A small-scale regularly packed circulating fluidized bed Part II: Mass transfer

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(Received April 26, 1993; revised November 29, 1993)

Abstract

The underlying objective of the present study is to increase gas–solids contact in a circulating fluidized bed by the introduction of obstacles in the riser portion. The presence of such obstacles leads to suppression of radial inhomogeneities in the solids mass flux and concentration, and break-up of solids clusters. At ambient conditions, gas–solids mass transfer was investigated for cocurrent upward flow of air and microsize solid particles (FCC, 70 μm diameter) over a regularly structured inert packing introduced into the riser part of a circulating fluidized bed unit. The packed section has a height of 0.48 m, a cross-sectional area of $0.06 \times 0.06 \text{ m}^2$, and contains regularly stacked 0.01 m diameter Perspex bars as the obstacles meant to enhance the gas–solids contact. Gas mass fluxes used were 1.4 and $2.7 \text{ kg m}^{-2} \text{ s}^{-1}$. Solids mass fluxes were varied in the range $0 \leq G_s \leq 12 \text{ kg m}^{-2} \text{ s}^{-1}$. Experimental mass transfer data were obtained by applying the method of adsorption of naphthalene vapor on FCC particles. A conservative estimate of the apparent gas–solids mass transfer coefficient k_g^* could be derived from the naphthalene vapor concentration profile along the packed section on the basis of a plug-flow-model interpretation, while assuming single-particle behaviour and neglecting intraparticle diffusion effects. Such k_g^* values appear to increase with increasing gas mass flux, but decrease with increasing solids mass flux (and consequently increasing solids volume fraction) probably due to the corresponding increase in particle shielding. Comparison of the present results with available literature data for similar solid materials suggests that the effect of the packing inserted into the CFB is significant: the Sherwood numbers derived from the present study are relatively high.

Introduction

The circulating fluidized bed (CFB) as a chemical reactor has some unique features, e.g. large gas throughput, essentially plug flow of gases and excellent inter-phase heat and mass transfer [1]. It is used industrially for high temperature, non-catalytic processes such as coal combustion, calcination, SO_2 absorption, and catalytic processes such as catalytic cracking and Fischer–Tropsch reactions. Due to the short gas-phase residence time, the CFB reactor is only applicable to fast reactions, such as high temperature desulfurization of gasifier product-gas with a regenerable sorbent, which is the main subject of a research project at the University of Twente as mentioned in Part I [2].

The gas–solids contact efficiency as reported by Dry *et al.* [3, 4] and White and Dry [5] for a small-scale CFB suggests that gas–solids contact is far from complete. This was attributed to poor distribution of gas and solids in the reactor, i.e. core–annulus flow and the existence of clusters or streamers. Results reported on the hydrodynamics of large industrial CFBs

by Saxton and Worley [6] and by Bartholomew and Casagrande [7] show an even larger degree of poor distribution.

The principle of the present investigation is that introduction of a regularly stacked packing suppresses radial inhomogeneities and therefore intensifies gas–solids contact. Other advantages and disadvantages of this packing are mentioned in Part I of this work, in which the hydrodynamic properties of a cocurrent gas–solids flow in a regularly packed circulating fluidized bed are discussed. The set-up used, similar to the present one, had a 0.48 m high square section of $0.06 \times 0.06 \text{ m}^2$ cross-sectional area which contained packing bars of 0.01 m diameter. Hydrodynamic results reported concern the pressure gradient, solids volume fraction and solids friction coefficient.

The present study (Part II) was carried out to determine the unknown gas–solids mass transfer in a regularly packed circulating fluidized bed (RPCFB) for FCC which is also the sorbent considered for the high temperature desulfurization process. Preliminary re-

sults, concerning gas–solids mass transfer, have already been reported in an earlier paper [8].

Previous work

Gas–solids mass transfer, and to a lesser extent heat transfer, is a relatively unexplored feature of circulating fluidized beds, and has not been investigated at all for a regularly packed circulating fluidized bed (RPCFB). Table 1 gives an overview of available previous work on gas–solids mass and heat transfer in circulating fluidized beds as well as in dilute cocurrent and countercurrent gas–solids contactors.

In general the following conclusions can be drawn from this summary:

- The experimental methods used are mainly based upon steady-state conditions.
- Fluid Cracking Catalyst appears to be a very uncommon solids material for laboratory experiments, despite its industrial importance. FCC has only been used by Dry *et al.* [3], all the other authors preferring to work with (much) coarser particles.
- A decreasing void fraction (increasing solids volume fraction β) has a negative influence on the (apparent) mass and heat transfer coefficient, probably due to an increase in particle shielding (Bandrowsky and Kaczmarzyk [9, 10], Kwauk *et al.* [11], Kato *et al.* [12] and Watanabe *et al.* [13]). This phenomenon is more pronounced for smaller particles.
- The obtained (apparent) mass and heat transfer coefficients approach asymptotically, at high void fractions and for relatively large particles ($> 300 \mu\text{m}$), the values predicted by the well-known correlation of Ranz and Marshall (Bandrowsky and Kaczmarzyk [9, 10] and Kato *et al.* [12]). The Ranz–Marshall correlation [14] describes the mass transfer between a single sphere and its surrounding undisturbed stagnant gas phase. Its analogue for heat transfer is derived after applying the Chilton–Colburn analogy [15].

In contrast to other literature, Dry *et al.* [3] defined the gas–solids contact efficiency as being the ratio of the amount of heat actually transferred from gas to solids to the amount needed to reach equilibrium between gas and solids. The experimental conditions applied were such that only heat transfer across the external gas film around particles was rate-controlling. While assuming that the average heat transfer coefficient can be calculated from the (analogue) Ranz–Marshall correlation (with slip velocity = terminal velocity), the value of the effective particle surface area was estimated by a simple steady-state heat transfer model. The surface efficiency was then defined as the ratio of the effective to the actual particle surface area. Dry *et al.* [3] did not take the axial voidage profile and its influence on

the local heat transfer into account. They assumed a constant heat transfer coefficient (independent of the solids volume fraction β) and used the average voidage to calculate the (average) effective particle surface area and the (average) surface efficiency.

