HEAT AND MASS TRANSFER IN REACTING FLOW SYSTEMS: AN APPROXIMATE THEORY

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Abstract—An approximate theory is developed to describe the combined effects of heat and multicomponent mass transfer in chemically reacting flow systems. The chemical potentials of the components are expressed in terms of the Planck potentials. The applicability of the theory is demonstrated for reacting Couette flow and its limitations are also discussed.

NOMENCLATURE

a _{i i} ,	phenomenological coefficients;
Å,	chemical affinity of reaction <i>j</i> ;
b_{ii}	phenomenological coefficients;
C ₁ ,	concentration of component k;
d.,	characteristic length of reaction i;
d.	distance between two plates in Couette flow;
f	phenomenological coefficients;
h.	enthalpy per unit mass;
Ĵ	reaction rate per unit volume of reaction <i>j</i> ;
J_{μ}	mass-transfer flux;
Ĵ.	heat-transfer flux;
Ŕ.	generalized flux;
Lin.	phenomenological coefficients;
n,	number of species of the mixture;
<i>p</i> ,	pressure;
q_{ai}	phenomenological coefficients;
Ö.	congruent transformation;
t,	time;
Ť,	temperature;
t _{an} ,	phenomenological coefficients;
Ŭ,	velocity;
ũ,	internal energy;
u,v,w,	velocity components;
V,	velocity vector;
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x, y, Cartesian coordinates.

Greek symbols

- θ , dimensionless temperature;
- λ , thermal conductivity;
- Λ, diagonal matrix ;
- μ_k , chemical potential of species k;
- *u*, viscosity coefficient ;
- v_{ki}, stoichiometric mass coefficient;
- π , scalar component of viscous stress tensor;
- ρ , density;
- σ , rate of entropy production per unit volume;
- τ, stress tensor;
- τ, dimensionless temperature profile component due to temperature difference;
- ϕ , dimensionless temperature profile
- component due to viscous dissipation;

$$\psi_k$$
, $= -\frac{r_k}{T}$ - Planck potential of component k;

 Ω , dimensionless dissipation factor.

Special symbols

- ()*, variable transformed by congruent
- transformation;
- (⁻), dimensionless variable.

1. INTRODUCTION

DURING the last decade there has been a considerable progress [1-4] in improving our understanding of reacting flow systems involving heat and multicomponent mass transfer. A difficult problem that arises in many of these studies is the possible effects of thermodynamic coupling, namely, how important are these effects, if at all? If and when should they be taken into account? And, most important, do they exist among chemical reaction rates? Unfortunately, most of these questions have not yet been resolved [5-9].

One reason, and not at all the only one, for the absence of agreement among the investigators in this field is the lack of an overall theory that takes into account momentum, energy and mass transfer in reacting systems, as well as all possible thermodynamic coupling effects. Such a general theory may allow a direct comparison with experiment without a priori neglecting the thermodynamic coupling effects, especially in systems involving large gradients of the proper fields (which contribute most to the expected values of thermodynamic coupling effects). Obviously, other types of couplings, such as between concentrations in the energy and species conservation equations, as well as between fields and the phenomenological coefficients, create additional difficulties. Therefore, no such general theory exists yet.

In this work we attempt to develop an approximate theory that will take some of these effects into account while neglecting some others. Thus, while assuming constant phenomenological coefficients, we take into account thermodynamic coupling between heat- and mass-transfer fluxes, thermodynamic coupling between the chemical reaction rates, and "normal" couplings between the concentration fields in the energy and the species conservation equations in dissipative flow systems with pressure gradients. The theory presented here is therefore of a limited value, nevertheless it represents the first analytical attempt to approximate such effects in reacting flow systems, that is to say it expands the Brown theory [10] from nonflow reacting systems to reacting flow systems.

At first, the mathematical treatment may appear straightforward, but as one proceeds through, entirely new problems arise, mainly those due to the convection and dissipation terms involved. Such a procedure eventually leads to a highly complicated theory which is hardly useful due to the many nonlinear terms involved. This theory is available elsewhere [11]. The present work gives a more limited, but workable theory whose applicability is demonstrated here for two fundamental cases that may be further used by such procedures as linear combinations. The results obtained indicate that a number of complex phenomena usually encountered in such systems as reacting boundary-layer flows, stagnation flows or reacting jet flows may be better understood by combining the results obtained here with other limited results for coupled flow systems [12, 13].

2. METHOD OF APPROACH AND FUNDAMENTAL FORMULATION

To start with we define a number of n independent generalized fluxes \bar{K}_i according to

$$\bar{K}_i \equiv \bar{J}_i - \frac{c_i}{h} \bar{J}_q \qquad (i = 1, 2, \dots, n).$$
⁽¹⁾

We then note that the rate of production of entropy per unit volume for heat and multicomponent mass transfer in reacting flow systems may be expressed as

$$\sigma = -\frac{1}{T^2} \overline{J}_q \cdot \operatorname{grad} T - \frac{1}{T} \pi \operatorname{div} \overline{V} - \frac{1}{T} \overline{\tau} : \operatorname{grad} \overline{V}$$
$$-\frac{1}{T} \sum_{j=1}^r J_j A_j - \sum_{j=1}^n \overline{J}_k \cdot \operatorname{grad} \frac{\mu_k}{T} \ge 0.$$
(2)

