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Ebullated bed fluid dynamics relevant to industrial hydroprocessing

Dominic Pjontek [a], Craig A. McKnight [b], Jason Wiens [b], Arturo Macchi [a,*]

[a] Centre for Catalysis Research and Innovation, Department of Chemical and Biological Engineering, University of Ottawa, 161 Louis Pasteur, Ottawa, ON, Canada, K1N 6N5

[b] Syncrude Canada Ltd., 9421-17 Avenue, Edmonton, AB, Canada, T6N 1H4

HIGHLIGHTS

- Overall phase holdups in an ebullated bed were measured at high gas holdups.
- Scale-down was based on geometric features, flow regimes and dimensionless groups.
- Increased liquid viscosity led to gas recirculation and solid holdups reduction.
- Freeboard gas holdups from solids-free estimate depend on bubble–particle interaction.
- Phase holdup correlations were provided for significant bubble coalescence inhibition.

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ABSTRACT

This study investigates the overall fluid dynamics of an ebullated bed operating at high gas holdup conditions to provide relevant observations for industrial residue hydroprocessors. Scaling approaches for three-phase fluidized beds were compared specifically for the scale-down of the industrially observed high gas holdup conditions. Five dimensionless groups, a binary approach for bubble coalescence behaviour in multi-component liquids, and geometric considerations are proposed to achieve dynamic similitude. Experiments were carried out in a 101.6 mm diameter co-current gas–liquid–solid fluidized bed operating at 0.1 and 6.5 MPa with liquids that do (e.g., 0.5 wt% aqueous ethanol) and do not (e.g., tap water) significantly inhibit bubble coalescence. A comparison of the overall phase holdups for two sizes of cylindrical particles (d_{SV} of 1.6 and 3.9 mm) at matching dimensionless groups provided a preliminary verification of the proposed scaling approach when bubble coalescence was sufficiently and consistently inhibited. The impacts of increased liquid viscosity (e.g., greater vacuum distillation tower residue feed fraction), varying superficial gas velocity (e.g., inlet gas flow rate and gas entrainment in the liquid recycle line), and varying superficial liquid velocity (e.g., liquid recycle pump speed) were experimentally studied due to their relevance for industrial ebullated bed hydroprocessors. For the studied operating conditions, gas holdups in the ebullated bed were much greater when bubble coalescence was sufficiently inhibited (up to approx. 45 vol%) compared to a coalescing system at equivalent conditions (up to approx. 25 and 35 vol% for 0.1 and 6.5 MPa, respectively). When increasing the liquid viscosity of the 0.5 wt% aqueous ethanol, a fraction of the gas was entrained in the liquid recirculation, increasing gas holdups and exhibiting operational similarities to industrial hydroprocessors. After adjusting the gas and liquid flow rates based on the gas recirculation, the increased liquid viscosity mainly reduced the solid holdups while gas holdups were approximately unchanged. Enhanced bubble break-up or bubble coalescence due to particles resulted in an overestimation or underestimation, respectively, of the freeboard gas holdups from a solids-free estimate based on the bed region phase holdups, where this model could not capture the complex bubble–particle interactions. Experimental results at high gas holdups conditions were used to correlate the bed and freeboard phase holdups based on the proposed dimensionless groups.

1. Introduction

The performance and optimization of industrial ebullated bed residue hydroprocessors, such as the LC-Finer^{SM}, are highly dependent on the overall fluid dynamic behaviour in the bed

* Corresponding author. Tel.: +1 613 562 5800x6939; fax: +1 613 562 5172.
E-mail address: arturo.macchi@uottawa.ca (A. Macchi).

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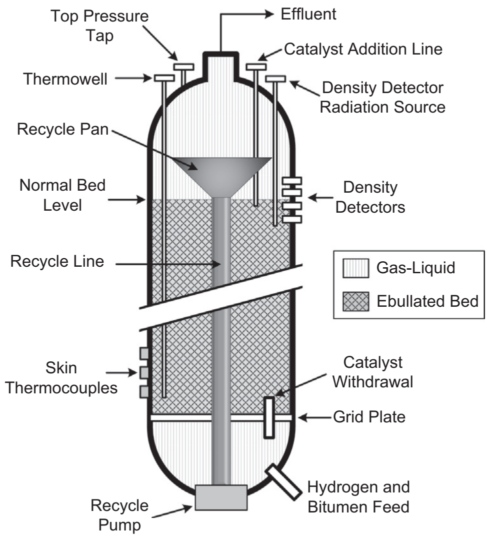

Fig. 1. Schematic of the LC-Finer^{SM} hydroprocessor (modified from McKnight et al., 2003).