This is in contrast with the method applied by the other authors listed in Table 1, who calculated the mass or heat transfer coefficient (and from that the Sherwood or Nusselt number) while assuming that the particle surface area is equal to $6/d_p$ and taking the local void fraction into account explicitly. Obviously, Dry *et al.* assume the validity of the Ranz–Marshall correlation for all experimental conditions applied, and calculate the effective particle surface area. However the other authors calculate the Sherwood or Nusselt number assuming a constant specific particle surface area. In the first case, the deviation from single particle behavior is attributed to a decrease in the particle surface area, whereas in the second case to a decrease in the heat or mass transfer coefficient and thus the Nusselt or Sherwood number.

Among the techniques mentioned in the above summary, the (semi) stationary ones should be preferred because (i) disturbances in the gas or solids flow rate as a result of tracer addition, are avoided and (ii) a fair amount of time is available to analyse the stationary tracer profile. Whether a mass or heat transfer method should be given preference is difficult to say. Both methods have their specific advantages and disadvantages. In a mass transfer method, the choice of tracer is free as long as it (i) adsorbs strongly and in large quantities onto the solid particles, (ii) diffuses rapidly into the particle (no intraparticle concentration profiles), (iii) does not adsorb significantly onto the construction material of the experimental set-up (i.e. adsorption onto the sample probe must be prevented) and (iv) can be detected easily and accurately at low concentration levels (ppm/ppb range). With respect to the last condition mentioned, it is known (Kwauk *et al.* [11]) that the concentration of halogen compounds can be measured down to the ppb range with the ECD (electron capture detection) method. In a heat transfer method, the tracer consists of heated gas or solids and the gas-phase temperature profile should be measured. These measurements require a specially constructed temperature probe which must not be influenced by the solids present. Other problems are (i) unavoidable heat leaks towards the wall, (ii) the possible presence of an intraparticle temperature profile (this can be avoided by selection of a solids material with high thermal diffusivity, small particle diameter and low porosity), (iii) the relatively small temperature range (1 to 2 decades) to operate in and (iv) the inaccuracy of temperature measurement, especially for small differences in temperature between gas and solids.

TABLE 1. Literature review of previous work on mass and heat transfer in a circulating fluidized bed and in dilute cocurrent and countercurrent gas-solids contactors

Authors	Set-up used	Applied method	Solids type and size	Reynolds number	Remarks
Kwauk <i>et al.</i> [11]	circulating fluidized bed, 0.024 m i.d., 1.5 m high	mass transfer (stationary), tracer: CCl_4	active carbon, $\bar{d}_p = 570 \mu\text{m}$	$Re \approx 38$	The apparent mass transfer coefficient varies from 0.02 to 0.07 m s^{-1} with decreasing solids volume fraction. Equilibrium is reached within 0.5 m from the entrance.
Dry <i>et al.</i> [3]	circulating fluidized bed, 0.09 m i.d., 7.2 m high	heat transfer (dynamic), tracer: heated gas pulse	Fluid Cracking Catalyst $\bar{d}_p = 71 \mu\text{m}$	$8 \leq Re \leq 18$	The surface efficiency, i.e. the ratio of the effective to actual particle surface area, varies from 0.01% to 0.13% and it is suggested that this may be related to the proportion of particles which actually behave as individuals. A typical value for a fluidized bed is 0.0035%.
White and Dry [5] Dry and White [4]	circulating fluidized bed, 0.09 m i.d., 7.2 m high	heat transfer (dynamic), tracer: heated gas pulse	sand, 140 and 275 μm	superficial gas velocity of 4, 6 and 8 m s^{-1}	The gas-solids contact efficiency improves with finer particles but this improvement is less than the corresponding increase in total (external) gas-solids surface area.
Watanabe <i>et al.</i> [13]	circulating fluidized bed, 0.021 m i.d., 1.8 m high	heat transfer (stationary), tracer: heated solid particles	glass beads: 177–250, 297–420, 420–590 μm ; activated alumina: 590–710 μm	$20 < Re < 200$	$0.4 < Nu < 5.1$
Bandrowsky and Kaczmarzyk [9, 10]	vertical pneumatic conveying 0.06 m i.d., 3.8 m high	heat transfer (stationary), tracer: heated gas	ceramic balls: 700–2600 μm	$180 < Re < 1800$	$Nu = 0.00114\bar{\beta}^{-0.5984} Re^{0.8159}$ with $0.00025 < \bar{\beta} < 0.05$
Kato <i>et al.</i> [12]	countercurrent flow in bed of open-end cylindrical screen packings	mass transfer (stationary), tracer: H_2O	activated alumina: 317–832 μm	$5 \leq Re \leq 150$	$Nu = 2.38 \times 10^{-3} Re^n \left(\frac{1-\bar{\beta}}{\bar{\beta}} \right)^{0.97}$ $n = 2.48 \left(\frac{1-\bar{\beta}}{\bar{\beta}} \right)^{-0.28}$; $0.001 < \bar{\beta} < 0.06$
Kiel [16]	gas-solids trickle flow reactor, countercurrent	mass transfer (stationary), tracer: H_2O	molecular sieves 640 μm and 2000–2360 μm	$30 \leq Re \leq 600$	The conservative estimates of the mass transfer coefficient and the Sherwood number amount to 40–80% of the theoretical values obtained by applying the Ranz-Marshall correlation.

In summary, the mass transfer method is more favorable than the heat transfer method because of the higher flexibility in tracer selection, and the broader range and higher accuracy of concentration measurement compared to temperature measurement. The method applied by Kwauk *et al.* [11] is in our view, the most reliable. However, one should be aware of the fact that intraparticle mass transport is, compared to heat transport, in general a (much) slower process. Therefore intraparticle transport will limit mass transfer processes sooner than heat transfer processes.

Theory

The principle of the method used here to determine the gas–solids mass transfer coefficient (k_g) has been described by Kwauk and coworkers [11], who investigated the adsorption of a halogen tracer (CCl_4) in a small-scale CFB unit at an extremely low concentration level (ppm to ppb range) to avoid saturation of the solids material during extended experimentation. In this way a (semi) steady-state adsorption process is obtained.

The gas–solids mass transfer coefficient k_g can be calculated from the measured axial gas-phase tracer-concentration profile using a simple one-dimensional model. Radial inhomogeneities, due to solids segregation over the column diameter and solids–wall interaction, are likely to be negligible due to the good radial (re-)distribution properties of the regularly stacked packing applied [16]. The model is derived from the basic differential equation of continuity for the tracer in the gas phase [eqn. (1)], while assuming steady-state conditions and plug flow behavior for the gas phase.

