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> MASS TRANSFER IN A PACKED BED OF RASCHIG RINGS AT LOW PECLET NUMBERS

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#### ABSTRACT

The mass transfer coefficients in non-irrigated packed beds of porous ceramic Raschig ring have been determined with the help of drying experiments in the constant rate period. Raschig rings of 6.8, 8.7, 10.4 and 16.2 mm outside diameter in packed beds of 50, 100, 150 and 200 mm height and 100 mm diameter have been investigated. For Peclet numbers greater than about 104 the Sherwood numbers were found to lie above those calculated for a single sphere as is to be expected. The factor, which was determined from the data of other workers and also from the present work, was found to be 1.9. In the low Peclet number range (Pe > 1000) the experimentally determined Sherwood numbers were found to decrease sharply with Peclet number and were found to lie much below the single sphere values. The bypass model, for the non-uniform distribution of gas, presented for a packed bed of spheres by E.U. Schlünder /1/ and H. Martin /2/ has been extended here to apply to a packed bed of Raschig rings also. The model explains the characteristic variation of Sherwood numbers in the low Peclet number range, for almost all the experimental data, to a good degree of accuracy.

#### Introduction

The heat and mass transfer in packed beds can be predicted from single particle calculations /3/.

$$Sh = f_{\psi} \cdot Sh_{p}$$
 (1)

 $f_{\psi}$  is a function of the void fraction and the shape of the particles. Sh for the single particle can be calculated by the equations proposed by Gnielinski /3/.

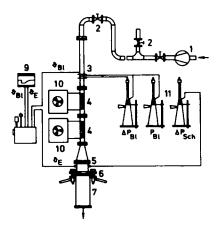
For spherical particles the factor  $f_\psi$  in equation (1) has been given by Schlünder /4/ to be

$$f_{\psi} = 1 + 1.5 (1 - \psi)$$
 (2)

In the high Peclet number region (Pe> $10^3$ ) this method was shown to predict the heat and mass transfer coefficients to within 15 per cent /3/, but in the low Peclet number region (Pe<10<sup>2</sup>) the experimental results begin to fall much below those calculated from equations (1) and (2). Kunii and Suzuki /5/ have summarized the heat transfer data of a number of authors and have shown that the experimental results in the low Peclet number region lie a few decades below the line predicted for a single sphere. Martin /2/ has applied a bypass stream model developed by Schlünder /1/, based on the non-uniform distribution of void fraction in a randomly packed bed of spheres, which explains this discrepancy between theory and experimental results to a good degree. In the following the same model will be extended to a packed bed of Raschig rings and will be compared with the experimental data obtained with four sizes of Raschig rings at various bed heights.

### Experimental

The mass transfer coefficients, for the packed bed of Raschig rings, were obtained by performing drying experiments and determining the rate of drying in the constant rate period.





Schematic diagram of the experimental set-up; 1 blower, 2 valve, 3 orificemeter, 4 heater, 5 filter, 6 quick coupling device, 7 packed bed, 8 thermostat, 9 recorder, 10 transformer, 11 Betz-manometer,  $\vartheta_{B1}/\vartheta_E$  thermocouples.

FIG. 1 is a schematic flow diagram of the experimental set-up used. A stream of preheated air was made to flow through a packed bed of porous, ceramic Raschig rings (which were previously wetted, under vacuum, with water). The packed columns, of various heights, were made of plexiglass pipe and also of porous ceramic pipe, which was also wetted with water, to ensure the temperature of the wall was nearly the same as that of the packing. A coupling device enabled the packed bed to be coupled to or removed from the set-up quickly. The rate of water evaporated during the constant rate period was determined by weight loss. The saturation concentration  $\tilde{X}_{s}$  was calculated from the adiabatic saturation temperature, which was determined from the equation

$$\vartheta_{\mathbf{I}} - \vartheta_{\mathbf{s}} = \frac{\widetilde{\Delta h}_{\mathbf{v}}}{\widetilde{c}_{pD}} \left\{ \left[ 1 + \frac{\widetilde{\mathbf{x}}_{\mathbf{s}}^{*} - \widetilde{\mathbf{x}}_{\mathbf{I}}}{1 - \widetilde{\mathbf{x}}_{\mathbf{s}}^{*}} \right]^{1} - 1 \right\}$$
(3)

