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Carine Julcour-Lebigue, Frédéric Augier, Harold Maffre, Anne-Marie Wilhelm, Henri Delmas. Measurements and Modeling of Wetting Efficiency in Trickle-Bed Reactors:Liquid Viscosity and Bed Packing Effects. Industrial and engineering chemistry research, American Chemical Society, 2009, 48 (14), pp.6811-6819. 10.1021/IE9002443 . hal-02353918

**HAL Id: hal-02353918**
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Submitted on 7 Nov 2019

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**To link to this article**DOI:10.1021/IE9002443
URL: http://dx.doi.org/10.1021/IE9002443

**To cite this version**: Julcour-Lebigue, Carine and Augier, Frédéric and Maffre, Harold and Wilhelm, Anne-Marie and Delmas, Henri (2009) Measurements and Modeling of Wetting Efficiency in Trickle-Bed Reactors: Liquid Viscosity and Bed Packing Effects. *Industrial & Engineering Chemistry Research*, vol. 48 (n°14). pp. 6811-6819. ISSN 0888-5885

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GENERAL RESEARCH

# Measurements and Modeling of Wetting Efficiency in Trickle-Bed Reactors: Liquid Viscosity and Bed Packing Effects

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An experimental parametric study on wetting efficiency is reported which evaluates the influence of liquid viscosity, as well as the effect of particle size(/shape) and bed porosity ($\varepsilon_B$), separately. A 10-fold increase of liquid viscosity improves slightly catalyst wetting (by about 10%), while an increase of either bed porosity or particle size has the opposite effect. Wetting efficiency is reduced by about 0.1 for an increase of $\varepsilon_B$ from 0.38 and 0.40 due to a change of particle size from 1.8 to $7 \times 10^{-3}$ m while the decrease is only 0.05 for a similar variation of $\varepsilon_B$ (0.38−0.41) with the same particles. The effect of particle shape (cylindrical/trilobe extrudates or spheres) appears very small in the investigated conditions. A new correlation for wetting efficiency is proposed, using a bounded function and only three dimensionless groups (liquid Froude and Morton numbers and bed porosity). This correlation is able to predict wetting efficiency with a very good precision on a large database, provided wetting liquids are used. Adding fines in the fixed bed is also examined, and its positive effect can be correlated with the size ratio between catalyst particles and fines.

## 1. Introduction

**1.1. Industrial Context.** The issue of partial wetting inside trickle bed reactors (TBRs) has been addressed for several decades, but it is still a subject of investigation due to the complexity of the phenomenon and its growing importance in oil industry. When superficial liquid velocities are below a critical value (usually about 0.01 m/s), the probability to wet only a fraction of the catalyst surface is high, resulting in some cases in a lower reaction efficiency. This is particularly true when the chemical reactions are limited by the diffusion of the liquid reactant (e.g., hydrodesulfurization), because partial wetting can affect liquid/solid mass transfer and apparent diffusivity inside catalyst particles.

Lab-scale reactors generally operate at a very low superficial liquid velocity in order to keep the same liquid hourly space velocity as in commercial plants. Reliable measuring techniques and prediction tools are thus needed by chemical engineers, either to avoid partial wetting of the catalyst or to be able to account for it when scaling up or down industrial processes.

**1.2. Measuring Techniques.** Partial wetting in trickle bed reactors has been experimentally investigated by five main techniques: the dynamic tracer technique,[1−4] chemical reaction method,[5,6] and, more recently, the hydrodynamic technique,[7,8] magnetic resonance imaging (MRI),[9] and dye adsorption technique.[10−12]

Except the last two ones, all of those approaches are overall and indirect and require a model of the reactor involving hydrodynamic, transfer, and/or kinetic parameters. The choice of some well-known reactive systems limits also the parametric range of the chemical method. The MRI method appears to be a very promising tool to get features and quantitative analysis of the gas−liquid flow, as well as a mapping of chemical conversion for reactive systems,[13−15] but it requires both complex equipment and a delicate treatment.

Recently Baussaron et al.[12] have compared three measuring techniques of wetting efficiency. The original hydrodynamic technique based on simple pressure drop measurements has been found unable to predict wetting efficiency with convenient accuracy in a wide range of operating conditions. The other ones, based on analysis of residence time distribution (RTD) and on dye adsorption on wetted surfaces, have given similar results. However the tracer technique is more delicate to operate as it requires a pretreatment of experimental signals as well as a precise knowledge of axial dispersion and effective diffusivity from separate experiments. Moreover, RTD signals acquired at low liquid flow rates often show long tails, and the fitting of the reactor model, necessary to calculate the wetting efficiency, is less accurate.

**1.3. Existing Correlations.** Several correlations have been proposed in the past to calculate the global wetting efficiency in TBR,[3,4,16−20] but they give very dispersed results over the same range of operating conditions. Those discrepancies can be explained by the various measuring techniques, whose precision was not fully examined, and also by the operating conditions, especially the bed prewetting procedure as pointed out by van Houwelingen et al.[10]

Recently Lappalainen et al.[21] have evaluated different models based on dimensional similitude and optimized their parameters using about 250 experimental points from literature, distinguishing the prewetting procedure and reconciliating some of the data. They have concluded that a wetting efficiency model does not describe all known trends suitably when including less than four dimensionless groups, despite the fact that the precision of data fitting is not really improved above two Π groups. Their final correlation is consistent with most of reported trends, but it is

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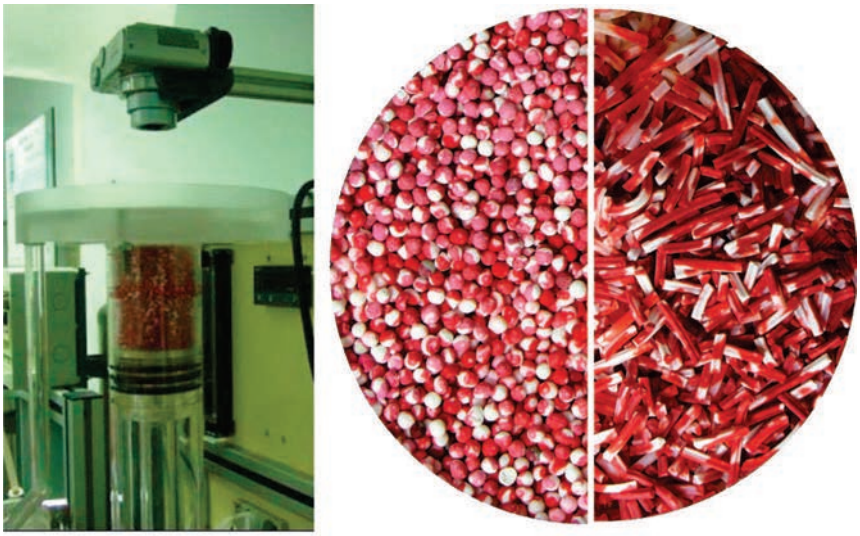


**Figure 1.** Photographs of (a) fixed bed column for dye-adsorption and (b) column cross-sections.

not bounded and may thus lead to unrealistic values when applied out the range of previously investigated conditions.

