HEAT TRANSFER FROM AN IMMERSED FIXED SILVER SPHERE TO A GAS FLUIDIZED BED OF VERY SMALL PARTICLES

by

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Results of unique heat transfer measurement in beds of fine, cracking catalyst particles, fluidized by air or helium gas, are compared with predictions from a theoretical model presented in the literature, and also with an earlier established empirical correlation. Moreover, the results have been related to dense phase flow conditions around a silver heat transfer probe by a simple turbulence model. A maximum heat transfer coefficient of h = 2300 W/m²K has been measured in a bed of 14μm (average diameter) particles, fluidized by helium gas. The data collected, and the model developed, can be used for the design of heat transfer tubes in fluidized beds of fine particles as for instance in fluid catalytic cracking (FCC) of crude oil heavy residues. The FCC is one of the most important conversion processes in the petroleum refineries.

Key words: *fluidization, heat transfer, heat transfer coefficient, immersed sphere, fine particles, fluid catalytic cracking fines, air, helium*

Introduction

Many industrially important processes are carried out in fluidized beds of fine particles (*e.g.* ferrite production from iron oxide, fluid catalytic cracking, *etc.*). These materials (Geldart's A-C powders) are often difficult to fluidize, unless special measures like the application of high fluidization velocities and/or the addition of coarse particles, are taken. The design of immersed heat exchanger tubes is based largely on the available information on heat transfer coefficients. Unfortunately however, the overwhelming quantity of experimental data on heat exchange between gas fluidized beds and an immersed surface has usually been obtained from measurements for beds of coarse particles ($d_p > 100$ μm). Moreover, the effect of the curvature radius of a heat exchanging surface is insufficiently considered in the literature. Just a few publications are dealing with heat exchange in a bed of particles finer than 100 μm, while none of them consider the heat exchange to an immersed spherical object [1-5]. In a previous article [6], the effect of the heat transfer probe dimension relative to the bed particle size has been investigated systematically, but for fluidized beds of coarse particles only. The present study examines the heat transfer between a single, immersed silver sphere (various diameters) and an air or helium fluidized bed of very fine, cracking catalyst particles.

It is important to note that the experimental data have been generated more than 30 years ago. They are recovered from conference proceedings [7] which are not available in the

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open literature. The work is still original and unique, but can neither be found back in on-line archives [8]. Over the past decades some publications appeared which are more or less related, demonstrating the relevance of the subject matter [8-12]. The present authors are currently preparing a more in-depth review article that will be published soon.

Heat transfer modelling

The main mechanism of heat exchange between a fluidized bed and an immersed surface is, as generally recognized, the convective contribution due to particle motion along the heat exchange surface. At moderate temperatures (T < 873 K) radiation will not contribute noticeably. Besides, for bed particles smaller than 500 μm the gas-convective contribution is usually unimportant, as can be estimated from the analogy with mass transfer, by applying the correlation of Prins *et al.* [13] for similar mass exchange conditions.

Xavier and Davidson [14] reviewed the currently published models for particle convective heat transfer. Not included is the model designed later by Martin [15]. This model deviates from the usual approach in which the gas-solid suspension is considered to be a continuous phase with composite so-called effective properties, see *e.g.* the early model concept of Mickley and Fairbanks [16]. Martin [15] applies a molecular-kinetic type of theory to the fluidized bed particles. His model contains a single unknown parameter, which has been determined by a comparison with experimental data. A comparison of the model predictions with experimental results obtained by various other investigators, turned out to be quite satisfactory. Variation of the heat transfer coefficient with bed particle size and bed voidage (or superficial gas velocity), as well as the effects of pressure and temperature, seem to be well predicted.

Results of heat transfer measurements reported in this paper will be compared with predictions from Martin's [15] model. Special attention will be paid to the particle size effect and the influence of the bed voidage (or superficial gas velocity). With respect to the first issue, the occurrence of an absolute maximum in the heat transfer coefficient, predicted by Martin's model [15] for particle diameters around 40 μm, will be verified. Concerning the influence of the bed voidage, experimentally determined actual values of ε and ε_d will be introduced into the model, instead of values estimated from the two-phase fluidization theory, as proposed by Martin [15].

