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Modelling of power law liquid–solid mass transfer in packed beds at Darcy regime

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Abstract

A model of liquid-to-particle mass transfer in packed beds with single phase non-Newtonian creeping flow is derived using a coherent approach which associates the Lévêque solution with a capillary-type representation of the porous structure. The model is compared to experimental data obtained with spherical and anisotropic parallelepipedal particles, respectively, as well as to literature results. The model allows to predict mass transfer in fixed beds of spheres and anisotropic parallelepipedal particles with a mean relative error of 11%. It is also shown that a better prediction of mass transfer is obtained for parallelepipedal particles if the surface area really offered to fluid flow is considered. Dimensionless numbers based on the pore dimension are shown to be more suitable than those based on a particle diameter.

Keywords: Mass transfer; Packed beds; Creeping flow; Non-Newtonian fluid

1. Introduction

Non-Newtonian liquid–solid mass transfer in packed beds remains a non-widely explored field. Few research contributions have been devoted to experimental studies and theoretical developments on the subject, as previously noticed by Chhabra [1,2]. The pioneering work of Kumar and Upadhyay [3] deals with mass transfer in fixed beds of spheres and fixed or fluidized beds of pellets; it is still considered as the reference work probably because of the quantity of data that are published. Few authors have published experimental data since. In particular, Coppola and Böhm [4] present mass transfer data in packed beds of screens; Wronski and Szembek-Stoeger [5] and Hwang et al. [6] propose new data concerning fixed beds and fluidized beds of cylindrical pellets respectively. All these works are exploring a range of Reynolds numbers which covers a part of the Darcy's regime and which is extending to the non-linear laminar flow regime. As far as theoretical aspects are concerned, Kawase and Ulbrecht [7,8] have proposed several developments for the prediction of the mass transfer coefficient. In [7], the authors use a boundary layer-type approach in order to obtain a dimensionless equation for Sherwood number at relatively high Reynolds numbers. The predicted mass transfer coefficients are of the order of magnitude of the experimental data produced by Kumar and Upadhyay. However, the difference between experimental and predicted mass transfer coefficients seems to increase at the extremal parts of the explored range of Reynolds numbers. Two years later [8], the authors publish an equation resulting from a completely different approach, associating the capillary-type approach of Blake and Kozeny with the Lévêque solution for the flow of a power law liquid in a cylindrical tube. Comparing again the obtained equation for the mass transfer coefficient with experimental data of Kumar and Upadhyay, they notice a better and satisfying fitting of the model with experiment, but obviously limited to the creeping flow regime. In 1992, Kawase [9] proposed a new version of his previous model [7] based on the boundary layer approach in order to improve its predictive capability in the case of beds of spheres. In this new version, he takes into account the tortuosity concept when defining the main stream velocity around a sphere and the characteristic length of the sphere.

In a previous work [10], we discussed the equations available in the literature concerning the case of Newtonian fluid flow in creeping flow regime conditions. In particular, we focused on the proposed models and emphasized the fact

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Nomenclature

A_{vd}	dynamic specific surface area (m^{-1})
A_{vs}	geometrical specific surface area (m^{-1})
d	tube diameter (m)
d_{part}	particle diameter (m)
d_{pore}	equivalent diameter of pore (m)
D	diffusion coefficient ($m^2\,s^{-1}$)
ERM	mean relative absolute error
h	height of a layer of particles (m)
J	mass transfer factor
k_d	mass transfer coefficient ($m\,s^{-1}$)
K	consistency index ($kg\,s^{n-2}\,m^{-1}$)
l	tube length (m)
l_c	equivalent length for mass transfer (m)
m	number of data
n	flow behaviour index
R	aspect ratio of flat plates, i.e. thickness to side ratio
Re	tube Reynolds number
Re_{part}	particle Reynolds number
Re_{pore}	pore Reynolds number
Sc	tube Schmidt number
Sc_{part}	particle Schmidt number
Sc_{pore}	pore Schmidt number
Sh	Sherwood number
Sh_{part}	particle Sherwood number
Sh_{pore}	pore Sherwood number
T	shear stress ($kg\,s^{-2}\,m^{-1}$)
u_0	superficial velocity ($m\,s^{-1}$)
u_{pore}	mean velocity in the equivalent pore ($m\,s^{-1}$)

