Chemical Engineering Science 143 (2016) 240–255

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Experimental and simulated solids mixing and bubbling behavior in a scaled two-section two-zone fluidized bed reactor

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HIGHLIGHTS

- Several hydrodynamic models were evaluated for a double-sized TS-TZFBR.
- JHM and TFM models describe bubbles and solids motion well at the two scales.
- The CCBM model is unable to predict solids mixing at the largest reactor scale
- Slugging is lower and axial mixing is better in the bigger reactor.
- The defluidization phenomena are more significant at the double-sized TS-TZFBR.

ARTICLE INFO

Article history:
Received 10 June 2015
Received in revised form
11 December 2015
Accepted 23 December 2015
Available online 6 January 2016

Keywords:
Fluidized bed scaling
CFD: Two fluids model
Bubble size distribution
Solids mixing

ABSTRACT

Digital Image Analysis techniques, a phenomenological Counter-Current Back-Mixing model (CCBM) and Two-Fluid Model (TFM) simulations were employed to evaluate the effect of scale on the Two-Section Two-Zone Fluidized Bed Reactors (TS-TZFBR) fluid dynamics, i.e. bubble characteristics, axial mixing of solids and defluidization phenomena. The reactor scaling did not affect the quality of the TFM bubble size predictions. A bubble size correlation previously proposed by the authors for TS-TZFBR units was able to predict the experimental axial bubble size evolution at the different reactor scales and gas velocities (u_{gas}/u_{mf}=1.5–3.0) with a relative error under 17%. The TFM simulated axial solid mass fluxes were same order as these obtained by Particle Image Velocimetry for every reactor size. However, the classical CCBM model was unable to predict the effect of scale on the solids axial mixing in a TS-TZFBR. The inclination angle of the defluidized bed regions found within the TS-TZFBR tapered zone, β, increased by $(u_{gas}/u_{mf})^{0.25}$ when duplicating the reactor size. Nevertheless, it did not exceed the prescribed upper limit of $\beta=80°$ for any of the conditions tested.

1. Introduction

Process intensification in chemical engineering aims to optimize safety, profits and capital or energy costs by reducing the chemical plant size (Stankiewicz and Moulijn, 2000). In this context, the Two-Zone Fluidized Bed Reactor (TZFBR) represents a potential tool for process intensification in the field of heterogeneous catalysis since its design provides two simultaneous catalytic processes in one single apparatus. The TZFBR technology has been proven to be effective at lab-scale for carrying out gas–solid catalytic reactions where the catalyst suffers from fast deactivation due to coke deposition and those catalytic oxidations where the catalyst can be used as oxygen carrier (Herguido et al., 2005).

The TZFBR reactor design has two separated gas inlets to the fluidized bed: an immersed gas inlet for the gaseous hydrocarbon reactant and a gas distributor at the bottom of the bed for the oxidizing agent. Heterogeneous catalytic reactions take place in the upper region of the TZFBR whereas catalyst regeneration takes place below the reactant injection point (Fig. 1a). This process integration is possible owing to the catalyst axial mixing, which is typical of fluidized beds. Under certain process conditions, the continuous coke burning within the lower bed zone in the presence of an oxidizer, i.e. diluted oxygen, may result in a long term steady state reactant conversion without net catalytic deactivation. The performance of the TZFBR has been successfully tested for light alkane dehydrogenations, ethanol steam reforming or methane aromatization

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http://dx.doi.org/10.1016/j.ces.2015.12.025
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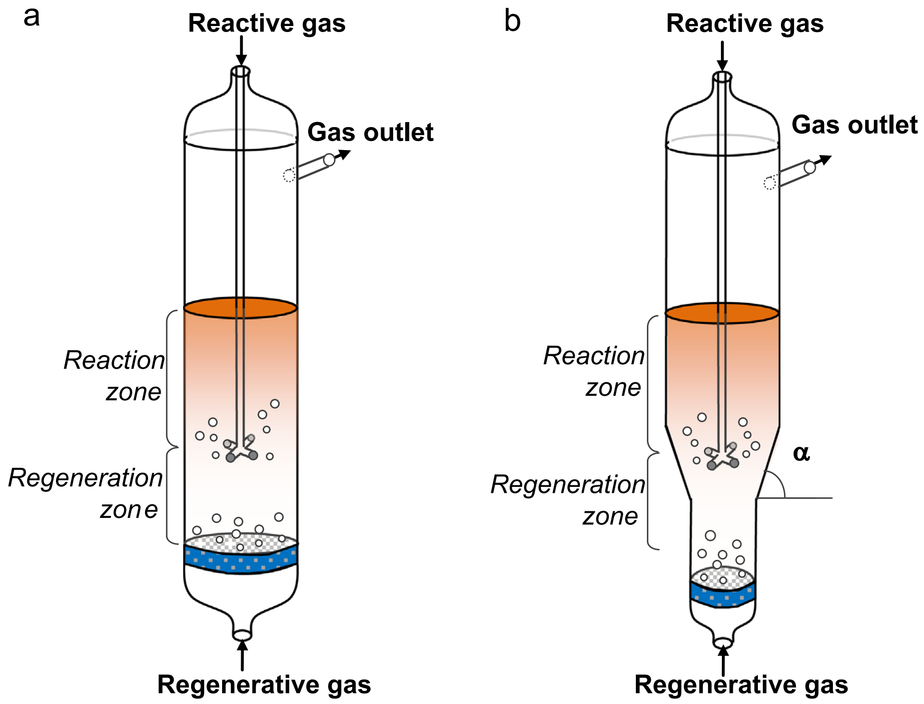