In this work, a previous experimental database (20 points with same type of alumina beads) has been completed to 90 points by using the same measuring technique (dye adsorption), the same setup device and operating procedure, to better understand the effect of the different operating parameters separately.

A special effort has been addressed to account for the parameters poorly investigated: liquid viscosity and the separate effects of particle diameter and bed porosity. Finally a new correlation has been developed, using a bounded function (ensuring $0 < f < 1$) and with special attention given to the choice of the most relevant dimensionless numbers according to the observed trends.

## 2. Experimental Section

**2.1. Trickle Bed Setup and Operating Procedure.** The experimental technique used to measure the catalyst wetting is based on the step injection of a colored liquid at the inlet of a bed of adsorbing particles and the image processing of the photographs of partially colorized particles.[10,11]

In their previous studies, those authors underlined the dramatic effects of the liquid distribution and bed prewetting procedure on the catalyst wetting efficiency. In the present work, only an optimized gas−liquid distributor and a prewetted bed have been used for the parametric study.

The trickle bed setup (Figure 1a) consists of two concentric columns, the inner one being allowed to move by means of a piston. This central tube of 0.057 m internal diameter is divided in 11 modules of 0.035 m height in order to get the axial evolution of the catalyst wetting. The liquid is uniformly injected on the bed cross-section by 49 capillaries of $1 \times 10^{-3}$ m diameter and 0.1 m high, while the gas is supplied independently through 24 openings of $2 \times 10^{-3}$ m in diameter. Two storage tanks, containing either the clear liquid or the colored one, are connected to the distributor by means of a three-way valve in order to easily switch from one liquid to the other one.

The bed of porous alumina particles is first prewetted with the (clear) liquid by flooding it for 5 min. It is then drained until only the residual liquid holdup remains. Afterward liquid (and gas) is (are) allowed to circulate in the bed over 30 min at the investigated velocities to ensure that stable hydrodynamic conditions are reached.

According to the terminology proposed by van Houwelingen et al.,[10] this corresponds to a "Levec pre-wetting" procedure.

Then, the clear liquid is replaced by a flow of the same liquid but containing a dissolved dye (Sudan red in heptane and gasoil), to colorize the surface of the particles which is wetted by the flowing liquid. Depending on the operating conditions, the flow of colored liquid is maintained for 5−30 min, so that all bed sections can be colored while preventing any color spreading due to dye diffusion inside the porous particles.

The bed is finally rinsed with the pure liquid before being carefully dismounted and photographed section by section. Details of the image processing used to calculate the average wetting efficiency of the bed can be found in the work of Baussaron et al.[11] It is based on an automated procedure using the Image Processing toolbox of Matlab, to divide the photographed cross sections of the bed (Figure 1b) into wet zones (red), dry zones (white), and interparticle spaces of dark color for which no distinction can be made between wet and dry. On every processed image, colored markers are selected for each group to identify its levels of color (2 criteria) and luminosity. Each pixel on the image is then classified into one of the three categories depending on the Euclidean distance between the pixel and the markers. The average value of the wetting efficiency is then calculated as the surface ratio of the red zones to the sum of the white and red zones.

**2.2. Investigated Parameters.** In the previous papers of Baussaron et al.,[12,20] many operating parameters were already investigated either by the dye adsorption technique or a tracer technique: liquid and gas flow rates, operating pressure and gas density, and particle shape and size, as well as the liquid−solid affinity, and a first correlation was proposed for wetting efficiency since deviation was too large with literature ones.[3,17,18]

This correlation based on five dimensionless numbers (Reynolds numbers for gas and liquid flows, Galileo number, and two capillary numbers based on gas/liquid and liquid/solid surface tensions) predicted the experimental wetting efficiencies with a maximum deviation of 13%.

However due to the very low number of tested values for particle diameter and bed porosity, the wetting efficiency was

**Table 1. Physical Properties of the Liquids (25 °C)**

| | $\rho_L$ (kg/m$^3$) | $\mu_L$ (Pa·s) | $\sigma_L$ (N/m) |
|---|---|---|---|
| heptane | 680 | $3.9 \times 10^{-4}$ | $20.1 \times 10^{-3}$ |
| gasoil | 830 | $3.4 \times 10^{-3}$ | $28.1 \times 10^{-3}$ |
| heptane/gasoil (50/50 vol %) | 780 | $1.2 \times 10^{-3}$ | $23.9 \times 10^{-3}$ |
| water[a] | 1000 | $1.0 \times 10^{-3}$ | $72 \times 10^{-3}$ |
| ethanol[a] | 790 | $1.1 \times 10^{-3}$ | $22.4 \times 10^{-3}$ |

[a] Used in previous work.

not very sensitive to the Galileo number. The effect of liquid viscosity described in the correlation through the Reynolds number was also not checked.

In order to complete the previous data bank, the following parameters are thus investigated in this work:

- viscosity of the liquid phase in the range $0.39-3.4 \times 10^{-3}$ Pa·s, by using heptane, gasoil (whose density, surface tension, and contact angle with alumina are very similar to those of heptane), and a mixture of the two (50 vol %),
- bed porosity by varying the bed loading with the same particles, to discriminate between the real effect of particle size and that resulting from the change in bed porosity due to the different particle sizes,
- particle diameter, from 1.8 to $7 \times 10^{-3}$ m for spherical particles (five lots),
- particle shape: spheres and cylindrical and trilobe-shape extrudates,
- addition of fine nonporous particles (silicon carbide, $0.9 \times 10^{-3}$ m diameter) varying the size ratio in between the alumina particles and the SiC particles (from 3 to 8). Only complete loading—to fully fill the interparticle spaces between the original beads—has been investigated.