Greek letters

$\alpha(\varepsilon)$	fraction of the diameter corresponding to a layer of spheres
$\dot{\gamma}$	shear rate (s^{-1})
ε	porosity
$\mu_{eff\ part}$	particle effective viscosity of non-Newtonian fluid ($kg\,s^{-1}\,m^{-1}$)
$\mu_{eff\ pore}$	pore effective viscosity of non-Newtonian fluid ($kg\,s^{-1}\,m^{-1}$)
τ	tortuosity

that theoretical models mostly deal with spherical particles. We used the approach followed by Kawase and Ulbrecht [8] based on combination of the analytical solution of Lévêque equation for mass transfer in short pipes, and a capillary representation of porous media. The main difference with the previous model of Kawase and Ulbrecht consists in the chosen capillary representation of porous media. Our model was shown to conveniently predict mass transfer in Darcy's regime in packed beds of particles presenting an isotropy in the main direction of flow, i.e. spheres and flat plates.

In the present work, an extension of this model to the case of non-Newtonian power law fluids is developed. Its validity is discussed and compared to experimental results obtained in beds packed with spheres and parallelepipedal plates, respectively.

2. Model development

2.1. General equation

As in [8], the starting point is the Lévêque solution for a developed laminar power law fluid flow in a short cylindrical tube of length l and diameter d:

$$Sh = 1.615\left(\frac{3n+1}{4n}\right)^{1/3}\left(\frac{Re\,Sc\,d}{l}\right)^{1/3} \quad (1)$$

where n is the flow behaviour index in the power law model. This equation is obtained from an analogy between heat and mass transfer, in particular between Sherwood and Nusselt numbers. The global Nusselt number corresponding to the characteristic length of the tube, l, is resulting from the integration of the local Nusselt number at the entrance of a tube with the hypothesis of a constant wall temperature [11].

This equation is applied to the equivalent pores defined in the capillary-type model of Comiti and Renaud [12]. The pore diameter is defined as:

$$d_{pore} = \frac{4\varepsilon}{(1-\varepsilon)A_{vd}} \quad (2)$$

where A_{vd} is the specific dynamic surface area defined as the ratio of the surface area offered to the fluid flow to the solid volume of porous material. It can be obtained, as well as a second structural parameter, i.e. the tortuosity τ, from permeametry measurements [12]. Its interest is to characterize the part of the surface area which is effectively active in convective mass transfer conditions.

The following equation is then obtained:

$$Sh_{pore} = 1.615\left(\frac{4\varepsilon}{(1-\varepsilon)A_{vd}l_c}\right)^{1/3}(Re_{pore}\,Sc_{pore})^{1/3} \times \left(\frac{3n+1}{4n}\right)^{1/3} \quad (3)$$

where l_c is the characteristic pore length for mass transfer and Re_{pore} and Sh_{pore} are, respectively, the pore Reynolds number and the pore Sherwood number. They are based on the pore diameter and the average fluid velocity u_{pore} defined in the capillary representation by:

$$u_{pore} = \frac{u_0\tau}{\varepsilon} \quad (4)$$

where u_0 is the superficial velocity of the fluid.

The pore Reynolds number is derived from the tube Reynolds number defined by Dodge and Metzner [13] for

non-Newtonian fluid flow:

$$Re_{\text{pore}} = \frac{\rho d_{\text{pore}} u_{\text{pore}}}{\mu_{\text{eff pore}}} = \frac{\rho d_{\text{pore}}^n u_{\text{pore}}^{2-n}}{K((3n+1)/4n)^n 8^{n-1}}$$

Using Eq. (2), we get

$$Re_{\text{pore}} = \frac{\rho \varepsilon^{2n-2} (\tau u_0)^{2-n}}{K((3n+1)/4n)^n 2^{n-3} (1-\varepsilon)^n A_{\text{vd}}^n} \quad (5)$$

The pore Schmidt number is calculated as

$$Sc_{\text{pore}} = \frac{\mu_{\text{eff pore}}}{\rho D}$$

where $\mu_{\text{eff pore}}$ is the effective dynamic viscosity deduced from (5).

