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Mass Transfer around Active Particles in Fluidized Beds

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1. Introduction

Fluidized beds are extensively used in a number of gas-solid applications where significant heat and/or mass transfer rates are needed. The design and modelling of such processes requires the precise knowledge of the heat and mass transfer coefficients around immersed objects in the fluidized bed. Thus, it is not surprising that since the early spreading of the fluidized bed technology a considerable experimental and theoretical activity on this topic has been reported in the literature, mostly focused on the heat transfer coefficient. A more limited effort was dedicated to the estimation of the mass transfer coefficient, because of the inherent difficulty of measuring this quantity in the dense environment of a fluidized bed. Unfortunately, a fluidized bed is one of those cases where the analogy between heat and mass transfer does not hold, so that measured heat transfer coefficients cannot be used to estimate mass transfer rates under similar operating conditions. In fact, the bed particles represent an additional path to heat transfer around an immersed object, while they only result in a decrease of the available volume for gas mass transfer (except for the very particular case when the bed particles can adsorb one of the transferred components). Strictly speaking, an analogy exists between mass transfer and the gas-convective contribution to heat transfer in a fluidized bed. On the other hand, the particle-convective contribution to heat transfer (and also the radiative one, if relevant) has not an analogous mechanism in a mass transfer process.

In this review paper we will focus our attention to the mass transfer coefficient around freely moving active particles in the dense phase of a fluidized bed. This case represents most situations of practical interest, whereas the case of a fixed object (with respect to a reference system bound to the reactor walls) is less frequently encountered in mass transfer problems, contrary to the heat transfer case. With active particle we mean a particle that is exchanging mass with the gas phase, because either a chemical reaction or a physical process (phase change) is taking place in or at the surface of the particle. Finally, we will mainly consider the case of mass transfer between the gas and one or few active particles dispersed in a fluidized bed of inert particles, as opposed to the case where the entire bed is made of active particles. This configuration is important for a number of processes like combustion and gasification of carbon particles, and most typically the inert particle size is smaller than the active particle size.

In the next sections we will thoroughly review the experimental and theoretical work available in the literature on mass transfer in the dense phase of fluidized beds, showing the

main achievements and the limitations for the estimation of the mass transfer coefficient. On the other hand, only few review papers addressing (partially) this topic have appeared in the literature (La Nauze, 1985; Agarwal & La Nauze, 1989; Ho, 2003; Yusuf et al., 2005), so that a more complete review of the previous literature available on mass transfer in fluidized beds is considered to be useful.

A convenient way to analyze and compare mass transfer data is to use the particle Sherwood number defined as $Sh = k_g \cdot d_a / D$. This quantity represents the average dimensionless gas concentration gradient of the transferring species at the active particle surface.

2. Mass transfer around isolated spheres in a gas flow

Before focusing on the dense phase of a fluidized bed we will briefly describe the mass transfer problem around an isolated sphere in a gas flow, as this is the starting point for further discussion on mass transfer in fluidized beds. This problem is relevant for particles or drops flowing in a diluted gas stream, like in spray-dry or entrained flow applications. It is important to note that in this case each particle moves isolated from the other particles and the analogy between heat and mass transfer processes around the particle is valid. An exact solution to the set of equations describing the boundary layer problem with mass and/or heat transfer around a sphere in a gas flow is not available, so that empirical or semi-empirical correlations are required to describe the experimental results.

Experimental data of mass and heat transfer coefficient for this system are mostly derived from evaporation of single liquid drops in a gas flow, due to the simplicity and accuracy in performing the measurements. In its pioneering experimental and theoretical work Frössling (1938) first proposed to correlate the mass transfer data (in the Reynolds number range of 2 to 1300) with the following expression derived by dimensional analysis:

$$Sh = 2.0 + K \cdot Re^{1/2} \cdot Sc^{1/3} \tag{1}$$

where $Re = \rho_g \cdot U \cdot d_a / \mu_g$, $Sc = \mu_g / D \cdot \rho_g$, and K is a constant, whose value was estimated to be 0.552. The first term on the right hand side represents mass transfer in stagnant conditions (diffusive term), while the second one accounts for the enhancement of mass transfer caused by the gas flowing around the particle (convective term). This expression is consistent with the theoretical requirement that Sh = 2 at Re = 0. It must be highlighted that the use of Eq. 1 (or similar ones) is based on the assumption that a steady boundary layer develops around the particle enabling the use of a steady-state mass transfer approach.

Ranz & Marshall (1952) used Eq. 1 to correlate both their own and previous mass and heat transfer data, and suggested a value K = 0.60 (for 0 < Re < 200). Successively, Rowe et al. (1965) also correlated with Eq. 1 their own and others' data available to that date and obtained K = 0.69 (for 20 < Re < 2000). This value of K is probably the most reliable one and with this value Eq. 1 is able to predict the heat and mass transfer data around an isolated sphere in a gas flow with a remarkable accuracy. Recently, Paterson & Hayhurst (2000) gave further theoretical background to this expression.

3. Mass transfer around active spheres in a fluidized bed: experimental data and correlations

In a fluidized bed the active particles are surrounded by a dense bed of inert particles and two different effects occur that influence the mass transfer process. First, the inert particles

decrease the gas volume available for mass transfer around the active particle. Second, the presence of the fluidized particles alters the gas fluid-dynamics and the formation of the boundary layer around the active particle. These two effects must be taken into account when interpreting the experimental data.

It is obvious that the experimental technique based on the evaporation of liquid drops for the measurement of the mass transfer coefficient is not feasible in the dense phase of a fluidized bed. Different techniques have been actually used in fluidized beds and reported in the literature, and they mostly belong to three categories: sublimation of solid particles, liquid evaporation from porous particles and combustion of carbon particles. A fourth technique has been recently reported based on chemical reaction on the surface of catalyst spheres. In the following we will examine these four groups separately, indicating advantages and drawbacks of each technique. The only other works found in the literature using a technique not belonging to these four groups to estimate the mass transfer coefficient are those reported by Hsu & Molstad (1955) and by Richardson & Szekely (1961) who studied the adsorption of carbon tetrachloride by a fluidized bed entirely made of activated carbon granules. These early studies, however, showed limited success and will not be examined further.

3.1 Sublimation of solid particles

This technique is based on the determination of the sublimation rate of one or more solid particles in the fluidized bed by the measurement either of their weight change or of the concentration of the sublimating component in the gas phase. Calculation of the mass transfer coefficient requires the knowledge of the vapour pressure and of the diffusion coefficient of the sublimating component at the operating temperature. If appreciable heat effects are associated to the sublimation process, the active particle temperature must be either independently measured during the tests or estimated with a heat balance coupled to the mass balance around the particle.