For all cases, wetting efficiency ($f$) was measured at liquid velocities from $0.2 \times 10^{-3}$ to $8 \times 10^{-3}$ m/s, extending the range of Baussaron et al.[12] ($V_{SL} \geq 0.5 \times 10^{-3}$ m/s).

These experiments were conducted at 25 °C and atmospheric pressure, with no gas flow.

The properties of the different liquids and solids investigated in the present study are reported in Tables 1 and 2, respectively. According to the Washburn technique, the first three liquids mentioned in Table 1 were found to perfectly wet the alumina particles.

## 3. Results and Discussion

**3.1. Validation of the Method.** Both from preliminary study in a 2D-monolayer of particles[11] and MRI visualization in a 3D trickle-bed,[22] the position of the rivulets in trickle flow was proved to be very stable, which validates the dye-adsorption method that integrates the wetting history in the final result.

It was also shown that for liquid velocities higher than $2 \times 10^{-3}$ m/s the particle wetting efficiency does not vary axially after a few centimeters in the bed (about 0.1 m, corresponding to modules 1−3) if the bed is prewetted, and an effective liquid distribution is provided.[11] It is still observed here for a much lower liquid velocity ($V_{SL} = 3 \times 10^{-4}$ m/s, Figure 2).

Thus in the present work, the global wetting efficiency is calculated as the average value of modules 4 and 5, showing standard deviation of less than 5%.

In the previous studies, two types of photographs were taken and compared:

- "section views" obtained along the column by taking photographs of the cross section on the top of each column slice,
- "volume views" of a representative sample of the pellets taken from each module and spread randomly on a perforated plate. The section views allowed the detection of some typical patterns of the trickle flow but could be biased for a quantitative measurement of wetting efficiency as they only showed the part of the beads facing the flow, so that the "volume views" were initially preferred.

However, the data obtained from analysis of the two photograph types were found to match within 10% at various axial locations.[12]

In the present work, some photographs were also taken after turning the column upside down, to analyze the sections that were not oriented toward the flow. The corresponding wetting efficiencies are also in very good agreement with the values obtained from direct section views (cf. Figure 3).

Finally in Figure 3, the wetting efficiencies obtained by averaging the values from section views 4 and 5 are compared to the previous data of Baussaron et al.,[11] showing a difference of less than 5% in the investigated range of liquid velocity.

As expected, wetting efficiency is found to be an increasing function of liquid velocity, with a very steep slope below $1 \times 10^{-3}$ m/s.

**3.2. Influence of Operating Parameters. 3.2.1. Liquid Viscosity.** Few studies investigated the effect of this parameter in the past, although many of the existing correlations predict a decrease of wetting efficiency when increasing liquid viscosity by including Reynolds number with a positive exponent.

Conversely Luciani et al.[23] reported an improved wetting of the particles when increasing the concentration of carboxymethyl cellulose in water, but as the solutions exhibited also a rather lower surface tension (and thus a better expected spreading of the liquid), they could not discriminate between the influence of each parameter separately.

Here heptane, gasoil, and a mixture of the two are chosen to investigate the influence of viscosity, as their other physical properties (density, surface tension, contact angle with alumina) are quite similar (cf. Table 1).

Figure 4 shows that increasing the liquid viscosity improves slightly the wetting of the particles, by less than 10% when viscosity is multiplied by a factor 8.5. This can be explained by an increased liquid holdup as reported for instance by Ellman et al.[24]

Figures also include predictions of the developed correlation, presented later in section 3.3. Its agreement with experimental data will also be discussed in that section.

**3.2.2. Particle Size and Bed Porosity.** Most of the studies conclude that there is a positive effect of size decrease on particle wetting,[1,9,18] even if some contradictory results are also found.[25,26]

In those studies, the number of particle sizes examined is rather low (2 or 3) and the probable change of bed porosity not always accounted for.

The wetting efficiency of five lots of spherical particles of same alumina type, but different diameters from 1.8 to $7 \times 10^{-3}$ m, is plotted as a function of liquid velocity in Figure 5.

Wetting efficiency is found to decrease when increasing particle size, by about 15% in between the two extreme sizes. The effect is more sensible for liquid velocities above $1 \times 10^{-3}$ m/s. Nevertheless, this result may also depend on the noticeable difference of bed porosity (0.40 for the largest particles and 0.38 for the smallest ones).

Thus, in Figures 6a and b the influence of bed porosity is studied separately by varying the bed loading with the same particles (spherical pellets or cylindrical extrudates).

As expected, increasing bed porosity leads to a lower wetting efficiency as a result of the lower number of particle−particle contacting points and thus less liquid menisci to drive the liquid

**Table 2. Physical Properties of the Alumina Particles**

| | $d_p$ (m) or $d_p$(m) × $l_p$ (m) | shape | $\varepsilon_B$ (−) | supplier |
|---|---|---|---|---|
| pellet A | $1.8-3.7 \times 10^{-3}$ | spherical | 0.369−0.410 | AXENS |
| pellet B | $4.5-7.0 \times 10^{-3}$ | spherical | 0.391−0.407 | CALDIC (4−8 grade D) |
| pellet C | $(1.25 \times 10^{-3}) \times (4.0 \times 10^{-3})$ | cylindrical | 0.367−0.398 | AXENS |
| pellet D | $(1.29 \times 10^{-3}) \times (8.0 \times 10^{-3})$ | trilobe | 0.372 | AXENS |

