

Heat and Mass Transfer at Liquid Gas Interphase

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ABSTRACT

Interphase heat transfer in gas/liquid systems plays an important role in chemical engineering and in power supply. A short introduction into the interphase transport phenomena is given for quantitative predictions of heat and mass transfer coefficients. Correlations and experimental data, known from the literature, are reported.

The discussion of the flow phenomena is concentrated to the fluid dynamic mechanisms, questions of thermodynamic equilibrium and also the influence of chemical reactions are not taken in account.

NOMENCLATURE

A	test section area	m ²
b	width	m
B	width in special type by /22/	m
d	diameter	m
D	diffusion coefficient	m ² /s
Eu	Euler-number	
g	acceleration due to gravity	m/s ²
Ga	Galilei-number	
n	exponent in equation (6)	
\dot{m}	mass flux	kg/m ² s
n	exponent in equation (4)	
Nu	Nusselt-number	
o	exponent in equation (6)	
Δp	pressure difference	N/m ²
Pr	Prandtl-number	
Re	Reynolds-number	
S	area rearranging factor def. by /4/	

Sc	Schmidt-number	
Sh	Sherwood-number	
t	time	s
V	volume	m ³
\dot{V}	volumetric flow rate	m ³ /s
w	velocity	m/s
We	Weber-number	
x	length	m
\dot{x}	quality	
Y	phase boundary distance	m
β	mass transfer coefficient	m/s
δ	boundary layer thickness	m
ϵ	turbulent diffusion coefficient	m ² /s
ν	kinematic viscosity	m ² /s
ρ	density	kg/m ³
τ	surface tension	kg/s

Subscripts

g	gas
l	liquid
p	particle or aerosol
d	mean liquid particle
o	without particle

INTRODUCTION

Heat and mass transfer in two or multiphase systems is a function of the interfacial area, of the fluid dynamic behaviour in each phase near the phase boundary, of the equilibrium conditions between the phases and in some cases also of interface active effects.

Generally, one can distinguish two flow conditions in two-phase systems, namely

- dispersed particles (bubbles or droplets) in a continuous phase or
- two continuous phases (falling film-flow).

The flow conditions - dispersed or continuous - are a function of the forces acting on the phases, like pressure drop, interfacial shear stress, buoyancy and surface tension.

Technical apparatus for heat and mass transfer in two-phase gas liquid systems, depending from their design features, work in a wide field of possible flow conditions whereby the interfacial area, its rearrangement and the turbulence near the phase boundaries are mostly artificially promoted. Theoretical deliberations and fundamental experiments vary often consider a single particle in an infinite fluid and then for the practice application, the difficulty arises how to take in account the influence of neighbouring particles and the interaction phenomena. Therefore, for layout of heat and mass exchanging apparatus very often pure empirical correlations are used. Especially in complicated fluid dynamic systems where the phases and the interfacial area are continuously renewed, mostly only phenomenological descriptions of the heat and mass transfer process are available.

FUNDAMENTAL ASPECTS

There are several models in the literature to describe the mechanism of heat and mass transfer between the phases. A detailed discussion of these models is given in the papers by Sideman and Pinczewski /1/ as well as by Johannisbauer /2/. Generally the models can be divided in three groups:

1. Two film theory
2. Penetration hypothesis
3. Turbulent diffusivity assumptions

The two film model assumes steady-state diffusion in the boundary layers of each phase near the interface. Assuming a boundary layer thickness δ the mass transfer coefficient β in each phase can be correlated with the diffusion coefficient D by the simple equation $\beta = D/\delta$. For most practical cases this model gives far too low mass transfer coefficients.

Higbie /3/ proposed the penetration model, according to which fluid parts having their origin from the turbulent area of the flow are penetrating the thin laminar layer at the phase boundary. During their rest at the interface there is unsteady mass transfer due to molecular diffusion with the other phase. Dankwerts /4/ developed from the penetration model a surface rearrangement model abandoning the idea that all fluid elements rest the same period at the phase boundary but giving an distribution function for the resting time. With Higbie's assumption of constant resting time t the mass transfer coefficient reads

$$\beta = 2 \cdot \sqrt{\frac{D}{t}} \quad (1)$$

and using Dankwerts theory with the area rearranging factor S which is the ratio of the produced new area per time unit and the total interfacial area one gets

$$\beta = \sqrt{D_L \cdot S} \quad (2)$$

The two film model and Higbie's penetration model were combined by Toor and Marchello /5/ in the so-called film penetration model.

