EFFECT OF PHYSICAL PROPERTIES OF THE WETTING LIQUID ON ITS DISTRIBUTION IN TRICKLE BED REACTORS*

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The effect is discussed of viscosity, surface tension, density and polarity of wetting liquid on its distribution in the catalytic bed of a reactor.

Recently¹⁻⁴ water was used as the wetting liquid in studies of liquid distribution on the porous catalytic packing of column type reactors. Here, the effect of variation of some of physical properties of the wetting liquid such as viscosity, surface tension, density and polarity on liquid distribution is studied.

The effect of physical properties of the wetting liquid on its distribution has been studied 5^{-8} only with polar liquids on non-porous packings while the effect of polarity has not been studied yet. It is possible to expect that physical properties of the wetting liquid affecting primarily the liquid transfer mechanism can be presented in agreement with the literature^{2,5,9,10} by use of two simplified models: *I*) Film (diffusion model) with the main part of liquid flowing downward on individual elements of packing in a narrow film. Beside the surface forces also gravitational forces and the geometry of the packing must be taken into account. 2) Channel model, with the origination of channels is the result of the random arrangement of the packing and of local changes of wettability of the packing but also physical properties of the liquid have a considerable effect first of all the surface tension. This transfer mechanism which is not suitable in the majority of catalytic and separation operations is controlled by surface forces.

It is also necessary to realize that viscosity affects the thickness of the liquid film on the surface of the packing element which can be observed in the reaction kinetics in the analysis of the system. Also the density of the wetting liquid can have a certain effect on distribution operation though its value is varying in a relatively narrow range as compared to viscosity and surface tension.

In quantitative evaluation only Staněk⁵ gives correlation coefficients characterizing the effect of physical properties of the liquid. Other authors^{6–8} state that this factor has no significant effect on quantitative description of its distribution in the studied range of physical properties of the wetting liquid.

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^{*} Part V in the series Liquid Distribution in Trickle Bed Reactors; Part IV: This Journal 40, 3145 (1975).

EXPERIMENTAL

The apparatus used for experimental verification of liquid distribution in the randomly packed bed of a reactor has been described together with the measuring procedures and the characteristics of the used packings in our recent studies¹⁻⁴.

Basical physical parameters of polar and non-polar wetting liquids are given in Table I. Polar wetting liquids were water, aqueous solutions of glycerol and solution of wetting agent in water. Nonpolar wetting liquid was a mixture of paraffin and naphthalenic hydrocarbons commercial mark Petropal (produced by Benzina) and a mixture of Petropal with cylindrical oil B 25 (Czechoslovak Standard 65 66 23).

The initial wetting densities were 1, 3, 7, 11 and 15 m³ m⁻² h⁻¹ with the uniform liquid source at maximum 11 m³ m⁻² h⁻¹.

RESULTS AND DISCUSSION

Effect of viscosity. The basical criterion for judging the degree of existence of the diffusion mechanism at liquid transfer is the dependence of the spreading coefficient on the initial wetting density which must be constant for verification of the diffusion mechanism of liquid transfer. At wetting the packings A, B, and C (symbols and characteristics of the packings used are given in the last paper of this series⁴) by water in the range $f_0 = 1-15 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$ the constant value of the spreading coefficient was proved².

The spreading coefficients for the packing A are given in Table II at substitution of water by aqueous solutions of glycerol, while the other experimental conditions

TABLE I Physical Properties of the Used Wetting Liquids, $t = 20^{\circ}$ C

Kind	Symbol	Density g cm ⁻³	Viscosity cP	Surface tension dyn cm ⁻¹	
Water		0.998	1.00	72.75	
25% (mass) aqueous solution of glycero	solution I	1.060	2.09	71.73	
47% (mass) aqueous solution of glycerol	solution II	1.119	5.25	69.82	
68% (mass) aqueous solution of glycerol	solution III	1.175	19.98	67.44	
0.09% (mass) aqueous solution	solution IV	0.998	1.00	49.00	
of wetting agent*					
Petropal		0.862	4.68	30.60	
Petropal + cylinder oil	solution V	0.867	7.83	39.70	
Petropal + cylinder oil	solution VI	0.870	10.03	39.75	

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Triethanolamine salt of mono- and dialkylphosphate acid (2-ethylhexanol).

remain constant. The spreading coefficient is evaluated from the modified Tour-Lerman relation¹. It is obvious that for solutions having concentrations of 47 to 68 mass % the spreading coefficient is decreasing with the decreasing flow rate. Certain tendency to chanelling was also observed visually with the above mentioned solutions of glycerol. At wetting with the 25% solution of glycerol the diffusion transfer mechanism was confirmed and the mean value of the spreading coefficient was equal to that at the wetting with water.

