Int. J. Heat Mass Transfer. Vol. 33, No. 9, pp. 2039–2044, 1990
Printed in Great Britain

0017–9310/90 $3.00 + 0.00
Pergamon Press plc

Influence of the interphase mass transfer on the rate of mass transfer—1. The system 'solid–fluid (gas)'

CHRISTO B. BOYADJIEV
Institute of Chemical Engineering, The Bulgarian Academy of Sciences, 'Akad. G. Bonchev' Str., Block 103, 1113 Sofia, Bulgaria

and

NIKOLAI L. VULCHANOV
Department of Mathematics, School for Mechanical and Electrical Engineering, 4 'H. Dimitar' Str., 5300 Gabrovo, Bulgaria

(*Received* 2 *February* 1988)

Abstract—A theoretical analysis of the influence of the direction of the interphase mass transfer on the rate of mass transfer is developed. The case of mass transfer between a solid plane surface and a fluid or gas flow in a boundary layer approximation is studied when the transport of mass and momentum at the phase boundary are coupled to account for non-linear effects in a direction normal to the main flow. Numerical results are reported and discussed.

1. INTRODUCTION

RECENTLY it was shown that according to the linear theory of mass transfer, the rate of mass transfer does not depend on the direction of the interphase transfer [1, 2].

The theoretical analysis of systems with intensive mass transfer [3–5] indicates that the large mass fluxes can initiate secondary flows colinear with the direction of mass transfer. From this it follows that a change in the direction of the interphase transfer could induce a change in the hydrodynamic flow and the rate of mass transfer.

The purpose of this study is to report a theoretical analysis of the influence of the direction of the interphase transfer on the rate of mass transfer between a smooth solid surface and a fluid or gas flowing past it. Numerical results for the rate of mass transfer in the approximation of a diffusive boundary layer [1, 6] will be presented and discussed.

2. THE MATHEMATICAL MODEL

The mass flux, J, across a plane surface of length L is defined [1] by the average value of the local mass flux I

$$J = Mk(c^* - c_0) = \frac{1}{L}\int_0^L I \, \mathrm{d}x \tag{1}$$

where the local diffusive mass flux, I, across the phase boundary, $y = 0$, according to the non-linear theory of mass transfer, has both a diffusive and a convective component

$$I = -MD\frac{\partial c}{\partial y} + Mc^* v_n = -\frac{MD\rho^*}{\rho_0^*}\frac{\partial c}{\partial y}, \quad y = 0. \tag{2}$$

In equation (2) v_n is the normal component of the velocity at the phase boundary, $y = 0$, and it is determined [3–5, 7] by the diffusive component of the mass flux

$$v_n = -\frac{MD}{\rho_0^*}\frac{\partial c}{\partial y}, \quad y = 0. \tag{3}$$

In equation (3) v_n can be considered as similar to the rate of the Stefan flux but v_n does not result from a phase change. It can also be thought of as an analogue to the local velocity of sucking or blowing from or into the laminar boundary layer [8] if the latter is limited by the mass transfer in the layer.

The analysis of equation (2) shows that the local mass flux, and consequently, the rate of mass transfer depend on two factors—the mass concentration and the concentration gradient at the phase boundary. The influence of the mass concentration at the phase boundary in the presence of a reversible process there ($c^* = 0$) is expressed by

$$\frac{\rho^*}{\rho_0^*} = 1 + \frac{Mc^*}{\rho_0^*}. \tag{4}$$

The above effect is of practical significance when

NOMENCLATURE

a_0, a_1, a_2	constants, see equation (15a)
c	concentration of the diffusing species [kg mol m^{-3}]
D	diffusivity of the diffusing species [m^2 s^{-1}]
f	non-dimensional function, see equation (16a)
I	local mass flux [kg (m^2 s)$^{-1}$]
J	average mass flux [kg (m^2 s)$^{-1}$]
k	mass transfer coefficient [m s^{-1}]
L	length of mass transfer surface [m]
M	molecular mass of the diffusing species [kg kg mol^{-1}]
n	outward normal to the surface $y = 0$ [m]
u	velocity in the x-direction [m s^{-1}]
v	velocity in the y-direction [m s^{-1}]
x	downflow coordinate [m]
y	coordinate, normal to the flow [m]
Pe	Peclet number for the diffusing species, see equation (12b)
Sc	Schmidt number for the diffusing species, see equation (9f)
Sh	Sherwood number for the diffusing species, see equations (7) and (12a)

Greek symbols

ε	$Sc^{0.5}$
ζ	non-dimensional coordinate, see equation (16b)
η	non-dimensional coordinate, see equation (9d)
θ	non-dimensional parameter, see equation (13)
μ	dynamic viscosity [kg (m s)$^{-1}$]
ν	kinematic viscosity [m^2 s^{-1}]
ρ	mass density [kg m^{-3}]
Φ	non-dimensional function, see equations (9a) and (9b)
Ψ	non-dimensional function, see equation (9c).