The up to now mentioned models start from the idea, that the fluid elements behave like stiff bodies and that the heat and mass transfer occurs through rigid phase boundaries which does not correspond to the physical reality. Therefore, numerous new models were developed describing the turbulence in the neighbourhood of the phase boundary. The models of Lamont /6/, Fortescue /7/, Ruckenstein /8/ or Barnejee /9/ regard an inner motion of the fluid elements near the phase boundary and take in account a velocity distribution in the flow. Whilst these models are considering the micro-structure of the turbulence near the phase boundary there are also other models, working with the eddy-diffusivity and describing the macro-structure of the turbulence. They mainly deal with turbulent falling film flow and define turbulent diffusion coefficients for momentum-, mass-, and heat flux. The correlations for these coefficients are coupled according to Boussinesq /10/ via the mean transport velocity. The method based on the eddy-diffusivity is well-known in single phase boundary layer theory. For mass transfer at the phase boundary the turbulent diffusion coefficient ϵ_m usually is assumed to be proportional to the square of the distance from the phase boundary.

$$\epsilon_m \sim y^2 \quad (3)$$

Lamourelle and Sandall /11/ basing on the correlation of King /12/

$$\epsilon_m = by^n \quad (4)$$

found for falling film flow

$$b \sim Re^{1,769} \quad n=2 \quad (5)$$

which is only valid for large values of $Re \cdot Sc$. For wavy falling film flow Javdani /13/ got an expression for ϵ_m by solving the momentum equation for wavy film flow. Limberg /14/ used a correlation for the turbulent momentum exchange coefficient from the literature and by assuming analogy between momentum-heat- and mass transfer he calculated the mass transfer via the velocity profile. Spalding's /15/ theory for the turbulent momentum exchange coefficient is the basis for Iribane's u.a. /16/ correlation of the mass transfer through a solid phase boundary for short inlet length.

All these theories are primarily only valid for regular wave motion in a falling film flow at large values of $Re \cdot Sc$. For irregular wave motion ($12 \leq Re \leq 400$) there is no theoretical solution for the heat and mass transfer which gives satisfactory agreement with measured data. Also in turbulent falling film flow, the prediction with theory is not too good. Recently Carrubba /17/ found that the velocity profile has no influence on the mass transfer and that in agreement with the usual assumption in the literature, the total mass flow density in the immediate neighbour-

hood of the solid phase boundary is approximately constant and equal to that at the gas liquid phase boundary in wavy falling film flow.

In an empirical way, the mass transfer often is correlated by

$$Sh = C \cdot Re^m \cdot Sc^n \cdot Ga^o \quad (6)$$

where the dimensionless numbers are defined as follows:

$$Sh = \frac{\beta \cdot \delta}{D} \quad (7)$$

$$Re = \frac{W \cdot \delta}{\nu} \quad (8)$$

$$Sc = \frac{\nu}{D} \quad (9)$$

$$Ga = x^3 g / \nu^2 \quad (10)$$

Equation (6) is mainly valid for separated flow like falling film flow. In the Galilei-number Ga , the distance from the flow inlet is denoted with x .

Surface active solutes like tensides can affect the stability of the phase boundary. Recently Palmer /18/ demonstrated, that these solutes have a profound stabilizing influence on convection induced by differential vapor recoil but no noticeable effect on the criteria for instability via the fluid inertia mechanism. Extensive work into the research of interfacial resistance was performed by Nitsch /19/, however, with liquid-liquid two-phase systems. Gradients in the surface tension due to concentration- or temperature differences can also produce a micro-convection near the phase boundary via the Marangoni-effect as shown by Beer /47/ with bubble boiling.

DISPERSED DROPLET SYSTEMS

The transport phenomena from droplets to a gas flow or verse vica are mainly of interest for spray dryers and spray evaporators, but also for absorption apparatus used for example to purify air or other gases. With small freely falling spray droplets the flow conditions around each droplet can be assumed purely laminar which results in a constant value of the Nusselt- or Sherwood-number ($Nu = 2.0$). The main transport resistance is on the gaseous side and the conduction or diffusion distances in the droplet are small. Mullin and Treleaven /20/ considered the effect of neighbouring particles on the mass transfer rate. Recently this effect of drop to drop interaction on the rate of drop- to fluid heat and mass transfer was studied by Miura u.a. /21/ by measuring

