A Lagrangian-Eulerian hybrid model for the simulation of poly-disperse fluidized beds: Application to industrial-scale olefin polymerization

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ABSTRACT

We present a generalization of the Lagrangian-Eulerian hybrid model for the numerical assessment of poly-disperse gas-solid flows (Schneiderbauer et al., Pow. Tech., 2016) to olefin polymerization fluidized beds. The main idea of such a modeling strategy is to use a combination of a Lagrangian discrete phase model (DPM) and a coarse-grained two-fluid model (TFM) to take advantage of the benefits of those two different formulations. On the one hand, the DPM model unveils additional information such as the local particle size distribution, which is not covered by TFM. On the other hand, the TFM solution deflects the DPM trajectories due to the inter-particle stresses. Moreover, sub-grid drag corrections are applied to account for the impact of the small unresolved scales on the gas-solid drag force. This hybrid approach further enables the efficient evaluation of gas-solid reactions at a particle level using DPM. In particular, at each DPM trajectory we consider the olefin polymerization accounting for the catalyst profile (activity over time), the pressure driven solubility of the reaction gases in the polymer particles, the particles crystallinity and the corresponding reaction masses and reaction heat. These, in turn, are mapped to the TFM approach, where they appear as additional mass and energy source terms. The predictive capability and numerical efficiency of this reactive hybrid modeling approach is demonstrated in the case of (i) an inert bi-disperse fluidized bed and (ii) in the case of an industrial-scale olefin polymerization fluidized bed. The results show that the model is able to correctly predict segregation in poly-disperse gas-solid flows. In addition, the model is able to predict the particle growth in a fluidized bed reactor as well as its impact on the hydrodynamics of the bed. The results further give a closer insight about the temperatures and the crystallinity of the polymer particles.

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1. Introduction

Polyolefins (POs), including polyethylene (PE), polypropylene (PP), ethylene-propylene elastomer (EPR), etc., have found widespread use in many modern human-life applications due to their excellent chemical, physical and mechanical properties, superior processability, good recyclability and relatively low cost [2] One common approach to produce polyolefins is employing gas-phase fluidized bed reactors, which are recognized as economically attractive [3] In those catalytic fluidized bed reactors (FBR), catalyst or prepolymerized particles are continuously fed into the reactor at a point above the gas distributor and react with the incoming fluidizing monomer(s) to form a broad distribution of polymer particles (e.g., 100 to 5000 μm) [4] The particulate flows inside most practically relevant gas-phase Olefin polymerization reactions manifest a broad particle size distribution [2, 5, 6, 7] Thus, it is important to understand the mixing and segregation of the particles in the process to evaluate its efficiency [8, 9] One of the most straight forward numerical methods to account for poly-disperse mixtures is CFD-DEM (CFD: computational fluid dynamics; DEM: discrete element method), where the primary fluid phase is governed by computational fluid dynamics and the secondary dispersed phase is pictured by a discrete element model [10, 11] However, since the total number of particles in fluidized bed reactors is extremely large, it may be impractical to solve the equations of motion for each particle. It is, therefore, common to investigate particulate

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flows in large process units using averaged equations of motion, i.e. two-fluid models (TFM), which include the inter-particle collisions statistically by kinetic theory based closures of the particle stresses [12, 13, 14] However, each representative particle diameter requires an additional momentum and continuity equation, which considerably raises the computational demand with increasing number of particle diameters [15, 16] One may restrict the calculations to spatially constant particle size distributions to evaluate the gas-solid drag force [2] Furthermore, the TFM approach requires considerably fine grids since the minimum stable sizes of clusters are around ten particle diameters [17] Thus, due to computational limitations a fully resolved simulation of industrial-scale gas-solid flows is still unfeasible. It is, therefore, a frequent practice to use coarse grids to reduce the demand on computational resources. However, such a procedure inevitably neglects the small (unresolved) scales, which leads, for example, to a considerable overestimation of the bed expansion in the case of fluidized beds of fine particles [18, 19, 20] Recently, several authors have applied TFMs to polymerization reactors [5, 6, 7, 21, 22, 23, 24]. However, only a few studies are available dealing with a fully coupled simulation of the hydrodynamics of the fluidized bed and the polymerization reactions including reaction heat [25] and particles growth [7] For example, Yan et al. [7] coupled the TFM approach with a population balance equation (PBE) to account for transport of the different particle sizes as well as a polymeric multilayer model (PMLM) to include particle growth. Such a modeling approach inevitably demands significantly enhanced computer resources to solve the additional transport equations for the PBE as well as the PMLM locally for a huge number of particles. Thus, most of the studies dealing with the chemistry in gas-phase reactors restrict themselves to global balance equations [26, 27, 28, 29] To sum up, the current understanding of the numerical simulation of industrial-scale gas-phase Olefin Polymerization reactors exhibits three computational demanding limitations. First, resolving each particle trajectory and each particle-particle collision requires on the one hand unaffordable computational resources (with respect to time and costs); on the other hand, using an averaged model (such as TFM) does not account for poly-disperse particle flows in first place. In particular, standard TFM approaches require a separate set of transport equations for each particle fraction under consideration. This inevitably leads again to excess computational costs in particle laden flows, which are characterized by broad particle size distributions. Second, the TFM approach additionally requires very fine numerical grids, which additionally increases the computational demands in the case of industrial-scale applications considerably. Third, an efficient and resource-saving computational approach is required to account for the effect of the polymerization heat and the related particle growth on the hydrodynamics of gas-phase reactors.