Subscripts

0	initial value
n	colinear with the outward normal.

Superscript

*	equilibrium value.

$$\frac{Mc^*}{\rho_0^*} > 10^{-2}. \tag{5}$$

When the process is irreversible ($c^* = 0$) it is theoretically absent. Obviously, this effect cannot depend on the direction of mass transfer. The concentration gradient at the phase boundary, i

$$i = \frac{\partial c}{\partial y}, \quad y = 0 \tag{6}$$

depends on the hydrodynamics and thus it changes if a change of the direction of mass transfer occurs because from equation (3) it follows that a change of the sign of i results in a change of the sign of v_n.

From equations (1) and (2) it follows directly that

$$Sh = \frac{kL}{D} = -\frac{\rho^*}{\rho_0^*}\frac{1}{c^*-c_0}\int_0^L \frac{\partial c}{\partial y}\,dx, \quad y = 0 \tag{7}$$

where $c(x, y)$ is the corresponding component of the solution of the following non-linear boundary value problem:

$$u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} = \nu\frac{\partial^2 u}{\partial y^2}; \quad x > 0, y > 0 \tag{8a}$$

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0; \quad x > 0, y > 0 \tag{8b}$$

$$u\frac{\partial c}{\partial x} + v\frac{\partial c}{\partial y} = D\frac{\partial^2 c}{\partial y^2}; \quad x > 0, y > 0 \tag{8c}$$

$$u = u_0; \quad x = 0, y > 0 \tag{8d}$$

$$c = c_0; \quad x = 0, y > 0 \tag{8e}$$

$$u = 0; \quad x > 0, y = 0 \tag{8f}$$

$$c = c^*; \quad x > 0, y = 0 \tag{8g}$$

$$v = -\frac{MD}{\rho_0^*}\frac{\partial c}{\partial y}; \quad x > 0, y = 0 \tag{8h}$$

$$u = u_0; \quad x > 0, y = \infty \tag{8i}$$

$$c = c_0; \quad x > 0, y = \infty. \tag{8j}$$

The problem, equation (8), can be cast into a non-dimensional form introducing the following variables and parameters:

$$u = 0.5u_0\varepsilon\Phi' \tag{9a}$$

$$v = 0.5\left(\frac{u_0\nu}{x}\right)^{0.5}(\eta\Phi' - \Phi) \tag{9b}$$

$$c = c_0 + (c^* - c_0)\Psi \tag{9c}$$

$$\eta = y\left(\frac{u_0}{4Dx}\right)^{0.5} \tag{9d}$$

$$\varepsilon = Sc^{0.5} \tag{9e}$$

$$Sc = \nu/D \tag{9f}$$

where η is the new independent variable, $\Phi(\eta)$ and $\Psi(\eta)$ the dependent ones, and Sc the Schmidt number. After inserting equation (9) into equation (8) and some standard manipulations, one is lead to the following non-linear two-point boundary value problem for a system of ordinary differential equations:

$$\Phi''' + \varepsilon^{-1}\Phi\Phi'' = 0; \quad \eta > 0 \tag{10a}$$

$$\Psi'' + \varepsilon\Phi\Psi' = 0; \quad \eta > 0 \tag{10b}$$

$$\Phi(0) = \theta\Psi'(0); \quad \eta = 0 \tag{10c}$$

$$\Phi'(0) = 0; \quad \eta = 0 \tag{10d}$$

$$\Psi(0) = 1; \quad \eta = 0 \tag{10e}$$

$$\Phi'(\infty) = 2\varepsilon^{-1}; \quad \eta = \infty \tag{10f}$$

$$\Psi(\infty) = 0; \quad \eta = \infty \tag{10g}$$

where a prime denotes differentiation in η and

$$\theta = \frac{M(c^* - c_0)}{\rho_0^* \varepsilon}. \tag{11}$$

The Sherwood number, defined by equation (7) can be calculated as

$$Sh = \frac{\rho^*}{\rho_0^*} Pe^{0.5}\,\Psi'(0) \tag{12a}$$

where Pe is the Peclet number, defined by

$$Pe = \frac{u_0 L}{D}. \tag{12b}$$

From equation (12a) it follows that a change of the direction of the interphase mass transfer could influence the rate of mass transfer only through the non-dimensional diffusive flux $\Psi'(0)$. The value of the latter is a function of ε and θ.

