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CFD modeling and simulation of industrial scale olefin polymerization fluidized bed reactors

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HIGHLIGHTS

- We derive subgrid corrections for coarse-grid simulations of HDPE fluidized beds.
- These corrections account for polydispersity of high density polyethylene (HDPE).
- These corrections are validated by a lab-scale experiment.
- The model is applied to an industrial scale fluidized-moving bed reactor assembly.
- The results discusses the separation of the reaction gases between both reactors.

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ABSTRACT

A two-fluid model for the numerical simulation of industrial scale olefin polymerization fluidized bed reactors is presented. However, a fully resolved simulation of industrial scale reactor is still nearly unfeasible. We, therefore, use sub-grid models (Schneiderbauer and Pirker, 2014) for the interphase drag and the solids stresses to account for the effect of the small unresolved structures on large resolved scales when using coarse grids. The sub-grid correction for the drag force is modified to consider the wide particle size distribution of high density polyethylene (HDPE). Furthermore, the sub-grid modification for the solids stresses is adapted to include the rheological properties of the polymer. On the one hand, the presented model is validated in the case of the coarse grid simulation of lab-scale bubbling fluidized bed by comparing bed expansion, bubble size and bubble rise velocities with experimental data. On the other hand, the model is applied to the coarse grid simulation of an industrial scale fluidized bed – moving bed reactor assembly. The numerical results demonstrate that our model reveals fairly good agreement with experimental data of average bed voidage, bubble diameters and bubble rise velocities. Finally, the impact of a barrier gas injection is studied, which is aimed to separate the fluidization gas from the gas in the moving bed reactor.

1. Introduction

1.1. Fluidized bed reactor technology for polyolefin manufacturing

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. It is believed that the degree of technological and scientific sophistication in relation to the polyolefin manufacturing has no equal among other synthetic polymer production processes. Polyolefins are commonly produced in low-pressure catalytic (e.g., Ziegler–Natta (Z–N), metallocenes, etc.) bulk, slurry and gas-phase reactors and they are recognized as economically attractive and environmentally friendly polymeric materials that account for more than half of the annual world-wide polymer production of approximately 200 millions metric tons [2]. Polyethylene (i.e., HDPE, LDPE

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and LLDPE) and polypropylene cover 60% and 40% of the total polyolefins production, respectively. In a catalytic fluidized bed reactor (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., 100–5000 μm) [3].

1.2. Enhancing polyolefins properties using fluidized bed-moving bed reactor assembly

The physical, chemical, mechanical and rheological properties of polymers are determined to a large extend by their chains microstructure. For example, high molecular weight polyolefins exhibit improved mechanical properties (e.g. toughness, strength, impact resistance and environmental stress cracking resistance, etc.). However, high molecular weight resins have higher melt viscosities and, therefore, it is more difficult to process them. On the other hand, polyolefins having a broad or bimodal molecular weight distribution (MWD) exhibit improved processability and, particularly, extrudability properties. The copolymer composition distribution (CCD) is another important distributed molecular property that influences the end-use properties (e.g. stiffness, hardness, shrinkage, transparency, optical properties, etc.). Thus, understanding and controlling the molecular properties in terms of process operating conditions is the most important step towards the production of polyolefins with desired application characteristics [4]. To exploit the great versatility of polyolefins and expand their applications, an optimal balance among the microstructural properties of the copolymer chains is required. This balance can effectively be achieved through the production of polymers with bimodal or/and multi-modal distributed molecular properties. Thus, polyolefins with a bimodal MWD can have both excellent mechanical properties, due to the high molecular weight tail of the distribution, and good processability due to the low molecular weight chains. The production of polyolefins with broad MWDs has been achieved by using two reactors, which are connected in series and operate under different operating conditions as well as use mixed catalyst systems. However, a certain segregation between high and low molecular weight polymer inevitably occurs in these systems, which negatively affects the operability of such processes [5]. To obtain an intimate mixture of different polyolefins, two gas phase reactors can be interconnected each other aiming at producing mixtures of polyolefins of widely different characteristics, thus, enlarging the envelope for properties of these polymers. In such a process, the polymerization is carried out in a gas-phase reactor having an interconnected polymerization zone, where the growing polymer particles (having bulk density approximately equal to 250 kg m^{-3}) flow through a first polymerization zone (FBR) under normal fluidization transport conditions. Then, they leave the FBR and they enter in a second polymerization zone, through which they flow in a densified form under the action of gravity (moving bed reactor, MBR), having polymer bulk density in the range of 340–420 kg m^{-3}). Subsequently, the polymer particles leave the MBR and are reintroduced into the FBR, thus establishing a circulation of polymer between the two polymerization zones. Depending on the aimed olefin, the polymerization in the FBR–MBR reactor assembly can be carried out by properly adjusting the polymerization conditions and the monomer(s) concentration in the two polymerization zones, so as to produce a wide variety of bimodal homopolymers and random copolymers.

