

LIQUID DISTRIBUTION IN TRICKLE BED REACTORS. I. SPREADING COEFFICIENT IN RANDOMLY PACKED POROUS BEDS

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The apparatus and method used are described for determination of spreading coefficient in a randomly packed porous bed. Values of this coefficient are given for the nickel catalyst.

Trickle bed reactor is one type of a unit in which heterogeneous catalytic reactions with contact of three phases – gas and liquid on a solid catalyst surface – can be materialized. In such reactor, reacting liquid flows downwards as a thin film on the catalyst pellets due to gravitation. Gas may flow either co or countercurrently, with the liquid product removed from the reactor bottom. This system has the advantage in perfect contact of reacting components and consequently, in high efficiency at minimum design requirements and costs.

Trickle bed reactors have been mostly used for refining of petrochemical raw materials and products (*i.e.* dehydrosulphurization and selective hydrogenation of hydrocarbon fractions)¹⁻³. For a more general application of trickle bed reactors, *e.g.* in organic technology, quantitative relations especially for mass and heat transfer are necessary. One of principal assumptions for derivation of relations for a concrete reaction, taking place in this reactor, is the knowledge of relations describing liquid distribution in the reactor packing. By distribution is understood the dependence of the density of liquid flow on the height and radius of the catalyst packing. Density of the liquid flow is defined as the volume of liquid flowing through an elementary area of the packing in a unit of time. If we know the mathematic relation describing liquid distribution, we can determine with a good accuracy the design parameters like the type and dimensions of the initial liquid source, optimum height of the catalyst packing, eventually are in a position to determine points of equal flow density and other dependences necessary for description of the given catalytic process.

Application of the liquid flow to catalytic reactions in trickle bed reactors has not yet been studied in detail. An attempt has been made here to describe the liquid distribution in random porous packing. In our study, we have used the same methods and relations as for absorption and distillation processes.

THEORETICAL

The liquid flow in a random packing can be realized by several different mechanisms. It is desirous from the view of optimum utilization of the packing catalyst surface that the downward liquid flow along individual elements of the packing takes place in a thin film and that the transfer of liquid from one packing element on to the other take place only at contact points of the catalyst particles. This mechanism is in absorption and distillation processes called either diffusive or film mechanism. Any deviation from the diffusive mechanism results in a substantially worsening utilization of the surface of catalytic charge. Obviously, it is necessary to select such conditions under which the flow of liquid along the random porous packing would correspond to the diffusive mechanism for which Cihla and Schmidt^{4,5} have proved validity of the partial differential equation (1) for axially symmetric cylindrical systems

$$\frac{\partial f(r, z)}{\partial z} = D \left[\frac{\partial^2 f(r, z)}{\partial r^2} + \frac{1}{r} \frac{\partial f(r, z)}{\partial r} \right]. \quad (1)$$

The basic criterion for determination of the diffusive flow of liquid along the packing is the dependence of the spread coefficient D on the initial wetting density which must be constant in the observed region.

Determination of this constant which determines at the same time liquid spreading on the packing surface, has been made experimentally from the measured radial distribution for 9.0×7.1 mm catalyst pellets (nickel on kieselguhr). The experiments were for evaluation of the spreading coefficient performed made so that the flow on the reactor wall be practically zero, which enabled solution of Eq. (1) with a zero boundary condition. It resulted, in this case in agreement with literature⁶, in equations describing distribution function

$$\frac{f(r, z)}{f_0} = \frac{2r_1}{a} \sum_{n=1}^{\infty} \frac{J_1(q_n R_1) J_0(q_n R)}{q_n J_1^2(q_n)} \exp(-q_n^2 T), \quad (2)$$

where q_n are roots of Eq.

$$J_0(q_n) = 0. \quad (3)$$

Relation (2) is valid for the case when liquid is brought in on the surface of the catalyst packing in the reactor axis, *i.e.* by a central source with radius r_1 . If the area of central source as compared to the reactor area (so-called hypothetical point source) can be neglected, limitation of relation (2) for $r_1 \rightarrow 0$ can be made, which results in simplifica-

tion⁷ of the equation describing the distribution function

$$\frac{f(r, z)}{f_0} = \sum_{n=1}^{\infty} \frac{J_0(q_n R)}{J_1^2(q_n)} \exp(-q_n^2 T), \quad (4)$$

where q_n are roots of Eq. (3).

