CHEMICAL ENGINEERING RESEARCH AND DESIGN 93 (2015) 213–223

Contents lists available at ScienceDirect

Chemical Engineering Research and Design

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

Interphase mass transfer of the high velocity bubbling fluidization regime

*Jean Saayman**, *Willie Nicol*

Department of Chemical Engineering, University of Pretoria – Main Campus, Corner Lynwood Rd & Roper St, Hatfield, Pretoria 0002, South Africa

ABSTRACT

Gas–solid fluidization experiments were performed in two separate experimental setups with similar dimensions. Fast X-ray tomography (XRT) was used in setup 1, while ozone decomposition experiments were performed in setup 2. Packing and operation characteristics for the two setups were close to identical. The hydrodynamic measurements from the XRT acquisitions were used to evaluate the interphase mass transfer characteristics obtained from the ozone decomposition results. Superficial velocities (U_0) spanning the bubbling up to the onset of the turbulent regime (U_c) were employed. Traditional specific interphase mass transfer (k_{be}) correlations are based on incipiently fluidized beds; however, results suggested that a distinction should be made between the low-interaction bubbling regime and the high-interaction bubbling regime. A change in mass transfer behaviour occurred around a U_0/U_c value of 0.25. An empirical correlation for k_{be} of the high-interaction bubbling regime is proposed. The correlation gave the best fit for the entire velocity range with an average error of 8%, although it is not recommended for $U_0/U_c < 0.17$. It was observed that the classical approach of penetration theory for interphase mass transfer, performed exceptionally well at low velocities ($U_0/U_c < 0.34$).

Keywords: Fluidization hydrodynamics; Ozone decomposition reaction; X-ray tomography; Interphase mass transfer

1. Introduction

Catalytic gas–solid fluidized bed reactors (FBRs) have been studied and used for over six decades. From novel laboratory demonstrations (Saayman and Nicol, 2011) to performing nanoparticle coatings (van Ommen et al., 2012), to being at the heart of large petrochemical companies (Duvenhage and Shingles, 2002; Steynberg et al., 1999), these reactors have many uses. From an engineering point of view, advantages include: efficient solids mixing, good gas–solid contacting and low pressure drop. A wealth of understanding of the hydrodynamics of FBRs and their effects on reactor performance has been gained, although there are numerous areas where fundamental understanding is lacking; examples include modelling, the applicability of two phase theory and the accuracy of interphase mass transfer correlation for different regimes. Many studies mainly focus on either a specific hydrodynamic parameter or the reactor performance. Few studies have followed an integrated approach, which creates difficulties in modelling an FBR. Depending on the operating velocity (U_0) several regimes exist in FBRs, most commonly used being the bubbling, turbulent or fast fluidization regimes. Each regime is characterized by its own hydrodynamic behaviours. The bubbling and fast fluidization regimes have enjoyed much academic attention due to the distinctness of the bubbles and the core annulus, respectively. The turbulent regime has better gas–solids contacting than the bubbling regime without the high solids circulation of the fast fluidization regime. These reasons make the turbulent regime a popular choice for industry. Commercial examples of turbulent reactors include FCC regenerators, zinc sulphide roasters and Mobil MTG, acrylonitrile, maleic anhydride, phthalic anhydride and ethylene dichloride reactors. Despite the turbulent regime being popular in industry, it has not received as much attention as the bubbling or fast fluidization regimes (Bi et al., 2000).

* Corresponding author.
E-mail address: jeansaayman@gmail.com (J. Saayman).

Available online 27 June 2014
http://dx.doi.org/10.1016/j.cherd.2014.06.023

Nomenclature

$A_{b(Cyl)}$	external surface area of cylindrical bubble [m^2]
A_{bed}	cross sectional area of the reactor [m^2]
a_I	inter-phase transfer surface [m^{-1}]
$C_{i,B}$	concentration in bubble [mol/m^3]
$\underline{C}_{i,E}$	concentration in emulsion [mol/m^3]
C_i	gas concentration of species I [kmol/m^3]
D_b	spherical-volume equivalent bubble diameter [m]
D_m	gas diffusion coefficient [m^2/s]
$d_{b(Cyl)}$	base diameter of cylindrical bubble [m]
d_p	Sauter mean particle diameter [m]
K_0	overall interphase mass transfer (catalyst volume based) [s^{-1}]
L_b	length/height of voids [m]
k_{be}	specific interphase mass transfer (bubble to emulsion) [m/s]
k_R	reaction rate constant (catalyst volume based) [s^{-1}]
Q	volumetric flow rate in test reactor [m^3/s]
R_i	reaction rate as a function of concentration [s^{-1}]
Sc	Schmidt number ($\mu/(\rho_g \cdot D_m)$) [–]
U_b	average bubble velocity, relative to distributor [m/s]
u_B	bubble phase reactor model gas velocity [m/s]
U_{br}	terminal rise velocity of a single bubble [m/s]
U_c	onset of turbulent regime velocity [m/s]
u_E	emulsion phase reactor model gas velocity [m/s]
U_{mf}	minimum fluidization velocity [m/s]
U_0	operating velocity [m/s]
V_b	volume of bubble/void [m^3]
W	solids volume of catalyst [m^3]
X	conversion [–]

