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MULTICOMPONENT BOUNDARY-LAYER CHARACTERISTICS—USE OF THE REFERENCE STATE

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Abstract—The possibility of using a constant property analysis for multicomponent diffusion systems where interaction between fluxes is not ignored, is investigated in this work. The results are compared with an exact multicomponent variable property analysis and a constant property analysis utilizing the "effective binary diffusivity" approximation.

In order to demonstrate and test this approach an isothermal evaporation problem of a multicomponent mixture into a free stream of inert gas is analysed. Also the case of flat plate transpiration mass transfer is considered.

Results show that also when the properties of the chemical species involved in the diffusion process are markedly different, the use of a constant property analysis with multicomponent diffusivity, evaluated at a reference state, is quite accurate.

NOMENCLATURE

c, molar density;
D, diffusion coefficient defined by equation (5);
D^B, binary diffusivity;
D^M, multicomponent diffusivity;
f, dimensionless stream function;
k, dimensionless mass-transfer coefficient (equation 24);
l, auxiliary function (equation 22);
m, mass flux, y directed when scalar;
M, molecular weight;
n, number of components;
r_{ij}, defined by equation (21);
u, velocity component, x direction;
v, velocity component, y direction;
w, mass fraction;
x, coordinate;
X, mole fraction;
y, coordinate, also an auxiliary function (equation 21);
z, auxiliary function (equation 21).

Greek symbols

η, similarity independent variable;
μ, dynamic viscosity;
ν, kinematic viscosity;
ρ, density;
ϕ, dimensionless mass concentration.

Subscripts and superscripts

0, interface;
∞, bulk;
i, j, running indeces;
m, mixture;
n, inert gas;
r, reference.

INTRODUCTION

PROBLEMS involving boundary layers of heat and mass transfer become quite cumbersome in the presence of multicomponent diffusion [1]. In this case the flux of each individual component depends on the mass fluxes of all other components. In other words, the local flux of each species is a complicated function of all local concentrations and local concentration gradients. The complete and exact treatment is not only complicated but may also consume so much computer time that it may become impractical, especially when the number of components is large [2].

The quest for simplicity leads, firstly to the use of constant property analysis as compared to variable property analysis. It has long ago been established that a reference state in which transport properties are considered constant but are evaluated at a suitable reference state is quite adequate for most practical cases [3]. The use of constant property analysis is not only simpler to perform but it is also usually more amenable to a generalized parametric studies as compared to variable property analysis.

In this quest for simplicity, multicomponent diffusion presents a special problem because of its complicated

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nature. As a common practice multicomponent diffusion is expressed approximately by a binary diffusion coefficient whereby the diffusive flux of each component is calculated using Fick's Law. This has been used for multicomponent mixture by separating the chemical components into two similar groups and using a single binary diffusion coefficient D_{ij}^{B} to express the diffusion of each component member of the group. A somewhat modified approach, but which is also based on the use of a single binary diffusion coefficient is given by Kendall [4]. Another common method is the use of an effective binary diffusivity D_{im}^{B} which expresses the diffusion of the ith component in the mixture in accordance with Fick's Law [5, p. 571]. These methods of treating multicomponent diffusion has been used also in recent advanced studies [2, 6].

The shortcoming in using these approximations lies first in the somewhat ambiguity in evaluation either the single binary coefficient D_{ij}^{B} or the effective diffusivity D_{im}^{B}. Secondly, as an approximate techniques, one looses the multicomponent interaction characteristics among fluxes of different species which may cause a serious error under certain conditions.

In this article we check the possibility of the use of a constant property analysis combined with multicomponent diffusion coefficients by the method of the reference state. Comparison is then made with the complete variable property analysis and with the simplified approach using the effective binary diffusion coefficient D_{im}^{B}.

The model problem which has been chosen to demonstrate and check this approach is the evaporation of a multicomponent mixture into uniform flow of an inert gas which is impermeable to the interface of the liquid mixture. This model may be realized in practice when a gas is used to dry a wet surface of volatile liquid mixture. The problem worked here may as well include the case of transpiration of several chemical species through a porous flat plate. This model for binary mixture has been extensively studied for the purpose of transpiration cooling [3, 7]. Some more pertinent recent studies regarding evaporation problems, although different than the one presented here, can be found in references [8–10].

ANALYSIS

The model considered here consists of a gas flowing with uniform velocity u and approaching a semi-infinite "flat plate" where the concentration, w_{i0}, of the $(n-1)$ evaporating chemical species is specified. The nth component, designated as the inert gas, is impermeable to the "flat plate". For the sake of generality we may also allow the free stream to contain initial concentration of the volatile gases.