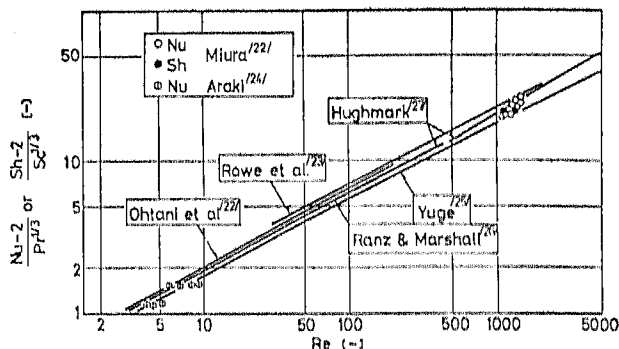


Fig. 1 Relationship between Re and $(Nu-2)/Pr^{1/3}$ or $(Sh-2)/Sc^{1/3}$ /21/

the transfer rate from drops. In their measurements for single drops they found good agreement with older experimental and theoretical data /22-27/ as shown in fig. 1. From this figure it also can be seen, that the Nusselt- or the Sherwood-number respectively can be expressed as a function of the Reynolds- and the Prandtl- or Schmidt-number. In continuing their research work the influence of neighbouring droplets was imitated by Miura using glass beads.

The influence onto the heat transfer depends from the glass bead diameter with respect to the droplet diameter and from the distance between glass bead and droplet. In fig. 2 the reduction of

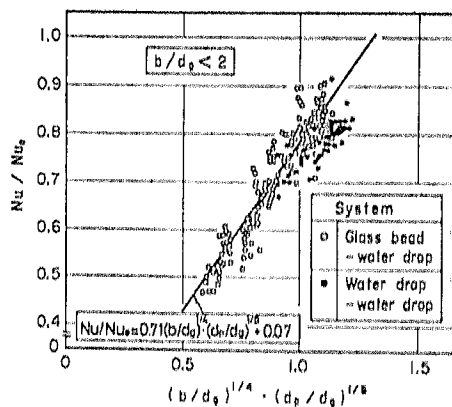


Fig. 2 Relationship between Nu/Nu_0 and $(b/d_0)^{1/4} \cdot (dp/d_0)^{1/6}$ /21/

the heat transfer behaviour due to the presence of other particles - glass beads or water droplets - is shown as a function of the mentioned geometrical conditions. In this figure Nu_0 is the Nusselt-number around a single droplet without neighbouring particles.

Miura u.a. /21/ gave the following correlations for neighbouring effects onto the heat or mass transfer from droplets.

$$b/d_0 \leq 2 \quad Nu/Nu_0 \text{ or } Sh/Sh_0 = 0.71(b/d_0)^{1/4}(dp/d_0)^{1/6} + 0.07 \quad (11)$$

$$b/d_0 > 2 \quad Nu/Nu_0 \text{ or } Sh/Sh_0 = 0.42(b/d_0)^{1/6} \cdot 0.4 \quad (12)$$

$$Nu/Nu_0 \text{ or } Sh/Sh_0 \approx 1.0 \quad (13)$$

$$db/B \leq 2 \quad Nu/Nu_0 \text{ or } Sh/Sh_0 \approx 1.0$$

$$db/B > 4 \quad Nu/Nu_0 \text{ or } Sh/Sh_0 \approx 0.57$$

With larger droplets also the heat and mass transport inside the droplet has to be taken in account. This transport is agitated by circulation effects as known since many years /28/. These circulations are due to the shear stress resulting from the gas velocity around the droplet.

At very high gas velocity, when the liquid dispersion is not performed by high pressure spray nozzles, but by adding the liquid in a continuous jet which is fragmented by the shear stress of the gas, filigrane lamina like particles may be formed. This is for example the case in venturi scrubbers which are used for absorption of aerosols and cleaning air from micro particles. Neumann /29/ studied the liquid fragmentation in venturi-nozzles and found extremely high interfacial areas.

Figure 3, which was performed with the technique of high-speed cinematography shows an example of such a liquid membran formed in a venturi-nozzle. The air-flow fragmentating the liquid had a velocity of 80 m/s. In Fig. 3 the liquid membran was photographed twice with a time difference between both exposures of 10⁻⁶ s. From the comparison of both exposures one can see that the membran expands due to the flow forces of the gas. Downward from the venturi-nozzle, the velocity difference between the liquid and the gas decreases, which results in a reduction of the gas forces and so a few inches after the membran formation started out of the liquid jet, the surface tension can prevail again and reduces the membran into a droplet form. Sometimes, the flow forces are high enough to disrupt the membran as shown in Fig. 4 which results in an formation of numerous very small droplets.