**Table 3. Operating Conditions of the Different Investigated Cases**

| case | $d_v$ (m) | $\varepsilon_B$ | $d_v/d_{SiC}$ | liquid | pellet | $V_{SL}$ range (m/s) | mean std deviation |
|---|---|---|---|---|---|---|---|
| 1 | $2.5 \times 10^{-3}$ | 0.37 | | heptane | A | $(0.5-8) \times 10^{-3}$ | 2.4% |
| 2 | $5.5 \times 10^{-3}$ | 0.41 | | heptane | B | $(0.5-6.5) \times 10^{-3}$ | 5.7% |
| 3 | $5.5 \times 10^{-3}$ | 0.41 | | water | B | $(0.5-6) \times 10^{-3}$ | 15.4% |
| 4 | $5.5 \times 10^{-3}$ | 0.41 | | ethanol | B | $(0.5-6) \times 10^{-3}$ | 12.6% |
| 5 | $3.65 \times 10^{-3}$ | 0.395 | | heptane | A | $(0.5-5) \times 10^{-3}$ | 4.6% |
| 6 | $2.85 \times 10^{-3}$ | 0.385 | | heptane | A | $(0.2-8) \times 10^{-3}$ | 2.7% |
| 7 | $1.8 \times 10^{-3}$ | 0.38 | | heptane | A | $(0.2-8) \times 10^{-3}$ | 4.0% |
| 8 | $1.8 \times 10^{-3}$ | 0.405 | | heptane | A | $(0.3-5) \times 10^{-3}$ | 3.0% |
| 9 | $2.1 \times 10^{-3}$ | 0.37 | | heptane | C | $(0.2-5) \times 10^{-3}$ | 3.5% |
| 10 | $2.1 \times 10^{-3}$ | 0.40 | | heptane | C | $(0.5-5) \times 10^{-3}$ | 4.5% |
| 11 | $1.8 \times 10^{-3}$ | 0.385 | | gasoil | A | $(0.15-3) \times 10^{-3}$ | 2.6% |
| 12 | $1.8 \times 10^{-3}$ | 0.38 | | heptane+gasoil | A | $(0.15-3) \times 10^{-3}$ | 5.0% |
| 13 | $4.5 \times 10^{-3}$ | 0.39 | | heptane | B | $(0.2-5) \times 10^{-3}$ | 2.8% |
| 14 | $4.5 \times 10^{-3}$ | 0.40 | 5 | heptane | B | $(0.2-5) \times 10^{-3}$ | 1.1% |
| 15 | $7.0 \times 10^{-3}$ | 0.40 | | heptane | B | $(0.2-8) \times 10^{-3}$ | 2.8% |
| 16 | $7.0 \times 10^{-3}$ | 0.40 | 7.8 | heptane | B | $(0.2-5) \times 10^{-3}$ | 1.5% |
| 17 | $2.85 \times 10^{-3}$ | 0.39 | 3.2 | heptane | A | $(0.2-5) \times 10^{-3}$ | 1.7% |
| 18 | $2.85 \times 10^{-3}$ | 0.37 | | heptane | D | $(0.5-5) \times 10^{-3}$ | 2.4% |

rivulets. The effect of the bed porosity is found to be similar for the two shapes of pellets. It can be noticed that wetting efficiency is reduced by about 0.1 for an increase of bed porosity from 0.38 and 0.40 due to a change of particle size from 1.8 to $7 \times 10^{-3}$ m (Figure 5) while the decrease is only 0.05 when varying $\varepsilon_B$ from 0.38 to 0.41 with the same particles (Figure 6).

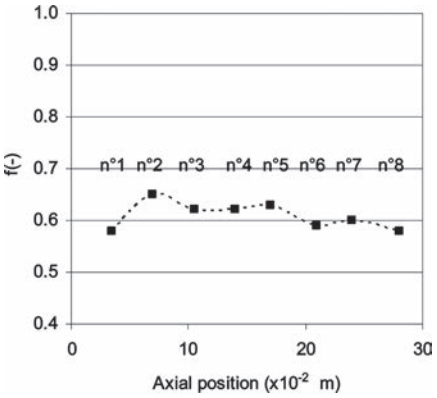


**Figure 2.** Axial evolution of wetting efficiency from the bed top (with corresponding module number). Case 6 from Table 3.

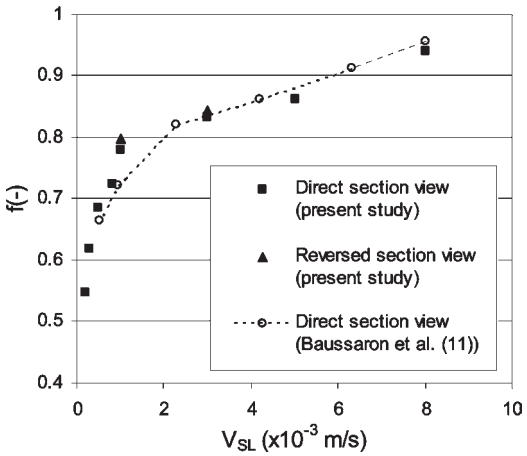


**Figure 3.** Wetting efficiency calculated from direct and reversed section views. Comparison with the results of Baussaron et al.[11] Case 6 from Table 3.