Hence

$$Sc_{\text{pore}} = \frac{K((3n+1)/4n)^n}{\rho D} \left[\frac{2(1-\varepsilon) A_{\text{vd}} \tau u_0}{\varepsilon^2} \right]^{n-1} \quad (6)$$

As it can be noticed, the Schmidt number depends on the pore velocity value.

The pore Sherwood number is given by

$$Sh_{\text{pore}} = \frac{k_{\text{d}} d_{\text{pore}}}{D} = \frac{4\varepsilon}{(1-\varepsilon) A_{\text{vd}}} \frac{k_{\text{d}}}{D} \quad (7)$$

The previous study concerning Newtonian flows [10] enabled to show that for stratified packed beds in which the particles are nearly axisymmetrical towards the flow, the characteristic length for mass transfer, l_{c}, can be expressed as:

$$l_{\text{c}} = \tau h \quad (8)$$

where h is the height of a layer of particles. For tightly packed beds of parallelepipedal particles, the thickness of the plates is a good estimation for h. For spherical particles, the thickness of a layer of particles depends on the spheres arrangement and thus depends on the porosity of the packing:

$$l_{\text{c}} = \tau \alpha(\varepsilon) d_{\text{part}} \quad (9)$$

For $0.26 \leq \varepsilon \leq 0.48$, $\alpha(\varepsilon)$ lies between 0.816 and 1.00 [10].

In this work, the validity of expressions (3) and (8) will be discussed and compared to experimental results.

2.2. Specific equation for packed beds of spheres

In packed beds of spheres, since the particle to particle contacts are mainly points, the specific dynamic surface area can be postulated to be identical to the geometrical surface area, A_{vs}. Then $A_{\text{vd}} = 6/d_{\text{part}}$, where d_{part} is the particle diameter. The equivalent expression of Eq. (7) based on the particle diameter is given by

$$Sh_{\text{part}} = 1.615 \left(\frac{3(1-\varepsilon)}{2\varepsilon^2} \right)^{1/3} \left(Re_{\text{part}} Sc_{\text{part}} \frac{d_{\text{part}} \tau}{l_{\text{c}}} \right)^{1/3} \times \left(\frac{3n+1}{4n} \right)^{1/3} \quad (10)$$

where

$$Re_{\text{part}} = \frac{\rho u_0 d_{\text{part}}}{\mu_{\text{eff part}}} = \frac{\rho \varepsilon^{2n-2} (u_0)^{2-n} d_{\text{part}}^n}{K((3n+1)/4n)^n 12^{n-1} (1-\varepsilon)^{n-1}}$$

and $\mu_{\text{eff pore}}$ is a function of interstitial velocity u_0/ε:

$$Sc_{\text{part}} = \frac{\mu_{\text{eff part}}}{\rho D} = \frac{K((3n+1)/4n)}{\rho D} \left[\frac{3(1-\varepsilon)(3n+1) u_0}{n \varepsilon^2 d_{\text{part}}} \right]^{n-1} \quad \text{and}$$

$$Sh_{\text{part}} = \frac{k_{\text{d}} d_{\text{part}}}{D}$$

Following the previous indications given in [10] concerning the different configurations of spheres packings, one may replace Eq. (10) by the following simplified expression:[3]

$$Sh_{\text{part}} = \frac{1.16}{\varepsilon} (Re_{\text{part}} Sc_{\text{part}})^{1/3} \left(\frac{3n+1}{4n} \right)^{1/3} \quad (11)$$

This equation can be applied to bed porosities that lie in the range $0.26 \leq \varepsilon \leq 0.48$.