These mechanisms guarantee not only a high area for heat and mass transfer between the phases but they also care for a violent rearrangement of the phase boundaries. The interfacial area is a strong function of the flow history as shown in fig. 5 and reaches

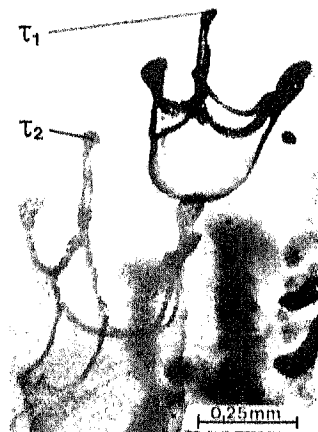


Fig. 3 Liquid membran in venturi-nozzles formed as parachute /29/



Fig. 4 Disruption of parachutes /29/

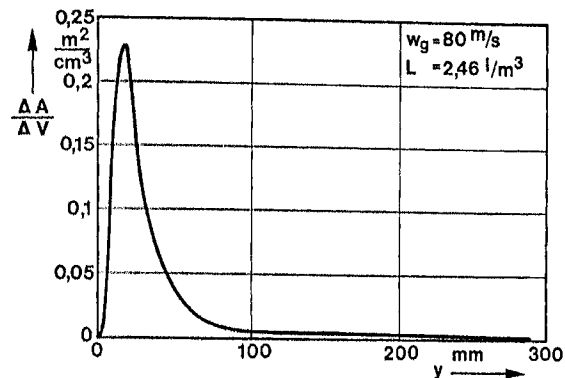


Fig. 5 Interfacial area as a function of flow route /29/

a maximum a few inches behind the point of water injection in the venturi-nozzles. From there it decreases rapidly again due to the decelerating of the gas flow in the expanding part of the nozzle and the decreasing velocity difference between liquid and gas. A theoretical description of this mechanism to predict the mass transfer and the separation efficiency of a venturi-scrubber seems to be without prospects and therefore Neumann described the product of mass transfer coefficient and superficial interfacial area Λ by an empirical correlation.

$$\frac{\beta \cdot \Lambda}{\dot{V}_g} = 94,9 \cdot Re^{0,1} \cdot Eu^{-0,2} \cdot We^{0,24} \cdot \frac{1-\tilde{\chi}}{\tilde{\chi}} \cdot \left(\frac{w_g}{w_s}\right)^{0,2} \quad (14)$$

with

$$\begin{aligned} w_g &= \text{gas velocity in venturi nozzle} \\ We &= \frac{d_p \cdot \rho_g \cdot w_g}{\sigma_l} & d_p &= \text{mean particle or aerosol diameter} \\ Re &= \frac{w_g \cdot d_g}{\nu_g} & \dot{V}_g &= \text{volumetric gas flow} \\ Eu &= \frac{\Delta p}{\rho_g \cdot w_g^2} & d_d &= \text{mean liquid particle diameter} \\ w_s &= \frac{d_p^2 \cdot \rho_p \cdot g}{18 \nu_g \cdot \rho_g} \\ \tilde{\chi} &= \frac{\dot{m}_g}{\dot{m}_g + \dot{m}_l} \end{aligned}$$

BUBBLE SYSTEMS

The momentum and mass transfer through the phase boundary of a bubble with and without changing the shape was discussed in detail by Glaeser and Brauer /30/. Only small bubbles have purely spherical form during rising in a liquid. Surface active materials like tencides can stabilize the spherical form of a bubble. With increasing diameter the bubble tends to oscillate between a spherical and an ellipsoidal form which results in a circulating flow inside the bubble. Still increasing the diameter the bubbles finally deviate completely from the spherical form, they have the shapes of mushrooms, umbrellas or flat discs.

An attempt to give an theoretical explanation for the effect of tencides onto the bubble behaviour was done by Lovich /31/. According to his theory, surface active molecules were transported to the lee-side of the bubble resulting in a concentration gradient of the tencides over the phase boundary. This produces a gradient in the surface tension, which acts against the fluid motion in the neighbourhood of the phase boundary.