When the non-linear effects are practically significant

$$\theta = \frac{M(c^* - c_0)}{\rho_0^* \varepsilon} > 10^{-2} \tag{13}$$

the sign of θ is determined by the direction of mass transfer. Setting $\Delta c = c^* - c_0$, if $\Delta c > 0$ the mass transfer is from the solid surface, while if $\Delta c < 0$ it is towards the surface.

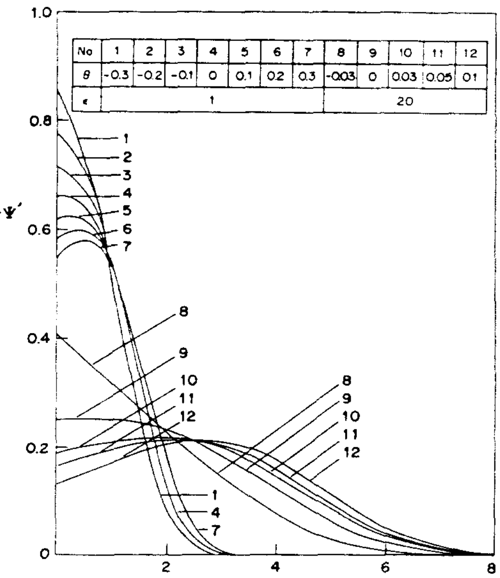

FIG. 1.

3. QUANTITATIVE ANALYSIS

The quantitative theoretical analysis of the influence of the direction of mass transfer on the rate of mass transfer was performed on the basis of numerical results for $\Psi'(0)$ computed from equations (10), utilizing a code for the numerical integration of two-point boundary value problems for systems of ordinary differential equations on semiinfinite intervals [9]. The results are listed in Table 1 and plotted on Figs. 1–3.

The data for $\Psi'(0)$, Table 1 and Fig. 1, show that if ε is increased, the concentration gradient at the phase boundary, the Sherwood number, respectively, is decreased. This is natural as far as it corresponds to a decrease of the diffusivity (an increase of the viscosity).

The velocity and concentration profiles are shown on Figs. 2 and 3, respectively, for gas ($\varepsilon = 1$) and liquid ($\varepsilon = 20$).

Table 1

	$\varepsilon = 0.1$		$\varepsilon = 1.0$		$\varepsilon = 2.0$		$\varepsilon = 10.0$		$\varepsilon = 20.0$	
θ	$\Phi''(0)$	$-\Psi'(0)$	$\Phi''(0)$	$-\Psi'(0)$	$\Phi''(0)$	$-\Psi'(0)$	$\Phi''(0)$	$-\Psi'(0)$	$\Phi''(0)$	$-\Psi'(0)$
0.0	133.0	1.03	1.33	0.664	0.332	0.535	0.0133	0.314	0.00333	0.250
0.03	128.0	1.03	1.30	0.650	0.327	0.515	0.0137	0.270	0.00332	0.190
−0.03	137.0	1.04	1.36	0.679	0.338	0.553	0.0135	0.384	0.00338	0.406
0.05	126.0	1.03	1.28	0.641	0.323	0.503	0.0131	0.248	0.00332	0.166
−0.05	140.0	1.04	1.38	0.689	0.342	0.572	0.0136	0.459	—	—
0.10	118.0	1.02	1.24	0.620	0.315	0.475	0.0130	0.207	—	—
−0.10	148.0	1.04	1.43	0.716	0.354	0.616	—	—	—	—
0.20	105.0	1.01	1.16	0.581	0.301	0.429	0.0128	0.160	—	—
−0.20	164.0	1.06	1.56	0.779	0.386	0.736	—	—	—	—
0.30	91.4	0.995	1.10	0.548	0.291	0.393	—	—	—	—
−0.30	181.0	1.07	1.71	0.855	0.437	0.936	—	—	—	—
∞	0.3	2.0	3.0	3.0	5.0	3.0	30.0	5.0	50.0	7.0

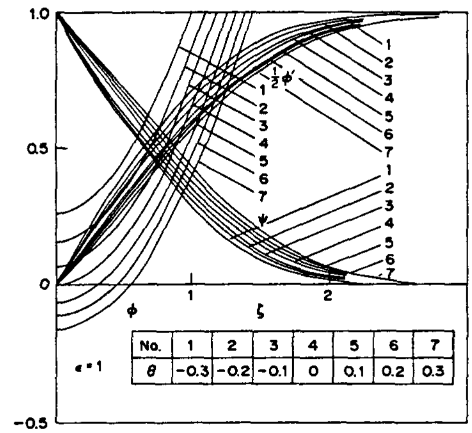

FIG. 2.