Most of the experimental data obtained with this technique have been collected using naphthalene as the sublimating component. This substance is conveniently available, non-toxic, easily mouldable, and sublimates at low but detectable rates at temperatures close to the ambient one. Further advantages are the possibility to measure the naphthalene vapour concentration by means of a flame ionization or infrared analyzer, and the small heat effect, so that the active particle temperature can be safely assumed to be close to the bed temperature. This technique was first applied to fluidized beds by Resnick & White (1949) and Chu et al. (1953). These authors used shallow beds composed of all active particles. To extend the range of the studies to smaller particles and deeper beds without approaching saturation in the gas phase, van Heerden (1952) diluted few naphthalene spheres in a bed of carborundum, coke or fly ash particles. In examining the experimental results, this author noted that Sherwood numbers below the theoretical minimum of 2 were obtained at low Reynolds numbers. This result was explained by the reduced volume available for diffusion because of the presence of the inert particles, and the use of an effective diffusion coefficient through the bed interstices was suggested. Hsiung & Thodos (1977) diluted few naphthalene spheres in a bed of inert particles of the same size and density. The inert particles were beads of styrene divinylbenzene copolymer, which were claimed not to adsorb appreciably naphthalene vapour after an initial exposure. The experimental results were correlated by the following expression (rearranged here in terms of the Sherwood number):

$$Sh = \left(0.040 \cdot Re_{mf} + 2.12 \cdot Re_{mf}^{0.41} + 0.62/Re_{mf}^{0.51}\right) \cdot Sc^{1/3} \quad (2)$$

where $Re_{mf} = \rho_g \cdot U_{mf} \cdot d_a / \mu_g$ is the Reynolds number at the minimum fluidization condition. An interesting outcome of this investigation was that Sh appeared to be independent of the total fluidization velocity, but only depended on the minimum fluidization velocity and on the particle size (and also on the gas properties). Prins et al. (1985) and de Kok et al. (1986) extended the investigation to the case where the inert particle size was different from that of the active particle. In particular, the size ratio between the active and the inert particles was varied in the range 3-200. These authors pointed out at two possible problems arising with the use of this technique: the risk of mechanical attrition of the active particles and of adsorption of naphthalene vapour by the bed material. Both these two problems would lead to an enhancement of the apparent mass transfer rate and, in turn, to an overestimation of the mass transfer coefficient. Careful selection of the operating conditions (T ≈ 65°C) and of the inert material (glass beads and dense alumina) could minimize these problems. The experimental results were in line with results of Hsiung & Thodos (1977). In particular, no effect of the fluidizing velocity on the mass transfer coefficient was observed over the complete range of bed materials. The experimental data of this work and those of Hsiung & Thodos (1977) were correlated by the following empirical expression (rearranged here in terms of the Sherwood number):

$$Sh = \left(0.105 + 1.505 \cdot \left(d_a/d_i\right)^{-1.05}\right) \cdot \left(d_a/d_i\right) \cdot \left[\left(1-\varepsilon_{mf}\right)/\varepsilon_{mf}\right] \cdot Re_{mfi}^{1-m} \cdot Sc^{1/3} \quad (3)$$

where $m = 0.35 + 0.29 \cdot \left(d_a/d_i\right)^{-0.50}$, and the Reynolds number at minimum fluidization was referred to the inert particle diameter $Re_{mfi} = \rho_g \cdot U_{mf} \cdot d_i / \left(1-\varepsilon_{mf}\right) \cdot \mu_g$. Coelho & Guedes de Carvalho (1988) made similar experiments with somewhat heavier naphthalene particles, and substantially confirmed results by Prins et al. (1985).

Pal'chenok & Tamarin (1985) and Tamarin et al. (1985) measured the mass transfer coefficients of single particles composed of a naphthalene spherical shell placed over a core made of different materials in order to vary the total density of the active particles. Bed materials of different size and density were used (glass spheres, corundum and fire clay). Experimental results indicated that for beds of large particles (d_i > 0.5 mm) the mass transfer coefficient was independent of the fluidization velocity. However, in the beds of fine particles the mass transfer coefficient slightly increased with the fluidization velocity and then levelled off at a maximum value. These authors proposed the following correlation for the maximum mass transfer coefficient:

$$Sh = 0.117 \cdot \left(d_i/d_a\right)^{0.13} \cdot \left(\rho_i/\rho_a\right)^{-0.15} \cdot Ar_i^{0.39} \cdot Sc^{1/3} \quad (4)$$

where the Archimedes number of the inert particles is $Ar_i = \left(\rho_i - \rho_g\right) \cdot \rho_g \cdot g \cdot d_i^3 / \mu_g^2$. Successively, Palchonok et al. (1992) suggested a new correlation valid over a wider range of particle sizes, for the special case $d_i = d_a$:

$$Sh = 2.0 \cdot \varepsilon_{mf} + 0.117 \cdot Ar_i^{0.39} \cdot Sc^{1/3} \quad (5)$$

A more general correlation was also presented by Palchonok (1998):

$$Sh = \left[\left(0.009 \cdot Ar_i^{0.5} \right) + \left(d_i / d_a \right)^{2/3} \cdot \left(2.0 \cdot \varepsilon_{mf} + 0.117 \cdot Ar_i^{0.39} - 0.009 \cdot Ar_i^{0.5} \right) \right] \cdot Sc^{1/3} \quad (6)$$

Joulié et al. (1986), Joulié & Rios (1993) and Joulié et al. (1997) carried out experiments of sublimation of naphthalene spheres with inert particles of two different densities (sand and zeolite). In their calculations they took into account the heat of sublimation and the difference between the active particle surface temperature and that of the bed. The experimental data were correlated using the following empirical expression:

$$Sh = 0.165 \cdot Ar_i^{0.55} \cdot \left(d_a / d_i \right)^{0.81} \cdot \left(\rho_a / \rho_i \right)^{2.70} \cdot \left[\left(U_{opt} - U_{mf} \right) / U_{mf} \right]^{1.91} \quad (7)$$

where U_{opt} represents the optimal gas velocity at which the maximum sublimation rate is found (U_{opt} was approximately 2 ~ 3 times U_{mf}). All the experiments correlated by Eq. 7 were carried out at $U = U_{opt}$. These authors, however, did not check the absence of adsorption effects by the bed materials used in their experiments. Oka et al. (1995) also carried out experiments of sublimation of naphthalene spheres in a fluidized bed of inert particles (corundum), and proposed a Frössling-type correlation:

$$Sh = 2.0 \cdot \varepsilon_{mf} + 0.97 \cdot Re^{0.60} \cdot Sc^{1/3} \quad (8)$$

The range of variation of the fluidization velocity and of the inert particle size in their experiments was rather small. This was true also for data reported by Donsì et al. (1998 and 2000), who found that the mass transfer coefficient was unaffected by the fluidization velocity for a particular bed particle size. They, however, correlated their data with a Frössling-type expression:

$$Sh = 2.0 + a \cdot \left(Re / \varepsilon \right)^b \cdot Sc^{1/3} \quad (9)$$

which predicts a significant influence of the fluidization velocity. In Eq. 9, a = 0.0086 and b = 1.34 according to Donsì et al. (1998), while a = 0.707 and b = 0.5 according to Donsì et al. (2000).