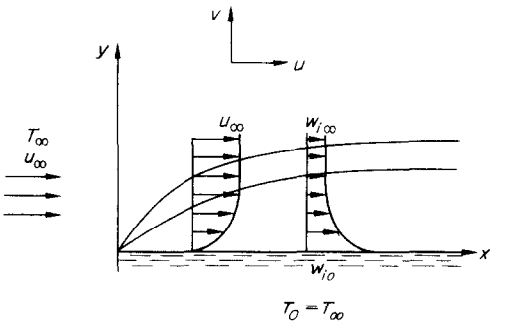

FIG. 1. Physical model and coordinate system.

For the convenience of presentation, the variable property analysis is presented first. Thus, using x as the longitudinal coordinate and y as the normal coordinate (see Fig. 1) the continuity and momentum equations read:

$$\frac{\partial(\rho u)}{\partial x} + \frac{\partial(\rho v)}{\partial y} = 0 \tag{1}$$

$$\rho u \frac{\partial u}{\partial x} + \rho v \frac{\partial u}{\partial y} = \frac{\partial}{\partial y}\left(\mu \frac{\partial u}{\partial y}\right). \tag{2}$$

For the conservation equation of each chemical species we use the accurate expression for multicomponent mixture of ideal gases where the mass flux of species i is given by [5, p. 569].

$$\mathbf{m}_i = \frac{c^2}{\rho} \sum_{j=1}^{n} M_i M_j D_{ij}^{M} \nabla X_j + \rho \mathbf{v} w_i \tag{3}$$

where D_{ij}^{M} is the multicomponent diffusion coefficient which is a function of the binary diffusion coefficients, of the concentration and of the molecular weights ([5], p. 570 and [11], p. 541). Equation (3) can alternatively be expressed more conveniently in terms of the mass fraction, namely

$$\mathbf{m}_i = \rho \sum_{j=1}^{n} D_{ij} \nabla w_j + \rho \mathbf{v} w_i \tag{4}$$

where

$$D_{ij} = M_i D_{ij}^{M} \sum_{k=1}^{n} \frac{w_k}{M_k} - \frac{M_i}{M_j} \sum_{k=1}^{n} D_{ik}^{M} w_k. \tag{5}$$

Using equation (4) the conservation equation for the chemical species takes the form of

$$\rho u \frac{\partial w_i}{\partial x} + \rho v \frac{\partial w_i}{\partial y} = -\frac{\partial}{\partial y}\left(\rho \sum_{j=1}^{n} D_{ij} \frac{\partial w_j}{\partial y}\right). \tag{6}$$

In considering this problem we assume that isothermal conditions prevail. Namely, the liquid mixture interface, is kept at constant temperature equal to that of the free tream temperature. In other words, a heating mechanism which prefix the mixture interface temperature is assumed to exist. Under these conditions

the energy equation can be discarded. Also effects of thermal diffusion and diffusion-thermo are neglected.

Equations (1), (2) and (6) are subjected to the commonly used similarity transformation as follows:

$$u = \frac{\rho_r}{\rho}\frac{\partial \Psi}{\partial y} \qquad v = -\frac{\rho_r}{\rho}\frac{\partial \Psi}{\partial x} \tag{7}$$

$$\eta = [\sqrt{(u_\infty/\nu_r x)}]\int_0^y \frac{\rho}{\rho_r}\,\mathrm{d}y \tag{8}$$

$$\Psi = [\sqrt{(\nu_r u_\infty x)}]f(\eta) \tag{9}$$

and

$$\phi_i = \frac{w_i - w_{i\infty}}{w_{i0} - w_{i\infty}}. \tag{10}$$

The resulting transformed momentum equation becomes

$$\tfrac{1}{2}ff'' + \left(\frac{\rho\mu}{\rho_r\mu_r}f''\right)' = 0 \tag{11}$$

while the diffusion equation, using also the identity

$$\sum_{i=1}^{n} w_i = 1,$$

reads

$$f\frac{\mathrm{d}\phi_i}{\mathrm{d}\eta} + 2\frac{\mathrm{d}}{\mathrm{d}\eta}\left[\left(\frac{\rho}{\rho_r}\right)^2 \sum_{j=1}^{n-1} \frac{D_{in} - D_{ij}}{\nu_r}\frac{w_{j0} - w_{j\infty}}{w_{i0} - w_{i\infty}}\frac{\mathrm{d}\phi_j}{\mathrm{d}\eta}\right] = 0. \tag{12}$$