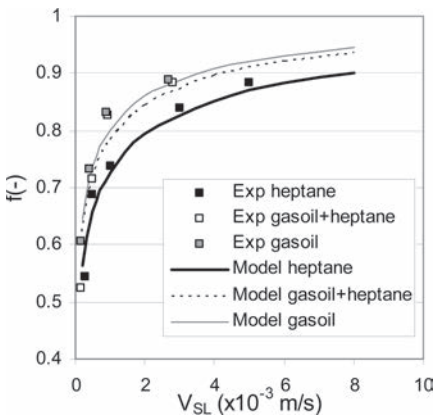


**Figure 4.** Effect of liquid viscosity. Cases 7, 11, and 12 from Table 3.

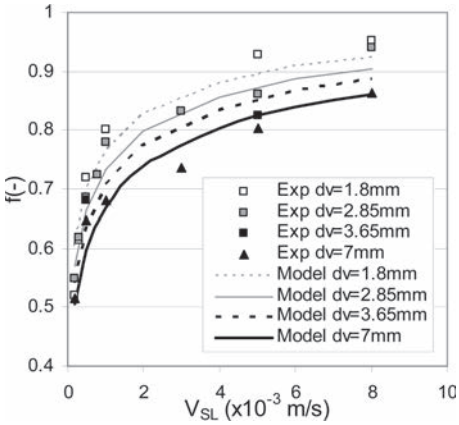


**Figure 5.** Influence of particle diameter. Cases 5, 6, 7, and 15 from Table 3.

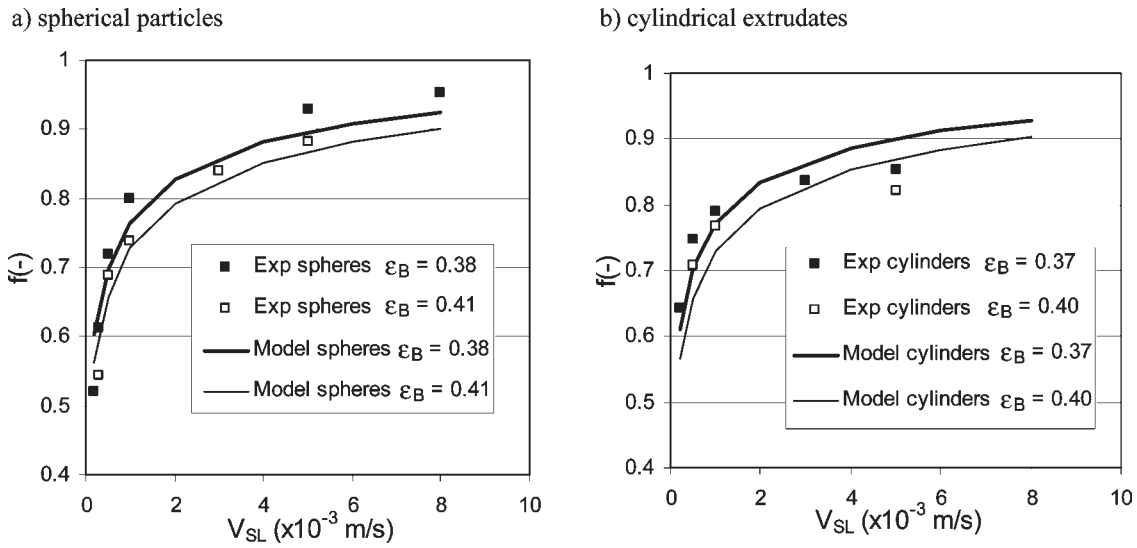


**Figure 6.** Influence of bed porosity. Cases 7−10 from Table 3.

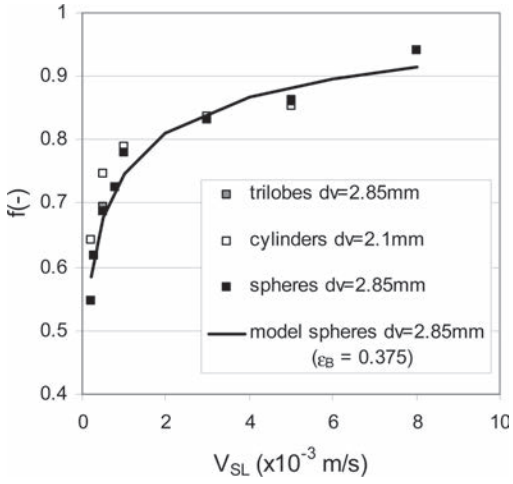


**Figure 7.** Effect of particle shape (with close equivalent volume diameter). Cases 6, 9, and 18 from Table 3.

This allows thus a conclusion about the real extent of particle size effect on the wetting of particles: an increase of 5−10% when particle size is divided by about 4.

**3.2.3. Particle Shape.** Catalyst particles with extruded shapes, as cylinders or trilobes, are very common in industry because they provide higher surface/volume ratios than spherical particles for the same specified pressure drop. It is usually admitted that the use of long extrudates may result in a worse liquid distribution due to a preferential orientation of the particles in a downward slant toward the reactor walls and thus to a lower global wetting efficiency of the bed. On the other hand, Trivizadakis et al.[27] have reported a noticeably greater dynamic liquid holdup (up to 40%) when using extrudates instead of spheres of comparable Sauter diameter. Figure 7 compares the wetting efficiency of cylindrical and trilobe shape extrudates to that of spherical pellets. Those extrudates being rather long compared to their diameter (cf. Table 2), the question could arise about the most suitable equivalent diameter to use for this comparison: based on the sphere of equivalent volume $d_v = (6V_p/\pi)^{1/3}$ or based on the sphere of same specific area $d_s = 6V_p/S_p$ (Sauter diameter). Those diameters give indeed a different weight to the particle length, resulting in $d_v = 2.1 \times 10^{-3}$ m (respectively $2.85 \times 10^{-3}$ m) and $d_s = 1.6 \times 10^{-3}$ m (respectively $1.85 \times 10^{-3}$ m) for the cylindrical (or trilobe shape) extrudates. In Figure 7, the different particles exhibit similar volume diameter $d_v$ and rather close wetting efficiency. This argues here for a negligible effect of particle shape. However corresponding experiments could not be performed with exactly the same bed porosity, and depending on the extent of size effect, this conclusion may not stand if another equivalent diameter (e.g., $d_s$) is chosen. According to the previous paragraph, the influence of particle size is rather moderate and the wetting efficiency variation from $d_v$ to $d_s$ should not be very significant. Thus, it has been decided to keep the equivalent volume diameter as a reference for further calculations.

It can be finally noticed that no significant dry zones have been observed with those long extrudates.

**3.2.4. Addition of Fines.** Al-Dahhan and Dudukovic[28] first reported the use of fine nonporous particles to improve the wetting efficiency of a catalyst bed. In this study silicon carbide (SiC) particles of $0.9 \times 10^{-3}$ m diameter are used to dilute beds of different packing sizes—$2.8 \times 10^{-3}$, $4.5 \times 10^{-3}$, and $7 \times 10^{-3}$ m—to fully fill the interparticle spaces between the original beads ($\phi = d_v/d_{SiC}$ ranging from 3.2 to 7.8). Only the wetted surface of alumina particles is measured, after the top layer of fines has been carefully removed. Results reported in Figure 8 show that for all packing diameters a positive effect of fines is observed, that is higher at a larger size ratio of alumina to SiC particles. The presence of SiC deeply modifies the bed topology in several ways, and two major positive effects are a decrease of the bed porosity (divided by 2 approximately) and an increase of the number of contact points between solid particles. This last factor has been pointed out by Gladden et al.[22] as a major parameter to explain local heterogeneities of wetting efficiency in TBRs. A detailed inspection of pictures reveals that the fraction of fully unwetted particles is less than 10% when $V_{SL} \geq 0.5$ mm/s. At $V_{SL} = 0.2$ mm/s, a few more unwetted particles have been observed without fines or when $\phi = 3.2$, but not at higher values of $\phi$.