On the whole, the naphthalene sublimation technique appears to be fairly accurate in measuring the mass transfer coefficient of active particles in a fluidized bed, provided experiments are carefully performed. The main sources of inaccuracy are: mechanical attrition of the naphthalene particles; adsorption of naphthalene vapour by the inert solids; errors in the measurement of the mass loss of naphthalene particles (due to extraction, weighting and re-injection of the particles in the bed); errors in the measurements of the naphthalene vapour concentration in the gas (discontinuous or inaccurate analyzers); errors in the estimation of the vapour pressure and diffusion coefficient of naphthalene; incorrectness of the assumptions for the bulk gas naphthalene vapour concentration and for the naphthalene particle surface temperature; change of particle size during the sublimation process.

Other solid substances have also been proposed as an alternative sublimating component, like p-dichlorobenzene (Riccetti & Thodos, 1961), ice (Joulié et al., 1986) and dry ice (Schlichthaerle & Werther, 2000). The use of these substances, however, appears to be more complicated than naphthalene. Besides the previously reported inaccuracies, in fact, these substances are more difficult to mould, and sublimation is often accompanied by appreciable heat effects.

3.2 Liquid evaporation from porous particles

This technique is based on the measurement of the evaporation rate of a liquid from the surface of one or more porous particles in the fluidized bed. As for the previous technique, calculation of the mass transfer coefficient requires the knowledge of the vapour pressure and of the diffusion coefficient of the evaporating component at the operating temperature. In addition, if appreciable heat effects are associated to the evaporation process, a heat balance must be coupled to the mass balance (or particle temperature measured independently), complicating the calculation of the mass transfer coefficient.

Kettenring et al. (1950), Riccetti & Thodos (1961), Bradshaw & Myers (1963), Petrovic & Thodos (1966, 1967), Wilkins & Thodos (1969), Yoon & Thodos (1972), Schlünder (1977) and Ciesielczyk (1996) studied the mass transfer coefficient in fluidized bed composed of all active particles, by means of porous particles filled either with water, nitrobenzene, n-decane, n-dodecane or n-tetradecane. Ziegler & Brazelton (1964) and Ziegler & Holmes (1966) studied the evaporation of water from a single fixed sphere immersed in a fluidized bed of seven different materials. Recently Tsotsas (1994a and 1994b) studied the evaporation of water from fluidized beds of all aluminium silicate spheres. This author pointed out that because of the porous nature of the particles particular attention should be paid for the additional contribution of particle-side kinetics, especially for the second drying period (falling-rate), as opposed to the first drying period (constant-rate) when the particle surface is still wet. Kozanoglu et al. (2001) analyzed the evaporation of water at reduced pressure in fluidized beds of silica gel or millet particles.

Up to now the only works available in the literature where this technique was applied to few freely moving active particles within a fluidized bed of inert particles are those reported by Vanderschuren & Delvosalle (1980) and Delvosalle & Vanderschuren (1985), and by Cobbinah et al. (1987). The first authors estimated with the aid of a simple model the mass transfer coefficient in beds of refractory silica and alumina particles, where wet particles were dried in beds of dry particles of the same material and size. Interestingly, the mass transfer coefficient was found not to vary with the fluidization velocity, but only with the particle nature and size. Cobbinah et al. (1987), instead, dried wet coarse alumina spheres within fluidized beds of fine sand. These authors also found that the mass transfer coefficient was independent of the fluidization velocity. However, this happened only for fluidization velocities large enough so that no segregation of the coarse and fine particles occurred.

The advantages of this technique are the availability of cheap particles and liquids, the easy measurement of the gas concentration of the evaporating component and the constancy of the active particle diameter with time. On the other hand, apart from possible inaccuracies in measurements and in parameters evaluation, several severe drawbacks are present for a practical application. First, the heat balance must be typically solved together with the mass balance or the particle temperature must be measured as a consequence of the non-negligible heat of evaporation. Second, internal resistances to mass transfer in a porous particle may add serious errors in the estimation of the external mass transfer coefficient, if they are not properly taken into account. Third, as it was the case for the sublimation technique, the evaporating component can be adsorbed over the inert bed particles. Finally, the presence of a liquid layer on the surface of the active particles may disturb smooth fluidization by the action of capillary forces.

3.3 Combustion of carbon particles

This technique is based on the determination of the combustion rate of one or more carbon particles burning in a fluidized bed. This can be accomplished by the measurement of the weight change of the carbon particles and/or of the concentration of CO_2 and CO in the gas phase during combustion. The assumption is made that the carbon particle temperature and size are large enough so that the combustion rate is controlled by external mass transfer of O_2 towards the carbon surface. Alternatively, the intrinsic carbon reactivity and intraparticle mass transfer resistance must be properly considered in the calculations and separately quantified. Carbon particles which leave after combustion a coherent ash layer should be avoided for the experiments, as this would add a further resistance to mass transfer of O_2. Calculation of the mass transfer coefficient requires the knowledge of the diffusion coefficient of O_2 at the operating temperature. In addition, the heat balance must be solved together with the mass balance (or the particle temperature must be measured during the experiment) as a consequence of the appreciable heat effects of combustion. A significant additional complication is associated with the two following still unsolved questions: what are the primary combustion products at the carbon surface, CO_2, CO or both of them and in which relative proportion? Where is CO oxidized to CO_2, nearby or far from the carbon particle? These two issues affect both the mass transfer coefficient and the particle temperature (a large fraction of the total heat release may be associated with CO combustion). These issues have been reviewed in detail by La Nauze (1985), Prins (1987), and Agarwal & La Nauze (1989).