Figure 1 shows a schematic representation of the packed section and a possible axial tracer concentration profile.

$$\frac{d(\Phi_v C)}{dz} = -k_g a \beta_{\text{dyn}} (C - C_i) A \quad (1)$$

where

- Φ_v volumetric gas flow rate
- C concentration of tracer in the gas phase
- z axial coordinate, taken positive in upward direction
- $k_g a$ average gas–solids mass transfer rate constant
average gas–solids mass transfer coefficient $\times$
specific particle surface area
- β_{dyn} dynamic solids volume fraction
- C_i gas-phase concentration of the tracer at the
gas–solids interface
- A cross-sectional area of the packed column

The gas (volume) flow Φ_v can be assumed to be constant because (i) the pressure drop across the packing is negligible and (ii) the gas-phase tracer concentration is kept very low ($<0.1\%$) to avoid saturation of the solids material. Additionally, the dynamic solids volume fraction (β_{dyn}) is considered to be independent of the axial coordinate z , and equal to the average dynamic solids volume fraction ($\bar{\beta}_{\text{dyn}}$) calculated from the solids hold-up measured in Part I [2]. This assumption seems reasonable because (i) the pressure profile measured in the packed section is linear and (ii) $\bar{\beta}_{\text{dyn}}$ is small for the experimental conditions applied and will therefore resemble that found for dilute-phase pneumatic conveying, whose value is in general constant, *i.e.* independent of the height. Based upon this, eqn. (1) becomes:

Fig. 1. Schematic representation of regularly packed column and axial tracer concentration profile.

$$U_g \frac{dC}{dz} = -k_g a \bar{\beta}_{\text{dyn}} (C - C_i) \quad (2)$$

If $C = C_1$ at $z = z_1$ and $C = C_2$ at $z = z_2$, the solution of eqn. (2) yields:

$$\frac{\Delta C_1}{\Delta C_2} = \exp \left[\frac{\bar{\beta}_{\text{dyn}} k_g a (z_2 - z_1)}{U_g} \right] \quad (3)$$

and after rearranging:

$$k_g a = \frac{U_g}{\bar{\beta}_{\text{dyn}} (z_2 - z_1)} \ln \left(\frac{\Delta C_1}{\Delta C_2} \right) \quad (4)$$

in which $\Delta C_1 = C(z = z_1) - C_i$ and $\Delta C_2 = C(z = z_2) - C_i$. The value of C_i , the gas-phase concentration of the tracer at the gas-solids interface of the particle, is basically unknown due to a lack of data on the intra-particle transport. Therefore the gas-phase tracer concentration in equilibrium with the amount adsorbed on the solids material (C^*) is used as an approximation. Based upon the fact that the load factor of the solids is only slightly increased during a single experiment (the absorption capacity of the solids flow is much larger than the amount of tracer available in the gas flow), the value of C^* is assumed to be constant over the column height during a single experiment and equal to the tracer concentration in the gas outlet; this however has to be verified experimentally. With this assumption the contribution of the transport resistance inside the particle is neglected and a conservative value of $k_g^* a$, viz. the apparent mass transfer rate constant, is calculated.

If individual behaviour of spherically shaped particles is considered, the specific mass exchanging surface area is given by:

$$a = \left(\frac{6}{d_p} \right) \quad (5)$$

In this case effects of particle shielding phenomena will appear in the values of the apparent mass transfer coefficient k_g^* . The value of $\bar{\beta}_{\text{dyn}}$ for the RPCFB is calculated from eqn. (6) derived from the hold-up measurements in Part I [2].

$$\bar{\beta}_{\text{dyn}} = 0.0084 G_s G_g^{-1.22} \quad (6)$$

One of the major model assumptions to be validated is the plug flow behavior of the gas phase. Unfortunately the degree of axial dispersion in the gas phase is unknown for (cocurrent) gas-solids flow in a RPCFB. However a conservative estimate of the degree of axial dispersion in the gas phase for a worst-case analysis, can be derived from the experimental data for countercurrent gas-solids flow of air and FCC (mean diameter: 70 μm) in a packed column of 0.015 m Pall rings (Roes and van Swaaij [17]) and in a baffle column (Noordergraaf

TABLE 2. Some experimental results of Roes and van Swaaij [17] and of Noordergraaf *et al.* [18] for countercurrent gas-solids flow of air and FCC in a packed column of Pall rings and in a baffle column, respectively

Authors	U_g (m s^{-1})	G_s (kg m^{-2} s^{-1})	Bodenstein number
Roes and van Swaaij [17]	0.2	0-6	0.9-1.2
Noordergraaf <i>et al.</i> [18]	0.12-0.24	80	1-2

et al. [18]). Some of their results relevant for this discussion are presented in Table 2. Based upon their results it seems reasonable to assume that the height of a gas-phase mixing unit will approximately be equal to the height of one packing layer at the high superficial gas velocities applied ($\geq 1.2 \text{ m s}^{-1}$). Above all, the main cause of axial gas dispersion in the countercurrent mode, the downward flow of solids, is far less in the cocurrent mode where the net solids flow is directed upwards. The assumption that one layer is one mixing unit means that the gas-phase behavior between the two tracer sample points ($= 8$ packing layers $\Rightarrow 8$ mixing units) resembles plug flow behavior closely as long as gas-phase conversion is not too high. In the extreme case of 95% conversion the error is still within 20%. Besides, assuming plug flow gives a conservative value for the calculated (apparent) mass transfer rate constant $k_g^* a$.

Axial dispersion in the solids phase is of no importance because the tracer loading of the solids does not change noticeably during a single experiment.

Experimental

Sorbent and tracer selection

The selection of our solids material for the physical adsorption experiments is based on its close resemblance to the sorbent used in our high temperature desulfurization research. For this reason the fluid cracking catalyst MZ-3S (see Table 3) was chosen. The tracer component to be used should meet the requirements of a high adsorption capacity on MZ-3S and a zero adsorption capacity on the applied construction materials (PVC and Perspex). At the same time this tracer component should be easily detectable at low concentration levels. Naphthalene fulfills all these requirements and was therefore selected. The naphthalene adsorption capacity of MZ-3S, measured with a thermogravimetric analyzer at 25 $^{\circ}\text{C}$ and a naphthalene gas-phase concentration of 60 ppm, appeared to be 18 wt. %.