and freeboard regions. Syncrude's LC-Finer^{SM} operates at elevated temperatures and pressures of approximately 440 °C and 11.7 MPa (McKnight et al., 2003), respectively, required for residue upgrading. A schematic of the unit is provided in Fig. 1. The inlet gaseous hydrogen and liquid atmospheric/vacuum distillation tower bottoms mixture are heated separately and then fed into the plenum chamber below the grid (i.e., gas–liquid distributor plate) using a horse-shoe/shroud distributor assembly. The feed is mixed with the recycled fluid, mainly consisting of unconverted liquid and some entrained gas from the freeboard region, before flowing through the risers and bubble caps located in the grid plate. Doped alumina cylindrical catalysts are fluidized by the co-current gas and liquid flow, where liquid can be considered the continuous phase while the hydrogen and catalyst constitute the dispersed phases. Above the ebullated bed, the liquid is recirculated to the plenum chamber using a recycle pan and pump. The liquid recirculation provides the necessary flow to fluidize the catalyst particles while also maintaining temperature uniformity throughout the reactor. It should be noted that the catalyst bed level is monitored using gamma-ray density detectors (shown in Fig. 1). These measurement devices have also been used to estimate freeboard gas holdups based on approximations for the gas and liquid densities at the reaction conditions (McKnight et al., 2003). Discrepancies between industrial measurements and typical experimental systems available in the literature generally arise from considerable differences in operating conditions, phase physical properties and column geometries. The high gas holdups observed in ebullated bed hydroprocessors at industrial operating conditions (McKnight et al., 2003) are difficult to predict or model due to the impacts of operating pressure and various interfacial phenomena. Experimental studies at industrially relevant fluid dynamic conditions are thus required to improve their design, optimization and regular operation. An appropriate scale-down method for the industrially observed high gas holdups must be identified, where scaling in general still presents an important challenge for gas–liquid–solid fluidized beds.

McKnight et al. (2003) discussed and identified key objectives to improve the LC-Finer^{SM} performance, noting that minimizing the bed and freeboard gas holdups requires further investigation to maximize pitch conversion. Freeboard gas holdup measurements in the industrial hydroprocessor (approx. 50 to 60 vol%) were considerably greater than predictions (approx. 15 to 25 vol%) for comparable operating conditions based on selected correlations from the literature (Hughmark, 1967; Tarmy et al., 1984). Safoniuk et al. (1999) hence proposed a scale-down approach based on dynamic similitude using the following dimensionless groups:

$$M=\frac{g(\rho_L-\rho_G)\mu_L{}^4}{\rho_L{}^2\gamma_{G-L}{}^3},\quad Eo=\frac{g(\rho_L-\rho_G)d_P{}^2}{\gamma_{G-L}},\quad Re_{L-S}=\frac{\rho_L\,d_P\,U_L}{\mu_L},\ \rho_P/\rho_L,\quad U_G/U_L \tag{1}$$

The method assumed that: (i) gas viscosity was negligible compared to the liquid viscosity, (ii) equilibrium interfacial properties were sufficient to characterize bubble coalescence behaviour, (iii) gas density was much lower than the liquid and solid densities, therefore it was only included in the buoyancy term ($g\,[\rho_L-\rho_G]$), and (iv) wall effects could be relaxed above a given column-to-particle size (d_C/d_P) ratio in the dispersed bubble flow regime. When attempting to match the proposed dimensionless groups for the LC-Finer^{SM} using a cold-flow experimental system with relaxed geometrical constraints, industrial freeboard gas holdups nearly doubled those obtained with the laboratory unit (McKnight et al., 2003). The significant discrepancy between industrial and the cold-flow system was attributed to the following possible reasons:

1. internal gas recycle via the liquid return line in the industrial unit,
2. inaccurate measurements of phase physical properties and holdups in the industrial unit,
3. inadequate and/or missing dimensionless groups for the fluid dynamic scale-down.

Although the first and second considerations could have significantly influenced the comparison, the large gas holdup differences were also believed to be due to difficulties simulating the high gas holdup conditions in a cold-flow unit. The authors suggested based on other experimental studies that the influences of interfacial phenomena for multi-component liquids (Macchi et al., 2001) and increased gas density due to elevated pressures (Luo et al., 1999; Macchi et al., 2003; Wilkinson et al., 1992) must be considered.

Few ebullated bed experimental studies which are relevant to the fluid dynamics of the LC-Finer^{SM} can be found in the available literature. Tarmy et al. (1984) and Ishibashi et al. (2001) measured the gas holdups in pilot scale coal liquefaction slurry bubble column reactors (i.e., reduced particle size and liquid flow rates compared to an ebullated bed) operating at pressures up to 20 MPa and temperatures up to 450 °C. They observed high gas holdups which were attributed to the large kinetic energy of the high pressure inlet gas and the presence of surface-active components. Luo et al. (1999) studied the bubble characteristics in a slurry bubble column operating at pressures up to 5.62 MPa and using Paratherm NF heat transfer fluid. The authors discussed the impact of operating pressure on the bubble break-up behaviour, thus reducing the maximum stable bubble size. The previous studies provide useful observations on bubble characteristics at elevated temperatures and/or pressures, however larger solid particles and increased liquid flow rates must be considered for an ebullated bed. Fan et al. (1987) and Song et al. (1989) investigated a 0.5 wt% aqueous *t*-pentanol solution in a cold flow ebullated bed at atmospheric pressure. The interfacial phenomena leading to bubble coalescence inhibition increased the gas holdups due to the reduced bubble size and rise velocities. Luo et al. (1997) studied pressure effects on the hydrodynamics and heat transfer in an ebullated bed at pressures up 15.6 MPa using Paratherm NF heat transfer fluid. The results provided valuable information on the fluid dynamic behaviour when increasing the pressure, however relevant high gas holdups were not observed and the superficial liquid velocity was restricted ($U_L<0.026$ m/s). Ebullated bed