Solution of equation (11) and (12) is subjected to the following boundary conditions:

$$\begin{aligned} &\eta = 0 \qquad f' = 0 \qquad \phi = 1 \\ &\eta \to \infty \qquad f' = 1 \qquad \phi = 0. \end{aligned} \tag{13}$$

A somewhat more complicated condition arises from the impermeability of the inert gas, namely

$$m_n = \rho \sum_{j=1}^{n-1} (D_{nj} - D_{nn})\frac{\partial w_j}{\partial y} + \rho v w_n = 0. \tag{14}$$

Using equation (14), the normal velocity is expressed in terms of the dimensionless stream function f as follows

$$f_0 = -2\left(\frac{\rho}{\rho_r}\right)^2 \sum_{j=1}^{n-1} \frac{D_{nn} - D_{nj}}{\nu_r}\frac{w_{j0} - w_{j\infty}}{w_{n0}}\frac{\mathrm{d}\phi_j}{\mathrm{d}\eta}. \tag{15}$$

Next we turn our attention to the solution of this problem using a constant property analysis but multicomponent diffusion coefficients, evaluated at a reference state. The reference concentration is chosen, conventionally, at

$$w_{ir} = \tfrac{1}{2}(w_{i0} + w_{i\infty}). \tag{16}$$

Then all properties, including the generalized diffusion coefficients given by equation (5), are evaluated at this reference concentration. There is no need to repeat the equations for constant property analysis because equations (7)–(15) can be used also for this purpose provided the reference properties ρ_r and μ_r are taken at the reference state (so far ρ_r and μ_r could have been arbitrarily chosen for the use of the exact solution). Thus one may realize that equations (11)–(15) become simpler because $\rho\mu/(\rho_r\mu_r) = \rho/\rho_r = 1$ and also the diffusion coefficients D_{ij} in equation (12) and (15) are considered constant (and can be taken outside the derivative operation) while they are concentration dependent (see equation (5)) for the exact analysis. Thus, although the formal appearance of the constant property equations seem almost identical to the variable property ones, they are much simpler and the effort in the analysis and the computation time for the latter is considerably reduced.

Finally we turn our attention to constant property analysis where the simplified binary diffusion coefficient D_{im}^B is used for all components excluding the inert one. In this case equation (6) takes the form

$$u\frac{\partial w_i}{\partial x} + v\frac{\partial w_i}{\partial y} = D_{im}^B\frac{\partial^2 w_i}{\partial y^2} \qquad i = 1 \ldots n-1 \tag{17}$$

while the transformed equation takes the form

$$f\phi_i' + \frac{2D_{im}^B}{\nu}\phi_i'' = 0. \tag{18}$$

The impermeability condition for the inert gas, using

$$m_n = m - \sum_{i=1}^{n-1} m_i \tag{19}$$

results in

$$f_0 = 2\sum_{i=1}^{n-1} \frac{D_{im}^B}{\nu}\frac{w_{i0} - w_{i\infty}}{w_{n0}}\frac{\mathrm{d}\phi_i}{\mathrm{d}\eta} \tag{20}$$

which is equivalent to equation (15) for the previous cases.

SOLUTION

Solution of the problems for f' and ϕ_i as a function of η was performed numerically using a standard Runge–Kutta integration routine. However few adjustments of the equations were needed in order to perform this operation efficiently.

In the case of the exact solution we introduced the auxiliary functions

$$y = \frac{\rho\mu}{\rho_r\mu_r}f'' \tag{21-A}$$

and

$$z_i = \sum_{i=1}^{n-1}\left(\frac{\rho}{\rho_r}\right)^2 \frac{D_{in} - D_{ij}}{\nu_r}\frac{w_{j0} - w_{j\infty}}{w_{i0} - w_{i\infty}}\frac{\mathrm{d}\phi_j}{\mathrm{d}\eta} \equiv \sum_{i=1}^{n-1} r_{ij}\frac{\mathrm{d}\phi_j}{\mathrm{d}\eta}. \tag{21-B}$$

Thereby equations (11) and (12) can be represented as

$$y' = -\tfrac{1}{2} f y \frac{\rho_r \mu_r}{\rho \mu} \tag{22-A}$$

$$l' = y \frac{\rho_r \mu_r}{\rho \mu} \tag{22-B}$$

$$f' = l \tag{22-C}$$

and

$$\phi_i' = \sum_{j=1}^{n-1} r_{ij}^{-1} z_j \tag{23-A}$$

$$z_i' = -\tfrac{1}{2} f \sum_{j=1}^{n-1} r_{ij}^{-1} z_j \tag{23-B}$$

which is in the form needed for the use of the Runge–Kutta procedure.