**3.3. Development of a New Correlation. 3.3.1. Characteristics of the Proposed Correlation. Experimental Database.** The experimental database used to develop the correlation includes the above-mentioned data as well as the data of Baussaron et al.[12] measured by the dye adsorption technique with the same types of alumina particles, resulting in 90 experimental points.

Table 3 reports the operating conditions corresponding to the different cases investigated (pellet diameter, porosity of alumina

**Table 4. Standard Deviations Obtained with the Different Π Group Combinations (Case of $n = 3$)**

| $C_0$ | $\varepsilon_B$ ($C_1$) | $Re_L$ | $We_L$ | $Stk_L$ | $Mo_L$ | $Fr_L$ | $Ga_L$ | std dvtn |
|---|---|---|---|---|---|---|---|---|
| 1.581 | −2.269 | −0.181 | 0.224 | 0 | 0 | 0 | 0 | 6.02% |
| 0.580 | −2.976 | 0.228 | 0 | 0 | 0.100 | 0 | 0 | 7.42% |
| 2.252 | −1.583 | 0 | 0.086 | 0.107 | 0 | 0 | 0 | 5.07% |
| 0.862 | −2.632 | 0 | 0.128 | 0 | 0.038 | 0 | 0 | 6.53% |
| 2.256 | −1.777 | 0 | 0.138 | 0 | 0 | 0 | −0.072 | 5.32% |
| 4.059 | 0.095 | 0 | 0 | 0.219 | −0.066 | 0 | 0 | 5.75% |
| 1.986 | −1.552 | 0 | 0 | 0 | 0.020 | 0.139 | 0 | 5.00% |

particle bed, size ratio of alumina/SiC particles when fines are used, fluid type, pellet shape, liquid velocity range, and standard deviation of the developed correlation). Cases 5−19 are new experimental results obtained in the present study.

**Choice of the Π Groups and Expression of the Correlation.** The feedback about the use of nonbounded correlations to estimate wetting efficiencies ($f$) is often disappointing, leading to nonphysical values of $f$ ($f > 1$) when applying them out of their domain of validity. Thus, the developed correlation is expressed as follows:

$$f = 1 - \exp[-C_0 \prod_{i=1}^{n} N_i^{C_i}] \quad (1)$$

where $C_0$... $C_n$ are fitted constants and $N_i$ are different dimensionless groups. The use of similar exponential functions has been already proposed by Mills and Dudukovic,[16] Ring and Missen,[29] and Gonzales-Mendizabal et al.[30]

According to the conclusions of the present study and those of the previous works, the following physical scales have to be accounted for: liquid superficial velocity $V_{SL}$, equivalent volume diameter of the particles $d_v$, bed porosity $\varepsilon_B$, properties of the liquid phase ($\rho_L$, $\mu_L$, $\sigma_L$), and ratio of alumina to SiC particles $d_v/d_{SiC}$ when fines are added. The gas phase effect has been neglected as measurements performed with either dye adsorption technique at atmospheric pressure or tracer method at various pressures with $N_2$ and $SF_6$ have showed that both the gas velocity and the gas density have a very low influence on wetting efficiency, rather positive at liquid velocity higher than $2 \times 10^{-3}$ m/s.[12] Thus, it would lead to an exponent of the $(1 + Re_G)$ group almost equal to zero. In the literature, there is actually no real consensus about the effect of gas velocity: Herskowitz and Mosseri[31] and Burghardt et al.[18] observed a decrease of wetting efficiency when increasing gas flow rate, while Al-Dahhan and Dudukovic,[4] Pironti et al.,[7] and Kundu et al.[8] found the opposite trend, especially at elevated pressure, as the consequence of an improved spreading of the liquid film despite a lower liquid holdup.

$N_i$ are then chosen in the following list: $Re_L$, $We_L$, $Stk_L$, $Mo_L$, $Fr_L$, $Ga_L$. The term $\varepsilon_B$ is considered as an additional independent number ($N_1 = \varepsilon_B$).