 $\tilde{\Delta h}_v =$  phase change enthalpy  $\tilde{c}_p =$  heat capacity

proposed by Schlünder /6/. The concentration  $\tilde{x}_{I}$  of the entering air was measured by a psychrometer and the concentration  $\tilde{x}_{O}$  of the leaving stream was calculated by a material balance. TABLE 1 shows the packing sizes and packed heights covered in the present experimental work.

TABLE 1							
Data	of	the	Raschig	Rings	and	Packed	Beds

d <sub>i</sub> (mm)	d (mm)	h (mm)	H (mm)	H/d <sub>o</sub>	A (m <sup>2</sup> )	$(m^2/m^3)$	ψ
5 d <sub>r</sub> =	6.78 0.91		50 107 150 200	7.37 15.78 22.12 29.49	0.3031 0.6062 0.9093 1.2124	799 729 779 779 779	0.690 0.711 0.690 0.690
1	8.65 0.94	•	47 93 143 193	5.43 10.75 16.53 22.31	0.2091 0.4182 0.6273 0.8364	572 578 564 557	0.714 0.711 0.718 0.721
	10.4 0.9 m	10.35 m	55 110 153 190	5.28 10:57 14.71 18.27	0.1775 0.3817 0.5358 0.6697	415 446 450 453	0.832 0.796 0.788 0.784
	16.2 2.05	15.6 mm	47 93 150 190	2.90 5.74 9.26 11.73	0.1177 0.2275 0.3452 0.4708	322 315 296 319	0.708 0.715 0.732 0.711

### The Bypass Stream Model for Raschig Ring Packings

This is an extension of the model, for spheres, proposed by Martin /2/ to Raschig ring packings.

It is a fact that the voidage in the immediate vicinity of the wallin a packed bed of Raschig rings is larger than that in the rest of the bed. This, therefore, leads to channels (regions) in the bed of different cross-section. The model consists of a bed of uniformly sized particles, with a void fraction  $\psi_1$  in a part  $(1-\Psi) \cdot f$  of its total cross-section f and another part, the by-pass cross-section  $\Psi \cdot f$  (near the wall), with a larger void fraction  $\psi_2$ . Since the two parts of the packed bed will offer different resistances to the flow, a nonuniform flow-rate distribution will result. The resistances can be calculated from the following equation (3) to (7) given by Brauer /7/.

$$\xi = \frac{\psi^2}{1-\psi} \cdot \frac{\Delta p}{\rho u^2} \cdot \frac{d_r}{H}$$
(4)

For Raschig rings the equivalent diameter  ${\rm d}_{\rm r}$  can be calculated from the equation

$$\mathbf{d}_{\mathbf{r}} = \mathbf{d}_{\mathbf{o}} \cdot \mathbf{E}^{\mathbf{n}}$$
(5)

where the exponent n is given as 1.9 /7/ and E is a function of the relative void volume  $\epsilon_r$  of a ring  $(\epsilon_r = (d_1/d_0)^2)$ 

$$E = \frac{1 - \varepsilon_r}{1 + (2/3) \cdot \sqrt{\varepsilon_r} - (1/3) \varepsilon_r}$$
(6)

For packings of granulated particles Ergun's equation for the resistance to flow in a packed bed is

$$\xi = \frac{A}{Re_{E}} + B$$
(7)

where A = 150 and B = 1.75

$$\operatorname{Re}_{\mathbf{E}} = \frac{1}{1-\psi} \cdot \frac{\mathbf{u} \cdot \mathbf{d}_{\mathbf{r}}}{v}$$
(8)

This equation can be used for Raschig rings provided  $d_r$  is defined as in equation (5). From equations (4), (5) and (8) we obtain another form of Ergun's equation which can be applied to both parts of the model packed bed.