To address the first concern, we follow our previous works [1, 16, 30, 31, 32] and present a hybrid model for the numerical assessment of poly-disperse gas-solid fluidized beds. The main idea of such a modeling strategy is to use a combination of a Lagrangian discrete phase model (DPM) and a TFM to take advantage of the benefits of those two different formulations. On the one hand, the local degree of poly-dispersity (i.e. the local particle size distribution), which is essential for the evaluation of the gas-solid drag force, can be obtained by tracking statistically representative particle trajectories for each particle diameter class. On the other hand, the computationally demanding tracking of the inter-particle collisions can be obtained from the inter-particle stresses, which are deduced from the TFM solution. These then appear in addition to the gas-particle drag as a body force in the equation of motion of each DPM-trajectory. Thus, the hybrid model represents a TFM simulation with additional DPM particles, which are used, for example, to provide a closure for the poly-disperse drag law. The second concern, is addressed by applying sub-grid modifications to the drag force and the kinetic theory stresses to account for the small unresolved scales

in the case of coarse grids. We refer this approach to as coarse grained TFM (cgTFM). Here, we employ sub-grid modifications presented in previous studies [2, 18, 19] Finally, by employing the above hybrid Lagrangian-Eulerian hybrid model, the polymerization rate as well as the corresponding reaction heat can be computed based on the representative Lagrangian particles. This, in turn, includes particle growth, since the diameter of those particles can be determined from the polymerization rate, which affects the particle size distribution (and thus the mean diameter) locally.

In this paper, we therefore demonstrate the predictive capability of such a hybrid model for the numerical assessment of poly-disperse gas-solid fluidized beds. First, we study segregation in an inert bi-disperse fluidized bed [9] The results show that the hybrid model is able to correctly predict the segregation of large particles as well as the pressure distribution in the bed. Second, we extend the hybrid approach to the numerical assessment of industrial-scale gas-phase Olefin Polymerization reactors by accounting for reacting enthalpies as well as particle growth by employing appropriate formulas for the reacting kinetics. The results are discussed with respect to temperature measurements in the corresponding real plant and the evolution of the particle size distribution with time. The following section discusses the sub-grid drag corrections [19] of the coarse grained TFM, the concept of the additional Lagrangian tracer particles and the polymerization model. Next, implementation issues are discussed in Section 4. In Section 5 results for the bi-dispersed fluidized bed and the polymerization fluidized bed are discussed. A conclusion section ends this paper.

2. Modeling of non-reactive poly-disperse gas-solid flows

2.1. Coarse grained two-fluid model (cgTFM)

Similar to our earlier study, Schellander et al. [16] we used a kinetic-theory based two-fluid model (TFM) to study gas-phase olefin polymerization fluidized beds. Since these equations have been extensively discussed in our previous works [13, 18, 19], we do not repeat all the details here and solely present the continuity and momentum equations for the solid phase below:

$$\frac{\partial}{\partial t} \epsilon_s \rho_s + \nabla \cdot (\epsilon_s \rho_s \boldsymbol{u}_s) = \mathcal{R}_s, \tag{1}$$

$$\frac{\partial}{\partial t}(\epsilon_s \rho_s \boldsymbol{u}_s) + \nabla \cdot (\epsilon_s \rho_s \boldsymbol{u}_s \boldsymbol{u}_s) = -\epsilon_s \nabla p - \nabla \cdot \left(\boldsymbol{\Sigma}_s^{\mathrm{kc}} + \boldsymbol{\Sigma}_s^{\mathrm{fr}} \right) + \beta (\boldsymbol{u}_g - \boldsymbol{u}_s) + \epsilon_s \rho_s \boldsymbol{g}. \tag{2}$$

Here, ρ_s, ϵ_s and $\boldsymbol{u}_s$ denote density, volume fraction and local-average velocity of the solid phase, respectively; $\mathcal{R}_s$ denotes the rate of generation of polymer due to chemical reactions; p is the gas phase pressure; $\boldsymbol{u}_g$ is the local-average velocity of the gas phase; β is the microscopic drag coefficient, which will be discussed later; $\boldsymbol{g}$ is the gravitational acceleration; finally, $\boldsymbol{\Sigma}_s^{\mathrm{kc}}$ and $\boldsymbol{\Sigma}_s^{\mathrm{fr}}$ are the stress tensors associated with the solids phase, where the frictional contribution, $\boldsymbol{\Sigma}_s^{\mathrm{fr}}$, arises from enduring or multi-particle collision events in dense areas. The kinetic-collisional part, $\boldsymbol{\Sigma}_s^{\mathrm{kc}}$, is closed using kinetic theory [12, 13] which requires an additional equation for the granular temperature. Since we thoroughly discussed the closures for the solid stresses and the granular temperature equation, we do not discuss them here. For further details we refer to our previous study [13] In the case of coarse grids it is common to use balance equations for the filtered counterparts of the local-average solids volume fraction ϵ_s and solids velocity $\boldsymbol{u}_s$ reading as [18, 19, 20, 33]

$$\frac{\partial}{\partial t} \rho_s \bar{\epsilon}_s + \nabla \cdot (\rho_s \bar{\epsilon}_s \tilde{\boldsymbol{u}}_s) = \tilde{\mathcal{R}}_s, \tag{3}$$