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Experimental validation of CFD hydrodynamic models for catalytic fast pyrolysis

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ABSTRACT

Hydrodynamics for catalyst fluidization and circulation for Catalytic Fast Pyrolysis (CFP) were investigated using cold flow experiments and used to validate predictions from computational fluid dynamics (CFD) simulations. Proprietary fresh and equilibrium CFP catalyst were tested in lab-scale fixed fluidized bed (FFB) and circulating fluidized bed (CFB) setup. Bed expansion and pressure drop were recorded for fresh catalyst in the fixed fluidized bed setup for a range of gas velocities (0–0.04 m/s) which encompassed minimum fluidization and bubbling regimes. Catalyst circulation in the circulating fluidized bed setup on the other hand, was quantified using mass holdup in the mock reactor, particle size distribution for this holdup and pressure profile for a range of transport gas velocities (0.094–0.561 m/s, 10–60 SLPM) and catalyst feedrates (3, 6, 12 kg/h; $G = 0.46, 0.92, 1.84$ kg/m^2-s) for a single-inlet and single-outlet mock reactor configuration. Flow patterns and gas jet penetration through the mock reactor provided qualitative observations that were used for evaluating CFD models. CFD code based on CPFD method (Barracuda Virtual Reactor®) was used to simulate these gas-solid fluidized beds based on three-dimensional grids. Grid independence was established for both fixed and circulating fluidized bed models such that an average grid volume of 23–29 mm^3 was sufficient to effectively predict quantitative and qualitative observables. Several CFD parameter sets were tested to evaluate combinations of drag models, blended acceleration model, particle-wall interactions and collision and stress models. Parker-drag model with blended acceleration and advanced settings for particle-wall interactions, collision and stress parameters is determined to provide the best agreement with experimental results.

1. Introduction

Catalytic Fast Pyrolysis (CFP) is a promising technology for producing renewable transportation fuels and chemicals from lignocellulosic biomass [1,2]. In the CFP process, circulating fluidized beds (CFBs) of catalyst particles are used to simultaneously pyrolyze biomass particles and catalytically upgrade the product vapors. CFBs used for CFP can range in solids volume fraction from risers and transport reactors on the dilute end of the spectrum, to bubbling beds on the other end [3]. The products of the CFP reactor are gases (CO, CO_2, and H_2O, plus oxygenate and hydrocarbon molecules with <5 carbon atoms), coke and char (which are burned in the regenerator to power the endothermic process) and a partially deoxygenated bio-oil. As a consequence of the catalytic deoxygenation that occurs in the CFP reactor, the bio-oil is much more stable and easier to handle than typical higher oxygen bio-oils made by strictly thermal pyrolysis processes. Downstream of the CFP reactor, the bio-oil is condensed, dewatered, filtered, hydrotreated to remove remaining oxygen, and fractionated to produce valuable fuels and chemicals.

Inaeris Technologies (formerly KiOR) is a world leader in the development and commercialization of the CFP process. Inaeris Technologies's CFP process is modeled on petroleum FCC; however, there are substantially important differences that make it difficult to apply the nearly seven decades of accumulated petroleum FCC knowhow. First, the catalyst is very different. The chemical functions are different: unlike FCC, catalytic deoxygenation is the principal cracking mechanism (although C–C bond cracking does occur as well). Secondly, the physical demands placed on the catalyst are also different. In FCC, the catalyst carries the heat from regenerator to riser needed to vaporize a heavy gas oil (VGO) or vacuum residue (resid or VR) and power the endothermic cracking process. In CFP, the catalyst must carry a much larger quantity of heat to pyrolyze solid biomass particles, then deoxygenate and crack the pyrolates. Inaeris Technologies's estimates for this heat requirement are three to six times the heat required in FCC to process an equivalent mass of VGO or VR. In order to satisfy this heat requirement, avoid overloading catalyst particles with coke, and provide ample surface area for catalyst-gas heat transfer, catalyst-to-

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biomass feed (CTF) mass ratios in CFP tend to be considerably higher than typical catalyst-to-oil (CTO) mass ratios in FCC.

In addition, the reaction residence times and mixing conditions are very different for CFP and FCC. In FCC, the use of advanced nozzles [4] and high activity catalysts have pushed riser residence time lower and lower, ultimately ending up below 1 s in short contact time (SCT) and even in the millisecond range for millisecond-catalytic cracking [5]. In CFP, this trend is not at all accepted as beneficial. While fast pyrolysis requires high particle heat-up rates to avoid excessive char formation—on the order of several hundred degrees per second [1]—time must be provided to achieve adequate mixing of catalyst and biomass. Optimizing the mixing of catalyst and biomass to achieve uniform fast pyrolysis conditions is much more challenging technically than mixing oil droplets and catalyst. Plus, catalytic deoxygenation reactions are generally slower than C–C cracking reactions [2].

For all of these reasons and more, Inaeris Technologies has been executing an aggressive research program into the relevant physics and chemistry of the CFP process and focusing on the development of a CFD model capable of reactor design, scale-up, troubleshooting and optimization. To achieve the model development objectives at an accelerated pace, Inaeris Technologies has been collaborating with CPFD Software, and has selected the Barracuda Virtual Reactor® software as the CFD package of choice.

The capability to make meaningful predictions about catalyst hydrodynamics (fluidization and circulation) in various configurations of CFB reactors is a core requirement for a successful CFD model. Furthermore, biomass transport, dispersion (agglomeration/deagglomeration), catalyst-biomass mixing, and catalyst-gas-biomass heat transfer add additional layers of complexity to this model. The chemistry layer - the complex reaction kinetics—can only be added once all these underlying layers are in place. For this reason, the Inaeris Technologies program has been structured in a fairly strict hierarchical fashion, as illustrated in Fig. 1. The two innermost layers—fluidization and catalyst circulation—are the subject of this paper. Batch mixing experiments of biomass and catalyst have been performed in parallel [6], but are beyond the scope of this paper. Currently, our program is addressing the stage of biomass transport and catalyst-biomass mixing in CFBs.