$$\frac{\Delta P}{H} = \frac{A(1-\psi_1)^2}{\psi_1^3} \cdot \frac{\eta \ u_1}{d_r^2} + \frac{B(1-\psi_1)}{\psi_1^3} \cdot \frac{\rho \ u_1^2}{d_r}$$
(9)  
$$\frac{\Delta P}{H} = \frac{A(1-\psi_2)^2}{\psi_2^3} \cdot \frac{\eta \ u_2}{d_r^2} + \frac{B(1-\psi_2)}{\psi_2^3} \cdot \frac{\rho \ u_2^2}{d_r}$$
(10)

where  $u_1$  and  $u_2$  are the superficial velocities in the two sections and given by

$$u_1 \equiv \frac{\hat{v}_1}{(1-\varphi)f}$$
 and  $u_2 \equiv \frac{\hat{v}_2}{\varphi \cdot f}$  (11)

The velocity ratio  $u_2/u_1 = w$  can be calculated from the equations (9) and (10) by the method given by Martin /2/, provided the particle diameter is replaced by  $d_r$ .

Now the individual changes in vapor concentration of the two streams can be denoted by

$$\phi_1 = \frac{\tilde{x}_{01} - \tilde{x}_s}{\tilde{x}_1 - \tilde{x}_s} = \exp(-NTU_1)$$
(12)

$$\phi_2 = \frac{\tilde{x}_{02} - \tilde{x}_s}{\tilde{x}_1 - \tilde{x}_s} = \exp(-NTU_2)$$
(13)

From a mass balance the average change in vapor concentration follows to be

$$\phi = \frac{\dot{v}_1}{\dot{v}} \phi_1 + \frac{\dot{v}_2}{\dot{v}} \phi_2 = \exp(-NTU)$$
(14)

where NTU is the number of transfer units for the combined system. From this it follows that

$$NTU = -\ln \phi = -\ln \left[ \frac{\dot{v}_1}{\dot{v}} \phi_1 + \frac{\dot{v}_2}{\dot{v}} \phi_2 \right]$$
(15)

$$= -\ln\left[(1-\dot{v}) \exp(-NTU_1) + \dot{v} \exp(-NTU_2)\right]$$
(16)

where

$$\dot{v} = \frac{\dot{v}_2}{\dot{v}} = \frac{1}{1 + \frac{1 - \phi}{\phi \cdot w}}$$
 (17)

With

$$Sh = \frac{\beta}{\delta} \frac{d_{O}}{\delta} = \frac{\tilde{V} \cdot NTU}{A} \cdot \frac{d_{O}}{\delta} = \frac{U \cdot f \cdot H \cdot NTU}{A \cdot H} \cdot \frac{d_{O}}{\delta}$$
$$= \frac{Pe}{A_{V} \cdot H} \cdot NTU = \frac{Pe_{\psi} \cdot \psi}{A_{V} \cdot H} \cdot NTU \qquad (18)$$

the Sherwood number for the combined system can now be written, by substituting for NTU from equation (16), as

$$Sh = -\frac{Pe_{\psi} \cdot \psi}{A_{\psi} \cdot H} ln \left[ (1-\dot{v}) exp(-NTU_{1}) + \dot{v} exp(-NTU_{2}) \right] (19)$$

Here  $NTU_1$  and  $NTU_2$  have been calculated from equation (18) as

$$NTU_{1} = \frac{A_{v1}^{H}}{Pe_{\psi_{1}}^{\psi_{1}}} \cdot Sh_{1} (Pe_{\psi_{1}}, Sc)$$
(20)

$$NTU_{2} = \frac{A_{v2} \cdot H}{Pe_{\psi_{2}} \cdot \psi_{2}} \cdot Sh_{2} (Pe_{\psi_{2}}, Sc)$$
(21)

where

 $\operatorname{Pe}_{\psi_1} = \frac{1-\dot{v}}{1-\varphi} \frac{\operatorname{Pe}}{\psi_1} \quad \text{and} \quad \operatorname{Pe}_{\psi_2} = \frac{\dot{v}}{\varphi} \frac{\operatorname{Pe}}{\psi_2}$ 

The Sherwood numbers for the two sections of the model packed bed have been calculated from the packed bed correlation of Gnielinski /3/ (as is valid for the range of high Peclet numbers). However, since for Raschig rings the value of  $f_{\psi}$  in eq. (1) is not known, it had to be determined.

