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## Residence times in fluidized beds with secondary gas injection

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#### Abstract

Experiments were carried out to determine the effects of secondary gas injection on the gas residence time and macromixing characteristics in a bubbling fluidized bed. Primary gas is introduced via a bottom distributor plate, while secondary gas is introduced via a fractal injector submerged in the bed. Results indicate that the average residence time decreases only slightly. Calculated overall reactor Péclet numbers indicate that the gas experiences less back-mixing with secondary gas injection. The bubble size was observed to decrease by up to 70%, indicating improved gas– solid contact. Taking this improved contact and plug flow behavior into account, the conversion in a fluidized bed with secondary gas injection is expected to increase significantly, particularly for mass-transfer limited reactions. © 2007 Elsevier B.V. All rights reserved.

Keywords: Secondary injection; Fluidized bed; Residence time distribution; Fractal injector; Experimental method

#### 1. Introduction

For several applications of fluidized beds it can be useful to manipulate the structure, and thereby introduce additional degrees of freedom, in order to intensify the process [\[1\].](#page--1-0) The secondary injection of gas directly inside the fluidized bed by using a fractal injector (right-hand side of [Fig. 2\)](#page--1-0) can significantly reduce the bubble size and decrease the rate of bubble coalescence [\[2,3\]](#page--1-0). This decrease in bubble size should increase the rate of gas exchange between the bubbles and the surrounding solids suspension, thus increasing conversion for a mass-transfer limited reaction. Previous experiments indicate that the total volume of gas present in bubbles is reduced as well, implying that there must be more gas in the dense phase, which results in a better gas–solid contact [\[2,3\]](#page--1-0). Injection of gas into a fluidized bed at locations throughout the reactor, instead of through the windbox alone, can also be used to effectively improve reaction selectivity and to maintain high reactant feed rates while still avoiding possible explosion limits [\[4,5\]](#page--1-0). All of these effects contribute to the intensification of fluidized bed processes.

The addition of gas higher in the bed affects the micro- and macro-mixing behavior in the fluidized bed. Around each injection point of the fractal injector (see [Fig. 2](#page--1-0)) there is increased

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micromixing due to the additional flow, which can improve the local gas–solid contact [\[6\].](#page--1-0) On the macro-scale, the whole flow pattern in the bed may be altered due to the distribution of the gas injection. This is important because a decrease in back mixing will increase the concentration gradient of a reactant, which is the driving force for a chemical reaction (with positive order kinetics), resulting in a higher conversion. Although the gas–solid contact time is needed to predict conversions in heterogeneously catalyzed systems [\[7,8\],](#page--1-0) the gas residence time distribution (RTD) is still useful to determine the gas holdup and the macromixing characteristics of the fluidized bed [\[9,10\].](#page--1-0) The purpose of the current study is to determine what effects secondary gas injection via a fractal injector has on the gas holdup (i.e. residence time) and the macromixing characteristics of a bubbling fluidized bed. The approach taken here is to measure the residence time distributions in such a system using pulseresponse experiments with an inert tracer at different primary to secondary gas flow ratios and at different total gas flow rates. The RTDs are then used to determine the average residence time and to calculate the Péclet numbers, which are a measure of the amount of macromixing (axial dispersion) in the system.

#### 2. Background

What effects do we expect to find with secondary gas injection on the residence time and mixing in the system? First,

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consider the effect of secondary gas injection on the average residence time,  $\tau$ , of the gas. Specifically consider the comparison of two identical fluidized beds with the same total flow rate, but one that has a portion of the total flow introduced higher in the bed. At first thought,  $\tau$  might be expected to decrease since gas is being injected higher in the reactor. However, the fact that the gas at the bottom of the reactor travels more slowly (as compared to the bed without secondary injection) cancels out the reduced average residence time of the fresh gas that is injected further up. This can be shown through a simple derivation adapted from Coppens [\[6\]](#page--1-0) (Fig. 1).