3. Model validation

3.1. Experimental study

3.1.1. Experimental apparatus

Mass transfer experiments were previously carried out by Hilal et al. [14] using the electrochemical polarographic technique with packed beds of 2, 3, 4 and 5 mm spheres, respectively, as well as a bed tightly packed with square based parallelepipedal plates of 1.045 mm of thickness and 5.00 mm in side (aspect ratio, $R = 0.209$). The power law fluid used was a mixture of potassium ferricyanide (1 mol m^{-3}), potassium ferrocyanide (10 mol m^{-3}), carboxymethyl cellulose (CMC) sodium salt (6 kg m^{-3}), $NaHCO_3$ (100 mol m^{-3}) and Na_2CO_3 (100 mol m^{-3}). At 25 °C, the density, ρ, of this solution was 1022 kg m^{-3}. The ferricyanide ion diffusion coefficient has been deduced from rotating disk electrode data and found equal to 5.04×10^{-10} m^2 s^{-1}. The raw experimental data, i.e. diffusion limiting currents, have been treated in a more accurate way. In particular, the axial dispersion phenomenon is accounted for in the computation of the mass transfer coefficient, k_{d}, following the procedure described by Seguin et al. [10]. The description of the fluid rheological behaviour was improved.

3.1.2. Rheological properties of liquid

The rheological behaviour of purely viscous non-Newtonian fluid is presently described by a series of power law equations. The range of investigation of the shear rate

[3] A typographic error was made in [10] concerning the value of the constant of the present equation which is 1.16 instead of 1.13.

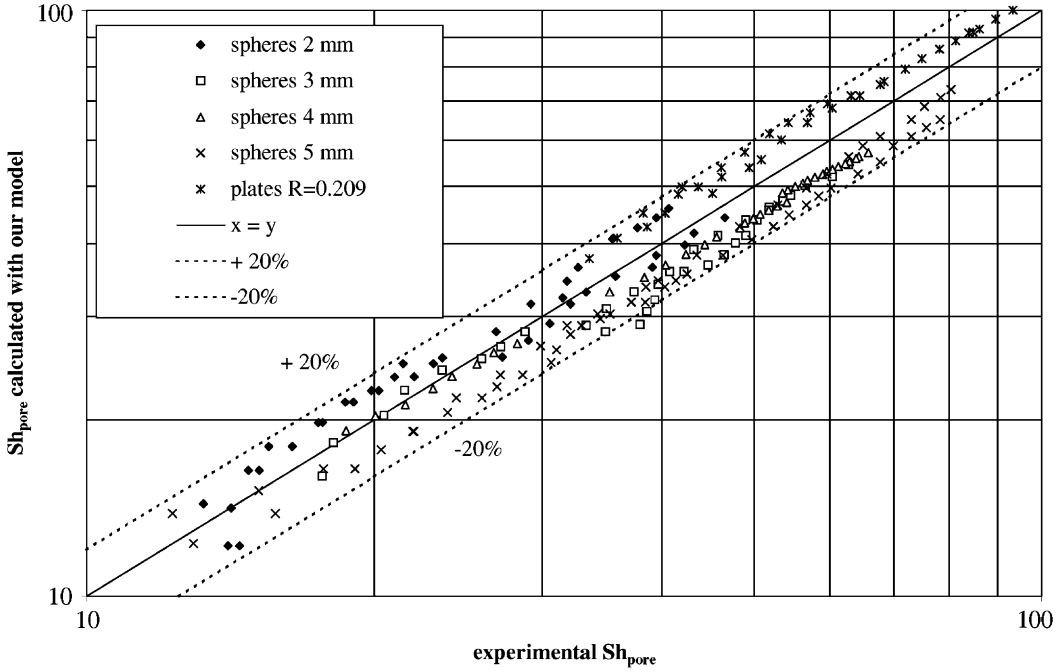

Fig. 1. Comparison between experimental and predicted values of mass transfer in packed beds.

is divided into three intervals. The rheological equation i may be written as

$$T = K_i \dot{\gamma}^{n_i} \quad \text{for } \dot{\gamma}_{i-1} < \dot{\gamma} < \dot{\gamma}_i \tag{12}$$

where T is the shear stress value corresponding to the shear rate value $\dot{\gamma}$. K_i and n_i are the consistency and the behaviour index in the considered range of shear rate, respectively.