From Table 1 and Figs. 1–3 it is seen that the change of the direction of mass transfer (the sign of θ) influences the value of the concentration gradient at the phase boundary $\Psi'(0)$, and consequently the mass transfer coefficient k or Sh. When the mass transfer is from the phase boundary to the main flow ($\theta > 0$), the non-linear effects result in lower mass transfer coefficients, while if the mass transfer is from the main flow to the phase boundary ($\theta < 0$) these effects result in higher mass transfer coefficients. For equal absolute values of θ, Sh increases more rapidly for $\theta < 0$ than it decreases for $\theta > 0$.

The data from Fig. 1 show that changes of θ practically do not influence the thickness of the diffusive boundary layer and the effect of the induced secondary flow is significant only in a layer close to the phase boundary with thickness approximately one-third of the thickness of the diffusive boundary layer. The non-dimensional thickness of this 'layer of non-linear mass transfer', η_0, can be determined approximately from Fig. 1 for $\varepsilon = 1$ and 20

$$\varepsilon = 1, \quad \eta_0 = 1 \tag{14a}$$

$$\varepsilon = 20, \quad \eta_0 = 2. \tag{14b}$$

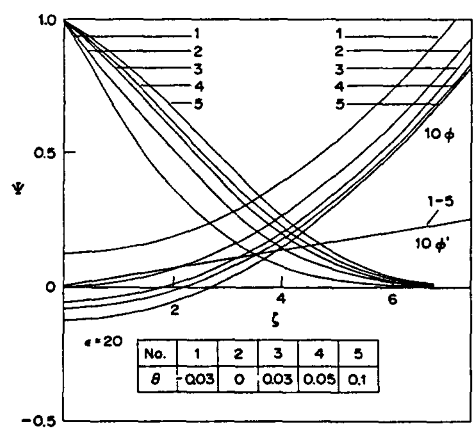

FIG. 3.

The dimensional thickness δ_0 of this layer can be calculated by means of equation (9d)

$$\delta_0 = \eta_0 \left(\frac{4Dx}{u_0} \right)^{0.5}. \tag{14c}$$

The quantitative difference in the non-linear effects for equal in absolute values but of opposite sign parameters θ is due to the fact that the velocity of the induced flow, v_n, is higher for larger in absolute value but negative θ. This can be explained if one assumes (for small η in the 'layer of non-linear mass transfer') that the normal velocity is proportional to Φ. The latter quantity can be expressed as [8]

$$\Phi(\eta) = a_0 + a_1\eta + a_2\eta^2 + \cdots \tag{15a}$$

By virtue of the boundary conditions, equations (10c)–(10e), one has

$$\Phi(0) = \theta\Psi'(0) + \tfrac{1}{2}\Phi''(0)\eta^2 - \frac{\Phi''(0)}{6\varepsilon}\Psi'(0)\eta^3 + \cdots \tag{15b}$$

If Φ_+ and Φ_- denote two solutions for Φ corresponding to $\theta = \theta_0$ and $-\theta_0 (\theta_0 > 0)$ at $y = 0$, then the condition

$$\Phi_+^2 - \Phi_-^2 < 0 \tag{15c}$$

is satisfied when

$$\frac{\Phi''(0)}{6\varepsilon}\eta^3 < 1. \tag{15d}$$

From Table 1 and Figs. 1–3 it is seen that the condition, equation (15d), is satisfied always when η is in the 'layer of non-linear mass transfer', see equation (14b).

The influence of the change of sign of θ on u or Φ', see equations (9a), (10c) and (13), is opposite to the one just described. The velocity u has higher values when $\theta > 0$. This shows that the non-linear effects result mainly from the normal velocity v_n, that is from the induced flow but not from the global change of the velocity field following from it.

The last line of Table 1 shows the values of η for which the boundary conditions for Φ and Ψ at infinity, equations (10f) and (10g) have been satisfied in the sense of the algorithm [9] to the accuracy required.

