developing a new industrial facility would be significantly reduced, making it more attractive to investors.

Several initial attempts have been made to use CFD to help with scale up. Lathouwers and Bellan (61) numerically studied the impact of reactor scale-up for biomass pyrolysis in a fluidized bed. Although no experimental validation was presented, they showed that increasing the bed size negatively impacted tar production and that shallow fluidized beds with high fluidization velocity had better scalability characteristics. Following a different approach, Van Ommen *et al.* (62) investigated the performances of different sets of scaling rules based on dimensionless numbers using CFD. They reported large variations in the void fraction and pressure data and noted that none of the scaling laws tested led to complete similarity between the two reactor sizes considered. No experimental validation was provided to corroborate the results. Even without experimental validation, these models suggest an interesting direction for pilot-scale work. In their review about fluidized-bed scale-up, Knowlton *et al.* (60) recognized that CFD is a promising approach, but given the state-of-the-art in numerical methods, experimental work is still required for successful scale up. With the right combination of computational advances and experimental validation, this may not always be the case. Limitations to that vision will be highlighted in the next section.

From Descriptive to Predictive CFD

The potential of CFD techniques is now widely acknowledged. However, the models are not advanced enough yet to be considered as a reliable and useful engineering tool for biomass gasification or pyrolysis. Two reasons for this are the lack of predictability and limited computational resources. In the following section, we give a brief overview of the different attempts to develop comprehensive CFD gasification and pyrolysis models, highlighting the strong

points and shortcomings of each method. Then, the obstacles that need to be overcome to get truly predictive results will be discussed.

Existing Models

Comprehensive CFD simulations of biomass gasification or pyrolysis are scarce. Although the challenges faced in coal conversion are slightly different from those encountered in biomass conversion, the numerical frameworks remain close, and tools developed for one type of fuel are expected to be applicable for the other with only minor changes of the numerical methods required. Therefore, advances in both domains will be reported here.

Simplified Models

The pressing need for predictive tools that are more sophisticated than engineering correlations to optimize or scale up existing technologies has led to the development of numerous simplified models that incorporate most physical and chemical phenomena into a streamlined system of one-dimensional differential equations. These equations represent the evolution of the biomass particles as they move along the reactor. For example, Gobel *et al.* (63) developed a mathematical model for a fixed-bed coal gasifier that included conservation of mass and energy and reaction kinetics in the gas phase and char based on chemical equilibrium and Langmuir–Hinshelwood correlations. Individual components of the model were either based on first principles or determined from thermo-gravimetric experiments. The approach was validated and implemented in a real plant. A slightly more detailed approach was followed by Radmanesh *et al.* (64) to model a bubbling fluidized-bed reactor under isothermal conditions. Three stages were simulated successively. The various product yields of biomass pyrolysis, including a tar pseudo-component, served as initial conditions in the one-dimensional simulation of the bed using a countercurrent back-mixing model describing the evolution of the bubble (gas) and emulsion (mixture of gas and solids) phases. This was further combined with a gas-phase chemistry model describing the tar conversion to permanent gases and refractory tar. Maistrenko *et al.* (65) developed a one-dimensional unsteady model for polydisperse combustion of coal in a fluidized bed with heterogeneous chemistry involving only a few permanent gas species.

The conservation equations along the reactor axis may be coupled to a one-dimensional description of the evolution of the reactive particle, usually assumed to be spherically symmetric. An example of such pseudo two-dimensional differential systems is found in Luo *et al.* (66), who studied wood-flash pyrolysis in a fluidized-bed reactor. All variables were assumed to be homogeneous in the bed radial and azimuthal directions. Very recently, Pierucci *et al.* (54) combined the semi-detailed chemistry model for biomass pyrolysis and gas-phase reactions of Ranzi *et al.* (67) with a one-dimensional model for moving bed gasifiers using a multi-zone description of the biomass particles. Validation

was performed using gas composition at the exit of a lab-scale reactor, and results showed acceptable agreement between simulated and experimental data.

Although some of the models above have been used successfully in the context of real industrial-scale processes, clearly the assumptions and simplifications made in their development prevent them from being extended to different conditions and configurations, and they provide only limited insight on the details of the small-scale physical phenomena responsible for some major behavior such as the amount of tar in the exit gas. Therefore, a more detailed consideration of those phenomena is essential.

