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Impact of catalyst density distribution on the fluid dynamics of an ebullated bed operating at high gas holdup conditions

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HIGHLIGHTS

- Impact of hydroprocessing catalyst PDD on fluidization behaviour was investigated.
- Marginal axial phase holdup variation observed for the L-S fluidized bed.
- G-L-S fluidized phase holdups were dependent on relative bubble sizes.
- Relatively small non-coalescing bubbles favour sharp bed-freeboard interface.

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ABSTRACT

Experiments were conducted to investigate the impact of particle density distribution on ebullated bed phase holdups and local fluidization behaviour when operating under high gas holdup conditions. Fresh and spent heavy oil hydroprocessing catalyst having relatively narrower and wider density distributions were compared. A 0.5 wt.% aqueous ethanol solution was used to obtain relatively high gas holdups as observed in many industrial reactors containing liquid mixtures with surface-active compounds. Axial pressure profiles were used to assess the degree of segregation on liquid-solid and gas-liquid solid fluidized beds. While marginal axial holdup variation occurred when operating the liquid-solid fluidized bed, introduction of gas significantly impacted the fluidized bed dynamics by rendering the bed-freeboard interface diffuse at low superficial liquid velocity as relatively large bubbles were formed. This was observed visually and experimentally based on the pressure profile curvature. At elevated liquid flow rates, the bed interface became more stable due to smaller bubbles being formed because of the greater shear stress at the gas-liquid distributor. Solid holdup was the most affected by the particle density distribution, where bed expansion/contraction was dependent of the liquid flow rate due to varying particle-bubble dynamics. Such information provided guidance on potential factors that can lead to the loss of bed-freeboard interface in the operation of heavy oil hydroprocessors such as the LC-Finer^{SM}.

1. Introduction

Physical properties (i.e., size, shape and/or density) distributions for solid particles can be encountered in industrial applications of three-phase fluidized bed reactors such as catalytic hydroprocessing of heavy oil residues (e.g., LC-Fining and H-Oil processes), Fisher-Tropsch synthesis, and waste water treatment (Fan, 1989). Even though solids particles may have uniform physical properties at the beginning of a process, variations may be progressively observed due to attrition, sintering, or chemical reaction. For example, due to uneven growth of biological film on supported media surface, particle size and/or density distribution can occur during the operation of a fluidized bed bioreactor (Fan et al., 1985). Variations in solid physical properties may adversely affect the normal operation of a process as particles may segregate or intermix, depending on the operating conditions, potentially influencing heat and mass transfer characteristics as well as reaction conversion. The impact of density driven solids mixing and/or segregation is investigated in this study.

The unit of interest in this study is the LC-Finer^{SM} resid hydroprocessor (see Fig. 1), which respectively operates at pressures and temperatures of approximately 11.7 MPa and 440 °C (McKnight et al., 2003). To maintain the catalytic activity, fresh

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Nomenclature

Ar_{L-S} liquid-solid Archimedes number, $Ar_{L-S} = \rho_L d_V^3 (\rho_S - \rho_L) g/\mu_L^2$
CV coefficient of variation
d_C column inner diameter (m)
d_P particle diameter (m)
d_{SV} Sauter-mean diameter (m)
d_V volume equivalent diameter (m)
g gravitational acceleration (m/s^2)
h_B bed height (m)
L_P particle length (m)
m mass of particle (kg)
$-\Delta P$ dynamic pressure drop (Pa)
Re_{Lt} liquid-particle Reynolds number based on terminal velocity, $Re_{Lt} = U_{Lt} \rho_L d_V/\mu_L$
T temperature (°C)
U_G, U_L gas and liquid superficial velocities (m/s)
U_{Lt} terminal settling velocity of particle, accounting for wall effects (m/s)
x_i mass fraction (–)
Δz vertical distance between differential pressure taps (m)

Greek symbols
ε_G, ε_L, ε_S global gas, liquid and solid holdups in the bed region (–)
ε_{G-FB} global freeboard gas holdup
μ_L liquid dynamic viscosity (Pa s)
ρ_G, ρ_L, ρ_S gas, liquid and solid densities (kg/m^3)
φ sphericity (–)

catalyst is fed to the reactor while spent (or equilibrium) catalyst is withdrawn at continuous intervals. During demetalization and catalytic cracking of heavy oil residue, heavy metals and coke will deposit into the catalyst pores, thus fouling and deactivating the catalyst due to pore volume reduction. As a result, a relatively wide particle density distribution will arise as a function of the particle residence time distribution within the reactor (McKnight and Nowlan, 1993). The catalyst bed level is monitored using gamma-ray density detectors above and below the bed-freeboard interface. Having a particle density distribution inside the reactor may influence the ebullated bed behaviour and render the bed level diffuse. Prior knowledge of bed-freeboard interface dynamics and spatial distribution of phase holdups would assist with the unit monitoring and control in order to prevent the loss of bed interface and resulting potential operational issues such as catalyst carry-over into the gas-liquid separator (recycle pan) and bed slumping.