The data of a number of workers /8 - 13/ on Raschig ring packings, have been collected and the Sherwood numbers have been plotted as a function of Peclet numbers in FIG. 2. The results of our own experiments have also been shown in this figure and an average value of  $f_{\psi}$  has been determined as a constant equal to 1.9.

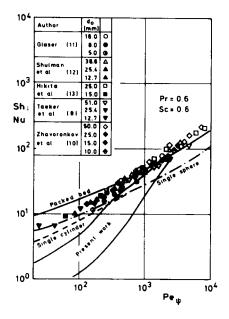


FIG. 2

Experimental data of heat and mass transfer in packed beds of Raschig rings

For comparison the (volume) specific surface area of the packing in the two parts of the packed bed, for use in equations (20) and (21) have been calculated as

$$A_{vi} = \frac{1 - \psi_i}{1 - \psi} \cdot A_v$$
 (22)

The values of  $\psi$  are given in TABLE 1 and  $\psi_1$  and  $\psi_2$  are calculated as

$$\psi_2 = \psi + \Delta \psi$$
$$\psi_1 = \frac{\psi - \varphi \psi_2}{1 - \varphi}$$

Roblee et al. /14/ have shown in their studies of radial porosity variations in packed beds of Raschig rings that the porosity of the wall zone  $(\psi_2)$  is about 12 - 15 per cent greater than that in the middle. Taking this as basis  $\Delta \psi$  is calculated to be 0.1. The bypass zone is assumed to be one particle diameter thick and the bypassed fraction of the cross-sectional area is then

$$\varphi = \frac{\pi D \cdot d_r}{\pi D^2/4} = 4 d_r/D$$

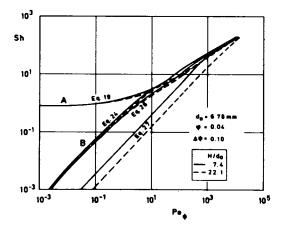


FIG. 3 Comparison of the course of several equations

Equation (19) is plotted in FIG. 3 as curve A. It can be seen that Sh decreases with decreasing Pe down to a Peclet Number of about 1 and then reaches a constant value, below that predicted for the single sphere. But when compared with the experimental results, for the packed bed, it still predicts a much larger value of Sh, particularly for Pe < 200.

Zabrodsky /15/ has suggested the influence of longitudinal heat conduction (diffusion), apart from non-uniform distribution of gas, to be responsible for the decrease in effective Nu (Sh). Taking longitudinal diffusion also into consideration, therefore,  $exp(-NTU_1)$  and  $exp(-NTU_2)$  can be worked out to be, by analogy /15/

$$\phi'_{i} = \exp(-NTU_{i}) = \exp\left(-\frac{1}{2}\operatorname{Pe}_{i}\cdot\frac{H}{d_{o}}\sqrt{1+\frac{4NTU_{i}}{\operatorname{Pe}_{i}\cdot H/d_{o}}}\right)$$
 (23)

Applying this to both parts of the model packed bed and substituting for  $exp(-NTU_1)$  and  $exp(-NTU_2)$  in equation (19) gives the equation for the combined effect of non-uniform gas distribution and longitudinal diffusion as

$$Sh = -\frac{Pe_{\psi} \cdot \psi}{A_{\psi} \cdot H} \ln \left[ (1-\vartheta) \phi_{1}^{\dagger} + \vartheta \phi_{2}^{\dagger} \right]$$
(24)  
(where  $\phi_{1}^{\dagger}$  and  $\phi_{2}^{\dagger}$  are given by eqn.(23)).