For a given value of the fluid superficial velocity, u_0, through the porous medium, the average shear rate at the pore wall is calculated using the expression of the average shear rate at a tube wall in laminar flow regime. It leads to the following equation proposed by Sabiri and Comiti [15]:

$$\dot{\gamma} = \frac{3n_i + 1}{2n_i} \frac{u_0 \tau}{\varepsilon^2} (1 - \varepsilon) A_{\mathrm{vd}} \tag{13}$$

The three rheological equations were determined from data obtained with two viscometers (Low Shear and Rheomat 115, Contraves):

$$T = \begin{cases} 0.0821\dot{\gamma}^{0.944} & \text{for } 1 < \dot{\gamma} < 17\,\mathrm{s}^{-1} \\ 0.131\dot{\gamma}^{0.793} & \text{for } 17 < \dot{\gamma} < 170\,\mathrm{s}^{-1} \\ 0.273\dot{\gamma}^{0.648} & \text{for } 170 < \dot{\gamma} < 1700\,\mathrm{s}^{-1} \end{cases}$$

These equations enable to evaluate the shear stress with an average relative error of about 1%.

3.2. Mass transfer model validation

In order to validate the model proposed in this work, the values of the pore Sherwood number calculated with Eq. (3) for fixed beds of spheres and parallelepipedal particles are compared to experimental values in Fig. 1. Positive and negative deviations of 20% of predicted values with experiments are illustrated with dotted lines. All the plotted values are comprised in the field delimited by these dotted lines. The values of the mean relative error between experimental and predicted data are given for each type of bed packing in Table 1:

$$\mathrm{ERM} = \frac{1}{m} \sum_{j=1}^{m} \left(\frac{|Sh_{\mathrm{pore}}^{\mathrm{experimental}} - Sh_{\mathrm{pore}}^{\mathrm{predicted}}|}{Sh_{\mathrm{pore}}^{\mathrm{experimental}}} \right)_j \tag{14}$$

One may emphasize the fact that experimental values of Sherwood number are themselves characterized by a non-negligible uncertainty due to the tedious character of the experiments and due to the number of experimental parameters, such as the molecular diffusion coefficient and the axial dispersion coefficient, taken into account in the successive steps of the calculations. Thus, a global uncertainty

Table 1
Mean relative errors between experimental and calculated data (Eq. (3)) (number of experimental data and ranges of explored pore Reynolds numbers)

Particles	ERM (%)	Number of data	Range of Re_{pore}
Spheres, $d_{\mathrm{part}} = 2$ mm	8.9	39	0.011–1.24
Spheres, $d_{\mathrm{part}} = 3$ mm	11.4	34	0.024–1.91
Spheres, $d_{\mathrm{part}} = 4$ mm	9.0	35	0.041–1.8
Spheres, $d_{\mathrm{part}} = 5$ mm	13.5	54	0.010–4.38
Plates, $R = 0.209$	10.8	39	0.207–14.6
All beds	10.9	201	0.010–14.6