For a theoretical tank with one input and one output, the average residence time is defined as the volume of the tank divided by the total volumetric flow rate (assuming no volume change due to reaction),

$$
\tau = \frac{V}{Q} \tag{1}
$$

With one secondary injection height, the reactor is split into two volumes,  $V_1+V_2=V$ , with a fraction p of the total flow going towards the windbox, and a fraction  $(1-p)$  being injected as secondary gas. Therefore, in this simple model of secondary injection, the volumetric flow rate into  $V_1$  is  $pQ$ , and the flow rate into  $V_2$  is  $pQ + (1-p)Q = Q$ . Since the primary gas must go through both volumes, the average residence time of the primary gas is calculated as the sum of the residence times for each tank:

$$
\tau_{\text{primary}} = \frac{V_1}{pQ} + \frac{V_2}{Q} \tag{2}
$$

Similarly, the average residence time of the secondary gas is:

$$
\tau_{\text{secondary}} = \frac{V_2}{Q} \tag{3}
$$

To obtain the overall average residence time we must use a weighted average of  $\tau_{\text{primary}}$  and  $\tau_{\text{secondary}}$  according to the fractions of the total gas that go through the primary and secondary flows. Thus, the total average residence time is:

$$
\tau_{\text{total}} = p \tau_{\text{primary}} + (1-p) \tau_{\text{secondary}} = \frac{V_1 + V_2}{Q} = \frac{V}{Q} \tag{4}
$$

This tanks-in-series model shows that the average residence time should not change at all. Strictly speaking, the above derivation is only valid for systems that behave ideally with uniform, additive flow. It does not account for possible deviations, such as bypassing (or stagnation) where some fluid elements travel at significantly different velocities than others (e.g. bubbles typically travel faster than the gas in the dense phase), and it assumes that there is no back-flow of gas from  $V_2$  to  $V_1$ . Previous research [\[2,3\]](#page--1-0) has shown that secondary gas injection reduces the size of the bubbles. As a result, the rise velocity of the bubbles decreases and the rate of gas bypassing drops. Thus, it is reasonable to expect that the average residence time should not change.



Fig. 1. Simplistic model for the average residence time with secondary injection. (a) Case with no secondary injection; (b) case with secondary injection.

Interestingly, Al-Sherehy et al. [\[4\]](#page--1-0) found that the addition of secondary gas as a jet (at velocities high enough for the jet to span the diameter of the column) with constant primary flow had only a very small negative effect on the residence time of the gas, even though their total gas flow was increasing with jet velocity in their experiments. In the current experiments, the secondary gas is distributed across the width of the reactor due to the uniform (albeit discrete) placement of the injection points on the fractal injector, and we maintain our total flow rate constant. Therefore, little or no negative effect on the residence time of the gas is expected.

Now consider how the macromixing of the gas in a fluidized bed with secondary injection could be affected. Since the bubbles are smaller (and thus travel with a slower velocity) and they have a lower rate of coalescence, the amount of mixing in the bed is expected to decrease. This decrease is because the bubbles themselves are a major driving force for the gas (and solids) mixing. Vigorously bubbling beds will have much more dispersion than beds where the bubble behavior is more controlled. Another effect is the nature of the secondary injection device. In this work a fractal injector with four levels of injection points is used. This way to inject gas can promote gas staging without the use of baffles [\[6\]](#page--1-0). Gas staging is beneficial because it causes the bed to be broken up into smaller mixing cells, and, as the number of cells increases, the overall flow behavior approaches that of plug flow. Controlling the bubble behavior and inducing gas staging are two reasons why we can expect secondary gas injection to shift the behavior of the gas closer to the ideal of plug flow.

### 3. Residence time theory

Gas residence times are frequently determined by using tracers that follow the flow. A complicating factor with secondary gas injection is that there are multiple flows. To determine the overall RTD of the gas, the residence times of both the primary and the secondary gas must be combined. Several researchers have developed a theory for residence times

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