CFD in Realistic Configurations

The next step toward predictive CFD tools in various configurations is to rely on first principles and directly solve the coupled conservation equations for all variables. The increased resolution comes with an increase in computational cost, and most studies can afford only two-dimensional domains, even for cold-flow configurations in which no energy or species conservation equation is solved. Numerical simulations of cold fluidized beds are numerous. A two-fluid approach is employed in the recent papers on segregation in poly-disperse fluidized beds by Gera *et al.* (68), Huilin *et al.* (69), and Fan and Fox (38), or in the work of van Wachem *et al.* (70), Patil *et al.* (71, 72), or Papadikis *et al.* (73) on closure formulation and validation. Others are based on a discrete particle approach (e.g., see Xu and Yu (74), Patankar *et al.* (44), Goldschmidt *et al.* (75) for the soft-sphere model; von Wachem *et al.* (76) for the hard-sphere model; Snider (77) for a volumetric approach to particle collisions; or the review paper of Deen *et al.* (78)). However, very few studies have tackled the coupled gas-solids hydrodynamic/reactive particle problem characteristic of biomass or coal thermochemical conversion systems. The following description of some of the most significant advances in this field gives a sense of the challenges still faced before CFD can become a predictable and reliable tool.

Fletcher *et al.* (55, 79) performed a three-dimensional simulation of coal combustion in an entrained flow biomass gasifier using the CFX package (80). Although the entrained flow reactor only involves a dilute particle-laden flow where collisions have been neglected, the simulation includes most of the important processes found in biomass gasification in fluidized beds and is worth mentioning here. Biomass particles are tracked down the reactor using a Lagrangian approach. A particle-size distribution is prescribed, and the corresponding initial particle diameters are assumed to remain constant during the simulation. Chemistry is included using a global kinetic model involving only a few species: CH₄, H₂, CO, CO₂, H₂O, N₂, and O₂, but considers devolatilization, heterogeneous char conversion, and gas-phase chemistry. Gas-phase turbulence is modeled using a RANS approach, and the effect of turbulence on both the particles and the chemical reactions is taken into account. Very limited validation was carried out, with only the qualitative exit gas composition compared with experimental data.

Lathouwers and Bellan (61, 81) proposed a comprehensive mathematical model to describe the dynamics of dense, reactive gas-solid mixtures and applied it to the simulation of biomass pyrolysis in a fluidized bed. Ensemble average equations are derived for each of the gas and various solids phases and appropriate closure models are formulated. Because the equations are derived using general moment methods, they do not require certain properties that are assumed in other two-fluid models such as equi-partition of granular energy among the particle classes. Chemistry is included in the form of the kinetic model for biomass pyrolysis developed by Miller and Bellan (51). The biomass particles are assumed to have a constant diameter throughout the simulation, while the porosity of the solids phase increases. No comparison with experimental data is presented; however, the qualitative response of the reactor in terms of tar yield as a function of parameters such as temperature is recovered. Parametric simulations are performed and demonstrate the ability of CFD models to identify optimal reactor operating conditions and assist in the scale-up process.

Zhou et al. (56, 82) coupled a soft-sphere discrete particle method with LES to describe coal combustion in a bubbling fluidized-bed reactor. The effect of particles on sub-grid scale gas flow and the turbulent gas-particle interaction force were taken into account. The kinetic model for char combustion is based on a distributed activation energy approach function of the heating rate. NO_x formation being a major concern for combustion systems, nitrogen species are considered along with CO , CO_2 , H_2O , and O_2 . The burning char evolves following a shrinking-core approach, in which particle density remains constant while their diameter decreases as combustion proceeds. Particles are assumed to be isothermal. Parametric simulations were conducted to study the heating and subsequent combustion of the initially cold coal particles introduced in the hot sand bed. Excess temperatures of the burning particles were found to be in agreement with values reported in the literature. Also, the simulations showed that the presence of large reactive particles significantly affected the particle flow structure and that momentum and energy were mainly exchanged through collisions between particles rather than through the gas phase.