Although sufficient experimental evidence is available [6] for the influence of the curvature radius of the heat exchanging surface on the heat transfer coefficient, unfortunately this parameter has no part in Martin's model [15]. Therefore we will also introduce a new heat transfer model based on the concept of the heat transfer to a quasi-continuous fluid, that is, to the suspension phase of the fluidized bed with its effective properties composed from those of the gas and the solid particles. Martin [15] criticizes this concept of the heat transfer to a quasi-continuous phase on the base of an estimate of the thermal boundary layer thickness, δ, around the heat exchanging surface. But that estimate, from which δ appears to be of the same order of magnitude as the bed particle diameter, is only correct for beds of relatively large particles.

The new heat transfer model proposed in this paper is a very simple one. It is based on the application of the Ranz-Marshall [17] equation derived for one phase flow along a single sphere in which the dimensionless Nusselt, Reynolds, and Prandtl numbers contain the effective properties of the emulsion phase (see list of symbols). Values for the effective density of the dense phase at various different fluidizing velocities can easily be obtained from the result of bed expansion measurements, if the original bed particle density is known. The effective heat capacity is assumed to be the same as the heat capacity of the bed particles. The effective heat conductivity, λ_e, can be calculated [18] from the widely accepted formula derived by Zehner and

Schlunder [19] (equation IX.51 of Ref. [20]). For the calculation of the effective viscosity of the emulsion phase, we propose:

$$\log(\eta_e) = 6.25\,(1-\varepsilon_d) - 3.88 \tag{1}$$

The equation was derived from the best fit of experimental data obtained by Matheson *et al*. [21], Langenberg-Schenk *et al*. [22], and Rietema [23]. Finally, the effective velocity of the emulsion phase is approximated with the relation for the average fluctuating velocity in an isotropic turbulent field, Davies [24], in which the energy dissipation per unit mass, E, is set equal to the power supplied to the emulsion phase by bubbles:

$$u = (Ed)^{0.33} \tag{2}$$

$$E = \mathrm{g}\,(U - U_d) \tag{3}$$

The approach of combining Kolmogoroff's theory of local isotropic turbulence with the energy input per unit mass of fluidum as a principle factor for the transfer process, has been successfully applied before, amongst others for aerated slurry systems, see *e.g*. Beenackers and van Swaaij [25]. The application for a fluidized bed should be considered as a first approximation only. Eventually, the heat transfer coefficient can be predicted by:

$$h = (1-\varepsilon_b)\frac{\lambda_e}{d}\left(2 + C\,\mathrm{Re}_e^{0.50}\,\mathrm{Pr}_e^{0.33}\right), \text{with} \tag{4}$$

$$\mathrm{Re}_e = \frac{\rho_e[\mathrm{g}d(U-U_d)]^{0.33}d}{\eta_e} \quad \text{and} \quad \mathrm{Pr} = \frac{\eta_e c_p}{\lambda_e} \tag{5}$$

A comparison of model predictions with experimental results should provide the value of C, the only unknown parameter of the model.

Experimental set-up and procedure

The experimental set-up used is shown in fig. 1. Air taken from the laboratory supply line, or helium gas supplied from a bottle battery, was passed through a water-filled bubble column to humidify the fluidizing gas up to 40% (relative humidity). Humidification of the fluidized gas was required to avoid the observed serious effect of static electricity on the heat transfer measurement. The fluidized bed column was constructed of glass (internal diameter 100 mm, length 750 mm) and contained a bed of 120 mm static height. The entering gas was distributed by a plate of sintered glass particles. After passing through bed and freeboard, the gas flow was led through a pair of cyclones to separate and recirculate the entrained bed particles.