Greek letters

β	Empirical mass transfer parameter ($\psi_B/(\Phi_0 U_0)$) [s/m]
ε_{mf}	gas volume fraction at minimum fluidization [–]
ρ_b	bulk density [kg/m^3]
ρ_p	particle density [kg/m^3]
ρ_g	gas density [kg/m^3]
μ_g	gas viscosity [Pa s]
σ_i	standard deviation of incoherence [Pa]
Φ_0	solids volume fraction ($1-\varepsilon$) [–]
Φ_{mf}	solids fraction of incipiently fluidized bed [–]
ψ_B	bubble phase volume fraction [–]

Based on observations of incipiently fluidized bubbling beds, the need for hydrodynamic descriptions of two-phase behaviour arose. The earliest well-known published works on the matter were those of Rowe et al. (1962), Rowe and Partrige (1963), Davidson and Harrison (1966,1963) and Lockett et al. (1967). The concept was developed further and gas exchange between the phases was explored (Stephens et al., 1967; Godard and Richardson, 1968; Drinkenburg and Rietema, 1972; Chavarie and Grace, 1975a; Sit and Grace, 1978, 1981). Ultimately, leaders in the field such as Kunii, Levenspiel and Grace proposed reactor models based on the theory (Chavarie and Grace, 1975b,c,d; Grace, 1984; Kunii and Levenspiel, 1990a,b). Generally, these reactor models and the two-phase theory best describe the hydrodynamic behaviour of bubbling fluidized beds (Abba et al., 2003; Chaouki et al., 1999; Thompson et al., 1999; Jafari et al., 2004). The theory entails that most of the gas reagents are contained in a lean, solids/catalyst-deprived phase that bubbles though a dense, solids-rich (emulsion) phase. This closely resembles the physical phenomena in the FBR. Since most of the gas throughput is present in the lean phase, the movement of gas into and out of the emulsion phase often dictates the performance of an FBR. Therefore the description of the interphase mass transfer becomes one of the crucial modelling variables. Most correlations for this transfer are derived on the basis of low-velocity/interaction bubbling regime behaviour with small U_0/U_c values of 0.02, where U_c is the onset velocity of the turbulent regime. In this low U_0/U_c regime the bubbles have near-ideal geometries and low interactions with each other. Despite the success of these models at lower velocities, the transfer correlations are not suited for higher velocity operations (Thompson et al., 1999; Wu and Agarwal, 2003; Sun and Grace, 1990; Campos et al., 1998).

Few attempts have been made to adapt interphase mass transfer correlations for the higher velocity bubbling regime or turbulent regime (Foka et al., 1996). Studies have been conducted where the performance of existing mass transfer correlation have been investigated. Using inert tracer-gas experiments Wu and Agarwal (2003) looked at the effects of temperature in a fluidized bed. This study was done in a 127 mm ID column with a bed of particles which was incipiently fluidized using pure nitrogen gas, while single argon gas bubbles were injected near the distributor at the bottom of the bed. These argon containing bubbles travelled upwards to the top of the bed where a tube extracted a sample of the bubble's gas as the bubble passed the tube. Experiments were conducted using particles from 264 μm to 463 μm and at temperatures of 298 K, 423 K, 573 K and 773 K. The Sit and Grace (1981) and Davidson and Harrison (1963) correlations were tested. Wu and Agarwal (2003) found that the Sit and Grace correlation performed better, but in some cases was not ideal. They incorporated a correction factor for the convection term.

When it comes to reactions in a freely bubbling bed, concentration gradients steepen due to reaction in the cloud phase and bubble interaction starts playing a big role. Thompson et al. (1999) used the data of Sun and Grace (1990) in a new transitional two-phase model. They had to incorporate a correction factor to the Sit and Grace (1981) correlation to fit the data. This might be due to the effect of the probabilistic transition factor present in the model. Campos et al. (1998) performed reactor performance experiments in a coke combustor at 1223 K and inferred mass transfer using two-phase theory. The researchers tested the overall mass transfer correlation of Kunii and Levenspiel and found that the correlations far over-predicted mass transfer. Although, Kunii and Levenspiel warn that the overall mass transfer correlation, which is based on a two-step mass transfer process and a three-phase model, cannot be reduced to a two-phase model for reacting systems (Kunii and Levenspiel, 1991). The book of Kunii and Levenspiel (1991) is also recommended for a general overview of the field of fluidization.

An integrated approach combining hydrodynamics and reactor performance is followed in this study with the focus on the upper end of the bubbling regime and the start of the turbulent regime. The aim is to investigate which theories in the literature are applicable and which do not hold in a