The first attempts to estimate the mass transfer coefficient during the fluidized bed combustion of carbon particles were reported by Avedesian & Davidson (1973), Chakraborty & Howard (1981), Pillai (1981), Tamarin et al. (1982) and Ross & Davidson (1982). These authors used the carbon particle burn-out time data to determine the particle Sherwood number in their experiments. As noted by La Nauze (1985), this method provides only the average Sherwood number during the burning time and does not allow any variation of Sh with the particle diameter. Further, the data analysis was based on a number of questionable assumptions regarding the carbon chemical kinetics, the CO oxidation reaction and the particle temperature. Avedesian & Davidson (1973), on the basis of their experimental results and of the estimation of the active particle Reynolds number (with the gas characteristic velocity in the dense phase assumed to be equal to U_{mf}), suggested that convective effects can be assumed to be negligible and that Sh for the burning particle should be close to its lower asymptotic value, i.e. at stagnant conditions. The authors proposed that in these conditions the limiting value of 2 should be multiplied by the bed voidage in the dense phase to account for the presence of the inert particles. If the two-phase fluidization theory (Davidson & Harrison, 1963) is assumed to hold, then $Sh = 2 \bullet \varepsilon_{mf}$. This result would be valid only in the case that the inert particle size is smaller than the active particle one. Basu et al. (1975) and Basu (1985) suggested that this value should be refined on account of the change of the local voidage with the radial distance from the active particle surface. This would result in a slightly larger value of Sh in stagnant conditions, because the voidage increases from the average bed value ε_{mf} far from the carbon particle to unity at the particle surface. Agarwal et al. (1988a), however, noted that given the poor accuracy of existing experimental data and the uncertainty in the estimation of physical properties, this refinement would be difficult to be verified. Chakraborty & Howard (1981) and Pillai (1981) proposed respectively the two Frössling-type correlations:

$$Sh = 2.0 \cdot \varepsilon + 0.69 \cdot Re^{1/2} \cdot Sc^{1/3} \tag{10}$$

$$Sh = \varepsilon \cdot \left(2.0 + 0.69 \cdot Re^{1/2} \cdot Sc^{1/3}\right) \tag{11}$$

They basically modified Eq. 1 (using the value suggested by Rowe et al. (1965) for the constant K) by introducing the mean bed voidage to account for the shielding effect of the particulate phase surrounding the burning particle. The arbitrary choice of the average bed quantities ε and Re, instead of the dense phase ones ε_{mf} and Re_{mf}, was not justified by the authors nor validated by their experimental data.

The first work in which the mass transfer coefficient was determined from combustion rate data was that reported by La Nauze & Jung (1982, 1983a) and Jung & La Nauze (1983). These authors performed experiments where single petroleum coke particles were burned in a fluidized bed and the mass loss of carbon was measured as a function of time. An independent measure of the chemical rate coefficient and of the particle temperature was available. Experimental results showed that particles with a size larger than 3 mm burned under external diffusion control and closely followed the shrinking sphere model. The authors correlated their results (assuming that no CO escaped the particle boundary layer) with a modification of Eq. 10:

$$Sh = 2.0 \cdot \varepsilon + 0.69 \cdot \left(Re/\varepsilon\right)^{1/2} \cdot Sc^{1/3} \tag{12}$$

The authors considered U/ε rather than U as the characteristic gas velocity relevant for convection around the burning particle. This correlation still predicts a large influence of the fluidization velocity, which however was not demonstrated in their experiments. In addition, the effect of the bed inert particle size was not taken into account. In a later paper La Nauze & Jung (1983b) suggested to change the first term from 2•ε to 2•ε_{mf}, on the basis of theoretical considerations. Additional experiments were also presented by La Nauze & Jung (1985, 1986), in which the oxygen-nitrogen fluidizing gas was substituted with an oxygen-helium mixture. In this condition the oxygen diffusivity was increased by a factor of about 2.5 times and, correspondingly, the mass transfer coefficient was found to be around two times larger.

Prins (1987) reported a comprehensive work where the burning rate of single graphite spheres in a fluidized bed was measured by following the CO_2 and CO concentrations in the flue gas. The author carefully considered and measured the effect of the burning particle temperature, CO/CO_2 primary ratio and CO oxidation, intraparticle oxygen penetration, and attrition, on the particle combustion rate. On the basis of the experimental results, it was concluded that a previously proposed correlation (Eq. 3) was able to correctly predict the mass transfer coefficient for the burning particle.

Guedes de Carvalho et al. (1991) measured with the same technique the burning rate of batches of coke or char particles in fluidized beds of sand or Pt doped catalyst beads at high temperature and at different pressures. In these conditions the combustion rate of carbon was controlled by external diffusion, and the intrinsic kinetics and intraparticle diffusion resistances were neglected. The particle temperature was not measured, but assumed to be 100 K above that of the bed. Experimental results indicated that the mass transfer coefficient was independent of the fluidization velocity. Comparing the combustion rates measured alternatively with beds of sand and of catalyst beads, the authors interpreted the

experimental data as an evidence of CO being the only primary product of carbon oxidation, followed by CO oxidation outside the particle boundary layer. On this basis, previously published mass transfer coefficient data were questioned and re-interpreted under the above assumption.

Salatino et al. (1998) measured the burning rate of single coal and biomass char particles in a fluidized bed of sand. The mass transfer coefficient was estimated on the basis of a simplified particle combustion model. The experimental results showed that Sh was not influenced by the fluidization velocity and oxygen inlet concentration, but only by the active and the inert particle size. The authors stressed the possible influence on the apparent burning rate of attrition and fragmentation of the char particle combined with fines post-combustion within the bed. Sh can be overestimated up to a factor of two (for porous and fragile chars, like those resulting from biomass devolatilization) if these phenomena are not taken into account. In a later paper, Scala et al. (2006) highlighted the significance of the shape of non-spherical particles on the correct evaluation of the exposed particle surface and of the mass transfer coefficient.

Paterson (2000) and Hayhurst (2000) noted that the implicit assumption of equimolar counter-diffusion of gaseous reactants and products around an active particle (which is typically made for the calculation of the mass transfer coefficient from experimental data) might not always be valid. For example, if CO is the only primary product of carbon combustion and it is further oxidized away from the carbon particle, assuming equimolar counter-diffusion (i.e. neglecting the Stefan flow) would lead to a 10 – 20% underestimation of the real mass transfer coefficient. The only case when the equimolar counter-diffusion approach can be safely used independently of the reaction stoichiometry is the case of very dilute gaseous reactant concentration. However, in this analysis it was assumed that the problem could be schematized as a pseudo-binary system and the presence of the other relevant gaseous species was not taken into account. Recently, Scala (2010a) considered the mass transfer coefficient for oxygen reacting with a spherical carbon particle in an atmosphere of O_2, N_2, CO_2, CO, and H_2O and analytically solved the complete set of Stefan-Maxwell equations under the assumption of negligible homogeneous reaction in the boundary layer. Results showed that under typical combustion conditions the use of the equimolar counter-diffusion mass transfer coefficient can lead to errors up to 10%.