Seven individual heat transfer probes have been constructed by cementing stiff Cr–Al thermocouples (1.5 or 0.5 mm diameter) to the centre of various sized silver spheres (4, 7, 9, 11, 15, 18, and 30 mm diameter). After the adjustment of desired experimental conditions, a single heat transfer probe, initially at a temperature of around 333 K, was introduced into the cold (T_b = 293 K) fluidized bed and kept at a fixed position in the centre while its temperature change was followed until it finally reached the bed temperature. A time average heat transfer coefficient could be determined for each cooling curve, as described in detail in our earlier paper [6]. Silica-alumina catalyst particles were used as a bed material. In this paper "FCC" is used as an acronym for the batch with an average bed particle diameter of 103 μm (this is the reference bed material), while FCC-fines indicates particles with an average bed particle diameter of 14 μm. No special measures have been taken to improve the fluidization behaviour of the cohesive FCC-fines. The particle density, ρ_p, of the bed material is 1830 kg/m^3, the heat capacity c_p = 1060 J/kgK and the heat conductivity λ_p = 0.36 W/mK ([14], p. 148). Several hundred measurements have been carried out to observe the influence of: the superficial gas velocity, the average bed particle

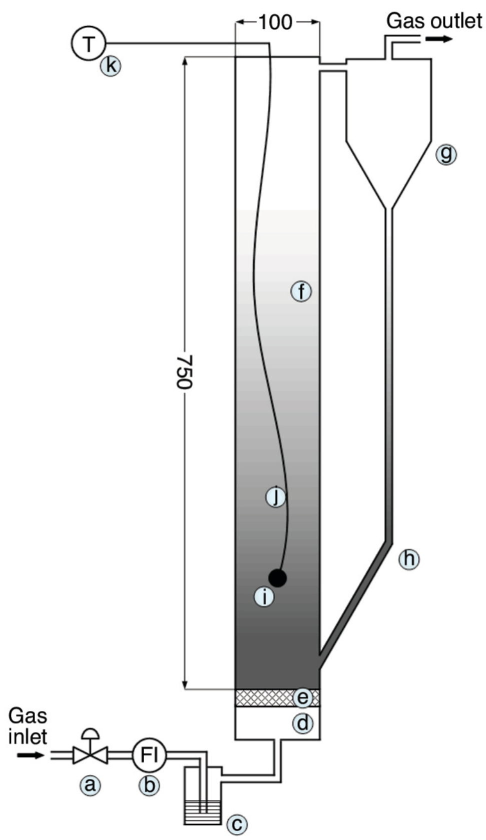

Figure 1. Sketch of the set-up of the fluid bed unit: *(a) reduction valve, (b) critical flow orifice, (c) humidifier, (d) gas box, (e) gas distributor plate, (f) fluidized bed, (g) cyclone, (h) return leg, (i) silver sphere, (j) thermocouple wire, and (k) digital temperature meter*

diameter, the type of fluidizing gas, and the diameter of the heat transfer probe. To find ε_d and ε_b and enable a proper interpretation of experimental results, the fluidized beds have been characterised further by collapse experiments performed by rapidly cutting off the gas supply and registration of the bed height during settling [26].

Results and discussion

Experimentally determined values of the total, bubble-phase and dense phase-void fraction, presented in fig. 2, are used for the model calculations of this paper. In figs. 3-5, results of the heat transfer measurements are compared with predictions from Martin's [15] model (solid blue curves) and the presently proposed simple model (dashed black and red curves). A value of 1.78 has been found for the parameter C of the present model, see eq. (4) by minimizing the average difference between predicted and experimental results for the air-fluidized-bed measurements.

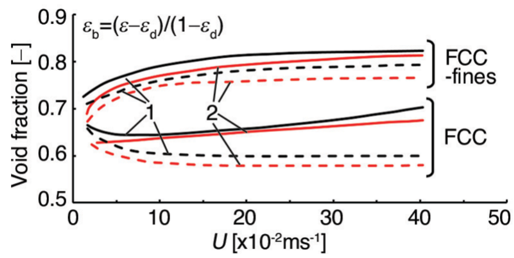

Figure 2. Results of bed-collapse experiments presented as a plot of total and dense phase voidage against the superficial gas velocity. Black (1) fluidization in air, red (2) fluidization in helium, solid lines - ε, dashed line - ε_d)