This equation is plotted in FIG. 3 as curve B. It can be seen that for Pe > 40 the two models predict almost the same values of Sh but for Pe < 40 the longitudinal diffusion model no longer shows a constant value of Sh but tends to zero. The experimental range covered in this work was from  $10 < Pe < 10^3$ . In this range, however, the experimental results still lie much below the values predicted by both these models.

In order to see if turbulent diffusivity was responsible for these low experimental values the equation (23) was modified to include this effect /15/.

Vol. 7, No. 4

 $Pe_{eff} = Pe_{i} (\delta/\delta_{eff})$ (25)

where 
$$\delta_{eff}/\delta = \delta_{so}/\delta + Pe/2$$

Substitution of Pe<sub>eff</sub> from equation (25) for Pe<sub>i</sub> in equation (23) and combining equations (23) and (24) gives the equation for the combined effects of non-uniform gas distribution, longitudinal diffusion and turbulent diffusivity. Curve C in FIG. 3 is a plot of this. It can be seen that turbulent diffusivity lowers the Sh by a maximum of 50 per cent in the range 1<Pe<10<sup>3</sup>. The experimental values, however, were found to be still far below these values. Another feature which can be observed from FIG. 3 is the effect of  $H/d_{\rm o}$  on these three curves (A, B and C). The  $H/d_{O}$  effects the Sh predicted by these models only to a small degree. The experimental results, on the contrary, show that increase of  $H/d_{a}$  decreases the Sh to a large extent.

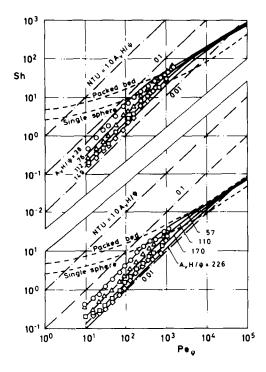
Martin /2/ has shown that in cases where the bypass crosssection is ineffective or has very little active surface area,  $A_{v2}$  (in Eq. 21) can be taken as zero giving a value of NTU<sub>2</sub> equal to zero.With this assumption the equation for the bypass model (Eq. 19) reduces to

$$Sh = -\frac{Pe_{\psi} \cdot \psi}{A_{\psi} \cdot H} \cdot ln \left[ 1 - (1 - \psi) (1 - e^{-NTU} 1) \right]$$
(26)

Curves D in FIG. 3 are for this equation and it can be seen that the spread of the curves for changing  $H/d_{o}$  ratio is quite large and corresponds to that observed experimentally.

## Experimental Results

FIGS. 4 and 5 show the experimental data for the four sizes of Raschig rings. The continuous lines represent the simplified model (Equation 26). There is a surprisingly good agreement with the experimental results indicating the existance of a bypass zone, which is responsible for the non-uniform gas distribution, but, presumably not contributing to the mass transfer process.



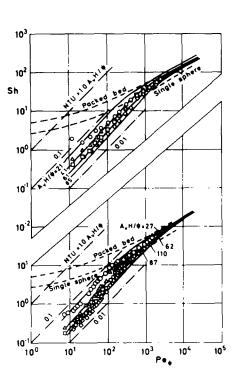


FIG. 4 Experimental results. For symbols see TABLE 2

FIG. 5 Experimental results. For symbols see TABLE 2

Index for FIG. 4, Lower Part					
Symbol	d (mm)	φ	Δψ	н/d <sub>о</sub>	A <sub>↓</sub> H/ψ
ο	6.78	0.04	0.10	7.4	57
		п	п	15.8	110
		n	11	22.1	170
◇ "		n	u	30.5	226
Index for FIG. 4, Upper Part					
0	8.65	0.04	0.10	5.4	38
	n	11	**	10.8	76
	11	"	17	16.5	112
$\diamond$	11	u	11	22.3	149

TABLE 2

Index for FIG. 5, Lower Part					
0	10.4	0.04	0.10	5.3	27
Δ	n	н		10.6	62
	11	n	"	14.7	87
$\diamond$	n	n	<b>11</b>	18.3	110
Index for FIG. 5, Upper Part					
0	16.2	0.08	0.10	2.9	21
	н	n	n	5.7	41
	H	u	"	9.3	61
$\diamond$	"	11	n	11.7	85