1.3. CFD Modeling of gas-phase Olefin Polymerization Reactors

Since the total number of particles involved in most practically relevant gas-phase Olefin Polymerization Reactors is extremely large, it is impractical to solve the equations of motion for each particle [6]. It is, therefore, common to investigate particulate flows in large process units by using averaged equations of motion [7,8], which are generally referred to as two-fluid models (TFM). For solids volume fractions up to 40% ($\epsilon_s \leq 0.4$), which is a typical range in fluidized beds and risers, the solids stresses arising from particle-particle collisions and the translational dispersion of the grains are commonly deduced by adapting the kinetic theory of gases [9–11]. The derivation of this kinetic theory of granular flows (KTGF), thereby, assumes that the collisions can be considered as binary and that multiple collisions are rare. In dense regimes, which are referenced as frictional, the particle-particle contacts are determined largely by enduring multiple frictional contacts [12] and, thus, kinetic theory does not apply. The closure of these dominant sustained multiple sliding frictional contacts in the dense regime is commonly based on critical state theory of soil mechanics ([13,14] and references cited therein). In the literature, there is a huge amount of studies dealing with frictional stress closures, which are based on Coulomb's law [13–18].

Due to computational limitations a highly resolved simulation of industrial scale reactors is still unfeasible [19–26]. It has been argued in the literature [27] that 10 particle diameters is about the size of the smaller clusters, and that the affordable grids are in fact much larger than that. However, such a procedure inevitably neglects small (unresolved) scales, which leads to a considerable overestimation of the bed expansion in the case of fine particles. Several authors have applied TFMs to polymerization reactors [28–34] and to scale biomass reactors [35–37]. In particular, Yan et al. [31] used the TFM approach to optimize the design of a multizone circulating polymerization reactor. Dehnavi et al. [29] verified the applicability of the TFM approach to lab-scale gasphase ethylene polymerization reactors by comparing the computed bed pressure drop with experimental data. Others [32,33] augmented the two-fluid models by a population balance to include particle size distributions. However, all of these studies did not consider those sub-grid contributions. Recently, numerous sub-grid modifications have been proposed to account for those unresolved scales [1,20,21,38–47].

In addition, polymer particles commonly manifest a broad particle size distribution and the particles are usually non-spherical. On the one hand, the drag force on the binary mixtures is considerably dependent on the particle size distribution [48,49]. To the authors knowledge solely sub-grid modifications in the case of bidisperse mixtures have been studied in detail [44]. However, recently Chen et al. [50] performed coarse-grid simulations of a poly-disperse riser by using an extended version of the EMMS model, where the local Sauter diameter was applied to the underlying mono-disperse Wen and Yu [51] drag model instead of incorporating more sophisticated poly-disperse correlations, such as Beetstra et al. [48,49] and Yin and Sundaresan [49]. On the other hand, the flowability behavior of the polymer particles is strongly connected to the degree of non-sphericity and the coefficient of inter-particle friction. In particular, the flowability plays a major role in dense regions in FBR and in MBR. Both effects have not been considered in those studies dealing with polymerization reactors.

In this study, we present a coarse grained TFM model, which can be applied to the coarse grid simulation of industrial scale polymerization reactors. Furthermore, we revised the sub-grid modifications for the drag and the solids stresses presented in our previous studies [1,40,41] to account for the physical properties of the high density polyethylene (HDPE). The model is validated against experimental data in the case of a coarse grid simulation of a lab-scale fluidized bed of HDPE particles. This validation includes the bed expansion, bubble number density and bubble rise velocities. Finally, the model is applied to an industrial scale FBR–MBR assembly.