Hayhurst & Parmar (2002) measured the burning rate of single freely moving graphite spheres and coal char particles in a fluidized bed of sand by following the CO_2 and CO concentrations in the flue gas. At the same time, the particle temperature was measured by a very thin and flexible thermocouple. The CO/CO_2 ratio as primary combustion products at the carbon surface was also estimated. The experimental results were quite scattered, and showed a slight decrease of Sh with the bed temperature (explained by the change of several physical properties of the system) and no clear trend with the fluidization velocity. The increase of Sh with the sand particle size was mainly attributed to the change of the minimum fluidization velocity. The experimental Sh data were correlated by the Frössling-type expression:

$$\mathrm{Sh} = 2.0 \cdot \varepsilon_{mf} + 0.61 \cdot \mathrm{Re}_p^{0.48} \cdot \mathrm{Sc}^{1/3} \tag{13}$$

where $\mathrm{Re}_p = \rho_g \cdot U_p \cdot d_a / \mu_g$, and U_p is the gas velocity in the particulate phase that was estimated from expressions available in the literature. Alternatively, the authors suggested

as a good approximation a modification of Eq. 12, where the bed values ε and Re are substituted by the dense phase ones ε_{mf} and Re_{mf}:

$$Sh = 2.0 \cdot \varepsilon_{mf} + 0.69 \cdot \left(Re_{mf}/\varepsilon_{mf}\right)^{1/2} \cdot Sc^{1/3} \tag{14}$$

More recently Dennis et al. (2006) showed that the interpretation of their own and of previous combustion rate measurements can suggest both a linear and a square root dependence of Sh from the active particle diameter, because of the scatter of data. The authors questioned the latter dependence (which is consistent with a Frössling-type expression) and preferred a linear one. This choice was justified by invoking the presence of a gas cushion underneath the active particle and a heap of defluidized sand resting on its upper surface.

Scala (2009) recently proposed a new indirect experimental technique to measure the product CO/CO_2 ratio at the surface of spherical coal char particles during fluidized bed combustion, based on the measurement of the burning rate of a single particle under low oxygen concentration conditions. In these conditions two advantages are obtained: the boundary layer mass transfer coefficient can be calculated without the need to account for high mass transfer rate and/or possible non-equimolar counter-diffusion corrections; heat effects are very limited and the char particle temperature can be assumed to be approximately equal to that of the bed. Scala (2010b) extended this technique and measured the primary CO/CO_2 ratio at the surface of coal char particles at different bed temperatures, oxygen concentrations, fluidization velocities, and inert bed particle sizes. In addition, the actual sphericity and temperature of the particle, as independently measured during the experiments, were taken into account in analyzing the data. Results showed that under all the experimental conditions investigated carbon was completely oxidized to CO_2 within the particle boundary layer. The experiments confirmed that the char particles burned under boundary layer diffusion control in the temperature range 800-900°C. The author concluded that single particle burning rate experiments can be used to estimate the particle Sherwood number in fluidized beds only if a high-reactivity fuel is used and if attrition can be assumed to be negligible. In fact, significant carbon attrition during the experiments would lead to a fictitious increase of the measured apparent particle Sherwood number.

In summary, despite the large amount of experimental data gained with the use of this technique, still some uncertainties exist on the accuracy of the estimated mass transfer coefficient values. Apart from possible errors in the measurements and in the parameters evaluation, the major limitations in the use of this technique are: the possible influence of intrinsic kinetics and intraparticle diffusion in the porous carbon on the overall combustion rate and on the particle conversion pattern; the correct evaluation of the carbon particle temperature; the assumption of the CO/CO_2 primary ratio and of the CO oxidation location; the influence of attrition and fragmentation of the carbon particles on the apparent combustion rate and on the particle number and size evolution; the change of carbon particle size with time by combustion; the use of non-spherical carbon particles.

3.4 Chemical reaction on the surface of catalyst particles

Recently, Venderbosch et al. (1998) proposed the oxidation reaction of CO with oxygen over a Pt catalyst as a model reaction to be used for the determination of mass transfer coefficients in gas-solid systems. It was shown with experiments in a fixed bed and in a riser

that this reaction is completely mass transfer controlled in the temperature range 450-500°C, even for very small active particles (< 100 μm) at low CO bulk concentrations. The authors highlighted a number of advantages of this technique like the easy preparation and stability of the catalyst particles and the absence of side reactions. They, however, did not apply the technique to fluidized beds.

Basically, this technique is based on the determination of the CO combustion rate on the surface of one or more catalyst particles dispersed in a fluidized bed. This can be accomplished by measuring the concentration of CO_2 and/or CO in the gas phase. Calculation of the mass transfer coefficient requires the knowledge of the diffusion coefficient of CO at the operating temperature. In addition, if high CO concentrations are used the heat balance must be solved together with the mass balance (or the particle temperature must be measured during the experiments) as a consequence of the appreciable heat effects of combustion.

Scala (2007) applied this technique to measure the mass transfer coefficient around freely moving active particles under bubbling/slugging fluidized bed conditions in a lab-scale reactor. In this work the mass transfer coefficient around one or few attrition-resistant Pt catalyst spheres immersed in an inert bed of sand was measured by following the CO oxidation reaction at 450°C at different fluidization velocities, catalyst sphere sizes and inert bed particle sizes. The experiments were performed using very low CO inlet concentrations (< 1000 ppm). In these conditions heat effects were negligible and the catalyst particle temperature could be assumed to be equal to that of the bed. Experimental results showed that Sh is not influenced by the fluidization velocity and by a change of regime from bubbling to slugging, whereas it increases with a square root dependence with the minimum fluidization velocity and with the active particle size. These results strongly suggest that the active particles only reside in the dense phase and never enter the bubble/slug phase. In addition, the reported 1/2 dependence of Sh on both U_{mf} and d_a is a double (independent) evidence of the applicability of the Frössling-type analysis to active particles dispersed in a fluidized bed. In particular, experimental data were excellently fitted by the following correlation (similar to Eq. 14, but with a slightly different coefficient):

$$\mathrm{Sh} = 2.0 \cdot \varepsilon_{mf} + 0.70 \cdot \left(\mathrm{Re}_{mf}/\varepsilon_{mf}\right)^{1/2} \cdot \mathrm{Sc}^{1/3} \tag{15}$$

A further interesting outcome of this work was that mass transfer around active particles in a fluidized bed of inert particles can be safely studied in a small lab-scale apparatus, since it appears not to be influenced by the bubble/slug fluid-dynamics, provided the local dense phase conditions are the same.