Fig. 1. (a) Scheme of a 3D TZFBR, (b) Scheme of a 3D TS-TZFBR.

(Medrano et al., 2013; Pérez-Moreno et al., 2012; Gimeno et al., 2010). The required residence time for particles in the two bed zones depends on the process variables such as catalyst type, reactor temperature, hydrocarbon conversion or coke burning kinetics and needs to be optimized for each process individually. In any case, the characteristic catalyst deactivation that takes place in the above mentioned processes becomes important after several minutes on stream (Froment, 2008; Lobera et al., 2008; Pérez-Moreno et al., 2013). Therefore, the solids residence time needs to be adapted accordingly, modifying either the gas velocity or the relative height of each bed zone.

Regarding energy consumption and the heat transfer potentials of fluidized beds, the heat generated by the catalyst oxidation in the lower TZFBR zone is used to perform the endothermic hydrocarbon conversion in the upper zone, thus, decreasing heat consumption in the reactor unit towards an autothermal regime. In contrast, a downstream separation unit may be required to remove the CO_x contents of the exhaust gas as a result of the in-situ coke burning.

Two critical issues affect the satisfactory TZFBR performance: the oxygen flow must be consumed in the lower bed zone before reaching the upper region and the hydrocarbon back-mixing, which is related to the mixing of solids, must be avoided. The first issue implies the need to optimize and carefully select the most suitable oxygen flow for each reaction condition. The second requires the control of the fluid dynamic regime in each bed region separately. On this regard, novel TZFBR configurations that incorporate two zones with different cross sectional areas, i.e. Two-Section TZFBR (TS-TZFBR), were proposed in a previous work (Julián et al., 2012). These improved reactor configurations allow the use of low regenerative-to-reactive gas flow ratios if required. Their main feature is the use of a tapered transition region between the two bed zones (Fig. 1b).

A comprehensive effort has been made to understand and characterize the lab scale TS-TZFBR bed hydrodynamics. Our previous works focused mainly on the study and prediction of the bubble size distribution along the vertical position of the bed (Julián et al., 2014a), on the experimental particle mixing between bed zones and the detection of defluidized regions (Julián et al., 2013), on the validation of a CFD tool to simulate the experimental hydrodynamic behavior (Julián et al., 2014b) and on the use of advanced imaging techniques (Particle Image Velocimetry, PIV, and Digital Image Analysis, DIA) to study the dense phase motion in detail within *pseudo-2D TS-TZFBR* configurations (Fig. 1b) without and with a horizontal tube bank (Julián et al., 2015) . Although the quantitative extrapolation of the phenomena observed in the 2D beds to real 3D fluidized beds is in general not trivial, the obtained results help to gain a better understanding and to validate numerical models, with which 3D beds can be investigated.

The motivation to develop a novel bubble size correlation for a TS-TZFBR arose from the lack of correlations in the existing literature that take into account both bed section enlargement and additional gas feed, simultaneously. Experimental findings from cold pseudo-2D measurements, which resulted in the so called JHM model (Julián et al., 2014a), were then used to validate Two-Fluid Model (TFM) simulations for different TS-TZFBR configurations (Julián et al., 2014b).

Analogously, the motivation to perform solids mixing studies was to find an operational window for some TS-TZFBR variables (superficial gas velocity, tapered section angle and immersed distributor location) that would allow a certain axial mixing rate between bed zones. Experimental mixing tests were performed using phosphorescent particles as optical tracers. The degree of axial mixing of solids was determined by means of transient tracer concentration profiles. Experimental results were compared to the axial mixing predictions of the two-phase Counter Current Back Mixing model (CCBM). Model parameters, viz. wake velocity (u_1), wake-emulsion mass transfer coefficient (K_w) and wake fraction (f_1), were estimated from classical hydrodynamic correlations (Julián et al., 2013).

Lastly, PIV/DIA studies allowed the motion of solids to be measured inside a pseudo-2D TS-TZFBR (Julián et al., 2012, 2015). Applying image processing algorithms to the high-resolution