Apparatus and experimental procedure

All the experiments have been carried out at room temperature and atmospheric pressure. Other conditions and properties of the solids material are given in Table 3.

The packed circulating fluidized bed set-up used was similar to that described in Part I [2]. For the mass transfer experiments the riser unit was additionally equipped (see Fig. 2) with a tracer-gas injection device (C) just below the packed section, while almost all the pressure taps in the set-up were replaced by gas sample probes. A special tracer-gas injection device was designed to ensure rapid mixing with the main stream. It consisted of a 0.03 m thick Perspex disk (0.12 m diameter) with a 0.06 m × 0.06 m square opening in the center, through which two horizontal and parallel, 6 mm o.d. copper tubes (see Fig. 3a) were passed, each provided with two rows of 9 evenly distributed downward-directed holes of 0.7 mm diameter for tracer-gas injection. The gas sample probe (Fig. 3b) was a 6 mm o.d. steel tube provided with a 12 mm long and 5.5 mm o.d. steel filter (pore size 5 μm) at the end.

Humidification of the main air stream (Fig. 2, stream I), to avoid undesired effects of static electricity, was achieved by injection of steam just before the lower part of the riser unit. Part of the total gas flow (Fig. 2, stream III) was saturated with naphthalene by leading it through a packed bed of naphthalene spheres (7) before adding it to the main stream through the tracer-gas injection device. The naphthalene feed line and the analysis section was heated to prevent naphthalene adsorption on the construction materials. The gas sample was withdrawn from the main gas stream by suction through the analysis line; typical sample flows range from 0.2 up to 0.8 ml s⁻¹, yielding a linear velocity at the porous sample tip of maximal 5 mm s⁻¹. Naphthalene concentrations were measured with a H.N.U. photoionization detector with a detection limit of <0.1 ppm

naphthalene. Regeneration of the catalyst was possible in a separate step by increasing the temperature of the fluidized bed (storage vessel provided with an electric heating coil (8)). Finally, the naphthalene in the fluid bed off-gas was removed by leading it through a packed bed of active coal granulates.

At the beginning of each mass transfer measurement, the regularly packed circulating fluidized bed was fed with the humidified main stream I only. Humidification was stopped just before the naphthalene adsorption experiment started, because the presence of water in large quantities interferes with the naphthalene analysis. However, the water which desorbed from the FCC material did not interfere noticeably because the water vapor concentration in the bulk of the gas flow is very low during the desorption period. Then, the naphthalene-saturated stream III (up to 25% of the total gas stream) was added to the main stream just below the packing section. The inlet concentration of naphthalene (up to 15 ppm) was controlled by adjusting the ratio of gas flow III to gas flow I. When the gas and solids flux were adjusted, the system was allowed to reach steady state. Finally, the naphthalene concentrations could be measured at sample points S7, S1, S2 and again at S7 (see Fig. 2), by opening the corresponding valve in the analysis line. Analysis of samples from points S3 to S6 usually yielded values too close to the equilibrium concentration, measured at sample point S7, to be accurate enough for ($\bar{\beta}_{\text{dyn}} k_g^* a$) calculation. This equilibrium concentration was constant for a single experiment but increased during a day's experimental program. The pressure drop along the column and the solids mass flux were determined immediately after the above-described adsorption experiment. As mentioned before, the naphthalene was desorbed during the night by heating the solids contents of the gently fluidized bed up to a temperature of 60–80 °C.

TABLE 3. Experimental conditions and properties of the solids material used

Gas flux range (kg m ⁻² s ⁻¹)	1.4 and 2.7
Solids flux range (kg m ⁻² s ⁻¹)	up to 12
Solids type	FCC (MZ-3S)
Particle size distribution:	
diameter (μm)	cumulative wt. %
< 149	99
< 105	79
< 80	45
< 40	8
< 20	1
Particle density (kg m ⁻³)	880
Mean particle diameter (μm)	70
Terminal velocity (m s ⁻¹)	0.11

Results and discussion

The apparent mass transfer rate constant based upon the reactor volume, $\bar{\beta}_{\text{dyn}} k_g^* a$ is calculated according to the rearranged eqn. (4):

$$\bar{\beta}_{\text{dyn}} k_g^* a = \frac{U_g}{(z_2 - z_1)} \ln \left(\frac{\Delta C_1}{\Delta C_2} \right) \quad (7)$$

where $\Delta C_1 = C(z_1) - C^*$ is the tracer concentration difference between sample points S1 and S7, $\Delta C_2 = C(z_2) - C^*$ is the tracer concentration difference between sample points S2 and S7, $(z_2 - z_1) = 0.08$ m is the distance between sample points S1 and S2, U_g is the superficial gas velocity calculated from the adjusted gas mass flux.

Fig. 2. Schematic diagram of the packed CFB set-up used for mass transfer measurements. The small-scale CFB consists of a riser (1) with a regularly packed square section (A), a tracer gas injection device (C) and two slide-valves (B) for solids hold-up measurements; two cyclones (2), a solids flux measuring device (3, 4), a fluid bed (5) for solids storage, a 45° inclined feedline (6) with a slide-valve for solids flow control, a packed bed of naphthalene spheres (7) and a heating coil (8).

The apparent mass transfer rate constant based upon reactor volume, $\bar{\beta}_{\text{dyn}} k_g^* a$, is plotted in Fig. 4 as a function of the solids mass flux. The results show a root-like dependency: $\bar{\beta}_{\text{dyn}} k_g^* a \propto G_s^{0.47}$. Figure 4 additionally shows that, except for this increase with the solids mass flux, the value of $\bar{\beta}_{\text{dyn}} k_g^* a$ also increases with increasing gas mass flux. Roughly, $\bar{\beta}_{\text{dyn}} k_g^* a$ values for $G_g = 2.7$ are 35% higher than for $G_g = 1.4 \text{ kg m}^{-2} \text{ s}^{-1}$, which indicates that $\bar{\beta}_{\text{dyn}} k_g^* a$ could also show a root-like dependence on the gas mass flux: $\bar{\beta}_{\text{dyn}} k_g^* a \propto G_g^{0.5}$. Combined, this means that:

$$\bar{\beta}_{\text{dyn}} k_g^* a = 12 G_s^{0.47} G_g^{0.5} \quad (8)$$

Figure 5 was derived from Fig. 4 by introducing the correlation for the solids volume fraction $\bar{\beta}_{\text{dyn}}$ (eqn. (6)) and substituting $a = (6/d_p)$. This operation, applied to eqn. (8), gives:

$$k_g^* = 0.017 G_s^{-0.53} G_g^{1.7} \quad (9)$$

The fact that k_g^* decreases with increasing solids mass flux (=increasing solids volume fraction) has been observed earlier by other investigators (see above) and is usually explained by a corresponding decrease in gas-solids contact due to solids agglomeration (particle shielding).