4. LIMITING CASES

In Table 1 there are no values of $\Phi''(0)$ and $\Psi'(0)$ for large values of ε and θ. They could not be computed utilizing the procedure [9] due to an increasing singular perturbation (or stiffness) of the solution of the boundary value problem, equation (10). Such limiting cases cannot be described by the theory of diffusive boundary layers but they can be explained if in equations (10) one introduces the following variables:

$$\Phi(\eta) = f(\zeta) \tag{16a}$$

$$\zeta = \eta/\varepsilon. \tag{16b}$$

Introducing equations (16) into equations (10), one readily obtains

$$f''' + ff'' = 0, \quad \zeta = 0 \tag{17a}$$

$$\Psi'' + \varepsilon f \Psi' = 0, \quad \zeta > 0 \tag{17b}$$

$$f(0) = \theta \Psi'(0), \quad \zeta = 0 \tag{17c}$$

$$f'(0) = 0, \quad \zeta = 0 \tag{17d}$$

$$\Psi(0) = 1, \quad \zeta = 0 \tag{17e}$$

$$f'(\infty) = 2, \quad \zeta = \infty \tag{17f}$$

$$\Psi(\infty) = 0, \quad \zeta = \infty. \tag{17g}$$

In equations (17) $\zeta \ll 1$ when $\varepsilon \gg 1$ and therefore f can be expressed by its McLorain series in the vicinity of zero (see equation (17d))

$$f(\zeta) = f(0) + \tfrac{1}{2} f''(0)\left(\frac{\eta}{\varepsilon}\right)^2 + \cdots \tag{18}$$

In the absence of non-linear effects ($\theta = 0$) from equations (17) it follows that $f(0) = 0$ and the problem always has a solution but when $\varepsilon \gg 1$. For the latter case

$$\tfrac{1}{2}\varepsilon f''(0)\left(\frac{\eta}{\varepsilon}\right)^2 = \tfrac{1}{2}\varepsilon \Phi''(0)\eta^2 < 10^{-2}. \tag{19}$$

Consequently, $\Psi'' = 0$ and the problem is reduced to stationary diffusion in a solid.

In the presence of non-linear effects ($\theta = 0$) from equations (17) it follows that $f(0) = 0$ and the problem always has a solution except when

$$f(0) \gg \tfrac{1}{2} f''(0)\left(\frac{\eta}{\varepsilon}\right)^2. \tag{20}$$

It is obvious, that in this case the diffusion is independent from the main hydrodynamic flow and from equations (10) and (18) it follows that

$$\Psi'' + \varepsilon\theta\Psi'(0)\Psi' = 0, \quad \zeta > 0 \tag{21a}$$

$$\Psi(0) = 0, \quad \zeta = 0 \tag{21b}$$

$$\Psi(\infty) = 0. \quad \zeta = \infty. \tag{21c}$$

The boundary value problem, equations (21), has no physically meaningful solution because a solution of the type

$$\Psi = \exp(-\varepsilon\theta\Psi'(0)\zeta) \tag{22a}$$

does not satisfy the boundary conditions for $\theta \geqslant 0$ ($\Psi'(0) < 0$), while if $\theta < 0$ equation (22a) satisfies equations (21) only if

$$\Psi'(0) = -\varepsilon\theta\Psi'(0) \tag{22b}$$

or, equivalently, $\varepsilon\theta = -1$. The last relation has no physical meaning in the boundary layer theory. It is possible that in this particular case the secondary induced flow, resulting from the non-linear mass transfer, dominates significantly the main flow and its convective transfer is the one that limits the process. If this is the case, it could generate process instability but the latter cannot be described in the approximation of the diffusive boundary layer theory and is a subject of a separate study.

5. CONCLUSIONS

The above theoretical analysis of the influence of the intensive mass transfer on the rate of mass transfer leads to some new conclusions concerning the phenomenon of non-linear mass transfer.

(1) The mass flux through the phase boundary is determined by two independent factors—the concentration gradient and the concentration of the species transported. The second one does not depend on the direction of mass transfer, while in the presence of an irreversible process the phase boundary does not exist.

(2) In the presence of a reversible process the effect of the concentration can dominate the effect of the concentration gradient.

(3) The change of the direction of mass transfer can influence the rate of mass transfer. The latter is higher (lower) than the one predicted by the linear theory, when the direction of mass transfer is to (from) the phase boundary.

(4) The decrease of the rate of mass transfer in the presence of intensive transfer from the phase boundary to the main flow is a new effect which cannot be predicted qualitatively from the non-linear theory of mass transfer.