Since f' and ϕ_i which are given at $\eta \to \infty$ rather that at $\eta = 0$ and f_0 which satisfy equation (15), are yet unknown, the solution was aided by an iterative technique using initial guesses for z_{i0}, f_0 and f_0''. Fast convergence was obtained utilizing the Newton–Raphson's procedure [12, pp. 2–56].

Solutions for the constant property analysis of both kinds, are less involved and more easily performed, hence they are not spelled out here. It is important however to mention that both cases, namely the one using multicomponent diffusivity and the other using the effective binary diffusion coefficient are almost comparable in terms of computation time. Whereas the solution for the variable property analysis needs much more time because properties have to be repeatedly evaluated at each step of the integration. When the problem involves many chemical components the exact analysis may indeed consume quite a large amount of time compared to the constant property analysis.

RESULTS

Results are reported for two ternary systems. The first system corresponds to evaporation of water and methanol mixture into a free stream of air (air is considered as one component). Total pressure and temperature were kept at 760 mm Hg and 75°C respectively. The other system consists of transpiration of hydrogen and carbon-dioxide, again, into a free stream of air. In this case the total pressure and temperature were kept at 760 mm Hg and 25°C respectively. The references [12–14] were consulted for the selection of properties of the individual species as well as their behaviour in the mixture.

Results for the mass transfer of each component are reported in terms of a dimensionless mass transfer coefficient which is defined and calculated as follows

$$k_i \equiv \frac{m_{i0}/\sqrt{(u_\infty \mu_r / x)}}{w_{i0} - w_{i\infty}} = \left(\frac{\rho}{\rho_r}\right)^2 \sum_{i=1}^{n-1} \frac{D_{ij} - D_{in}}{\nu_r} \frac{w_{j0} - w_{j\infty}}{w_{i0} - w_{i\infty}} \left(\frac{\mathrm{d}\phi_j}{\mathrm{d}\eta}\right)_0 - \frac{w_{i0} f_0}{2(w_{i0} - w_{i\infty})}. \tag{24}$$

Equation (24) is valid also for the constant property analysis (with multicomponent diffusion) provided all properties are evaluated at the reference concentration.

For the case where the effective binary diffusivity approximation is used, a simpler expression is obtained which reads:

$$k_i = -\frac{D_{im}^B}{\nu} \frac{\mathrm{d}\phi_i}{\mathrm{d}\eta} - \frac{1}{2} \frac{w_{i0}}{w_{i0} - w_{i\infty}} f_0. \tag{25}$$

The effective binary diffusivity, D_{im}^B, for the diffusion of water and methanol in the first case and CO_2 and H_2 in the second one were chosen as the binary coefficient of each component in air. This is consistent with the exact solution when the volatile components are in low concentration in the air.

The results for the methanol–water–air system is reported in Fig. 2. In this figure the dimensionless mass-transfer coefficient of methanol ($i = 1$) and water ($i = 2$) is given as a function of the relative concentration of methanol for constant interfacial air concentration ($i = 3$). Note that we obtained results only for the case where the free stream contained only air ($w_3 = 1$).

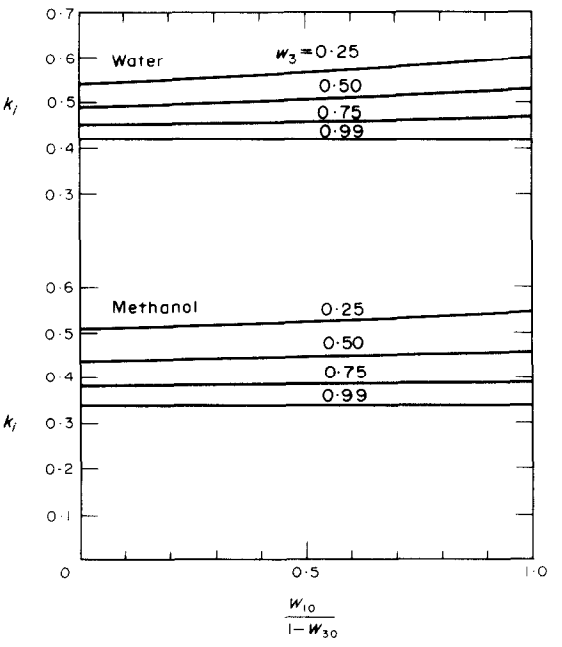

FIG. 2. Dimensionless mass-transfer coefficient for the methanol–water–air system. Exact and approximate solutions.