In the case of unlimited packing, i.e. when $f(\infty, z) = 0$, relation by Tour-Lerman⁸ can be also used for calculation of the distribution function

$$f(r, z) = k \exp(-h^2 r^2), \quad (5)$$

where k and h are constants dependent on the packing height, total liquid flow rate and the type of catalyst packing. By introducing Eq. (5) into the diffusive equation by Cihla and Schmidt (1) we obtain $h^2 = 1/(4Dz)$ and $k = K/(Dz)$, where K is the constant dependent on the total liquid flow rate in the reactor only. Under assumption of the hypothetic point source the relation holds

$$\iint \lim_{z \rightarrow 0} f(r, 0) d\varphi dr = K \iint \delta(r) d\varphi dr = K \int_0^{2\pi} d\varphi \int_{-\infty}^{\infty} \delta(r) dr = Q. \quad (6)$$

By integration, we obtain

$$4\pi K = Q. \quad (7)$$

By modification of relation (5) and by making use of values of the constants h , k and K , we obtain for distribution in the region of an unlimited packing a very simple Eq. (8), from which the spreading coefficient can be determined graphically since this equation can be easily linearized by the logarithm of relation

$$f(r, z) = Q/(4\pi Dz) \exp[-r^2/(4Dz)]. \quad (8)$$

EXPERIMENTAL

The apparatus and measurements. The apparatus (Fig. 1) consisted of a glass cylinder of inside diameter 0.25 m 1, base 2, distributor and a device enabling its installation in the column 3, doser (charger) and a controller 4, remover 5. The base was divided into 13 removing sections in a form of centric annular rings. Each annulus was filled with a helix to prevent passage of packing elements which could block holes for the liquid exit. Dimensions of individual annular rings are given in Table I. For concentration of the charge was chosen the central point source of diameter 0.004, 0.006, 0.01 and 0.016 m according to the varying quantity of the charged media. As wetting liquid was chosen water with its temperature kept at 20°C.

After random packing of the required height of catalyst, the column was intensively wetted by liquid with maximum wetting of both the packing and column walls. Due to this initial wetting,

the steadying time of the regime in the column was considerably shorter and reproducibility of the results was increased. The initial wetting densities were: 15, 11, 7, 3 and $1 \text{ m}^3/\text{m}^2$ hour. All measurements were performed from higher to lower values. The range of initial wetting densities within the mentioned limits comes most often into consideration in catalytic reactions in a trickle bed reactor.

Evaluation of experimental data. The accuracy with which the spreading coefficient is determined from an experimentally found profile of liquid flow in individual sections (at a given height of the packed bed) will depend on the number of sections in the base and on the area of the section. This way, two contradictory phenomena appear. The number of experimental data (points) rises with the number of annular rings, but on the other hand the accuracy of the found values of each point decreases with their increasing number due to feasible fluctuation of the liquid stream from the elements of packing situated in the neighbourhood of the removing sections. Therefore a certain compromise must be found between these two phenomena.

An error caused by fluctuation is decreasing by increasing number of visible pellets in an individual section. The number of visible pellets is determined on a circular area by relation⁹

$$p = 3/2(1 - \varepsilon) (d_k/d_p)^2 . \quad (9)$$

For the given experimental arrangement the number of visible pellets on the circular area p as well as their number in individual sections m (Table II) was calculated (d_p was substituted by $d_{ekv} = 8.8 \text{ mm}$). The experimentally found overall number of visible pellets (p_{12}) made 645, the calculated number was 645.8. It is obvious from Table II that the error caused by fluctuation of one stream is greater than 5% in the 1st and 2nd removing sections only. For this reason the measured values of the liquid flow rates through individual sections were used directly without further correlations.

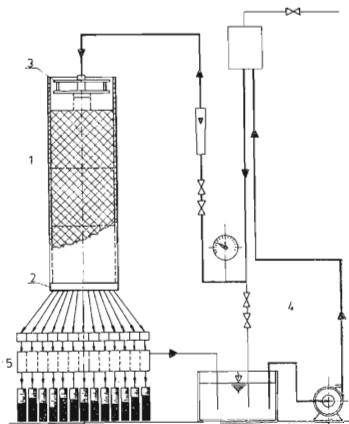


FIG. 1

Block Diagram of Experimental Apparatus

1 Glass column with packing; 2 base; 3 liquid source; 4 charging and controlling sections; 5 removing section.

RESULTS

For determination of the spreading coefficient was used the experimentally found radial distribution. Experiments were arranged so that the zero boundary condition was fulfilled; this was attained by an appropriate choice of the packing height. In this case the maximum height of the catalyst packing was 0.2 m at a zero wall flow. The spreading coefficient was expressed by non-linear regression analysis by Marquart¹⁰ from relations (2), (4) and (8). As the first estimate of the spreading coefficient for the non-linear regression its value found from relation (8) by the least square method was used (linearization was made by using coordinates $\ln f$ against r^2). The spreading coefficient was determined at the initial wetting densities 1 to 15 m³/m² h. The measurement was made for each value of the initial wetting density twice, where for repeated measurements of the radial distribution the packing