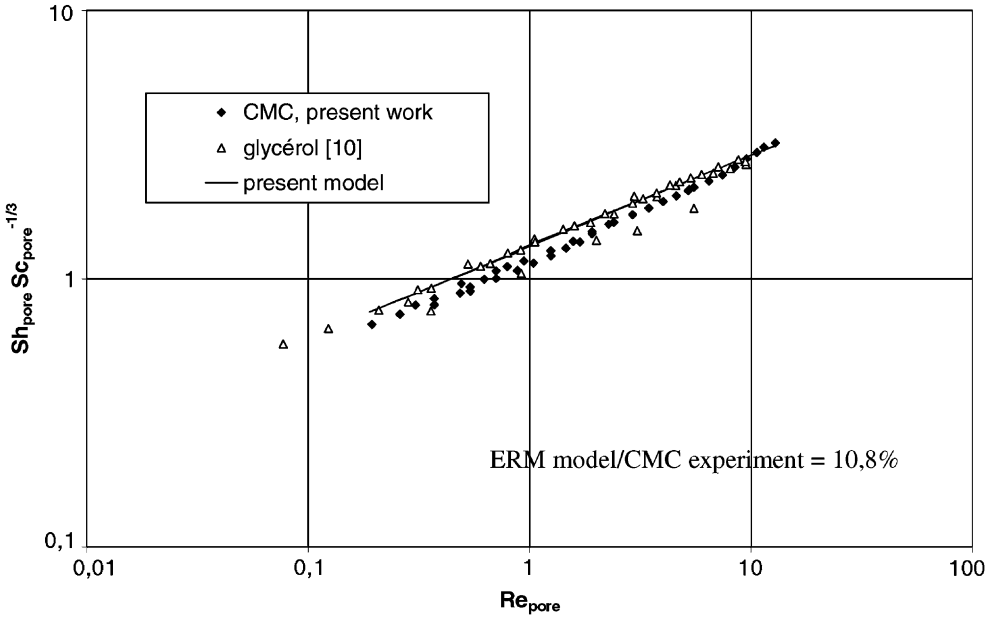

Fig. 2. Verification of the continuity of non-Newtonian fluid data with Newtonian fluid data in packed beds of parallelepipedal particles.

of 10% can be estimated, it is of the same order of the calculated values of ERM.

One can notice that the Sherwood number values predicted by the model are in satisfying accordance with the experimental ones. Moreover, the proposed dimensionless representation allows to unify Newtonian and non-Newtonian data on a same representative graph for spheres as well as for anisotropic particles such as plates with an aspect ratio, $R = 0.209$. An example of comparison of experimental data obtained with Newtonian and non-Newtonian fluids is given in Fig. 2 for the bed of plates. These two observations demonstrate the pertinence of the choice of dimensionless criterions based on the capillary representation of the porous media.

3.3. Comparison with previous works

The literature cited in the introduction section concerns mostly the packings of spherical particles and pellets. Dimensionless numbers formerly used by authors are always related to the particle diameter. The use of pore dimension is often impossible, due to the lack of information on the dynamic specific surface area and on the tortuosity. Then, in the following we compare experimental correlations from literature, in terms of particle dimensionless numbers, with a set of our results obtained with spherical particles. Finally, we discuss the pertinence of using these experimental correlations for the case of parallelepipedal particles.

3.3.1. Comparison of experimental data

We compare a set of our data obtained with spherical particles, namely the 5 mm diameter spheres, with correlations that fit data obtained by Kumar and Upadhyay [3] and by Wronski and Szembek-Stoeger [5], respectively. Kumar and Upadhyay have tested successive packings of spherical particles with different diameters ranging from 0.01 to 0.03 m, and also packings of cylindrical particles. The tested fluid was 1% aqueous CMC solution ($n = 0.85$). They found that a empirical equation, previously published in [16], well correlate their data:

$$\varepsilon J = \frac{0.765}{Re_{\text{part}}^{0.82}} + \frac{0.365}{Re_{\text{part}}^{0.386}} \tag{15}$$

for particle Reynolds numbers comprised in the range 0.1–80. J is a mass transfer factor defined as

$$J = \frac{k_{\text{d}}}{u_0} Sc_{\text{part}}^{2/3} \tag{16}$$

In [5], Wronski and Szembek-Stoeger published their experimental data obtained with cylindrical pellets (4 mm in diameter and of 4.77 mm mean height) in a large range of particle Reynolds numbers. Aqueous solutions with different percentages of CMC were tested (0.5, 1.0 and 1.5%). The corresponding correlation is

$$\varepsilon J = \frac{1}{0.097(1-\varepsilon)^{0.3} Re_{\text{part}}^{0.3} + 0.75(1-\varepsilon)^{0.61} Re_{\text{part}}^{0.61}} \tag{17}$$

with $\varepsilon = 0.37$ for tested cylindrical pellets, and $0.0001 < Re_{\text{part}} < 50$ and $10^3 < Sc < 6.3 \times 10^5$.