Fig. 3. Schematic representation of the tracer-gas injection device and the gas sample probe. (a) (I) bottom-view of injection device, (II) vertical cross-section of injection device; (b) gas sample probe.

Fig. 4. The apparent mass transfer rate constant based upon reactor volume, $\bar{\beta}_{dyn} k_g^* a$ as a function of the solids mass flux for two values of the gas mass flux. ($\square$) $G_g = 1.4 \text{ kg m}^{-2} \text{ s}^{-1}$; $\bar{\beta}_{dyn} k_g^* a = 15 G_s^{0.44}$; ($\blacklozenge$) $G_g = 2.7 \text{ kg m}^{-2} \text{ s}^{-1}$; $\bar{\beta}_{dyn} k_g^* a = 20 G_s^{0.50}$.

The solids velocity u_s is defined by eqn. (10). Substitution of $\bar{\beta}_{dyn}$ by eqn. (6) shows that u_s is independent of the solids mass flux applied.

$$u_s = \frac{G_s}{\rho_s \bar{\beta}_{dyn}} = \frac{G_g^{1.22}}{0.0084 \rho_s} \quad (10)$$

The slip velocity ($u_{slip} = u_g - u_s$, see eqn. (11)) is therefore only a function of the applied gas mass flux and nearly proportional to G_g . The approximation in eqn. (11) is valid because $\bar{\beta}_{dyn} \ll \epsilon_p (= 0.39)$ and $0.0084 \rho_s \gg \rho_g (1 - \epsilon_p)$ for the experimental conditions applied.

$$u_{slip} = u_g - u_s = \frac{G_g}{\rho_g (1 - \epsilon_p - \bar{\beta}_{dyn})} - \frac{G_g^{1.2}}{0.0084 \rho_s} \approx \frac{G_g}{\rho_g (1 - \epsilon_p)} \quad (11)$$

Combination of eqn. (9) with eqns. (6) and (11) eliminates G_s and G_g :

Fig. 5. The apparent mass transfer coefficient k_g^* as a function of the solids mass flux for two gas fluxes. ($\square$) $G_g = 1.4 \text{ kg m}^{-2} \text{ s}^{-1}$; $k_g^* = 0.033 G_s^{-0.57}$; ($\blacklozenge$) $G_g = 2.7 \text{ kg m}^{-2} \text{ s}^{-1}$; $k_g^* = 0.098 G_s^{-0.48}$.

$$k_g^* = 0.0016 \frac{G_g^{1.1}}{\sqrt{\bar{\beta}_{dyn}}} = 0.0011 \frac{(u_{slip})^{1.1}}{\sqrt{\bar{\beta}_{dyn}}} \quad (12)$$

Equation (12) then shows that the increase in k_g^* due to an increase in the gas mass flux at a certain fixed value of the solids mass flux, is mainly caused by an increased slip velocity between gas and particles and to a lesser extent by a decreased solids volume fraction. Unfortunately, the data reported in Figs. (4) and (5) show much scatter, due to inaccuracies of the measuring technique and fluctuations in the applied conditions. Nevertheless it is clear that values of k_g^* can become quite high upon proper selection of operating conditions. The contribution of the packing applied to the value of k_g^* , i.e. the difference between the k_g^* values obtained in a RPCFB and in a CFB, has not been measured explicitly. The influence of the packing must therefore be concluded from a comparison with literature data.

To compare our results with literature data, usually presented as the Sherwood number as a function of the Reynolds number, the value of Sh should be calculated according to:

$$Sh = \frac{k_g \cdot d_p}{D_{\text{naph}}} \quad (13)$$

According to Perry and Chilton [19], the diffusion coefficient of naphthalene is given by:

$$D_{\text{naph}} = 5.13 \times 10^{-6} \left(\frac{T(K)}{273.15} \right)^{1.75} \quad (14)$$

The already-mentioned particle shielding phenomenon is clearly illustrated in Fig. 6, where Sh is plotted as a function of the solids volume fraction for a gas mass flux of 1.4 and 2.7 kg m⁻² s⁻¹. It shows that Sh decreases exponentially with increasing $\bar{\beta}_{\text{dyn}}$.

Comparison with literature data

Comparison of the presently observed mass transfer data with those for packed and fluidized beds available from the literature, is possible in the well-known diagram of Sherwood/Nusselt numbers as a function of Reynolds numbers (Kunii and Levenspiel [20]; Nelson and Galloway [21]). Sherwood and Nusselt numbers are interrelated by the Chilton–Colburn analogy (Chilton and Colburn [15]). The Sherwood numbers for our packed circulating fluidized bed then appear to fit nicely into the band of literature data for fluidized and packed beds (see Fig. 7). Additionally, the gas–solids mass transfer or heat transfer results of Kwauk *et al.* [11], Dry *et al.* [3, 4], Watanabe *et al.* [13] and Bandrowski and Kaczmarzyk [9], all measured under CFB conditions, are shown. The results of Kwauk *et al.* [11] and of Dry *et al.* [3, 4] had to be reinterpreted as shown in the

Fig. 6. The Sherwood number as a function of the solids volume fraction measured in a RPCFB with bar diameter of 0.01 m. ($\square$) $G_g = 1.4 \text{ kg m}^{-2} \text{ s}^{-1}$, $Sh = 0.021 \bar{\beta}^{-0.55}$; ($\blacklozenge$) $G_g = 2.7 \text{ kg m}^{-2} \text{ s}^{-1}$, $Sh = 0.058 \bar{\beta}^{-0.50}$