More recently, Yu et al. (83) developed a two-fluid model based on the kinetic theory of granular flow and coupled it with a multi-step chemistry model to study coal gasification in a two-dimensional bubbling fluidized-bed gasifier. Turbulence is captured using a k - ϵ RANS model, and the competition between kinetics and turbulent mixing is considered. The coal particles are assumed to be monodisperse spheres of constant density. Evolution of the major permanent gas species are included, as well as C_2H_6 , C_6H_6 , and H_2S . Calculated mole fractions of the major species in the exit gas were shown to be in good agreement with experimental data for different running conditions. A comparative summary of these studies is given in Table 1.

Table 1. Comparison of the models used in comprehensive CFD studies of biomass or coal conversion devices

<i>Authors</i>	<i>Lathouwers et al.</i>	<i>Zhou et al.</i>	<i>Yu et al.</i>	<i>Fletcher et al.</i>
<i>Reference</i>	(61, 81)	(56, 82)	(83)	(55, 79)
Fuel	Biomass	Coal	Coal	Biomass
Application	Pyrolysis in fluidized-bed	Combustion in fluidized-bed	Gasification in fluidized-bed	Gasification in downdraft gasifier
Dimensions	2	2	2	3
Multiphase	Eulerian	Lagrangian	Eulerian	Lagrangian
Turbulence	RANS	LES	RANS	RANS
Chemistry	Global, 3-component wood, tar, char, gas	Multi-step, NO _x species	Multi-step, CO, CO ₂ , O ₂ , H ₂ O, H ₂ , CH ₄ , char	Multi-step, CO, CO ₂ , O ₂ , H ₂ O, H ₂ , CH ₄ , char
Validation with experiments	None	None	Major species composition of exit gas	Very limited (exit gas composition)

An attempt to integrate a more comprehensive description of the complex chemistry taking place in the gas phase into a CFD code is given in Gerun *et al.* (84). They studied the impact of tar formation on the temperature and velocity patterns in the oxidation zone of a two-stage downdraft gasifier using a semi-detailed mechanism for a tar model compound, phenol, in a two-dimensional axisymmetric domain. No solids were considered, and turbulence was modeled using a RANS approach coupled with an eddy dissipation model. A very partial comparison with experiments was done for the average gas temperature profile inside the reactor and tar concentration after the oxidation zone.

Model Verification and Validation

One of the major obstacles preventing a wider use of CFD tools at both the research and industrial level is the lack of thorough verification and validation of the existing models. Verification involves confirming the correct implementation of the model from a numerical point of view, while validation aims to assess the ability of the model to represent the actual physical process considered (85). Grace and Taghipour (86) provide a critical analysis of the current standards for fluidized-bed CFD model validation, highlighting the fact that virtually none of the existing models, although claimed to be validated with experimental data, have enough credibility to be applied beyond the model development stage. The validation process usually follows a hierarchical approach, with the building blocks of the CFD model being first tested in simple configurations involving a limited range of physical or chemical phenomena, or for which theory can provide

analytical results. Examples of these simpler test cases include transient interface levels for batch sedimentation of particles (87), Reynolds number at minimum fluidization conditions to validate the drag force model (88), or hopper discharge rate to evaluate the particle friction model (89).

On a more global scale, most experimental data on the hydrodynamics of fluidized beds are obtained in pseudo-two-dimensional gas-fluidized beds (41, 75, 76, 90, 91). It must be noted that because of the chaotic nature of fluidized-bed reactors, only flow statistics are meaningful enough to compare between simulations and experiments. Images from these experiments are either used for visual qualitative comparison or post-treated to extract quantitative data such as average void fraction or porosity across the bed, bed height, and bubble average diameter and velocity. Additional measurements can be done for bed expansion (92, 93) or pressure fluctuations (70). Fluidization and segregation has been studied extensively for binary mixtures (94) and continuous particle size distributions (95). Measures have been developed to quantify the extent of segregation in a fluidized bed (93). To measure the gas turbulence in the freeboard of a fluidized bed being induced by bubble bursting at the surface of the bed, Solimene *et al.* (96) recently developed laser diagnostics to study the evolution of vortices generated by a single bubble bursting at the surface of the bed. These experiments provide quantitative measurements of vortex displacement and concentration of a tracer species. As illustrated above, the hydrodynamics of a fluidized bed are experimentally characterized only on a global scale, often for simplified, two-dimensional systems. Gas and particle velocity fluctuations throughout the bed or three-dimensional measurements of the bubble dynamics are not available, nor are systematic studies over ranges of fluidization conditions, for non-spherical particles or evolving particle size and density distributions, both characteristic of biomass systems.