The empirical equations (15) and (17) are plotted in Fig. 3 and are compared to:

- the experimental data obtained with 5 mm in diameter spheres presented in this work,

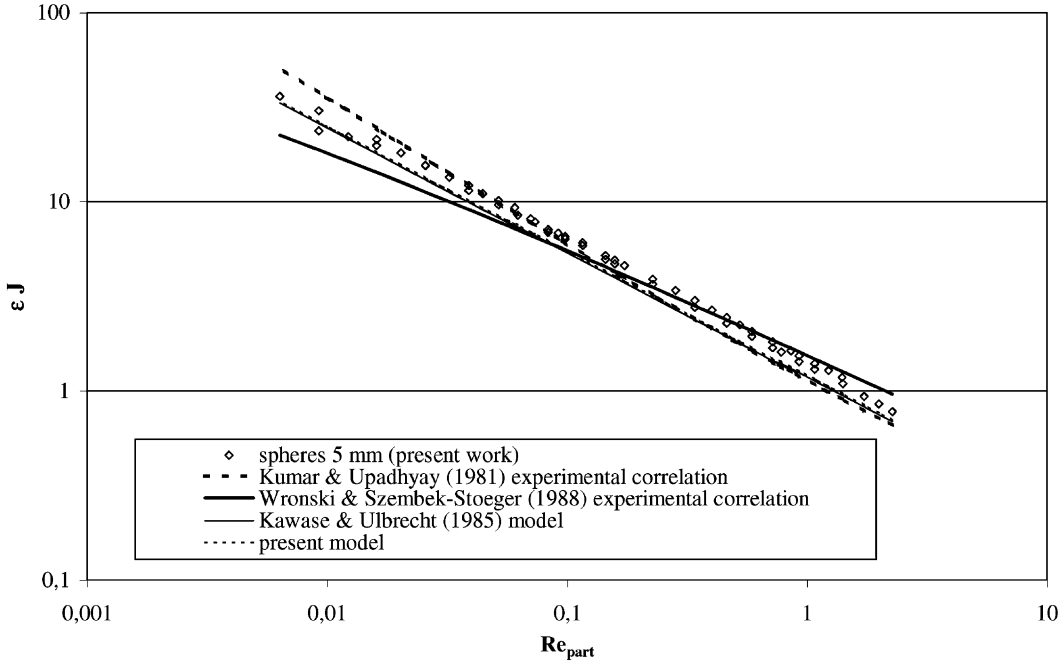

Fig. 3. Comparison of experimental correlations with present data (packed beds of 5 mm in diameter spheres).

- the simplified Eq. (11) of the present model available for spheres, and
- the theoretical equation derived by Kawase and Ulbrecht [8]:

$$\varepsilon J = 1.8\{\varepsilon(1-\varepsilon)\}^{1/3}\left(\frac{3n+1}{4n}\right)^{1/3} Re_{\text{part}}^{-2/3} \qquad (18)$$

In the main part of the explored Reynolds number range, our data are comprised in a domain delimited by the cited experimental correlations. These correlations are somewhat moving away in opposite directions from our data for low values of the particle Reynolds number. However, one should take into account that, in particular, the experimental data of Wronski and Szembek-Stoeger are scattered in a field defined between $\pm 0.3\ \varepsilon J$.