Appendix. The reported data agree well with the (shaded) literature band except for the results of Dry *et al.* [3, 4], which are extremely low and deviate strongly not only from our results for FCC but also from the results reported by Kwauk *et al.* [11] and Watanabe *et al.* [13]. For the FCC-material this large deviation is remarkable because the solids material used is similar to ours. Differences however exist in the experimental method used (steady-state mass transfer vs. dynamic heat transfer), the presence of a regularly stacked packing in our case and the column geometry. Dry and White [4] also reported results concerning heat transfer measurements with fine (140 μm) and coarse (275 μm) sand particles. The surface efficiencies calculated ranged from 0.1% to 1%. Reinterpretation of these data according to the method outlined in the Appendix shows that the corresponding Nu number is at least a factor 100 to 1000 lower than the Nu number calculated according to the Ranz–Marshall equation (see Fig. 7). Comparison with other literature data (Watanabe *et al.* [13] and Kwauk *et al.* [11]) shows that the results of Dry and White [4] are again far lower. In addition to the difference in experimental method, the internal diameter of the riser section was only 0.021 and 0.024 m in the case of Watanabe *et al.* and Kwauk *et al.* respectively, and 0.09 m in the case of Dry and White. The flow of gas and solids in the 0.021 and 0.024 m i.d. riser, compared to the 0.09 m one, is probably more symmetrical and would therefore yield a higher gas–solids contact efficiency and Nu/Sh number.

The regularly structured packing can be considered as a set of parallel riser sections each with an internal diameter equal to the bar diameter (0.01 m). In this context it seems reasonable to assume that the difference between Dry's results and the present ones is mainly due to the effect of the packing which breaks up the radial inhomogeneities and solids clusters, and hence increases the gas–solids contact efficiency. The existence of radial inhomogeneities in their set up used for the heat transfer experiments, has indeed been reported by Dry [22]. Bartholomew and Casagrande [7], and Saxton and Worley [6] also reported them for large-diameter CFB installations.

Conclusions

The adsorption of naphthalene vapor on FCC particles in a regularly packed circulating fluidized bed was investigated. Values of the mass transfer coefficient were derived from the decrease in the axial naphthalene vapor concentration by applying a simple plug flow model. It was assumed that the contribution of the transport resistance inside the particle is negligible, consequently the (apparent) mass transfer coefficients

Fig. 7. Sherwood and Nusselt numbers as a function of Reynolds number for different gas-solids systems. Shaded area represents literature data for fluidized and packed beds (Kunii and Levenspiel [20]; Nelson and Galloway [21]).

presented are conservative estimates. From the results obtained it can be concluded that:

(i) The apparent mass transfer coefficients (k_g^*) and the Sherwood numbers in the packed section of a circulating fluidized bed are relatively high and increase with increasing gas mass flux.

(ii) However, k_g^* and Sh decrease with increasing solids mass flux because of the corresponding increase in solids hold-up and particle shielding.

(iii) The following relationship may be used to calculate the mass transfer coefficient in a RPCFB from the adjusted flow conditions:

$$k_g^* = 0.017 G_s^{-0.53} G_g^{1.7} \quad (9)$$

It should be noted however, that the validity of this correlation has not yet been demonstrated sufficiently.

(iv) The above equation can be rewritten to give:

$$k_g^* = 0.0011 \frac{(u_{slip})^{1.1}}{\sqrt{\beta_{dyn}}} \quad (11)$$

by using the results of an earlier hydrodynamic study [2]. This means that if, at a certain fixed value of the solids mass flux, the gas mass flux is increased, k_g^* will increase mainly because of a corresponding increase in the particle slip velocity and only to a lesser extent because of the corresponding decrease in the solids volume fraction.

(v) The introduction of packing elements into a circulating fluidized bed may enhance the apparent mass transfer rate (based upon the reactor volume) $\beta_{dyn} k_g^* a$, firstly due to the increase in the dynamic solids volume fraction β_{dyn} which appears to dominate the effect of particle shielding on k_g^* , and secondly because of the related increase in gas velocity (and

slip velocity) as a result of the smaller cross-sectional surface.

(vi) Comparison of the Sh values with literature data obtained for a comparable solids material in a CFB (Dry *et al.* [3]), suggests that the influence of the packing is significant. It seems to break up the radial inhomogeneities and solids clusters, and hence to increase the gas-solids contact efficiency. Additional mass transfer experiments in the CFB as well as in the RPCFB are needed to further substantiate this conclusion.

(vii) The obtained Sh values fit nicely in the shaded literature band for packed and fluidized beds.

Acknowledgements

We wish to thank F. Horvath and P.J. Bots for their valuable contribution to the design of the packed CFB set-up and for performing the experimental work. The financial support of the Netherlands Agency for Energy and the Environment (NOVEM) is gratefully acknowledged.

List of symbols

a	specific particle surface area = $6/d_p$ ($\text{m}^2 \text{m}^{-3}$)
A	cross-sectional area of the packed section (m^2)
C	concentration of tracer in gas-phase (kmol m^{-3})
C_i	concentration of tracer at particle interface (kmol m^{-3})
C^e	concentration of tracer in gas-phase at equilibrium with solid-phase (kmol m^{-3})

C_p	heat capacity of air ($\text{kJ kg}^{-1} \text{K}^{-1}$)
d_p	average particle diameter (m)
D_{naph}	gas-phase diffusion coefficient of naphthalene ($\text{m}^2 \text{s}^{-1}$)
D_{CCl_4}	gas-phase diffusion coefficient of CCl_4 ($\text{m}^2 \text{s}^{-1}$)
E_{gs}	gas-solids contact efficiency
h_f	heat transfer coefficient for the external gas film ($\text{W m}^{-2} \text{K}^{-1}$)
G	mass flux ($\text{kg m}^{-2} \text{s}^{-1}$)
k_g	gas-solids mass transfer coefficient (m s^{-1})
k_g^*	apparent gas-solids mass transfer coefficient (m s^{-1})
$k_g a$	gas-solids mass transfer rate (s^{-1})
$k_g^* a$	apparent gas-solids mass transfer rate (s^{-1})
L	length, distance between injection and sample point (m)
Nu	Nusselt number, $(h_f d_p) \lambda_g^{-1}$
Re	Reynolds number, $(\rho_g (u_g - u_s) d_p) \eta_g^{-1}$
Sh	Sherwood number, $(k_g^* d_p) D_{\text{naph}}^{-1}$
T	temperature (K)
T_0	gas temperature at the bottom of the test section (K)
u	local velocity (m s^{-1})
u_{slip}	local slip velocity between gas and solids (m s^{-1})
U	superficial velocity (m s^{-1})
z	coordinate in vertical direction (m)