The mass transfer through the interface of a spherical stable bubble can be correlated by the equation /12/.

$$Sh = 2 + \frac{0.651 (ReSc)^{1/2}}{1 + (ReSc)^{1/4}} \quad (15)$$

A more detailed description of the mass transfer from or into spherical bubbles needs the numerical solution /11/ of the mass transfer-differential equation. This then gives also information about the local distribution of the Sherwood-number around the bubble and one finds, that on the front side of the bubble the Sherwood-numbers are much higher than on the backside.

When the bubble changes its form the mass transfer coefficient is highly dependent from the local conditions around the bubble. A detailed theoretical analysis by Glaeser and Brauer /30/ showed a tremendous variation in the Sherwood-number via the perimeter of the bubble as pointed out in Fig. 6. Glaeser and Brauer regarded also the influence of the turbulent diffusivity and in Fig. 6 results with and without this correction factor are presented. The differences in the local Sherwood-number around the bubble become greater with increasing Reynolds-number as demonstrated in Fig. 7.

Interesting informations for a better understanding of the heat transfer from condensing vapour bubbles to a subcooled liquid can be drawn from holographic observations performed by Nordmann /34/. The inclination of the interference fringes in Fig. 8 can be regarded as temperature gradients in a first rough approximation. For a quantitative evaluation due to the three-dimensional almost spherical form of the bubbles the so-called Abel-correction has to be made with which one gets the temperature distribution in the immediate neighbourhood of the phase boundary as also demonstrated in Fig. 8. Assuming a laminar sublayer on the liquid side of the interface the heat transfer coefficient immediately can be deduced from the temperature profile.

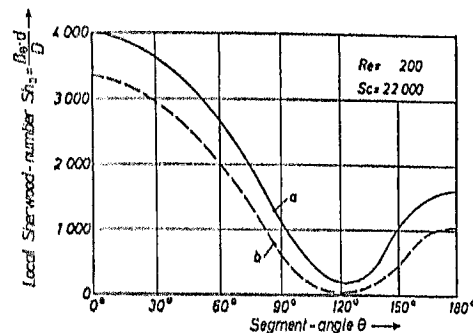


Fig. 6 Local Sherwood-number vs. Segment-angle θ with (a) and without (b) turbulent mass transfer /30/

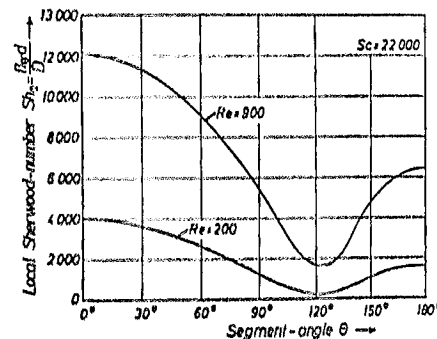


Fig. 7 Local Sherwood-number vs. Segment-angle θ Influence of Reynolds-number /30/

A condensing bubble has a fast moving phase boundary due to its volumetric reduction. This results in a high turbulence in the surrounding liquid. Fig. 9 allows to compare the interfacial conditions at slow condensation rates with low subcooling of the liquid and at high condensation velocities due to large heat transfer rates and large subcooling with violent interface movement. A comprehensive theoretical description of the heat and mass transfer from and to bubbles with unsteady phase boundaries, as it is the case during recondensation, is not yet available in the literature.