TABLE I
Geometrical Parameters of the Base Reactor

Section	Mean radius m	Area 10 ⁴ m ²	Dimension- less radius	Section	Mean radius m	Area 10 ⁴ m ²	Dimension- less radius
1	0.0085	9.075	0.07	8	0.0857	56.543	0.68
2	0.0222	14.671	0.18	9	0.0964	65.063	0.77
3	0.0327	21.595	0.26	10	0.1071	72.320	0.85
4	0.0434	29.283	0.35	11	0.1160	51.004	0.92
5	0.0540	35.608	0.43	12	0.1225	46.158	0.98
6	0.0647	44.729	0.52	13	0.1255	—	1.00
7	0.0754	48.519	0.60				

TABLE II
Number of Visible Pellets on the Area of Reactor Base (p) and Number of Pellets in Individual Sections (m)

Section	d_k/d_p	p	m	Section	d_k/d_p	p	m
1	3.9	12.1	12.1	7	18.3	266.2	62.7
2	6.3	31.6	19.5	8	20.7	340.7	74.5
3	8.6	58.8	27.2	9	23.1	424.2	83.5
4	11.1	97.9	39.1	10	25.6	521.1	96.9
5	13.5	144.9	47.0	11	27.2	588.1	67.0
6	16.0	203.5	58.6	12	28.5	645.8	57.7

TABLE III

Dependence of the Spreading Coefficient Determined from Given Equations by the Method of Non-Linear Regression on Initial Wetting Density

Spreading coefficient evaluated from Eqs D_1 -(8); D_2 -(2); D_3 -(4). a , b repeated measurements.

f_0 $\text{m}^3 \text{m}^{-2} \text{h}^{-1}$	D_1, m		D_2, m		D_3, m
	a	b	a	b	a
1	0.0021	0.0018	0.0021	0.0019	0.0022
3	0.0020	0.0020	0.0020	0.0020	0.0020
7	0.0022	0.0023	0.0022	0.0022	0.0022
11	0.0019	0.0021	0.0018	0.0020	0.0020
15	0.0018	0.0018	0.0020	0.0021	0.0023
∅	0.0020	0.0020	0.0021	0.0020	0.0021

was removed from the reactor and was packed again. Results of those experiments are given in Table III.

In spite of the fact that individual measured values of the radial distribution have had a relatively large dispersion (about 20% rel.), reproducibility of determination of the spreading coefficient D is very good for the given initial wetting density as can be seen from Table III by comparison of paragraphs a and b . It can be also seen from Table III that the used Eqs (2), (4) and (8) are for determination of the spreading coefficient equivalent. It is thus advantageous to use the modified Tour-Lerman's relation (8) because of its simplicity. It follows from the dependence of the spreading coefficient on the initial wetting density (Table III) that the liquid flow rate through a randomly packed porous bed in a given range of wetting densities is in accordance with the diffusive mechanism.

LIST OF SYMBOLS

a	radius of the reactor (m)
D	spreading coefficient (m)
$d_{\text{ekv}} = 1.241(V_p)^{1/3}$	equivalent diameter of packing element (m)
d_k	reactor diameter (m)
d_p	diameter of packing element (m)
$F = f/f_0$	dimensionless wetting density
f	wetting density ($\text{m}^3 \text{m}^{-2} \text{h}^{-1}$)
$f_0 = Q/(\pi a^2)$	initial wetting density ($\text{m}^3 \text{m}^{-2} \text{h}^{-1}$)
h	constant by Tour and Lerman (m^{-1})
J_0	Bessel function first type, zero order
J_1	Bessel function first type, first order
K	constant
k	distribution constant according to Tour and Lerman (m^{-2})

m	number of visible catalyst pellets on the area of the removal section
p	number of visible catalyst pellets on circular area
Q	liquid feed ($\text{m}^3 \text{h}^{-1}$)
q_n	roots of transcendental equations
$R = r/a$	dimensionless radius
r	radius in cylindrical coordinates (m)
$R_1 = r_1/a$	dimensionless radius of the liquid source
r_1	radius of liquid source (m)
V_p	volume of a packing element (m^3)
$T = Dz/a^2$	dimensionless spreading coefficient
z	bed height (m)
δ	function characterizing the liquid source
ε	porosity of packing

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