Thus the r.h.s. of the abscissa corresponds to a binary mixture of methanol and air while the left one corresponds to a binary mixture of water and air. Figure 2 reveals that the dimensionless mass transfer coefficient, which is proportional to the flux divided by the driving force $(w_{i0} - w_{i\infty})$ is practically unaffected by the change in methanol–water ratio in the mixture. On the other hand increasing interfacial concentration of air reduces the dimensionless coefficient because the increase of air concentration has an additional effect of reducing the wall blowing value v_0.

One may realize that Fig. 2 contains only one set of results and there is no distinguishable differences among the exact solution and the two approximate ones. The numerical values of the calculations reveal maximum discrepancy of no more than 3 per cent and hence were omitted in Fig. 2. Thus, although transport properties of methanol water and air are different they still do not differ to the extent that any of the above constant property analyses will show a significant error.

Our second system was chosen so that the properties of its species are drastically different, especially with respect to their binary diffusivities. Figure 3 indeed shows that the results here are markedly different. Focusing attention on the variation of the dimensionless mass-transfer coefficient for CO_2 $(i = 1)$ we may observe the following: At air $(i = 3)$ concentration of 0·99 all solutions are almost the same. However, when the concentration of air decreases we can see that the multicomponent constant property analysis is quite close to the exact solution (less than 5 per cent discrepancy) whereas the results using the effective diffusivity approximation are quite erroneous. Note that at the r.h.s. of the abscissa the two approximate solutions are exactly the same (binary mixture of CO_2 and air) whereas their slight difference from the exact solution exhibit the error in the constant property analysis. As expected the dimensionless mass-transfer coefficient for Hydrogen is higher than for CO_2. On the other hand, the values of this coefficient, as calculated by the constant property analysis exhibit again less than three per cent error and therefore the approximate analyses are not included in the figure. This fact is explained simply because air and CO_2 are not markedly different in their properties and thus Hydrogen diffuses almost according to its binary diffusion coefficient either in air or in CO_2. On the other hand, CO_2 diffuses in air–H_2 mixture. The differences in properties of air and H_2 suggest that a more exact multicomponent diffusive law is essential.

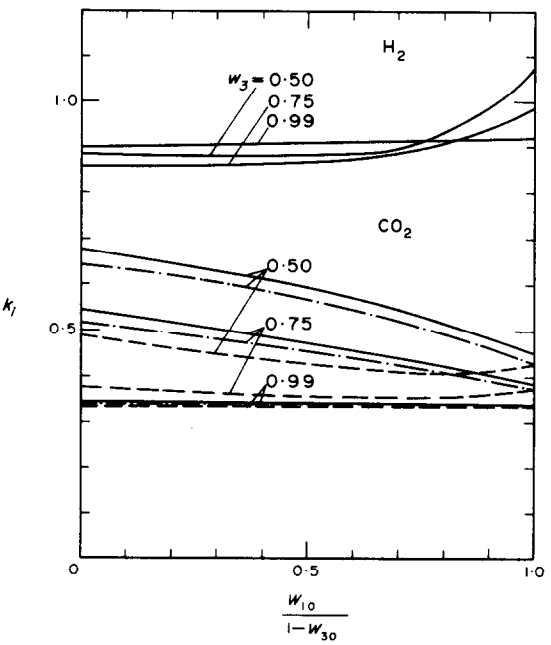

FIG. 3. Dimensionless mass-transfer coefficient for the CO_2–H_2–air system.
——, Exact solution; —·—, "Multicomponent" constant property solution; – – –, Effective binary diffusivity solution.

SUMMARY AND CONCLUSIONS

In this work the application of a constant property boundary-layer analysis with a multicomponent diffusion law is tested. It has been shown to yield very accurate results as compared to an exact analysis also when the transport properties are markedly different. The use of the effective binary diffusivity have been shown to be inferior to the constant property analysis using multicomponent diffusion law (although when variation in properties is not large any method is adequate).