The degree with which the channeling mechanism exists has not been profound in the region of studied wetting densities $(0.998 - 1.175 \text{ gcm}^{-3})$ and viscosities (1.00 - 19.48 cP). This conclusion has been verified, among other, by the usual equilibrium wall flow reached in the range of experimental accuracy with wetting by water and by 68% aqueous solution of glycerol on packings A, B and C (Table III). Also the scatter of experimental liquid distributions at wetting of various heights of packings A by aqueous solutions of glycerol has not been greater than the theoretical curve when comparing with wetting by water (Table IV). This curve has been calculated on basis of the mathematical model proposed by Kolář and Staněk^{5,11} based on the assumption of liquid transfer due to diffusion mechanism. Determination of individual constants is given in our recent publication³ (at evaluation of the dimensionless spreading coefficient the average value of D was substituted for $f_0 = 1-15 \text{ m}^3$. $m^{-2} h^{-1}$).

Effect of surface tension. The spreading coefficients at wetting by aqueous solution of wetting agent are also given in Table II which confirms the diffusion mechan-

TABLE II

Timid		f_0 , m ³ m ⁻² h ⁻¹					
Liquid	15	11	7	3	1	Ø	
Water	0.1942	0.2011	0.2200	0.2233	0.1814	0.2000	
Solution I	0.2304	0.2092	0.1680	0.1600	0.2360	0.2007	
Solution II	0.1933	0.1797	0.1736	0.1452	0.1689	0.1722	
Solution III	0.1991	0.1682	0.1575	0.1539	0.1472	0.1650	
Solution IV	0.1961	0.1874	0.1761	0.2082	0.1917	0.1919	
Petropal	0.1495	0.1372	0.1369	0.1621	0.1503	0.1472	
Solution V	0.1292	0.1244	0.1213	0.1445	0.1913		
Solution VI	0.1069	0.0954	0.1292	0.1454	0.1800	-	

Spreading Coefficient $D \cdot 10^2$ (m) in Dependence on the Initial Wetting Density for Water, Aqueous Solutions of Glycerol and Wetting Agent, Petropal and Solutions of Petropal with Cylinder Oil (Packing A)

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ism of liquid transfer which can be expected with liquids of the lowered surface tension. In this experimental arrangement the mean value of the spreading coefficient is practically identical with its value obtained at wetting the packing A with water. Also the equilibrium wall flow on packings A, B and C was equal with these both types of wetting liquids (Table III).

Higher degree of uniformity of the flow is manifested in the catalytic packing at wetting by liquid with reduced surface tension by a lower scatter of experimental data from the theoretical curve in comparison with other liquids (Table IV).

Thus it can be concluded that the mathematical model by Kolář and Staněk describes with sufficient accuracy the distribution of polar liquids with both the increased viscosity and with the decreased surface tension in the region of studied properties $(1\cdot00-19\cdot48 \text{ cP}, \text{ or } 49\cdot0-73\cdot0 \text{ dyn cm}^{-1})$ at application of porous packings.

TABLE III

Wall Flow (%) in the Region of Equilibrium Liquid Distribution for Packings A, B, C at Wetting by Water Solutions III, IV, and Petropal, Point Central Source, Column \emptyset 0.159 m

Packing type	Water	Solution III	Solution IV	Petropal	
А	15.9	17.7	15.7	17.1	
в	9.1	7.7	11.7		
С	11.8	10.3	14.5	-	

TABLE IV

Standard Deviation of Experimental Data in the Region of Limited Packed Bed from the Theoretical Curve Calculated According to the Model by Kolář and Staněk at Wetting the Packing A by a Central Point Source with Various Types of Liquids (Column \emptyset 0.254 m)

Bed height	Water	Solution I	Solution II	Solution III	Solution IV
0.5	0.3013	0.2942	0.3144	0.2967	0.2231
0.8	0.3162	0.2764	0.2825	0.3109	0.1817
1.1	0.3299	0.3002	0.2547	0.2944	
1.5	0.1438	0.2023	0.2202	0.2311	0.1713
1.8	0.1887	0.2361	0.1894	0.1674	

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Wetting by non-polar liquid. In studies of the effect of polarity of the wetting liquid on the distribution process, the liquid Petropal was used having beside other also a relatively small viscosity and surface tension.