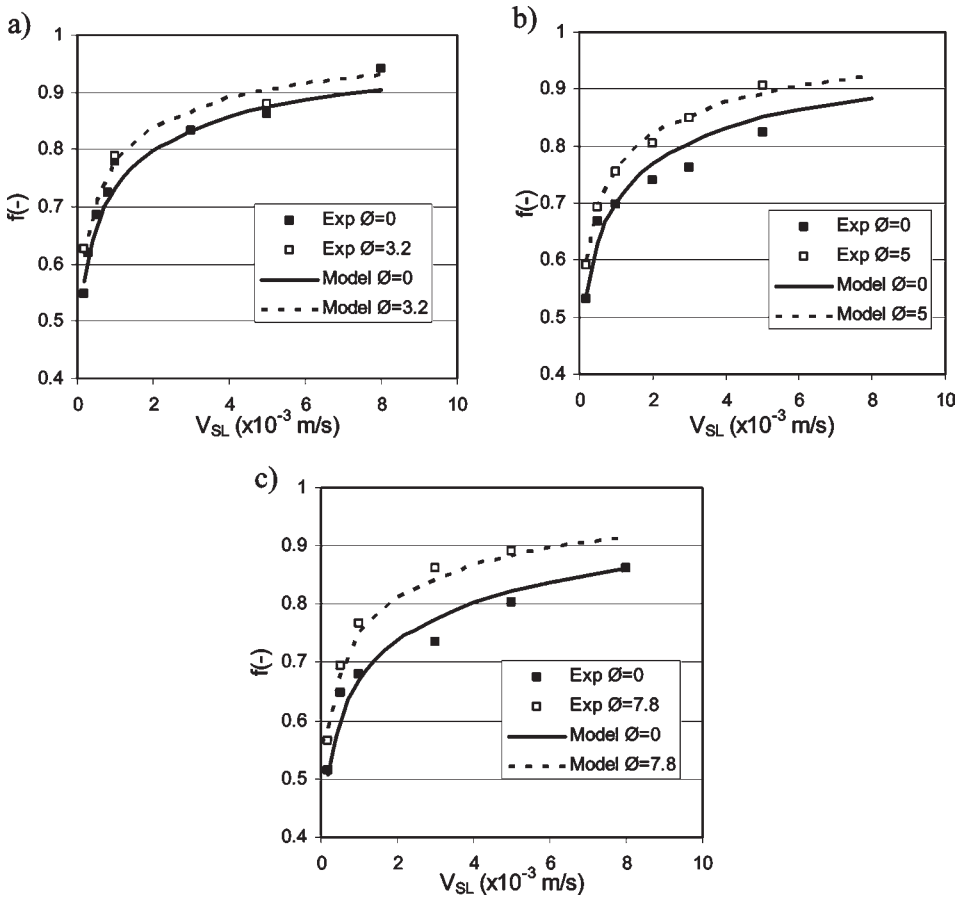


**Figure 8.** Effect of silicon carbide fines: alumina particles of (a) $d_v = 2.85 \times 10^{-3}$ m, (b) $d_v = 4.5 \times 10^{-3}$ m, (c) $d_v = 7 \times 10^{-3}$ m. Cases 6 and 13−17 from Table 3 ($\phi = 0$ corresponds to the bed without fines, $\phi = d_v/d_{SiC}$ otherwise).

**3.3.2. Parameter Optimization.** The case $n = 3$ ($\varepsilon_B + 2$ dimensionless numbers) is studied first to predict wetting efficiencies without SiC. The mean standard deviation is minimized using a generalized reduced gradient algorithm. Among the possible couples of dimensionless numbers, best results are obtained using Morton and Froude numbers (cf. Table 4). In particular, the use of the Reynolds number, often proposed in literature to represent the effect of liquid phase velocity, appears in fact rather inconvenient, because the associated effects of both the viscosity and the particle diameter are not consistent with the experimental trends.

The following correlation is then obtained:

$$f = 1 - \exp[-1.986 {Fr_L}^{0.139} {Mo_L}^{0.0195} {\varepsilon_B}^{-1.55}] \qquad (2)$$

resulting in a mean standard deviation of 5% between experimental and predicted values.

In this expression, the three dimensionless groups ($Fr_L$, $Mo_L$, and $\varepsilon_B$) are representative of three independent global features: respectively, the flow behavior at the pellet scale, the physical properties of the fluid, and the bed topology. Larger combinations of groups have been also tested (up to $n = 4$), but they have not improved the optimization criteria. As the effect of bed porosity on wetting efficiency did not prove to depend on the shape of the particles, the exponent of $\varepsilon_B$ has been supposed independent of the particle shape.

Available correlations using exponential functions have been compared with the developed one. The two relations proposed by Mills and Dudukovic[16] account for four dimensionless groups ($Re_L$, $Fr_L$, $We_L$, $a_t d_p^2/\varepsilon_B$) and are potentially able to represent the impact of all the physical parameters considered. The correlation of Ring and Missen[29] is only based on the superficial velocity of the liquid phase. Standard deviations with the considered data are about 10% for the relations proposed by Mills and Dudukovic and 13% for that of Ring and Missen. The present correlation thus improves the fitting, in particular due to a better description of the viscosity effect. It is interesting to note that a very simple relation like the one of Ring and Missen gives a not so bad prediction of the data. It underlines the preponderant effect of liquid velocity.

**3.3.3. Further Examination of the Correlation. Case of Wetting Liquids.** Corresponding parity diagram and individual standard deviations are given in Figure 9 (for the case of wetting liquids only) and Table 3, respectively.

For nonspherical particles, the deviation between experimental and calculated points is slightly modified (±0.5%) if $d_v$ is replaced by $d_s$ in the correlation. This allows indeed the assessment of the negligible effect of the particle shape discussed before.

As shown in Figures 4−6, the correlation is able to predict the wetting efficiency evolution when increasing liquid velocity and viscosity, bed porosity, and particle diameter.

The good agreement between experimental and calculated values confirms the relevance of the exponential function used in the expression of $f$.

**Case of Water and Ethanol.** The influence of the liquid wetting properties observed by Baussaron et al.[11] at very low liquid velocity (lower than $2 \times 10^{-3}$ m/s) is not actually well described by the correlation (the mean standard deviation is about 14% for ethanol and water). Despite the fact that the correlation predicts a negative influence of the surface tension $\sigma_L$ in accordance to the trend found by Baussaron et al.—$f$(water) < $f$(ethanol) < $f$(heptane)—this effect is balanced by the viscosity change in the Morton number. This may suggest that for those cases an additional parameter is required which is specific to the liquid−solid affinity. The contact angles ($\theta$) measured by the Washburn technique are: 65° and 39° for water and ethanol, respectively, and 0° for heptane (reference) and gas oil. The smaller the contact angle, the stronger the affinity of the liquid for the solid and the thinner and wider the liquid film. However, for $V_{SL} > 2 \times 10^{-3}$ m/s, the wetting efficiencies measured with heptane are only 5% higher than those measured with water. Thus the weight of this parameter is not the same in the whole velocity range.

A corrective term accounting for the contact angle and a critical Froude number is proposed here, that can further improve the predictions of the correlation:

$$N_4^{C_4} = (\cos\theta)^{1/(1+5Fr/Fr_c)} \qquad (3)$$

where $Fr_c$ is a critical Froude number equal to $7.4 \times 10^{-5}$ (corresponding to $V_{SL} = 2 \times 10^{-3}$ m/s for the considered particle size). Note that $C_4$ is here not a constant, but a decreasing function of the Froude number (bounded between 0 and 1) and that $N_4 = 1$ for heptane and gasoil which are considered as perfectly wetting liquids.

Figure 10 shows the parity diagram of the correlation (with and without the corrective factor) for the experiments of Baussaron et al. with different liquids. All data points are found to be still slightly overpredicted by the modified correlation, even with heptane. This bias is not well understood, leading to give to those points a weight penalty for the correlation regression.