(5) The change of the rate of mass transfer resulting from a change of the direction of the intensive mass transfer is due, most of all, to the convective effect of the secondary induced flow. This effect is significant close to the phase boundary inside the 'layer of non-linear mass transfer' of thickness approximately one-third of the thickness of the diffusive boundary layer.

(6) The non-linear theory of mass transfer cannot predict the rate of mass transfer for very large Schmidt numbers (diffusion in a solid) or in the presence of very high concentration gradients. In the latter case the secondary induced flow fully dominates the main flow and the approximations of the theory of the diffusive boundary layer are not valid.

Acknowledgement—This project was completed with the financial support of the Committee for Science at the Council of Ministers under Contract No. 154.

REFERENCES

1. Chr. Boyadjiev and V. Beshkov, *Mass Transfer in Liquid Film Flows*. Publishing House of the Bulgarian Academy of Sciences, Sofia (1984).
2. Chr. Boyadjiev and Pl. Mitev, On the concentration boundary layer theory at a moving interface, *Chem. Engng J.* **14**, 225–228 (1977).

3. Chr. Boyadjiev, Nonlinear mass transfer in falling films, *Int. J. Heat Mass Transfer* **25**, 535–540 (1982).
4. Chr. Boyadjiev, Influence of some nonlinear effects on the mass transfer kinetics in falling liquid films, *Int. J. Heat Mass Transfer* **27**, 1277–1280 (1984).
5. Chr. Boyadjiev and N. Vulchanov, Non-linear mass transfer in boundary layers—1. Asymptotic theory, *Int. J. Heat Mass Transfer* **31**, 795–800 (1988).
6. V. G. Levich, *Physico-chemical Hydrodynamics*. Prentice-Hall, Englewood Cliffs, New Jersey (1962).
7. N. Vulchanov and Chr. Boyadjiev, Non-linear mass transfer in boundary layers—2. Numerical investigation, *Int. J. Heat Mass Transfer* **31**, 801–805 (1988).
8. H. Schlichting, *Granzschicht Theorie*. G. Braun, Karlsruhe (1965).
9. N. Vulchanov and M. Konstantinov, Numerical integration of asymptotic TPBVP's for ODE's. EQUADIF 6, Abstracts, p. 250. Brno, CSSR, 25–30 August (1985).

INFLUENCE DU TRANSFERT DE MASSE A L'INTERPHASE SUR LE FLUX DE MASSE—1. LE SYSTEME "SOLIDE-FLUIDE (GAZ)"

Résumé—On développe une analyse théorique de l'influence de la direction du transfert de masse à l'interphase sur le flux massique. Le cas du transfert de masse entre une surface plane solide et un fluide ou un gaz dans une approximation de couche limite est étudié lorsque les transferts de masse et de quantié de mouvement à la frontière de phase sont couplés pour tenir compte des effets non linéaires dans une direction normale à l'écoulement principal. Des résultats numériques sont rapportés et discutés.

EINFLUSS DES STOFFTRANSPORTS AN DER PHASENGRENZE AUF DIE GESCHWINDIGKEIT DER STOFFÜBERTRAGUNG—1. DAS SYSTEM FESTKÖRPER-FLUID (GAS)

Zusammenfassung—Der Einfluß der Richtung des Stofftransports an der Phasengrenzfläche auf die Geschwindigkeit des Stoffübergangs wird theoretisch untersucht. Der Stofftransport zwischen einer festen ebenen Oberfläche und einer Fluid- oder Gasströmung wird unter Anwendung der Grenzschichtnäherung analysiert, und zwar für den Fall, daß Stoff- und Impulstransport an der Phasengrenze gekoppelt sind. Dadurch werden nicht-lineare Effekte in einer Richtung senkrecht zur Hauptströmung berücksichtigt. Die numerischen Ergebnisse werden vorgestellt und diskutiert.

ВЛИЯНИЕ НАПРАВЛЕНИЯ МАССОПЕРЕНОСА НА ЕГО СКОРОСТЬ—1. СИСТЕМА "ТВЕРДОЕ ТЕЛО–ЖИДКОСТЬ (ГАЗ)"

Аннотация—Теоретически анализируется влияние направления массопереноса на межфазной границе на его скорость. С целью учета распространения эффектов в направлении, перпендикулярном основному течению, исследуется случай массопереноса между твердой плоской поверхностью и течением жидкости или газа в приближении пограничного слоя при взаимосвязанном переносе массы и импульса у межфазной границы. Приводятся и обсуждаются численные результаты исследования.