The situation is even more critical for reactive systems, for which very little detailed experimental data is available. Average exit gas flow rate and composition can be easily measured and compared to CFD results (83). Radmanesh *et al.* (64) measured the evolution of the gas composition along a fluidized-bed reactor during biomass gasification for various conditions in terms of bed temperature and equivalence ratio. Only total dry gas yield and major permanent gas species were provided. Van Paasen *et al.* (97) provide comprehensive tar measurements from biomass gasification in fixed and fluidized beds. However, gas samples were not taken inside the reactor, but after a cyclone, which might not be directly comparable with CFD simulations. Numerous studies of wood devolatilization have been conducted to measure devolatilization time and char yield for various feedstock, particle sizes and shapes in fluidized beds, by Sreekanth *et al.* (98), Di Blasi and Branca (99), Wang *et al.* (100) and de Diego *et al.* (101) for large wood cylinders or cuboids. Using one single model for devolatilization time, Sreekanth *et al.* (98) showed that those measurements were consistent with each other. Jand *et al.* (102) performed devolatilization of a finite number of smaller wood particles in a fluidized bed. Conditions were designed so that the devolatilization chemistry occurred inside the particle, therefore limiting the extra-particle and gas-phase conversion of volatiles to permanent gases and secondary tar. Char

yield measurements, permanent gas compositions, and amount of tar in the exit gas were reported.

However, even if models describing multiple particles should remain an extension over single-particle models, the one-at-a-time particle feeding approach used in most of these experiments causes significant problems for CFD, preventing the corresponding experimental data from being used for rigorous model validation. On one hand, Eulerian techniques are not designed to handle a finite number of particles, as the average solid fraction on which the conservation equations are based becomes ill defined in the limit of very few particles. On the other hand, most Lagrangian methods assume a small particle diameter compared to the gas-phase computational grid. This assumption breaks down when large particles are gasified in moderate size fluidized beds. In that case, two-way coupling between gas and particles needs to be revised (103). Moreover, single particles injected on top of a bed can remain on top, sink to the bottom of the bed or be caught in a recirculation region. These different trajectories involve different types of physical processes and different heat transfer modes. Therefore, it is very unlikely that Euler-Lagrange CFD simulation of a single-particle gasification matches what happened in the experiments. A statistical treatment needs to be adopted, such as Monte-Carlo simulations, where the same process is simulated a large number of time from slightly different initial conditions (104). Also, very little is known about the evolution of biomass particle size, density, porosity and composition during gasification, even if those variables are expected to greatly impact both the hydrodynamics of the bed and products release as a function of time.

Another important aspect of CFD validation using reactive bed experimental data, such as exit gas composition, is the fact that these data are the results of a large number of different processes that cannot be distinguished from one another. Therefore, a good agreement for the output gas composition between a CFD simulation and some set of experimental data may be the result of error compensation. Again, a reasonable way to validate a CFD model is to proceed hierarchically from much simpler configurations involving only a few processes and considering a wide variety of experimental results. However, even this approach might prove difficult. For instance, many kinetic data on biomass devolatilization may not have been obtained under rigorous kinetic control, casting doubt on their validity (46).

Computational Resources and Modeling Challenges

Simulating industrial-size fluidized-bed reactors while considering all physical and chemical processes involved is a fantastic task, even supposing that all appropriate models have been developed. The multi-scale nature of the flow requires either a very fine mesh resolution or adequate sub-grid scale models. Cold Lagrangian simulations, originally developed for two-dimensional beds (40), have very rarely been applied to three-dimensional beds and usually involve only a few thousands particles, very far from the actual number found in realistic systems and not sufficient to derive good statistical data. To circumvent this computational limitation and move toward more realistic simulations, particles

with similar characteristics may be grouped into parcels (44). Still, the problem is exacerbated by the wide particle-size distribution functions usually found in biomass samples.

To deal with particle-size distributions, Lagrangian methods require even more notional particles to correctly represent the distribution function, while Euler methods often consider separate conservation equations for each particle class. This approach quickly becomes complicated, as closure terms and interactions must be defined for each particle class and assumptions have to be made on particle velocity distributions (105). Simple binary mixtures were investigated and compared to experimental data using this approach (106). A very promising method called quadrature method of moments, or QMOM, represents the particle size distribution as a collection of weighted delta functions for which separate evolution equations can be solved (107). Particle mixing and segregation in a fluidized bed with a continuous particle size distribution were investigated and the results compared favorably with detailed DPM simulations (38).