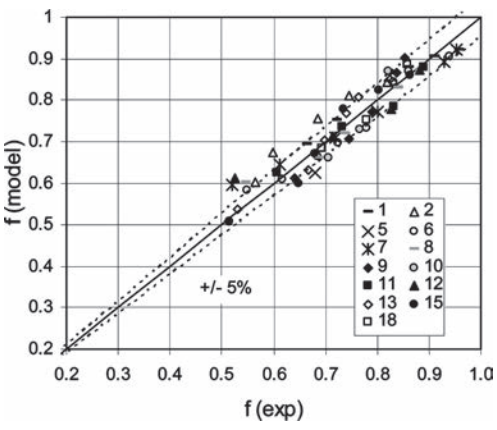


**Figure 9.** Parity diagram for wetting liquid/alumina systems. See Table 3 for experimental conditions.

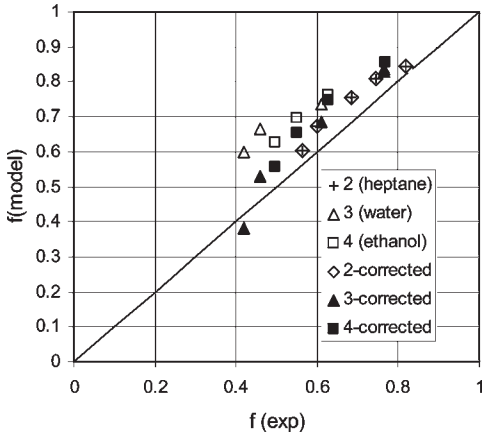


**Figure 10.** Parity diagram of original and modified correlation for heptane, ethanol, and water (data from Baussaron et al.[11]). See Table 3 for experimental conditions.

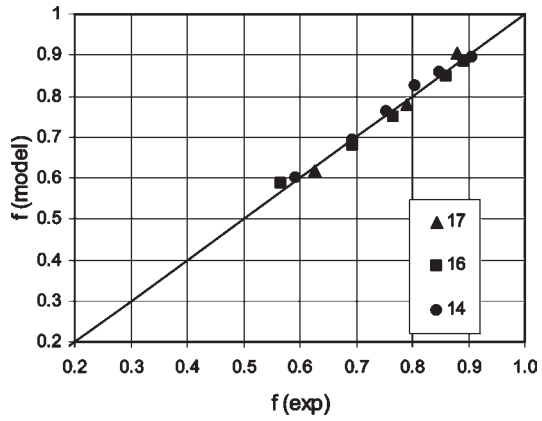


**Figure 11.** Parity diagram of the correlation including group $N_5$ when adding SiC. See Table 3 for experimental conditions.

**Addition of SiC Fine Particles.** As discussed in section 3.2.4, the effect of SiC particles is complex and difficult to model. Accounting only for the modification of bed porosity due to the addition of SiC particles is not adequate to describe the effect of fines. The number of contact points between catalyst and SiC particles is thought to be the predominant factor, but this parameter is very difficult to calculate or measure. According to the observed trend, the effect of fines has been described using an additional dimensionless parameter, written as a function of the size ratio between catalyst and SiC particles: $N_5 = (1 + \phi)$ where $\phi = 0$ when fines are not used and $\phi = d_v/d_{SiC}$ otherwise. Data regression leads to $C_5 = 0.11$, and the corresponding parity diagram is shown in Figure 11.

## 4. Conclusions

The experimental work has provided wide information on wetting efficiency by using a direct method, based on the adsorption of a dye at the wetted surface of particles. The influence of liquid phase viscosity has been investigated, as well as the separate effects of bed porosity, particle size, and shape. The parametric study performed when adding SiC particles has also allowed the correlation of the effect of fines with the size ratio between catalyst particles and fines.

The experiments performed for this study and other existing data obtained with the same procedure have been used to optimize a new model for wetting efficiency. It is based on a bounded function (decreasing exponential) and involves only three dimensionless groups (liquid Froude and Morton numbers and bed porosity) to describe classical experiments without fines and with a wetting liquid. This correlation is able to predict wetting efficiency with a very good precision for a large database. It is thus particularly suited for the small downward pilot reactors used in refineries and petrochemical research and development centers. The prediction of wetting efficiency in case of poorer liquid/solid affinity is still a challenge because it is difficult to dissociate contact angle and surface tension effects. A corrective term accounting for the contact angle and a critical Froude number has been proposed that can further improve the correlation predictions in this case. The effect of fines can be accurately described by including an additional parameter, written as a function of the size ratio between catalyst and SiC particles.

A complementary aspect to be investigated in the future would be the influence of bed anisotropy to verify the observed trends regarding the effect of bed porosity.

## Acknowledgment

The authors wish to thank Alain Muller and Jean-Louis Nadalin (LGC Toulouse) for the design and implementation of the experimental setup.

## Notations

$a_t$ = packing external surface area per unit volume of reactor, 1/m
$C_i$ = fitted constants of eq 1, −
$d_{SiC}$ = SiC (inert fines) diameter, m
$d_v$ = equivalent volume diameter, $d_v = (6V_p/\pi)^{1/3}$, m
$f$ = wetting efficiency, −
$g$ = gravitational constant, m/s$^2$
$N_i$ = Π groups or dimensionless parameters of eq 1, −
$V_{SG}$ = superficial gas velocity, m/s
$V_{SL}$ = superficial liquid velocity, m/s
$\varepsilon_B$ = porosity of the alumina particle bed, −
$\phi$ = fines parameter, $\phi = 0$ when not used and $\phi = d_v/d_{SiC}$ otherwise, −
$\mu_G$ = gas viscosity, Pa·s
$\mu_L$ = liquid viscosity, Pa·s
$\theta$ = contact angle (measured by the Washburn technique), deg
$\rho_G$ = gas density, kg/m$^3$
$\rho_L$ = liquid density, kg/m$^3$
$\sigma_L$ = liquid surface tension, N/m
$Fr_L$ = liquid Froude number, $Fr_L = (V_{SL}^2/gd_v)$, −
$Ga_L$ = liquid Galileo number, $Ga_L = (d_v^3 g\rho_L^2/\mu_L^2)$, −
$Mo_L$ = liquid Morton number, $Mo_L = (g\mu_L^4/\rho_L\sigma_L^3)$, −
$Re_G$ = gas Reynolds number, $Re_G = (V_{SG}d_v\rho_G/\mu_G)$, −
$Re_L$ = liquid Reynolds number, $Re_L = (V_{SL}d_v\rho_L/\mu_L)$, −
$Stk_L$ = liquid Stokes number, $Stk_L = (\mu_L V_{SL}/gd_v^2\rho_L)$, −
$We_L$ = liquid Weber number, $We_L = (\rho_L V_{LS}^2 d_v/\sigma_L)$, −

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