On the whole, this technique appears to be suitable for the accurate determination of mass transfer coefficients in fluidized beds in the temperature range 450-500°C. In fact, it allows to overcome most of the difficulties and uncertainties associated with other available techniques. The following advantages over the other techniques can be identified: a) only the heterogeneous oxidation reaction $CO + 1/2O_2 = CO_2$ at the catalyst particle surface is active and no parallel homogeneous or side reactions are present; b) the reaction is completely controlled by external mass transfer and no influence of intrinsic kinetics or intraparticle diffusion is present; c) at low CO concentrations heat effects are negligible and the catalyst particle temperature can be assumed to be equal to that of the bed; d) the CO conversion degree can be easily and accurately calculated by measuring CO and/or CO_2

concentrations at the reactor outlet; e) both CO and CO_2 are not adsorbed significantly by typical inert bed materials; f) at low CO concentrations, the Sherwood number can be calculated without the need to account for high mass transfer rate and/or non-equimolar counter-diffusion corrections; g) the Pt catalyst can be easily prepared and is very stable with respect to the oxidation reaction; h) the catalyst particles can be made with a spherical shape and keep a constant size during the experiments; i) by careful selection of the catalyst support material the particle attrition rate in the fluidized bed can be made negligible; j) properties of the different gaseous species and solid particles can be easily and accurately estimated. On the contrary, apart from possible inaccuracies in measurements and in parameters evaluation, the most critical issue is the selection of an attrition-resistant catalyst support. The occurrence of significant catalyst attrition would lead to an overestimation of the Sherwood number because of the continuous generation of new catalytic surface within the bed. Finally, if high CO concentration are used in the experiments, the catalyst particle temperature should be measured or estimated, and the high mass transfer rate and/or non-equimolar counter-diffusion corrections should be taken into account.

4. Mass transfer around active spheres in a fluidized bed: modelling studies

Contrary to the large amount of experimental work, only few theoretical investigations on mass transfer of active particles in fluidized beds of inert particles have appeared in the literature.

Tamarin (1982) applied the steady-state boundary layer theory for flow past a sphere to describe mass transfer to an active particle in a fluidized bed of inert particles. The average velocity gradient and the average tangential stress at the particle surface were determined with suitable simplifying assumptions. Correction to the gas velocity near the active particle was introduced to account for fluctuations of the impingement velocity and possible stagnant zones near the points of contact of adjacent particles. By relating the average stress to the particle weight, the following expression was obtained:

$$Sh = 0.248 \cdot \left(d_a / d_i\right)^{1/2} \cdot Ar_i^{1/3} \cdot Sc^{1/3} \tag{16}$$

La Nauze & Jung (1983b) and La Nauze et al. (1984) questioned the suitability of a steady-state approach to describe mass transfer around an active particle in a fluidized bed. They proposed an unsteady-state model where mass transfer in the dense phase was assumed to occur because of a gas convective component (due to gas percolating through the bed at minimum fluidization condition) and a particle convective component (due to packets of bed particles coming into contact with the active surface, and whose motion is induced by the bubbles). Solution of the unsteady equation with a suitable estimation of the gas renewal frequency at the surface of the particle gave:

$$Sh = 2 \cdot \varepsilon_{mf} + \left[\left(4 \cdot \varepsilon_{mf} \cdot d_a / \pi \cdot D\right) \cdot \left(U_{mf} / \varepsilon_{mf} + U_b\right)\right]^{1/2} \tag{17}$$

The authors also suggested that when the active particle size becomes comparable to the inert particle size the particle convective component can be neglected and the term U_b can be dropped from Eq. 17. Guedes de Carvalho & Coelho (1986) noted that in the derivation of Eq. 17 La Nauze and coworkers should have used the molecular diffusivity instead of the effective diffusivity, so that the correct equation should read:

$$Sh = 2 \cdot \varepsilon_{mf} + \varepsilon_{mf} \cdot \left[\left(4 \cdot d_a / \pi \cdot D \right) \cdot \left(U_{mf} / \varepsilon_{mf} + U_b \right) \right]^{1/2} \quad (18)$$

They also noted that in this formulation the gas renewal frequency by packets was likely to be overestimated, because a continuous stream of bubbles near the active particle was assumed. This last point was partly addressed in an alternative formulation by La Nauze & Jung (1985, 1986), where the particle convective gas renewal frequency was directly related to the bubble frequency. By matching the model with experimental data a proportionality parameter was estimated giving:

$$Sh = 2 \cdot \varepsilon_{mf} + \left[\left(4 \cdot \varepsilon_{mf} \cdot d_a / \pi \cdot D \right) \cdot \left(U_{mf} / \varepsilon_{mf} + 6.93 \cdot U_b \cdot \varepsilon_b \cdot d_a / d_b \right) \right]^{1/2} \quad (19)$$

It must be noted, however, that the points raised by Guedes de Carvalho & Coelho (1986) still apply to this later model formulation.

Coelho & Guedes de Carvalho (1988) presented a steady-state model of mass transfer around a large active particle in a packed or fluidized bed of smaller particles. A continuum approach was adopted to describe the flowfield and a potential flow solution was obtained. The assumption was made that no hydrodynamic boundary layer establishes around the particle. Transverse dispersion was considered to be more suitable to describe mass transfer rather than molecular diffusion. An analytical solution was obtained for the particular case of a thin concentration boundary layer around the sphere, with suitable simplifying assumptions. A numerical solution was necessary for the more general equation, and an empirical approximation was derived by matching the numerical model results and bridging the two exact solutions for the stagnant and thin boundary layer cases. For a fluidized bed, assuming that the active particle resides only in the dense phase, the expression becomes (Guedes de Carvalho et al., 1991):

$$Sh = \varepsilon_{mf} / \tau \cdot \left[4 + 0.576 \cdot Pe^{0.78} + 1.28 \cdot Pe + 0.141 \cdot \left(d_i / d_a \right) \cdot Pe^2 \right]^{1/2} \quad (20)$$

where the Peclet number was defined as $Pe = U_{mf} \cdot d_a \cdot \tau / \varepsilon_{mf} \cdot D$, and τ represents the bed tortuosity. In a later paper, Guedes de Carvalho & Alves (1999) examined the more general case where longitudinal dispersion is non-negligible, under the same assumptions of the previous model. The numerical solution was again approximated by an empirical expression representing the product of the solution for advection plus molecular diffusion and the enhancement brought about by convective dispersion. For a fluidized bed the expression becomes:

$$Sh = \varepsilon_{mf} / \tau \cdot \left(4 + 4/5 \cdot Pe^{2/3} + 4/\pi \cdot Pe \right)^{1/2} \cdot \left[1 + 1/9 \cdot \left(d_i / d_a \right) \cdot Pe \right]^{1/2} \quad (21)$$