Incorporating detailed chemistry into a CFD code is also very computationally expensive, as it introduces many additional conservation equations for each of the species considered, which in turn requires lengthy evaluation of the chemical source terms and may lead to space and time scales much smaller than those of the hydrodynamic processes. Once reactive time-scales dominate, the time steps and grid spacing are dramatically restricted. Virtually all CFD studies of reactive fluidized beds have considered only global conversion steps or multi-step kinetic schemes involving only a few major species. Very recently, Ranzi et al. (67) proposed the first semi-detailed kinetic model for biomass conversion containing several hundreds species and reactions. This model employed a lumped approach and a limited number of model compounds to describe biomass devolatilization where detailed mechanistic relationships are developed to describe the decomposition of the model compounds. However, the size of such mechanisms prevents them from being used directly in CFD. Several approaches to handle complex chemistry in CFD simulations have been developed in the context of hydrocarbon combustion systems that could potentially be transferred to gasification processes. The first one is to reduce *a priori* the detailed mechanism to a smaller size in terms of species and reactions based on homogeneous simulation results, so that only the chemical pathways relevant to the current study are retained (108). Then, computing the chemical source term can be optimized and accelerated using an on-the-fly storage and retrieval technique called in-situ adaptive tabulation, or ISAT (109). In each computational cell, the chemical source term and corresponding chemical state are stored as the simulation proceeds and are re-used instead of re-computed if a similar chemical configuration is found later in the simulation. The applicability of the method for reactive fluidized beds was demonstrated by Xie et al. (110) for silane pyrolysis.

Another tabulation technique consists of solving the evolution equations for a few variables only, chosen so that these variables map the entire chemical composition space with good accuracy. Detailed chemistry is tabulated *a priori* using these variables, which drastically reduces computational time. Tabulation methods have been developed and extensively validated for gas phase combustion

systems (111), in which a mixture fraction linked to the local ratio of fuel and oxidizer, or a progress variable describing the local extent of the global combustion reaction are easily defined. Such techniques cannot be directly applied to gasification processes, but must be adapted to the specific chemistry occurring in the fluidized bed.

One of the major obstacles to predictive CFD capabilities of biomass gasification or pyrolysis is the intrinsic complexity of the biomass itself. Biomass composition varies widely depending on the feedstock, age, geographic location, and even time of year. It was also shown repeatedly that biomass is not just the sum of its major components; lignin, cellulose, and hemicellulose (46). Biomass particles are highly anisotropic (112), contain trace components such as metal that can act as chemical catalysts (113) and moisture that may delay gasification (102). Biomass gasification is, therefore, a perfect example of a multi-scale problem, in which phenomena localized at the smallest scales are responsible for large-scale behavior. Reactor optimization requires these processes to be fully understood and characterized. Some detailed modeling studies at the scale of the biomass or coal particle take into account their complex structures (114, 115). Such investigations, coupled with experimental observations, are essential to develop larger scale statistical models suitable for use in CFD, because it is not currently conceivable to include that amount of detail in large-scale simulations.

Conclusions

Computational fluid dynamics methods, as an essential complement to experimental investigations, has a considerable potential to help meet our present and future needs for efficient energy production and conversion systems. However, we have shown that a lot of challenges still lay ahead in the near future for biomass thermochemical conversion processes to obtain reliable, predictive simulations that can be used as stand-alone tools for reactor design and optimization. Many areas of biomass conversion modeling can benefit from advances in other fields, such as combustion systems, in which CFD is a very active research topic. However, the modeling of biomass conversion systems combines two of perhaps the most challenging aspects of CFD, namely dense particulate flows and a complex and very specific chemistry. Until now, biomass chemical models have been based on global, over-simplified mechanisms obtained through experimental correlations. Because detailed and accurate chemistry is a key element in understanding and controlling output efficiency and tar formation, only significant progress in this area, coupled with major advances in numerical methods for multiphase flows and viable verification and validation strategies, will enable us to seize the opportunities provided by the ever-growing computational resources.

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