Greek letters

β	solids volume fraction
$\bar{\beta}$	average solids volume fraction
β_{dyn}	dynamic solids volume fraction i.e. fraction of solids carried by the gas
$\bar{\beta}_{\text{dyn}}$	average dynamic solids volume fraction i.e. fraction of solids carried by the gas
ϵ_p	volume fraction of packing
λ_g	thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)
Φ_v	volumetric gas flow rate ($\text{m}^3 \text{s}^{-1}$)
ρ	density (kg m^{-3})
η	dynamic viscosity ($\text{kg m}^{-1} \text{s}^{-1}$)

Subscripts

g	gas phase
s	solids phase

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Appendix: Reinterpretation of literature data

Kwauk et al.

Kwauk et al. [11] used adsorption to study gas-solids contact in hydrodynamically dispersed particulate systems. They investigated the adsorption of a halogen tracer (CCl_4) on active carbon particles of $570 \mu\text{m}$ diameter in a small-scale circulating fluidized bed unit of 0.024 m diameter and 1.5 m length at extremely low tracer concentration ($\leq 3 \text{ ppm}$). This was done to avoid saturation of the solids material during extended experimentation. The ECD (electron capture detection) method was used to determine the concentration of the halogen tracer in the ppm to ppb range.

The experimental results, i.e. the axial tracer concentration and voidage profile, were interpreted with

a plug flow model similar to that described in this paper. Kwauk *et al.* [11] neglect the value of the particle surface concentration C_i , due to the lack of data on the intraparticle transport. This gross simplification caused Kwauk *et al.* [11] to report a decrease in the k_g value with height. When C_i is assumed to be equal to C_{out} (the tracer concentration in the outlet stream which is in equilibrium with the absorbent), the phenomenon of a decreasing k_g value with height vanishes. The newly calculated k_g values are shown in Table A.1.

Table A.1 shows clearly that the apparent mass transfer coefficient decreases with increasing solids volume fraction. The Sherwood numbers are also plotted in Fig. 7 and compared with other literature data. They appear to fit nicely in the literature band.

Dry *et al.*

Dry *et al.* [3] measured the gas–solids contact efficiency for a Fluid Cracking Catalyst (71 μm) in a circulating fluidized bed. Their measurement technique involved the use of heated gas pulses as an interacting tracer and detection of this hot gas by a rapid-response thermocouple. The gas–solids contact efficiency was then defined as the ratio of the amount of heat actually transferred from gas to solids, to the amount needed to reach equilibrium between gas and solids. Their experimental conditions were such that only heat transfer across the external gas film was rate-controlling. The experimental results indicated which fraction of the gas introduced at the bottom of the bed emerges at the outlet, without having come into contact with any solid particles.

The experiments were conducted in a circulating fluidized bed rig containing a 0.09 m i.d. steel riser unit with a height of 7.2 m. Superficial gas velocities

of 2–8 m s^{-1} and solids mass fluxes up to 200 $\text{kg m}^{-2} \text{s}^{-1}$ were applied. The results showed that the gas–solids contact appeared to be almost 100% at 2 m s^{-1} for a solids mass flux exceeding 100 $\text{kg m}^{-2} \text{s}^{-1}$, but decreased steadily as gas velocities increased. At 8 m s^{-1} , it appeared that contact efficiencies are significantly less than 100%, even at solids mass fluxes approaching 200 $\text{kg m}^{-2} \text{s}^{-1}$.

By means of a simple model for steady state heat transfer, Dry *et al.* estimated the ratio of the effective to actual particle surface area. This resulted in a surface efficiency which ranged from 0.01% to 0.13%. They suggested that this could be related to the fraction of particles which are free to move as individuals at any instant, as opposed to particles which make up loose agglomerates such as streamers or dense wall regions.

The concept of defining a surface efficiency is unusual. Generally the specific surface area (a) is assumed to be equal to that of a single particle ($=6/d_p$) and the results for the contact efficiency are then used to estimate the value of the heat transfer coefficient (h_t) and the Nusselt number (Nu). Based upon the experimental results of Dry *et al.* [3] the value of h_t , Nu and Re can easily be calculated, using eqns. (A.2), (A.3) and (A.4) respectively, and assuming single particle behavior. The results of these recalculations are shown in Table A.2. Equation (A.2) is obtained by rearranging eqn. (A.1), the result of the steady-state energy balance derived by Dry *et al.* [3].

$$E_{gs} = \frac{T_0 - T}{T_0 - T_s} = 1 - \exp\left[-\frac{h_t a \beta L}{U_g \rho_s C_p}\right] \quad (\text{A.1})$$

$$h_t = \frac{U_g \rho_s C_p}{a \beta L} (-LN(1 - E_{gs})) \quad (\text{A.2})$$

$$Nu = \frac{h_t d_p}{\lambda_g} \quad (\text{A.3})$$

TABLE A.1. Maximum k_g values reported by Kwauk *et al.* [11], together with the recalculated values for k_g and corresponding Sherwood and Reynolds numbers

U_g (m s^{-1})	Kwauk <i>et al.</i>		Reinterpreted results		
	β (%)	$[k_g]_{\text{max}}$ (m s^{-1})	$[k_g]_{\text{recalc}}$ (m s^{-1})	Sh (–)	Re (–)
2.0	2	0.07	0.083	4.8	38
1.9	3	0.06	0.054	4.1	38
1.8	7–10	0.03	0.034	2.1	38
1.7	17–21	0.02	0.018	1.4	38

where

Sh	$(k_g d_p) D_{CCl_4}^{-1}$
d_p	$0.57 \times 10^{-3} \text{ m}$
D_{CCl_4}	$8.34 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ at 293 K and 1 bar (Reid <i>et al.</i> [23])
Re	$(\rho_g u_{\text{slip}} d_p) \eta_g^{-1}$
ρ_g	1.21 kg m^{-3}
η_g	$1.81 \times 10^{-5} \text{ Pa.s}$
u_{slip}	$u_g - u_s \approx 1 \text{ m s}^{-1}$ (assumed)