The objective of this study is to develop CFD models for describing CFP hydrodynamics and rank and validate them using cold flow experiments. This is essentially an application paper and focuses on fundamentals necessary to achieve this goal. Given this sharp degree of focus, the experiments in this study are restricted to a single commercial CFP catalyst and an “e-cat” (equilibrium catalyst, per FCC parlance) prepared in-house by testing the catalyst in Inaeris Technologies's proprietary pilot unit (KCR). Details concerning this catalyst and its development and manufacture—including the manufacturer—are proprietary and cannot be disclosed. However, significant details on the cold flow experimental setup and modeling work are included in this paper, allowing a thorough review of the results and conclusions.

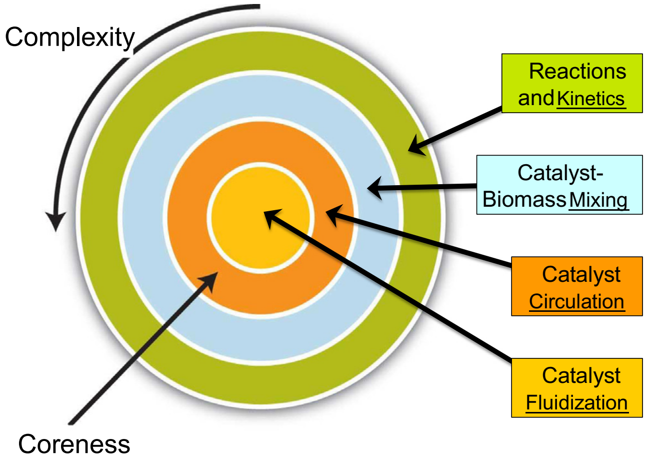

Fig. 1. The hierarchy followed in the Inaeris Technologies model development program. This paper focuses on the two innermost layers: catalyst fluidization and circulation.

The current study entails two lab-scale cold-flow setups: a fixed fluidized bed (FFB) unit and a CFB unit. We report both experimental and computed bed expansions and pressure drops for fresh catalyst in the FFB unit, for gas velocities ranging from minimum fluidization U_{mf} to values well into the turbulent regime. Next, we compare and discuss experimental flow regime maps for both fresh catalyst and e-cat circulated in the CFB setup, and quantitatively compare the CFD-predicted values for catalyst mass holdup, pressure drop and extent of classification (reduction in 0–40 μm fines content) against the experimental measurements. Based on these all of considerations, we rank different CFD parameter sets for their relative worth in predicting both quantitative (bed expansion, pressure profiles, catalyst holdup, particle size distribution) and qualitative observables.

2. Experimental details

2.1. Catalysts

Fig. 2 shows the particle size distribution (PSD) of the fresh CFP catalyst, along with a typical microscopic picture of the fresh catalyst. This particle size analysis is based on the laser scattering method [7]. Visually, it is clear that the sphericity of this catalyst is quite high for a commercial catalyst produced in a production-scale spraydryer. The catalyst has a very low content of particle clusters, twinned particles and other morphology defects.

Also shown in Fig. 2 is the PSD of the KCR e-cat used in this study. This catalyst was prepared by deactivating the fresh catalyst and testing it in one of Inaeris Technologies' KCR units. These are pilot-scale units based on the Davison Circulating Riser (DCR) design, with proprietary modifications made to convert the unit from FCC to CFP service. This e-cat shows a narrower size distribution due to pre-sieving before the KCR test, and additional classification that occurred during the test. US Sieve sizes No. 325 (44 um) and No. 100 (149 um) were used for pre-sieving to remove both <40 um fines and 150 + um oversized particles. Most of the 0–40 μm fines have been removed from the e-cat: 0–40 μm content is 2 v%, vs 12 v% in the fresh catalyst.

2.2. Fixed fluidized bed (FFB) setup and experimental procedure

Fixed-fluid-bed experiments were carried out in a cylindrical column with 7.6 cm id and 34 cm height, made from 0.65 cm thick Lexan tubing. The complete apparatus is shown schematically in Fig. 3. Fluidizing N_2 gas is first passed through a bed of ceramic beads, then through a sintered metal distributor plate at the inlet of the main cylindrical column. The beads and sintered metal plate (20 μm average pore size) were designed to ensure that N_2 gas flow is uniform across the cylindrical cross-section. In keeping with best practices for distributor design, ΔP across the sintered metal plate ranged from 30% to 80% of bed weight, with higher values corresponding to higher velocities.

In the FFB unit, N_2 gas flowrate is controlled by a mass flow controller (Omega, 0–15 SLPM range) having a repeatability of ±1% of full scale. Per Omega documentation, standard temperature of 21.1 °C (70 °F) and absolute pressure of 101.3 kPa was used for reporting SLPM values. Gage pressure transducers (±0.08% accuracy) are located along the wall of the cylindrical column at 5 cm intervals above the distributor plate. A reference gage pressure transducer is installed 4 cm below the distributor plate.

The FFB distributor plate was carefully checked before each experimental run to ensure the pores had not been blinded by catalyst particles. First, validation runs were performed by flowing N_2 through an empty column (no catalyst) at four velocities ($U = 1, 2, 3$ and 4 cm/s) and checking ΔP against standard values. If this test passed, an additional check was to visually verify uniform bubbling of gas through a

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