It is a well-known phenomenon that, for a certain bed material, the heat transfer coefficient passes through a maximum with increasing superficial gas velocity, due to the competition between the enhancing effect of increased bed turbulence and the unfavourable influence of increased bed porosity. Considering the experimental data points of figs. 3 and 5, a flat maximum can be observed around $U = 0.25$ m/s in case of the FCC bed material. For FCC-fines, however fig. 4, the maximum has not been completely reached. Apparently, high fluidization velocities are required to attain full bed turbulence for such a cohesive powder. Martin's model [15] predicts a maximum in the heat transfer coefficient, approximately at $U = 0.1$ m/s, for the air fluidized beds only (solid blue curves, figs. 3 and 4). However, none of the dashed curves in figs. 3-5 shows a maximum within the velocity range $0.05 < U < 0.45$ m/s. For ultra-fine particles neither Martin's nor the present model is able to predict the precise form of the h *vs.* U curve accurately. As can be seen in figs. 3-5, our simple model is deviating significantly from the experimental data in the region of low superficial gas velocities (below 0.05 m/s). On the other hand, at superficial gas velocities close to 0.5 m/s, the model still predicts the heat transfer coefficients fairly well.

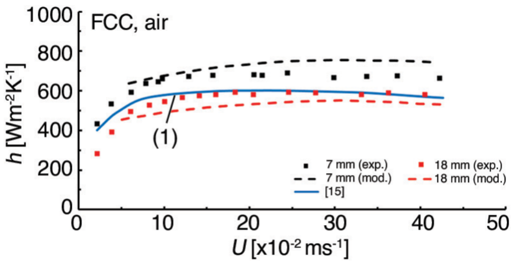

Figure 3. Heat transfer coefficient as a function of the superficial gas velocity for a 7 mm and 18 mm diameter sphere, in an air fluidized bed of FCC at a relative humidity of 40 %; dashed lines have been calculated from the present model; the line (1) shows the prediction by Martin's model [15]

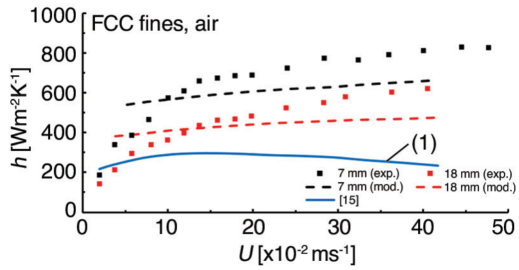

Figure 4. Heat transfer coefficient as a function of the superficial gas velocity for a 7 mm and 18 mm diameter sphere, in an air fluidized bed of FCC -fines at a relative humidity of 40%; dashed lines have been calculated from the present model; the line (1) shows the prediction by Martin's model [15]

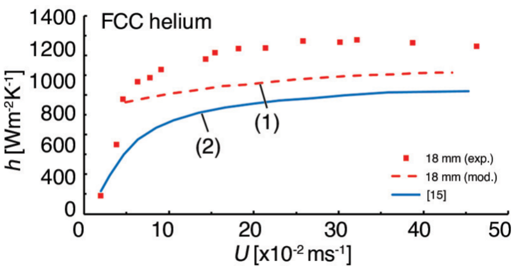

Figure 5. Heat transfer coefficient as a function of the superficial gas velocity, of a 18 mm diameter sphere in a helium fluidized bed of FCC at a relative humidity of 40 %; the red line (1) has been calculated from the present model; the line (2) represents the prediction by Martin's model [15]

Nevertheless a significant extrapolation to much higher superficial gas velocities should be discouraged. The model has been developed primarily for the turbulent fluidization regime. It should not be used for superficial gas velocities close to, or below the minimum bubbling velocity and neither for a situation where the turbulent bed has largely disappeared due to excessive entrainment (finally ending up pneumatic transport).