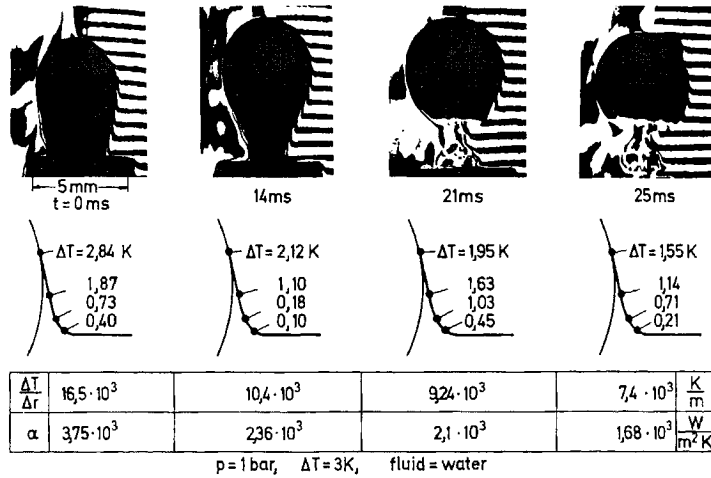


Fig. 8 Temperature gradients during bubble growth and condensation /34/

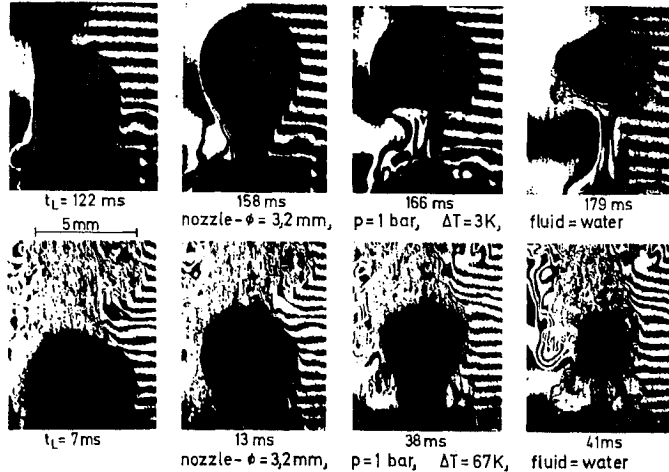


Fig. 9 Growth and condensation of bubbles produced with nozzles Influence of subcooling. /34/

LIQUID FILMS

There is a tremendous amount of papers in the literature dealing with fluid dynamics, heat and mass transfer in falling film systems. In most practical applications the falling liquid film is thick enough to form waves and as shown in Fig. 10 /35/ these waves produce vortices which finally results even in a reversed flow in the valleys of these waves. For a detailed fluid dynamic description of a wavy film flow and its heat and mass transfer therefore the wave length, the phase velocities and the amplitude of the waves have to be known, which usually are expressed as a function of the Weber-number. Furthermore, the stability of the falling film has to be carefully checked which may be influenced by the Marangoni-effect and by interaction with the surface of the solid wall. The stability of evaporating falling films for example was researched by Coulon /36/ and Dammann /37/. On both sides of the phase boundary vortices can be formed which influence the heat and mass transfer, too.

The mass transfer through the moving interface of a falling liquid film was measured by a great number of authors. In Fig. 11 /17/ the experimental results of Kamei and Oishi /38/, Lamourelle and Sandall /39/, Hikita /40/, Emmert and Pigford /41/ and Malewski /42/ are compared and the equations (16), (17), (18) represent these data.

$$Sh_{\infty} = 2,24 \cdot 10^{-2} Re^{0,8} Sc^{0,5} \quad (16)$$

für $12 \leq Re \leq 70$ und $Sc \geq \frac{2,32 \cdot 10^4}{Re^{1,6}}$

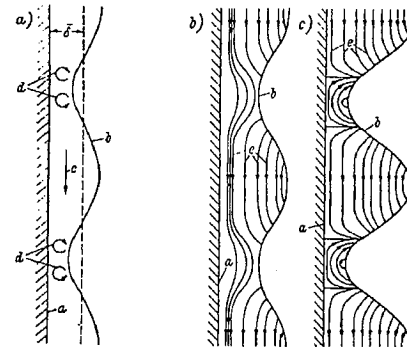


Fig. 10 Characteristics of wavy films /35/

$$Sh_{\infty} = 8,0 \cdot 10^{-2} \cdot Re^{0,5} \cdot Sc^{0,5}$$

für $70 \leq Re \leq 400$ und $Sc \leq \frac{1,82 \cdot 10^3}{Re}$ (17)

$$Sh_{\infty} = 8,9 \cdot 10^{-4} \cdot Re^{1,25} \cdot Sc^{0,5}$$

für $400 \leq Re$ und $Sc \leq \frac{1,47 \cdot 10^7}{Re^{2,5}}$ (18)

Carrubba /17/ developed a theory for predicting heat and mass transfer coefficient in wavy film flow, when the transport resistance is on the liquid side. He used the eddy-diffusivity model and added in the diffusion equation a turbulent diffusivity coefficient m to the molecular diffusion coefficient. An example of the variation of the reduced turbulent coefficient with respect to the distance from the interphase is shown in Fig. 12. As well-known from single phase boundary layer theory the eddy-diffusivity is a strong function of the Reynolds-number.

Recently the interfacial heat and mass transfer under the turbulent motion of fluids was also researched by Kolar /43/. He mainly regards the structure of the turbulence near the interface and comes to a general expression for the mass transfer based on the local degree of turbulence and the thickness of the laminar layer.