Previous studies have mainly focused on binary-solids mixtures in gas-solid and liquid-solid fluidized beds (Asif, 2004, 2002; Di Maio and Di Renzo, 2016; Epstein et al., 1981; Formisani et al., 2008; Gibilaro and Rowe, 1974; Rowe et al., 1972; Wakeman and Stopp, 1976), and to a lesser extent ternary-solids mixtures (Escudié et al., 2006; Olaofe et al., 2013; Wang and Chou, 1995). Limited research on gas-liquid-solid fluidized beds having solid mixtures has been found in the open-literature (Chun et al., 2011; Fan et al., 1985; Kim et al., 2017; Rim et al., 2013, 2014) where the focus is mainly on the impact of the gas phase on particle layer inversion; with Fan et al. (1985) also reporting gas holdups. Furthermore, academic experimental studies that have focused on investigating the fluid dynamics of three-phase fluidized bed hydroprocessors have used particles of relatively uniform density distribution (Fan et al., 1987; Jiang et al., 1997; Luo et al., 1997; Song et al., 1989; Kama et al., 1999; Ruiz et al., 2004a, 2004b, 2005; Sanchez et al., 2008; Pjontek et al., 2015). Finally, fluid dynamic models for three-phase fluidized beds usually do not have a parameter for the particle density distribution, but may still account for a resulting axial solids holdup profile (Eccles, 1993; Larachi et al., 2001; Fan and Yang, 2003; Schweitzer and Kressmann, 2004; Martínez et al., 2010; Cheng et al., 2014; Ye et al., 2015).

As it is thus of both academic and industrial interest, this work experimentally investigates the impact of a particle density distribution on gas-liquid-solid fluidized beds phase holdups and bed behaviour when operating under high gas holdup conditions relevant to the LC-Finer^{SM} hydroprocessor. Experimental bed behaviour and interface sharpness are discussed and related to gas-liquid distributor design and bubble-particle interaction. Finally, thoughts are provided on the potential impact of the particle density distribution on the loss of bed-freeboard interface at LC-Finer^{SM} operating conditions.

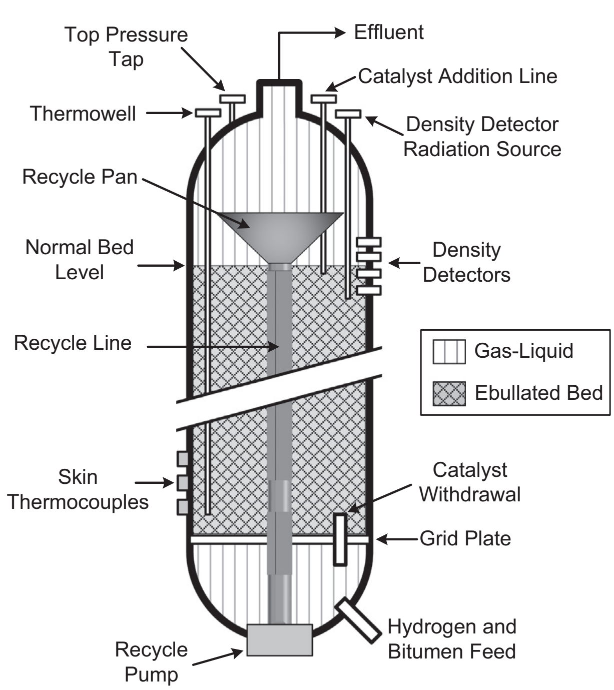

Fig. 1. LC-Finer^{SM} schematic (Pjontek et al., 2015).

2. Materials and methods

2.1. Experimental setup and procedure

Experiments were performed at ambient temperature and pressure in a clear polyvinyl chloride column with a maximum expanded bed height of 2.7 m and an inner diameter of 0.152 m, adequately large to minimize wall effects on overall phase holdups (Wilkinson et al., 1992). Gas was sparged in the plenum chamber of the column (i.e., below the distributor plate) via a sintered pipe with 10 μm diameter pores. The combined gas–liquid mixture then flowed through a perforated distributor plate with 16 holes of 4 mm diameter. A mesh placed on top of the distributor was used to prevent particles from entering the plenum chamber. At the top