All the models presented so far assume that the active particle resides only in the dense phase of the fluidized bed. Agarwal (1987) questioned this assumption and, basing on the particle movement patterns under the action of the raising bubbles, developed a model to estimate the probability (p) of the particles being in the dense phase. The author claimed that up to 20% of the life-time of the active particle may be associated to the bubble phase, depending on the density of the particle. On the basis of this particle circulation model Agarwal et al. (1988a, 1988b) developed a mass transfer model for a large active particle in a

bed of smaller inert particles. They assumed that the active particle resides alternately in the bubble and emulsion phases, so that:

$$Sh = p \cdot Sh_e + (1-p) \cdot Sh_b \tag{22}$$

where the relevant Sherwood numbers in the emulsion and bubble phases were estimated from a boundary layer model as:

$$Sh_e = K_p \cdot (C_D/8)^{1/3} \cdot (\tau/\varepsilon_{mf} \cdot Re_{mf})^{2/3} \cdot Sc^{1/3} \tag{23}$$

$$Sh_b = 2 + K_p \cdot (C_D/8)^{1/3} \cdot Re_b^{2/3} \cdot Sc^{1/3} \tag{24}$$

where $Re_b = \rho_g \cdot 3 \cdot U_{mf} \cdot d_a / \mu_g$ and $K_p = 0.69 \cdot \left[(1+Re \cdot Sc)^{1/3} - 1\right] \Big/ (Re \cdot Sc)^{1/3}$. The drag coefficient C_D and the parameter K_p were evaluated using Re_{mf} and Re_b respectively in the emulsion and bubble phase. Though the assumption underlying this model is somewhat fascinating, it must be noted that no direct evidence exists that (non-gas-emitting) active particles enter the bubble phase during their motion in the fluidized bed.

5. Comparison of available correlations with experimental data

The available empirical/theoretical correlations reported in the previous sections were checked for their accuracy in predicting the experimental mass transfer data of Scala (2007), which we consider to be the most complete and accurate reported in the literature to date. As a preliminary screening, all the correlations which predict a (direct or indirect) influence of the total fluidization velocity on Sh were discarded (Eqs 8-13, 17-19), as they are incompatible with the experimental data. Moreover, Eq. 5 predicts no influence of the active particle size on Sh and Eqs 4 and 6 predict a decrease of Sh with the active particle size, contrary to the experimental data. These correlations were also discarded. Finally, eq. 22 is based on the assumption that the active particle resides partly in the bubble phase, contrary to the experimental evidence, and therefore was not considered further.

Figures 1 and 2 report the comparison of the remaining correlations with the experimental data. As regards the empirical and semi-empirical correlations (Figs 1A and 2A), Eq. 15 excellently fits the experimental data. It is noted that Eq. 14 gives practically the same results as Eq. 15, and fits equally well the experimental Sh data. The purely empirical correlation by Prins et al. (1985), Eq. 3, fits very well the experimental Sh data at varying active particle size (Fig. 2A), but fits worse the Sh data at varying inert particle size (Fig. 1A). It must be noted, however, that for $d_i > 700$ μm the experiments are outside the range of validity of Eq. 3, as reported by the authors in their paper. Eqs 2 (Hsiung & Thodos, 1977) and 7 (Joulié et al., 1997) both significantly overpredict the Sherwood number. In Eq. 7 we have conservatively considered $U_{opt} = 2 \bullet U_{mf}$. Should the ratio U_{opt} / U_{mf} be > 2, the discrepancy of the predicted Sh with the experimental data would have been even larger. It is also worth to note that in Eq. 7 (but also in Eq. 4) the active particle density is explicitly present. In the experiments performed by Scala (2007) this variable was varied in a relatively limited range ($1050 < \rho_a < 1950$ kg/m^3), and within this range it appeared not to influence significantly Sh. It is our opinion that if particle segregation is avoided, the active particle density has no importance on mass transfer, but this speculation needs further experimental confirmation.

In Figs 1B and 2B the available theoretical correlations are compared with the experimental data. Equation 16 (Tamarin, 1982) significantly overpredicts the data. The two models by Guedes de Carvalho and coworkers (Eqs 20 and 21) give practically the same results and underpredict the data, except for the largest inert particle sizes. In these equations a value of $\tau = 1.414$ was used as suggested by the authors. A change of the bed tortuosity value in the range $1.0 < \tau < 1.414$, however, gave only a very limited benefit to the comparison with the experimental data. On the whole, it appears that no available fluidized bed mass transfer theoretical model is able to correctly predict the Sherwood number.

6. Conclusions

Four experimental techniques have been used so far to measure the mass transfer coefficient around active particles dispersed in a fluidized bed. The technique based on liquid evaporation from porous particles appears to be affected by severe limitations and its use is not recommended. The other three techniques, namely sublimation of solid particles, combustion of carbon particles, and chemical reaction on the surface of catalyst particles, all appear to be suitable for measuring Sh, provided experiments are carefully performed.

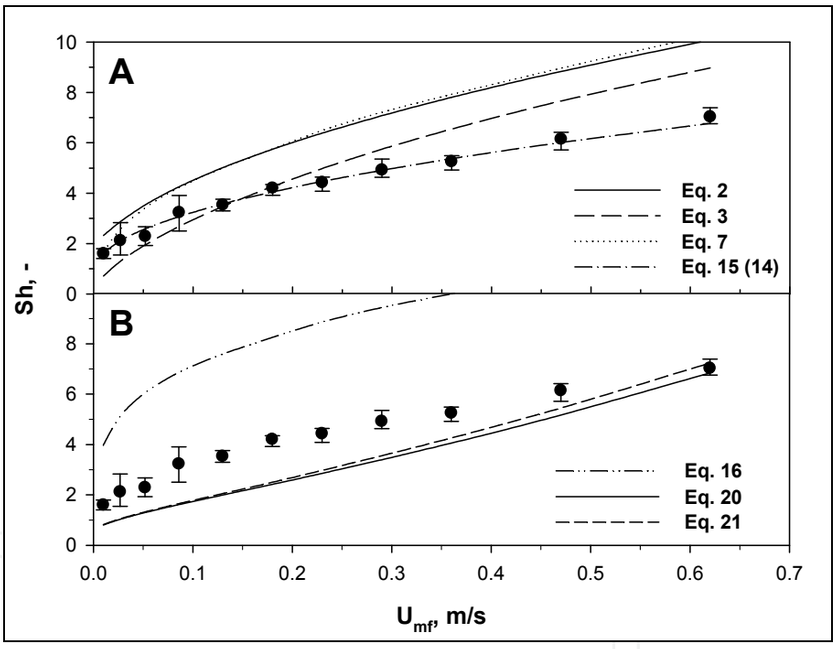


Fig. 1. Comparison of Sherwood number correlations with experimental data taken from Scala (2007). A) Empirical and semi-empirical correlations. B) Theoretical correlations. Average Sherwood number as a function of minimum fluidization velocity (d_a = 4.6 mm)

In particular, the most critical issues are connected to the need of minimizing particle attrition during the experiments, and of properly accounting for temperature differences between the active particle and the bed due to heat effects. For the technique based on the combustion of carbon particles a further critical aspect is the assumption of the CO/CO_2 primary ratio and of the CO oxidation location. Among these three techniques, we consider that based on chemical reaction on the surface of catalyst particles to be particularly simple and accurate, and for this reason we recommend its use whenever possible.