## S.P. Rao and V. Gnielinski

From FIGS. 4 and 5 one can see that the apparent Sherwood numbers become inversely proportional to the bed height (at least for bed heights greater than 50 mm) at lower Peclet numbers. This implies that NTU becomes independent of the bed height, attaining a constant finite value so that full equilibrium would not have been reached even if the bed height would have been increased unlimitedly. This can also be seen from the simplified equation (26) which gives the limiting value of NTU, which is directly related to the relative bypass stream:

One could overcome this limitation in performance of the bed if a radial mixing of main flow and bypass flow could be brought about. Assuming complete radial mixing after a certain bed height and letting this mixed outlet gas flow through a second bed should give twice the NTU of a single stage. So with the same amount of packing material a considerably higher performance can be expected if the packing is subdivided into several stages with radial mixing after each stage. Further experimental work is needed to investigate this assumption.

### Conclusions

In the range of intermediate and low Peclet numbers the experimental values of the Sherwood number lie a few decades below those predicted from single particle correlations (assuming a homogeneous system) showing that a heterogeneous system like a packed bed cannot be treated as a quasi-homogeneous system, particularly in this region. The model of non-uniform gas distribution in a packed bed of spheres proposed by Schlünder /1/ and Martin /2/ is found to explain this behaviour for Raschig rings also, when it is assumed that the packing surface in the bypassed zone is inactive.

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## Notation

A <sub>v</sub>	$m^2/m^3$	Specific surface area of the packed bed
A	m <sup>2</sup>	Surface area of the packed bed
c pD	J/kmol K	Molar heat capacity
D	m	Diameter of the packed bed
$\tilde{\operatorname{dh}}_v$	J/kmol	Molar phase change enthalpy
d <sub>o</sub>	m	Outside diameter of the Raschig rings
d <sub>i</sub>	m	Inside diameter of the Raschig rings
<sup>d</sup> r	m	Diameter of the Raschig rings defined by Eqn.(4)
Е	-	Function defined by Eqn. (5)
f	m <sup>2</sup>	Cross-sectional area of the packed bed
fψ	-	Factor in Eqn. (1)
h	m	Height of Raschig rings
Н	m	Height of packed bed
u	m/sec	Superficial velocity of air
v	m <sup>3</sup> /hr	Volumetric flowrate of air
ĩ	-	Molefraction of water vapour in air at inlet
۶ <sub>0</sub>	-	Molefraction of water vapour in air at outlet
ĩs	-	Adiabatic saturation concentration of water vapour in air (molefraction)

Greek Symbols					
ß	m/hr	Mass transfer coefficient			
δ	m <sup>2</sup> /hr	Diffusivity of the air			
<sup>δ</sup> eff	u	Effective diffusivity of the packed bed			
<sup>δ</sup> so	11	Diffusivity of the static packing (zero flowrate)			
η	kg/m sec	Dynamic viscosity of the air			
ν	m <sup>2</sup> /hr	Kinematic viscosity of the air			
ρ	kg/m <sup>3</sup>	Density of the air			
ξ	-	Frictional resistance in the packed bed			
φ	-	Fractional (by-pass) cross-sectional area of the packed bed			
ψ	-	Porosity of the packed bed			
Δp	kp/m <sup>2</sup>	Pressure drop across the packed bed			
v		Relative by-pass stream $(\dot{v}_2^{\prime}/\dot{v})$			
ø	-	Relative change in vapour concentration			
ø'	-	Defined by Eqn.(23)			
Dimensionless Numbers					
Sh		Sherwood number ( $\beta d_0/\delta$ )			
Pe		Peclet number based on empty tower cross-			
		section (u $d_0/\delta$ )			
<sup>Pe</sup> ψ		Peclet number in the packed bed (u d $_{ m O}/\delta$ $\psi$ )			
Sc		Schmidt number $(v / \delta)$			
Subscripts					

i	1 or 2
1	Main cross-section with porosity $\psi_1$
2	By-passed cross-section with porosity $\psi_2$

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