The advantages of using this multicomponent constant coefficient may be summed up along the following points. First, by comparison with the exact solution we may outline advantages such as:

(1) Simplicity: the description of a constant property is simpler and is more amenable to a parametric study. It's solution is also much more easily obtained.

(2) Save of numerical efforts: In our example the computing time of the constant property analysis was about 1/5 as compared with the variable property analysis. Also without referring to the specific example used here it is clear that the number of operations for the variable property analysis, where properties have to be evaluated at each step of the numerical integration is much higher. This number of operations is highly dependent on the number of components. When the number of components is high the save in computation time could be indeed drastic.

On the other hand the advantages of this method as compared with the effective binary diffusivity method are as follows:

(1) It is much more accurate when the differences among the binary diffusivity coefficients of the various components are large.
(2) The evaluation of the multicomponent constant diffusivity in its reference state is clearly and uniquely defined.
(3) It is almost comparable to the method of the constant effective binary diffusivity method in terms of numerical efforts. This last point indeed suggests that this method could be used with no trade-off penalty making its use very highly recommended.

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CARACTERISTIQUES DE COUCHE LIMITE A PLUSIEURS COMPOSANTES—UTILISATION DE L'ETAT REFERENCE

Résumé—On considère ici la possibilité d'utiliser une analyse de propriété constante pour des systèmes de diffusion à plusieurs composants où l'intéraction entre les flux n'est pas ignorée. Les résultats sont comparés avec ceux d'une analyse exacte à propriété variable et d'une analyse à propriété constante utilisant l'approximation de la "diffusivité binaire effective".

De façon à démontrer et d'éprouver cette approche, est considéré le problème d'évaporation isotherme d'un mélange à plusieurs composants dans un écoulement libre de gaz inerte. On étudie aussi le cas du transfert massique par transpiration sur une plaque plane.

Les résultats montrent que lorsque les propriétés des espèces chimiques sont très différentes dans le processus de diffusion, l'usage d'une analyse à propriété constante, avec une diffusivité complexe évaluée à un état de référence, est très pertinent.

BESTIMMUNG DER MEHRKOMPONENTEN-GRENZSCHICHT-CHARAKTERISTIK ÜBER EINEN REFERENZZUSTAND

Zusammenfassung—In dieser Arbeit wird die Möglichkeit der Anwendung eines Modells mit konstanten Stoffeigenschaften für die Untersuchung von Mehrkomponenten-Diffusionssystemen, bei denen die Wechselwirkung zwischen den Strömungen nicht vernachlässigt werden darf, untersucht und die Ergebnisse mit denen einer exakten Theorie für mehrere Komponenten und variable Stoffeigenschaften sowie eines Ansatzes für konstante Stoffeigenschaften bei Berücksichtigung einer Näherung durch das "effektive binäre Diffusionsvermögen" verglichen.

Um diese Näherungsmethode zu veranschaulichen und zu prüfen, wird bei einer Mehrkomponentenmischung in einer freien Strömung von Inertgas das isotherme Verdampfungsproblem und an einer ebenen Platte der Fall des Stoffaustauschs durch Verdunstung untersucht.

Die Ergebnisse zeigen, daß selbst wenn die Stoffeigenschaften stark unterschiedlich in den Diffusionsvorgang eingehen, das Modell mit konstanten Stoffeigenschaften—bezogen auf einen Referenzzustand—bei Mehrkomponentendiffusion das Problem sehr genau beschreibt.

ИСПОЛЬЗОВАНИЕ ОПРЕДЕЛЯЮЩЕГО СОСТОЯНИЯ ДЛЯ ПОЛУЧЕНИЯ ХАРАКТЕРИСТИК МНОГОКОМПОНЕНТНОГО ПОГРАНИЧНОГО СЛОЯ

Аннотация — В работе при анализе многокомпонентной диффузии, когда нельзя пренебречь взаимодействием потоков, используется допущение о постоянстве свойств. Результаты сравниваются с данными точного анализа с учётом переменности свойств при многокомпонентной диффузии и анализа с допущением постоянных свойств и использованием «эффективного коэффициента бинарной диффузии».

Для демонстрации и проверки данного метода рассматривается задача изотермического испарения многокомпонентной смеси в свободный поток инертного газа. Также рассматривается случай переноса массы при пористом вдуве из плоской пластины.

Результаты показывают, что при заметном различии свойств химических компонент, участвующих в диффузионном процессе, анализ с допущением постоянства свойств и использованием коэффициента многокомпонентной диффузии, рассчитанного для определяющего состояния, даёт достаточно точные результаты.