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Review

Developments in the understanding of gas–solid contact efficiency in the circulating fluidized bed riser reactor: A review ☆

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ABSTRACT

In the last several decades, circulating fluidized bed reactors have been studied in many aspects including hydrodynamics, heat and mass transfer and gas–solid two phase contacting. However, despite the abundance of review papers on hydrodynamics, there is no summary paper on gas–solid contact efficiency to date, especially on high density circulating fluidized beds (CFBs). This paper gives an introduction to, and a review of the measurement of contact efficiency in circulating fluidized bed riser. Firstly, the popular testing method of contact efficiency including the method of heating transfer experiment and hot model reaction are discussed, then previous published papers are reviewed based on the discussed methods. Some key results of the experimental work are described and discussed. Gas–solid contact efficiency is affected by the operating conditions as well as the particle size distribution. The result of the contact efficiency shows that the CFB riser is far away from an ideal plug flow reactor due to the characteristics of hydrodynamics in the riser. Lacunae in the available literature have been delineated and recommendations have been made for further work.

1. Introduction

Fluidization is a process which involves the flow of solid particles in contact with liquid, gas, or both gas–liquid flow. Regimes for gas–solid fluidization may include particulate, bubbling, slugging (abnormal state), turbulent, fast fluidization and pneumatic transport [1,2]. In a fast fluidized bed, solids must be continuously fed into the bed (the riser) at or near the bottom and the entrained solids by the high velocity gas flow are captured at the top and sent back to the bottom of the riser through a recirculation system outside of the riser to maintain an "uninterrupted" solid circulation. Therefore, the fast fluidization and the corresponding fast fluidized bed was referred to as "continuous fluidization" and "circulating fluidized bed", respectively [3,4]. Benefits of circulating fluidized bed reactors include significantly reduced gas and solid backmixing, improved contact efficiency, and continuous process coupled with higher product capability [3–5].

Industrial applications of circulating fluidized beds (CFBs) started in the 1950s [6], and rapidly expanded in the last five decades. It is clear to see that two main waves of tremendous developments of CFBs associated with fluid catalytic cracking (FCC) and circulating fluidized bed combustion (CFBC). As discussed by Zhu and Bi [7], there are many distinctive differences in the operating conditions of these two industrial processes. Some key differences are listed in Table 1. It shows that typical FCC units operate at a gas velocity ranging from 6 to 28 $m\cdot s^{-1}$ and a solid circulation rate (G_s) of 400–1200 $kg\cdot m^{-2}\cdot s^{-1}$, while typical CFBC reactors operate at lower gas velocity from 5 to 9 $m\cdot s^{-1}$ and much lower G_s from 10 to 100 $kg\cdot m^{-2}\cdot s^{-1}$, resulting in much lower solid hold-up in CFBC reactors. Moreover, bed geometry, solid inventory and solid feeding device are also significantly different between FCC and CFBC.

Table 1
Typical operating conditions of FCC and CFBC units [7]

Operating conditions	FCC	CFBC
Particle density/$kg\cdot m^{-3}$	1100–1700	1800–2600
Particle mean size/mm	0.04–0.08	0.1–0.3
superficial gas velocity/$m\cdot s^{-1}$	6–28	5–9
Net solid flux/$kg\cdot m^{-2}\cdot s^{-1}$	400–1200	10–100
Solid holdup	3%–15%	<1%
Average solid residence time/s	2–4	2–4
Cross-section geometry	Circular	Rectangular or square
Reactor diameter/ m	0.7–1.5	4–8 equivalent
Height to diameter ratio	>20	<5–10
Solid inventory	High	Low
Solid exit structure	Smooth	Abrupt
Solid feeding device	Mechanical valves	Non-mechanical valves

In reviewing the research history, the earlier reported studies throughout the 1970s to early 1990s almost entirely focused on hydrodynamics of CFBs operating with low solid circulation rate of

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$100\ \mathrm{kg \cdot m^{-2} \cdot s^{-1}}$ concentrating on operating conditions used for CFB coal combustion while little research was published for the FCC riser except for an earlier report from Shell [8]. Previous experiments have clearly demonstrated that a CFB reactor with low solid flux is hydrodynamically characterized by an extremely non-uniform flow structure, with a dense bottom region and dilute upper region in the axial direction [9,10] and a 'core-annulus' flow structure in the radial direction [2,11]. This non-uniform flow structure and the relatively dilute solid concentration (usually less than 10%) result in many disadvantages, such as serious gas by-passing through the core dilute region and extensive backmixing of solids in the wall region, consequently resulting in lower gross gas–solid contact efficiency and poor selectivity of chemical reactions [12,13]. Also, there are a reduction in heat transfer coefficients between heat transfer surfaces and suspension and somewhat greater temperature gradient than in dense beds [4,11]. These limitations as suggested by the previous researchers greatly affect the application of CFBs' to processes with slow reaction rates or requiring high heat transfer rates.

To raise the attention of the CFB working community, Bi and Zhu [14] proposed the concept of HDCFB/HFCFB (high density/flux circulating fluidized bed) in contrast to LDCFB/LFCFB (low density/flux circulating fluidized bed). It was pointed out that although many commercial CFB setups are operating under high density conditions such as FCC, majority of the fundamental studies had been carried out in relatively low density circulating fluidized beds (LDCFB) and there had been little reported fundamental effort on the high density circulating fluidized bed (HDCFB). Such high density includes both high flux and high hold-up with the criterion of solid circulation rate reaching $200\ \mathrm{kg \cdot m^{-2} \cdot s^{-1}}$ and the average solid holdup of 10% as the boundary to demarcate the two conditions [15,16].