The existence of three phases of flow - region of unlimited packing, limited packing and equilibrium distribution were similarly verified as with wetting by polar types of liquids³ at the flow of this liquid through the randomly packed bed of catalysts A.

The spreading coefficient was evaluated in the region of infinite packing by the modified Tour-Leman relation. Its value is independent of the initial wetting density and is significantly smaller (0.001472 m) as compared to the spreading coefficients for water and aqueous solutions of glycerol or wetting agent at the use of the same catalyst. With the increasing viscosity (mixture of Petropal and cylinder oil) at this type of liquid there takes place a relatively significant effect of the chanelling mechanism of liquid transfer verified by the dependence of the spreading coefficient on the initial wetting density (Table II). For this reason other experiments with these types of liquids were left out.

For description of distribution of non-polar liquids on the randomly packed bed of catalyst was used the model according to Cihla and Schmidt¹² which described with a relatively good accuracy the given process for the polar liquid³.



FIG. 1

Comparison of the Theoretical Distribution Curve Calculated According to the Model by Cihla and Schmidt with Experimental Points

Point central source, packing A, liquid Petropal, column diameter 0.251 m; \bigcirc ______ z = 0.4 m, \bullet _____ z = 1.2 m.



Comparison of the Theoretical Curve Calculated According to the Model by Cihla and Schmidt with the Experimental Data

Circular shower, packing A, liquid Petropal, column diameter 0.251 m, z = 0.8 m.

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This model has been proved suitable (Figs 1 and 2). The greatest deviation of experimental data from the theoretical curve was at the use of the point central source ($\sigma = 0.5363$; for the circular shower $\sigma = 0.4818$ and for the uniform source $\sigma = 0.1800$ for z = 0.6 to 1.5 m). Its relatively high value is firstly affected by the spread of experimental data, secondly by the error in calculation at high local wetting densities in the centre of the packing at smaller heights where the absolute error is the largest.



FIG. 3

Wall Flow in Dependence on the Dimensionless Bed Height

Point central source, packing A, liquid Petropal O ——— column diameter 0.084 m, • ——— column diameter 0.159 m.





Wall Flow in Dependence on the Dimensionless Bed Height

Point central source, packing A, liquid water $\bigcirc -\cdots -$ column diameter 0-084, • ---- column diameter 0-159, • ----column diameter 0-251 m.



FIG. 5

Comparison of Calculated and Experimental Data of Wall Flow in Columns 0.084 and 0.159 at Wetting by a Central Point Source, Liquid Petropal

○ Column diameter 0.084, ● column diameter 0.159 m.

The mathematical evaluation according to Cihla and Schmidt must be considered as the first approximation. If it were necessary to describe the operation more accurately, the multiparameter models should be used out of which perhaps the most suitable would be the model according to Kolář and Staněk.

The wall flow is affected by a number of factors which were discussed in our last $study^4$. The effect of physical properties of the wetting liquid, most of all polarity, is manifested at the flow of liquid from the wall of the equipment. The bed height on which the liquid reaches the wall of the reactor (region of limited-finite packing) as well as the height from which the wall flow begins to be constant varies with the type of liquid.

By studying the dependence of the wall flow on the dimensionless bed height in the given region d_k/d_p , it has been determined that, as long as the type of the wetting liquid and packing does not change, the equilibrium wall flow in columns of various diameters is reached at the constant value Z. This dependence is obvious first of all from Figs 3-4, where are given the limiting dimensionless bed heights at the reached equilibrium distribution for water and Petropal. The obtained limiting dimensionless bed heights (for water $Z_L = 5$, Petropal $Z_L = 9$, pellets of Nickel catalyst kieselguhr) enable the calculation of the minimum bed height at which the equilibrium liquid distribution is reached for columns of various diameters ($z = Z_L d_k$). The equilibrium wall flow was in the range of experimental errors for all types of used liquids the same (Table III).

For calculation of the wall flow at wetting by nonpolar liquids the relations given in paper⁴ can be used (Fig. 5).

LIST OF SYMBOLS

$d_{\mathbf{k}}$	column diameter, m
d	particle diameter, m
Ď	spreading coefficient, m
f	wetting density, $m^3 m^{-2} h^{-1}$
f_0	initial wetting density, $m^3 m^{-2} h^{-1}$
$F = f/f_0$	dimensionless wetting density
R	dimensionless diameter of column defined as the ratio of radius of the section where
	the wetting density is measured to the total column radius
Wn	wall flow, %
z	bed height, m
$Z_{\rm L}$	dimensionless limiting bed height

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