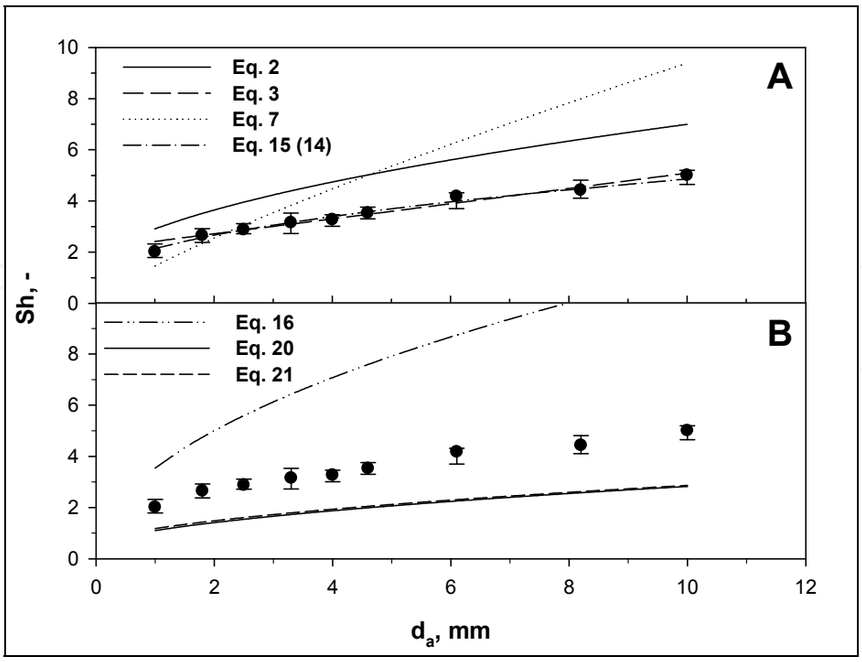


Fig. 2. Comparison of Sherwood number correlations with experimental data taken from Scala (2007). A) Empirical and semi-empirical correlations. B) Theoretical correlations. Average Sherwood number as a function of active particle size (500 μm < d_i < 600 μm)

Comparison of experimental data reported by Scala (2007) with empirical and semi-empirical correlations available in the literature showed that a Frössling-type correlation (Eq. 15) was able to fit excellently the data. The empirical correlation (Eq. 3) proposed by Prins et al. (1985) also fitted satisfactorily the data, but only when d_i < 700 μm. On the contrary, no available theoretical model could correctly predict the Sherwood number.

These results strongly suggest that in order to analyze the dependencies of Sh on the operating variables a Frössling-type analysis is the only one based on sound physical grounds. The first diffusive term accounts for mass transfer around the active particle in stagnant conditions. Contrary to the fixed bed case, however, this term has no strict physical meaning here, since a fluidized bed cannot exist in stagnant conditions (U = 0). So this term must be considered only as an asymptotic behaviour. For a free sphere in stagnant conditions Sh = 2. Two effects must be taken into account when we consider a fluidized bed. First, the inert bed particles decrease the volume available for mass transfer, which depends on the average void fraction around the active particle. This value can be assumed as a first approximation equal to ε_{mf}. A second effect, discussed by Coelho & Guedes de Carvalho (1988), implies that an effective diffusion coefficient D /τ should be used, where τ is the bed tortuosity accounting for the hindering effect of the granular bed on gas diffusion. So in principle the diffusive term should written as: $Sh = 2 \bullet \varepsilon_{mf} / \tau$. It must be noted, however, that to our knowledge no estimation of tortuosity exists for the dense phase of a fluidized beds. Given the theoretical rather than practical significance of this term, it is suggested here that $Sh = 2 \bullet \varepsilon_{mf}$ is a reasonable approximation for the diffusive term.

As regards the convective term, theoretical considerations suggest a 1/2 dependence on the Reynolds number and a 1/3 dependence on the Schmidt number. The first point here is the discrimination of the relevant particle Reynolds number to be used. On the basis of the

experimental findings, the relevant gas velocity around the active particle is the dense phase gas interstitial velocity. In most operating conditions this velocity can be safely approximated with U_{mf}/ε_{mf}. So the correct Reynolds number to be used in a Frössling-type expression should be Re_{mf}/ε_{mf}. Analysis of experimental data reported by Scala (2007) strongly support this point. In addition, the reported 1/2 dependence of Sh on both U_{mf} and d_a is an evidence of the soundness of this approach and also of the applicability of the Frössling-type analysis to a fluidized bed.
In spite of the relatively well understood behaviour of Sh with the main operating variables, several issues still remain open to debate, namely the effect of the active particle shape and density on mass transfer coefficient, and the value of Sh for the case $d_a/d_i < 1$.

7. Notation

a	constant in Eq. 9, -
Ar	Archimedes number, -
b	constant in Eq. 9, -
C_D	drag coefficient in Eqs 23 and 24, -
d	particle diameter, m
D	diffusion coefficient, m^2/s
g	gravity acceleration, m/s^2
k_g	mass transfer coefficient, m/s
K	constant in Eq. 1, -
K_p	parameter in Eqs 23 and 24, -
m	parameter in Eq. 3, -
p	probability of active particles being in the dense phase (Eq. 22), -
Pe	Peclet number, -
Re	Reynolds number, -
Sc	Schmidt number, -
Sh	Sherwood number, -
T	temperature, K
U	fluidization velocity, m/s

Greek letters

ε	bed voidage, -
ρ	density, kg/m^3
τ	bed tortuosity, -
μ	viscosity, kg/m•s

Subscripts

a	active particles
b	bubble or bubble phase
e	emulsion phase (Eqs 22 and 23)
g	gas
i	inert bed particles
mf	at minimum fluidization conditions
mfi	at minimum fluidization conditions referred to inert particles (Eq. 3)
opt	at optimal conditions (Eq. 7)

p particulate phase (Eq. 13)

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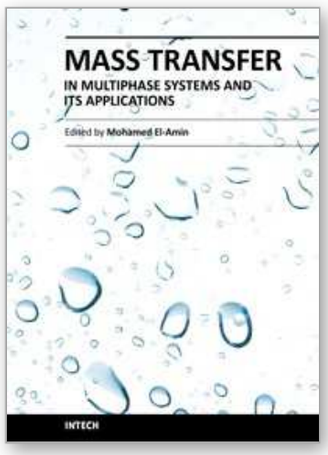
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This book covers a number of developing topics in mass transfer processes in multiphase systems for a variety of applications. The book effectively blends theoretical, numerical, modeling and experimental aspects of mass transfer in multiphase systems that are usually encountered in many research areas such as chemical, reactor, environmental and petroleum engineering. From biological and chemical reactors to paper and wood industry and all the way to thin film, the 31 chapters of this book serve as an important reference for any researcher or engineer working in the field of mass transfer and related topics.

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