TABLE A.2. Results reported by Dry *et al.* [3] and the values of h_t , Nu and Re numbers calculated from these results

U_g (m s^{-1})	G_s ($\text{kg m}^{-2} \text{s}^{-1}$)	$\bar{\beta}$ (–)	E_{gs} (%)	$\hat{h}_t$ ($\text{W m}^{-2} \text{K}^{-1}$)	Nu (–)	Re (–)
2	12	0.023	76	0.26	7.0E-04	8
2	31	0.083	86	0.10	2.7E-04	9
4	25	0.011	51	0.54	1.5E-03	11
4	58	0.033	72	0.32	8.7E-04	14
4	125	0.065	90	0.29	8.0E-04	14
6	52	0.012	61	0.97	2.7E-03	14
6	105	0.028	81	0.74	2.0E-03	16
6	146	0.039	84	0.58	1.6E-03	17
6	200	0.057	91	0.52	1.4E-03	18

$$Re = \frac{\rho_g u_{slip} d_p}{\eta_g} = \frac{\rho_g (u_g - u_s) d_p}{\eta_g} \quad (A.4)$$

where

E_{gs}	gas-solids contact efficiency (–)
T	gas temperature (K)
T_0	gas temperature at the bottom of the test section (K)
T_s	solids temperature (K)
h_f	heat transfer coefficient for external gas film ($W m^{-2} K^{-1}$)
a	specific particle surface area = $6/d_p$ $m^2 m^{-3}$
d_p	mean particle diameter = $71 \mu m$
β	solids volume fraction (–)
L	length, distance between injection and sample point = $6.92 m$
U_g	superficial gas velocity
ρ_g	density of air = $1.21 kg m^{-3}$
C_p	heat capacity of air = $1 kJ kg^{-1} K^{-1}$
λ_g	thermal conductivity of gas = $0.026 W m^{-1} K^{-1}$
u_g	local gas velocity = $U_g/(1 - \beta)$ ($m s^{-1}$)
u_s	local solids velocity = $G_s/(\rho_s \beta)$ ($m s^{-1}$)
G_s	solids mass flux ($kg m^{-2} s^{-1}$)
ρ_s	solids density = $1370 kg m^{-3}$
η_g	dynamic viscosity of air = $1.81 \times 10^{-5} Pa s$

Equations (A.1) and (A.2) are only valid for a constant value of the product ($h_f a \beta$), i.e. a constant value of β . This condition has to be verified first. The heated gas pulse was injected 0.4 m above the bottom of the fast bed and the gas temperature was monitored about 7 m higher with a thermocouple probe in the bend at the top of the riser. Based upon the axial position of the injection and sample point and the experimental conditions applied, it seems very unlikely that the solids volume fraction β was constant between these two points. The value of h_f depends (strongly) on β , which means that h_f also changes between the injection and sample point. Equations (A.1) and (A.2) are therefore, strictly speaking, not valid but due to a lack of data (Dry *et al.* did not measure the axial porosity and temperature profile) we have to assume in the first approach that the product of ($h_f a \beta$) is constant. Our own results already showed that the value of ($k_g a \beta$) levels off with increasing solids mass fluxes (Fig. 4), which could indicate a constant value for ($h_f a \beta$) at high solids mass fluxes. The calculated value of ($h_f a \beta$) is an average for the riser section, so that after correction of β and a , an average heat transfer coefficient $\bar{h}_f$ is obtained.

The values of Nu are also presented in Fig. 7 and compared with other literature data. It shows that the reported data are located at a factor of 100 to 1000 below the shaded literature band.

Dry and White [4] recently published the contact efficiencies measured for two grades of sand (140 and $275 \mu m$). The experimental set-up, the measurement technique and the interpretation method were identical to those applied for the FCC material. From the experimental results, the values of h_f , Nu and Re can easily be calculated using the method outlined above. The results of these recalculations are shown in Tables A.3 and A.4 for fine and coarse sand respectively. The values of Nu are also presented in Fig. 7 and compared with literature data for similar solids material. As in the case of the FCC material, the reported data are located far below the other literature data and the shaded literature band.

TABLE A.3. Results reported by Dry and White [4] and the values of h_f , Nu and Re numbers calculated from these results. Solids material: fine sand, $d_p = 140 \mu m$, $\rho_s = 2650 kg m^{-3}$

U_g ($m s^{-1}$)	G_s ($kg m^{-2} s^{-1}$)	β (–)	E_{gs} (%)	$\bar{h}_f$ ($W m^{-2} K^{-1}$)	Nu (–)	Re (–)
4	50	0.013	66	1.34	7.2E-03	25
4	90	0.049	84	0.62	3.3E-03	33
4	130	0.074	90	0.51	2.7E-03	34
4	220	0.088	91	0.45	2.4E-03	32
6	70	0.007	51	2.46	1.3E-02	21
6	170	0.034	77	1.06	5.7E-03	40
6	250	0.070	84	0.65	3.5E-03	48
6	380	0.123	91	0.47	2.5E-03	53

TABLE A.4. Results reported by Dry and White [4] and the values of h_f , Nu and Re numbers calculated from these results. Solids material: coarse sand, $d_p = 275 \mu m$, $\rho_s = 2650 kg m^{-3}$

U_g ($m s^{-1}$)	G_s ($kg m^{-2} s^{-1}$)	β (–)	E_{gs} (%)	$\bar{h}_f$ ($W m^{-2} K^{-1}$)	Nu (–)	Re (–)
4	33	0.019	59	1.53	1.6E-02	63
4	66	0.059	76	0.77	8.1E-03	70
4	88	0.089	79	0.56	5.9E-03	74
6	59	0.013	68	4.11	4.4E-02	81
6	99	0.022	73	2.83	3.0E-02	82
6	161	0.048	79	1.58	1.7E-02	93
6	205	0.115	85	0.80	8.5E-03	112
8	33	0.005	49	8.61	9.1E-02	102
8	80	0.008	64	7.93	8.4E-02	81
8	106	0.013	70	5.72	6.1E-02	94
8	161	0.022	79	4.64	4.9E-02	99