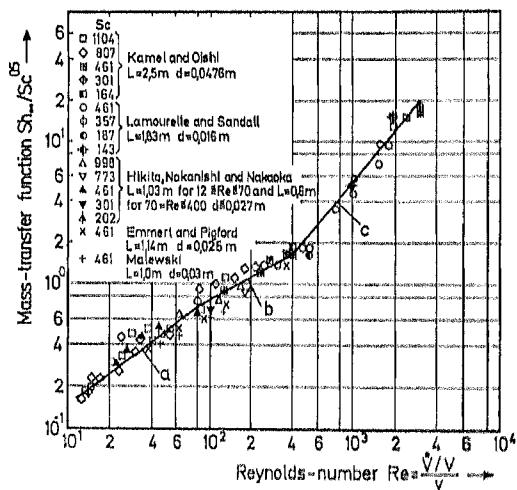


Fig. 11 Mass transfer-function $Sh_{\infty}/Sc^{0,5}$ vs. Reynolds-number /17/ Measurements recalculated by /17/ with equation (16) for a, (17) for b, (18) for c

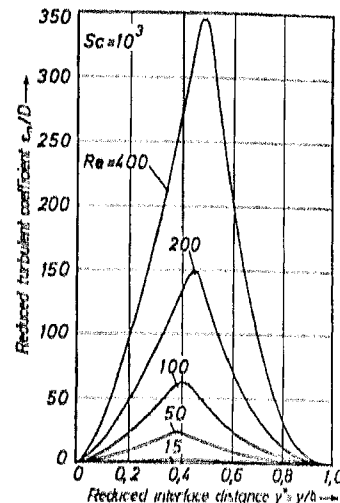


Fig. 12 Reduced turbulence mass-transfer coefficient ϵ_m/D vs. reduced coordinate $\eta' = \eta/\delta$ /17/

TRANSPORT BEHAVIOUR IN PRACTICAL AND INDUSTRIAL EQUIPMENTS

Liquid/gas interphase heat transfer phenomena occur in a wide variety of apparatus in the chemical industry as for example in bubble column, packed and fluidized beds, spray columns and jet-mixers. For their optimal layout, a reliable prediction of the heat and mass transfer exchanging area and of the transfer coefficients is needed. Systematic criteria for selecting gas/liquid contact apparatus with respect to their interfacial area were elaborated by Nagel u.a. /44/. He correlates the interfacial area A per unit of volume V with the dissipated energy E which is due to the pressure drop in the apparatus. The interfacial area is a function of the gas flow rate \hat{V} as shown in Fig. 13 for a pebble bed reactor with co-current gas/liquid flow.

This unique relationship between the interfacial area and the energy dissipation, both referred to the unit of volume, is valid for such different kinds of heat and mass transfer apparatus like jet nozzles, bubble columns, packed beds, venturi-scrubbers and stirred mixers. From Fig. 14 /45/ the demand for a special apparatus design depending from the desired heat and mass transfer area can be deduced. As expected high interfacial areas and with this also high mass transfer is linked with large energy consumption by the circulation pump or the blower. Interesting is the comparison between the venturi-scrubber and the tubular reac-