The diameter of a heat transfer cylinder or sphere has an important effect on the heat transfer rate [6, 27]. This has now been confirmed again by the results of the present measurements for fine bed particles, as reported in figs. 6 and 7. They show that the probe diameter dependency of the heat transfer coefficient approximately fits a proportionality of $h \sim d^{-0.2}$, which is in good agreement with the earlier results obtained for coarse bed materials [6]. The present model predicts this dependency reasonably well, as can be seen in figs. 3, 5, and 7. At the time Fluidization VI was held (1998), this was a unique feature of the model: the influence of the immersed surface radius was never considered in fluidized bed heat transfer modelling [28, 29]. Later on, correlations which account for the heat transfer probe (sphere) diameter became more common [9,11,12, 30-34]. The fluidized bed heat transfer coefficient increases for smaller bed particle diameters, as a consequence of an increased contact surface area per unit bed volume which shortens the transfer paths between particles and heat transfer surface. This effect however, will be suppressed by the considerable increase in dense phase porosity which occurs upon entering the regime of ultra-fine bed particles. Martin [15] suggests that for the fine particles regime the heat transfer rate will gradually become limited by the particle volumetric heat capacity. In that case, the heat transfer coefficient would fall again with further decreasing particle size. Therefore, this model predicts a maximum in the *h vs.* d_p curve, viz. around 40 μm for air fluidized beds of glass beads (and of FCC as well). The effect of the bed particle size can be recognized by comparing results for FCC (d_p = 103 μm) and FCC-fines (d_p = 14 μm). Figure 3 shows that Martin's model, like the present model, predicts the values of the heat transfer coefficient rather well in case of FCC as a bed material. However, in contrast with the present model, Martin's model prediction fails in the case of FCC-fines as illustrated by fig. 4. Much too low values of h are given by the solid blue curve, while the dashed curves fit the experimental data points more successfully. Figure 6, in which results are presented obtained from the helium-fluidized-bed measurements, also indicates that higher maximum values of the heat transfer coefficient are measured if the average bed particle size is reduced from 103 μm (FCC) to 14 μm (FCC-fines). Martin's model seems not to be valid for very fine particles.

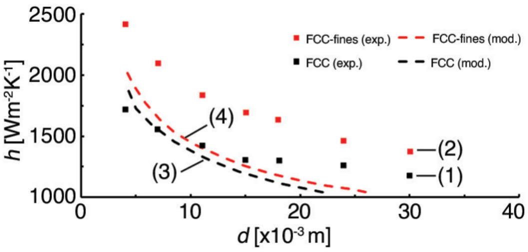

Figure 6. Maximum heat transfer coefficient as a function of the immersed sphere diameter in beds fluidized by helium gas at a relative humidity of 40%; black squares (1): FCC-fines, d_p = 14 μm, U = 0.45 m/s; red squares (2): FCC, d_p = 103 μm, U = 0.35 m/s; dashed lines are predicted by the present model: black (3) for FCC-fines, red (4) for FCC; Martin's model [15] predicts a constant value of 330 and 1000 W/m²K for FCC-fines and FCC, respectively

Application of helium as a fluidizing gas causes a drastic increase in the heat transfer coefficient. A maximum value of 2300 W/m²K has been measured for a 4 mm diameter sphere immersed into a helium fluidized bed of FCC-fines fig. 6. As an average, the measured values of h appear to be roughly 2.2 times higher than in case of air fluidized beds of the same material corresponding to a factor $(\lambda_{\mathrm{He}}/\lambda_{\mathrm{air}})^{0.45}$. This influence of the gas conductivity is relatively high, but has been observed several times before; the often applied empirical correlation of Zabrodsky [35] predicts $h \sim (\lambda_{\mathrm{He}}/\lambda_{\mathrm{air}})^{0.6}$. The present model underpredicts the experimental values for helium fluidized beds by approximately 30%. It should be noted that these predictions strongly rely on the estimate of the effective emulsion-phase conductivity according to equation IX. 51 of [20]. It could be subject of future work to determine λ_e by proper experiments.

The results of the present measurements for air fluidized beds of FCC and FCC-fines have been plotted in fig. 6 as values of $\mathrm{Nu}_{\max}\mathrm{Ar}^{-n}f_T^{-1}$ *vs*. the ratio of heat transfer sphere and bed particle diameter d/d_p, for a comparison with the earlier obtained [6] empirical correlation for fixed heat transfer spheres, eq. (6). Here the subscript *max* always refers to the maximum values observed when varying the superficial gas velocity:

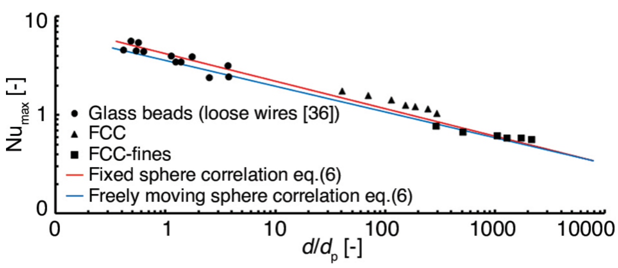

Figure 7. Comparison of data points from the present measurements and those of Turton *et al.* [36] (for fine loose wires), with Prins *et al.*'s [6] correlation for heat transfer from an air fluidized bed to a single immersed sphere

$$\mathrm{Nu}_{\max}\,\mathrm{Ar}^{-n} f_T^{-1} = 4.175\left(\frac{d}{d_p}\right)^{-0.278} \tag{5}$$

$$n = 0.087\left(\frac{d}{d_p}\right)^{0.128} \tag{6}$$

For (almost) freely moving spheres a similar equation was derived [6], differing only marginally in the values of the constants, and represented in fig. 7 by the lower line. Shown in fig. 7 as well are results of measurements by Turton *et al.* [36], who determined heat transfer coefficients to immersed, loose wires.

All the data points, including these of Turton *et al*. [36], are in close agreement with the prediction of the empirical correlation eq. (6). This demonstrates the usefulness of the simple correlation to estimate maximum heat transfer coefficients for air fluidized beds over an extended range of experimental conditions.

Conclusions

- Martin's [15] model underpredicts the maximum heat coefficient for an air fluidized bed of ultra-fine particles (d_p = 14 μm) almost by a factor 3. No indication has been found confirming Martin's suggestion that h would decrease considerably upon entering the regime of ultra-fine particles.
- A tentative model based on emulsion phase flow conditions around the immersed sphere, and leading to a Ranz-Marshall type equation with a constant C = 1.78, gives satisfactory predictions of the level of maximum heat transfer.
- This tentative model appears to be applicable for air fluidized beds of FCC particles and FCC fines, within the superficial gas velocity range from 0.05 to 0.5 m/s. In contrast with any other heat transfer model, it accounts for the variation of immersed sphere diameter.
- No model predicts the measured, extremely high heat transfer coefficients for helium fluidized beds accurately; the present model predicts values which are about 30 % too low.
- The validity range of an earlier obtained [6] empirical correlation for maximum heat transfer in air fluidized beds has been extended considerably by the results of Turton *et al*. [36] for fine, loose wires, and by the results of the present measurements.

Acknowledgment

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Nomenclature

Ar – Archimedes number (= $gd_p^3\rho_p/\rho\nu^2$), [–]
C – constant in eq. (4), [–]
c_p – specific heat of the bed particles, [Jkg^{-1}K^{-1}]
d – diameter of the heat transfer sphere, [m]
d_p – average bed particle diameter, [m]
E – energy dissipation per unit mass, [Js^{-1}kg^{-1}]
f_T – temperature correction factor, [–]
g – acceleration of gravity, [ms^{-2}]
Nu_{max} – Nusselt number in eq. (6), (= $h_{max}d_p/\lambda$) [–]
n – exponent of the Archimedes number, eq. (6a) [–]
Pr – Prandtl number, eq. (5), [–]
Re – Reynolds number, eq. (5), [–]
T_b – fluidized bed temperature [K]
U – superficial gas velocity, [ms^{-1}]
U_d – interstitial gas velocity in the dense phase, [ms^{-1}]
u – average fluctuation velocity of the dense phase, [ms^{-1}]

Greek symbols

ε – total void fraction of the fluidized bed [–]
ε_b – volume fraction of the bubbles [= $(\varepsilon - \varepsilon_d)/(1 - \varepsilon_d)$] [–]
ε_d – void fraction of the dense phase [–]
η_e – effective viscosity of the dense phase [kgm^{-1}s^{-1}]
λ – heat conductivity of the fluidizing gas, [Js^{-1}m^{-1}K^{-1}]
λ_e – effective heat conductivity of the dense phase [Js^{-1}m^{-1}K^{-1}]
λ_p – heat conductivity of the bed particles [J s^{-1}m^{-1}K^{-1}]
ν – kinematic viscosity of the fluidizing gas, [m^{2}s^{-1}]
ρ – density of the fluidizing gas, [kgm^{-3}]
ρ_e – effective density of the dense phase [= $(1-\varepsilon_d)\,\rho_p$] [kg m^{-3}]
ρ_p – density of the bed particles, [kgm^{-3}]

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