Studies on HDCFB were first carried out in the late 1990s. Recently, high density circulating fluidized beds have become one of the focal points in the field of CFB systems. [10,16–18] Numerous studies under high solid flux have shown that the hydrodynamics are quite different in comparison with low flux and low density CFB risers operated with $G_s < 200\ \mathrm{kg \cdot m^{-2} \cdot s^{-1}}$ [15,19–25]. Fig. 1 shows a typical flow structure in high density circulating fluidized bed. Some key findings include:

(1) In the axial direction, the average solid holdup for HDCFB is up to 10%–30% with the densest solid holdup up to almost 40% at the bottom section of the riser, much higher than LDCFB with solid holdup usually lower than 1% in the developed region. In addition, a fairly uniform axial profiles of solid holdup is achieved across the whole bed under extremely high solid circulation rate of $1000\ \mathrm{kg \cdot m^{-2} \cdot s^{-1}}$, at higher solid holdup.

(2) In the radial direction, local solid concentration under HDCFB is also much higher than that under LDCFB. As shown in Fig. 1, the radial solid distribution becomes steeper with higher flux. Work by other researchers [27] also showed that the downflow of particles in the wall region almost disappeared under high density conditions leading to a reduction of axial gas dispersion under HDCFB. Based on these current studies, it can be concluded that in HDCFB reactors, more favorite hydrodynamics with high solid flow rates and more uniform dense suspension along the axial direction may result in improved reactor performance with better gas–solid contacting efficiency and higher conversion per unit volume and will be very useful for applications requiring higher solid/gas feed ratios and uniform solids and gas residence time, and processes where high gas–solid contacting efficiency is crucial [28,29]. The contact efficiency between gas and solids is closely related to the hydrodynamics and mass and heat transfer behaviors in the CFB reactor, and it has a significant influence on the overall system performance.

Until now, most of the research concentrate on hydrodynamics as summarized by several authors. While several hundreds of papers have been published on the hydrodynamics of fluidized beds, fewer results (all summarized in Table 2) have been reported on the gas–solid contacting. The contact efficiency of gas–solid in a circulating fluidized bed is not well understood, partially due to measurement difficulties and the lack of a rigorous definition for the contact efficiency. A clear understanding of the gas–solid contacting will help in the design of CFB reactors. This paper attempts to present efforts made in developing fundamental understanding of gas–solid contact efficiency in CFB reactors, especially in HDCFB reactors.

2. Experimental Testing Methods

In a general sense, contact efficiency is related to gas bypassing due to the nonuniform structure especially the nonuniform flow structure in the radial direction in both LDCFB and HDCFB reactors. As described by Dry *et al.* [12], in a CFB reactor, not all the gas fed into the reactor would come into close contact with the solids: some of the reactant gas would experience intimate contact with the dispersed particles, and some may emerge at the outlet of the reactor without having made substantial contact with any solids at all. Of the gas which does come into close contact with the solids, a fraction would be converted and the rest would exhaust unchanged. The contact efficiency then can be defined based on the conversion of a special species under carefully chosen conditions. Gas-phase conversion measured across the reactor would reflect a combination of hydrodynamic and reaction behaviors. The technique developed so far for measuring the contact efficiency can be divided into two categories: the indirect method using gas–solid heat transfer testing and the direct method using catalytic ozone decomposition as a model reaction, as shown in Table 2.

The former is a reasonable way because heat transfer is largely controlled by the gas and solid contacting behavior. The results of heat transfer between gas and solids can be used to characterize the gas–solid contacting efficiency and the mass transfer performance. Therefore, earlier studies on contact efficiency were almost all conducted using heat loss from a pulse of hot gas [12,34,38]. Typical apparatus and the testing probe is schematized in Fig. 2 and detailed information can be found in the related papers.

The latter is more attractive because it employs real chemical reaction to evaluate the reactant conversion in CFBs and the chemical reaction itself can supply direct information on reactor performance. A carefully chosen chemical reaction is the key success for this method. Because of its simplicity in reaction kinetics (very close to first-order reaction), negligible heat effect of reaction due to the low concentrations involved, and the availability of a simple and accurate measurement method, a heterogeneous catalytic reaction—ozone decomposition catalyzed by ferric oxide—was very often employed as a model reaction to investigate reaction coupled with mass transfer in fluidized bed reactors [30,36]. A typical ozone generation and measurement system is shown in Fig. 3. The underlying theory was elaborated by Ouyang *et al.* [45] and Li *et al.* [41].

3. Contact Efficiency by Heat Transfer Measurement Method

3.1. Definition of gas–solid contact efficiency

For the heat transfer method, gas temperatures were usually measured by a rapid-response thermocouple. Assuming that the heat loss to the CFB wall can be negligible, an energy balance across a given section of the reactor gives:

$$\text{Heat loss from the gas stream} = \text{Heat transferred to the solids phase} \quad (1)$$

$$c_{pg}\rho_g U_g A(T_{g,1} - T_{g,2}) = h_{gs} a(\overline{T_g} - \overline{T_p}) A \Delta Z \quad (2)$$

where c_{pg} is the specific heat of gas, ρ_g is the gas density, A is the cross-section area of the bed, $T_{g,1}$ is the gas temperature at the top of a given riser and $T_{g,2}$ is the gas temperature at the bottom of a given bed section,