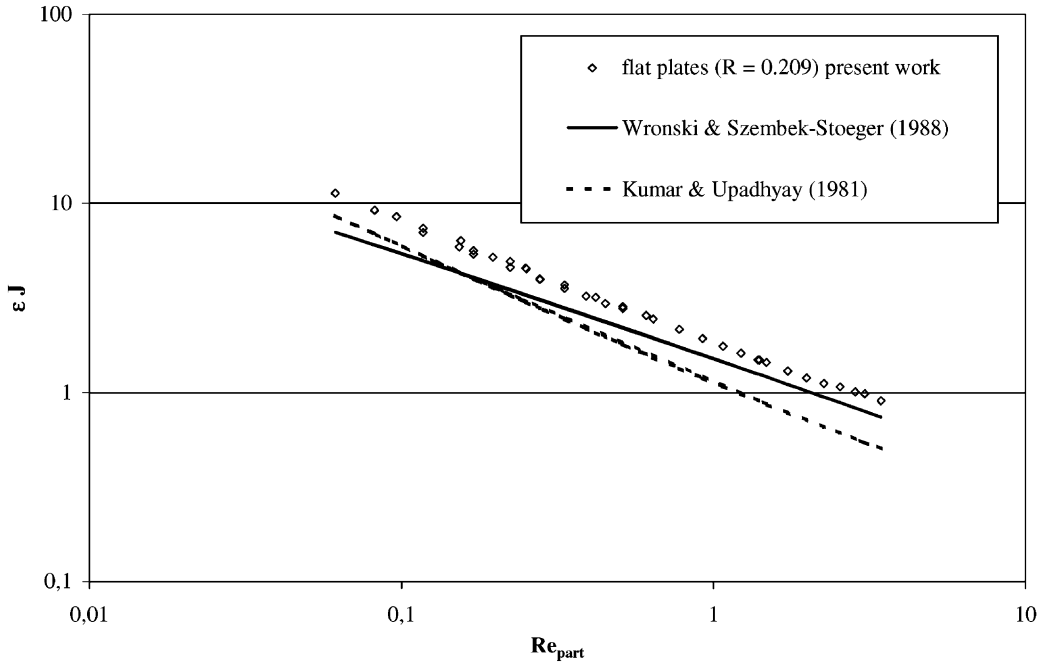

Fig. 4. Use of particle dimensionless numbers in the case of the parallelepipedal particles—comparison of data with values calculated from correlations.

Table 2
ERM values between experimental data obtained with flat plates ($R = 0.209$) and correlations

ERM (%)	Dynamic surface area based equivalent diameter	Geometrical surface area based equivalent diameter
Experimental data for plates/calculated values with Eq. (15), Kumar and Upadhyay [3]	18.2	34.5
Experimental data for plates/calculated values with Eq. (17), Wronski and Szembek-Stoeger [5]	7.7	22.8

As can be expected, Eq. (18) proposed by Kawase and Ulbrecht is equivalent to our simplified equation for spherical particles.

3.3.2. Discussion on using particle dimensionless numbers for parallelepipedal particles

For non-spherical particles such as cylinders or parallelepipedal particles, a convenient definition of the particle diameter has to be found. Kumar and Upadhyay [3] have tested for cylindrical particles the equivalent particle diameter based on the geometric surface area and equivalent particle diameter based on the geometric volume area. They found that the first one is more proper to gather pellets data on the same line than spherical particles data. Wronski and Szembek-Stoeger also used the equivalent particle diameter based on the geometric surface area to define the particle diameter of their pellets.

In Fig. 4, we have plotted our data obtained with flat plates ($R = 0.209$) in terms of εJ as a function of Re_{part}, defining d_{part} as the equivalent particle diameter based on the surface area. We present in the same figure a plot of the correlation proposed by Kumar and Upadhyay [3] and that proposed by Wronski and Szembek-Stoeger, with $\varepsilon = 0.35$. The values of J have also been calculated by using an equivalent diameter based on the dynamic surface area of a particle really offered to the fluid flow, taking into account the overlapping. In Table 2, the values of the mean relative error between our experimental data and those calculated using these two definitions of the equivalent diameters are presented.

A part from using the equation of Wronski and Szembek-Stoeger with a particle diameter based on the dynamic specific area, the mean deviations between experimental values and calculated ones are much larger than the mean relative error obtained between our data and the presently proposed model, Eq. (3), i.e. 10.8% (Table 1). However, calculating a particle diameter from a dynamic surface area has no particular physical sense. This demonstrates the unsuitability of dimensionless equations based on particle diameter, for particles the shape of which is significantly different from that of the sphere.

4. Conclusion

The interest of the approach followed in this work to predict solid non-Newtonian fluid mass transfer in creeping flow regime in packed beds is to use a capillary-type representation and to define dimensionless numbers with the characteristic parameters of the pore. Using dimensionless numbers based on the particle diameter for non-spherical particles supposes that an equivalent diameter has to be calculated. Because no definition of equivalent diameter seems to be actually satisfying, the use of a pore diameter, as proposed in this work, is more suitable. In particular, the proposed equations allow to predict mass transfer in beds of spheres or anisotropic parallelepipedal particles with a global mean relative error of 10.9%, which is the order of magnitude of the experimental data uncertainties. Moreover, the followed procedure does not require adaptative parameters.

Available experimental data, from this work or from literature cited in this paper, concern non-Newtonian fluids characterized by flow behaviour indexes ranging from 0.648 to 0.944. Next complementary works would be of help to consider the influence of smaller values of the flow behaviour index as well as the influence of other non-spherical particle shapes on the validity of the available equations.

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