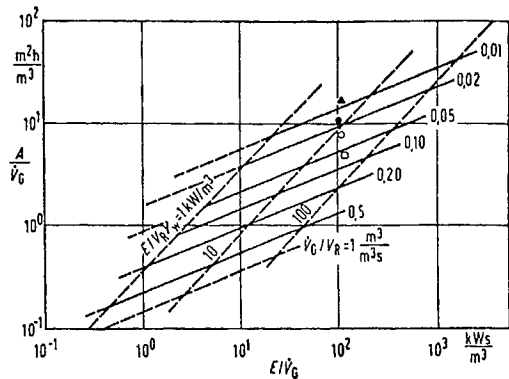


Fig. 13 Reduced interfacial area as a function of the reduced gas-flow rate /44/

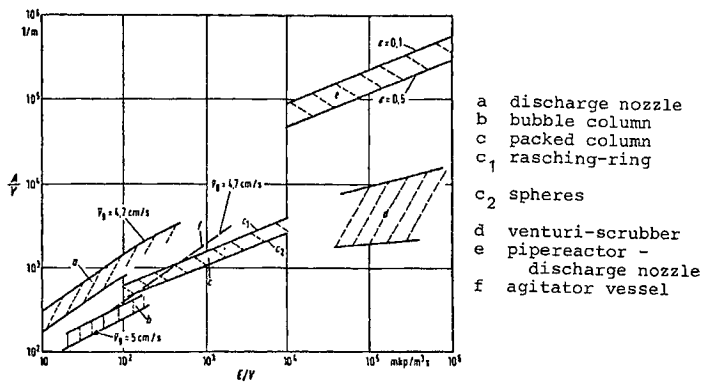


Fig. 14 Comparison of gas/liquid-reactors /45/

tor/jet nozzle for which Fig. 14 shows, that the venturi-scrubber - needing high energy - is not always an optimal design for mass transfer processes. Here, however, one has to distinguish carefully whether there is mass transfer resistance on both phases, as it is the case in the example of Fig. 14 or whether the mass transfer is only controlled by the fluid dynamic behaviour on the

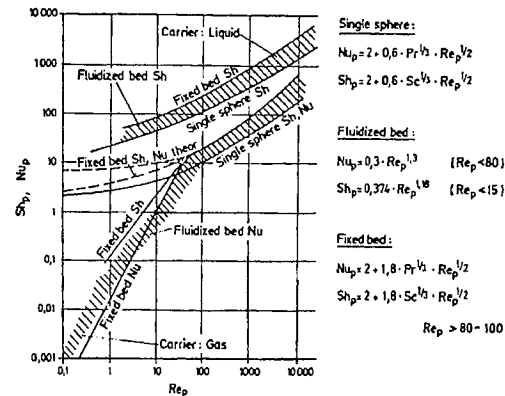


Fig. 15 Deviation of fluid to particle heat and mass transfer in fixed and fluidized beds from single sphere data /46/

gaseous phase as for example with the separation of submicron particles or aerosols from gas flow. Under the later mentioned conditions the high turbulence and the large shear stress in the venturi-scrubber has quite a benifite to the separation effect.

A critical review of the trends in research and industrial application of fluidization including general remarks onto the interfacial mass transfer in these apparatus was recently presented by Reh /46/. In his comparison of heat and mass transfer behaviour he unfortunately does not consider gas liquid systems, but only liquid/solid and gas/solid systems. From his comparison he states - as shown in Fig. 15 - a remarkable drop of fixed bed and fluidized bed transfer data for gas/solid systems at low Reynolds-numbers compared with those of single sphere transfer data.

CONCLUDING REMARKS

There is such a large number of theoretical and experimental papers dealingwith fundamental questions of heat and mass transfer at gas/liquid interfaces, that it is completely impossible to discuss even a small fraction of them in a representative way within a short review paper. However, facing the task of making a layout for an industrial plant operating in two-phase conditions, one realize that there is still a great lack of experimental data and theoretical correlations which are valid under the complicated conditions of industrial plants. Therefore, large scale experiments based on the experience of fundamental research are needed to get a better prediction for two-phase flow interfacial transfer behaviour.

This, however, does not mean, that there is no further need for fundamental research. We still do not have a sufficient understanding of the interfacial phenomena even in simple phase geometries for developing theoretical models and correlations of general validity. Optical methods like the interferometry or the laser-doppler-anemometry should help to give more detailed information.

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Interface Conditions in Boiling of Saturated Binary Mixtures

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ABSTRACT

One of the methods of increasing the heat transfer coefficient in boiling is to add trace amount of a second component to a pure system. Many of the mixtures give heat fluxes much higher than those for either pure component at the same temperature difference. This marked effect of the addition of a small amount of a second component to a pure system suggests that the mass diffusion plays an important role in the heat transfer process. Apparently, the diffusion problem will have to be coupled with the present theory to predict satisfactorily the film boiling behaviour of liquid mixtures.

Stable film boiling of a saturated binary mixture is analysed by the two-phase boundary layer theory for the case of a binary mixture flow on a heated horizontal plate. Both diffusion and heat conduction in the liquid phase are taken into account. The conditions at the interface are investigated. The analysis shows that increases in film boiling heat flux may be obtained for mixtures of liquids. The increased heat transfer is shown to occur at relatively small values of wall superheat. The interface concentration of more volatile component reduces as Schmidt number or volatility of the additive increase.

NOMENCLATURE

B	dimensionless quantity, equation (47)
Q_p	specific heat at constant pressure, J/Kg.K
δ	diffusivity, m^2/sec .
F_1, f	dimensionless velocity functions
g	acceleration due to gravity, m/sec^2
Ah_v	latent heat of vaporization, J/kg