Using now the definition of \bar{K}_i together with the Gibbs-Duhem equation

$$h \operatorname{grad} \frac{1}{T} + \frac{1}{\rho T} \operatorname{grad} p - \sum_{k=1}^{n} c_k \operatorname{grad} \frac{\mu_k}{T} = 0, \qquad (3)$$

the rate of production of entropy may be re-expressed as

$$\sigma = -\frac{1}{T} \sum_{j=1}^{r} J_{j} A_{j} - \frac{1}{T} \pi \operatorname{div} \overline{V} + \sum_{k=1}^{n} \overline{K}_{k} \operatorname{grad} \left(-\frac{\mu_{k}}{T} \right) - \frac{J_{q}}{h\rho T} \operatorname{grad} p - \frac{1}{T} \overline{\tau} \operatorname{:} \operatorname{grad} \overline{V} \ge 0.$$
(4)

For isotropic systems, the total rate of entropy production originates from three independent contributions, according to the tensorial order of the fluxes and driving forces involved, i.e.

$$\sigma_0 = -\frac{1}{T} \sum_{j=1}^r J_j A_j - \frac{1}{T} \pi \operatorname{div} \overline{V} \ge 0$$
(5)

$$\sigma_1 = -\sum_{k=1}^n \bar{K}_k \cdot \operatorname{grad}\left(-\frac{\mu_k}{T}\right) - \frac{\bar{J}_q}{h\rho T} \cdot \operatorname{grad} p \ge 0$$
(6)

$$\sigma_2 = -\frac{1}{T}\,\overline{\tau}: \operatorname{grad}\,\overline{V} \ge 0. \tag{7}$$

If we limit ourselves to incompressible fluid flows, or to cases where the Stokes hypothesis is valid, the scalar contribution to σ becomes

$$\sigma_0 = -\frac{1}{T} \sum_{j=1}^{r} J_j A_j$$
(8)

where, as an approximation

$$J_{j} = -\sum_{j'=1}^{r} l_{jj'} \frac{A_{j'}}{T} \qquad (j = 1, 2, ..., n)$$
(9)

$$\bar{K}_{i} = -\sum_{j=1}^{n} a_{ij} \nabla \psi_{j} + f_{ip} \frac{\nabla p}{\rho T} \qquad (i = 1, 2, ..., n)$$
(10)

$$\frac{\overline{J}_{q}}{h} = \sum_{j=1}^{n} q_{qj} \nabla \psi_{j} + \frac{tqp}{T} \nabla p.$$
(11)

Here the chemical affinity $A_{j'}$ of reaction j' is defined as

$$A_{j'} \equiv \sum_{k=1}^{n} v_{kj'} \mu_k.$$
 (12)

Using the above definitions the source terms in the species conservation equations become

$$\dot{m}_{i} = \sum_{j=1}^{r} v_{ij} J_{j} = \sum_{j,j'=1}^{r} \sum_{k=1}^{n} v_{ij} l_{jj'} v_{kj'} \left(-\frac{\mu_{k}}{T} \right) = \sum_{k=1}^{n} b_{ik} \psi_{k} \quad (i = 1, 2, \dots, n)$$
(13)

where the phenomenological coefficients b_{ik} are defined by

$$b_{ik} \equiv \sum_{j,j'}^{r} v_{ij} l_{jj'} v_{kj'} \qquad (i,k=1,2,\dots,n).$$
(14)

Using equations (1) and (10) one obtains⁺

$$\nabla \cdot \bar{K}_{i} = \sum_{j=1}^{n} a_{ij} \nabla^{2} \psi_{j} + f_{ip} \frac{\nabla^{2} p}{\rho T} - f_{ip} \frac{\nabla p \cdot \nabla T}{\rho T^{2}} - f_{ip} \frac{\nabla p \cdot \nabla \rho}{\rho^{2} T}$$
$$= \nabla \cdot \bar{J}_{i} - \frac{c_{i}}{h} \nabla \cdot \bar{J}_{q} \qquad (i = 1, 2, ..., n).$$
(15)

The divergence of J_i and J_q can be expressed by the familiar species conservation and energy equations. Equation (15) may then be recast as

$$\sum_{k=1}^{n} a_{kj} \nabla^2 \psi_j + \frac{f_{kp}}{\rho T} \left[\nabla^2 p - \frac{\nabla p \cdot \nabla T}{T} - \frac{\nabla p \cdot \nabla \rho}{\rho} \right] = \sum_{i=1}^{n} b_{kj} \psi_j - \rho \frac{dc_k}{dt} + \frac{c_k}{h} \left[\rho \frac{d\tilde{u}}{dt} + \bar{\tau} : \operatorname{grad} \bar{V} + p \operatorname{div} \bar{V} \right] \qquad (k = 1, 2, \dots, n).$$
(16)

Onsager's reciprocal relations imply that for isotropic systems, obeying linear laws

$$a_{kj} = a_{jk}$$
 $(j,k = 1,2,...,n)$ (17)

$$l_{jj'} = l_{j'j}$$
 $(j,j^1 = 1,2,...,r).$ (18)

The definition of b_{ki} and the symmetry relation of $l_{ii'}$ yield

$$b_{kj} = b_{jk}$$
 $(j,k = 1,2,...,n).$ (19)

The matrices $[a_{kj}]$ and $[b_{kj}]$, being symmetric, can be diagonalized simultaneously by a congruent transformation Q that is defined by

$$\tilde{Q}\tilde{a}Q = U \tag{20}$$

$$\tilde{Q}\tilde{b}Q = \Lambda \tag{21}$$

where Λ is a diagonal matrix defined by

$$|\vec{b} - \Lambda_k \vec{a}| = 0. \tag{22}$$

Using the transformation

$$\psi_k^* = [Q_{kj}]^{-1} \psi_j \tag{23}$$

$$f_{kp}^* = [\tilde{Q}_{jk}]f_{jp} \tag{24}$$

$$c_k^* = [\tilde{Q}_{jk}]c_j \tag{25}$$

we finally obtain an expression for the energy equation, in terms of the Planck potentials

$$\nabla^{2}\psi_{k}^{*} + \frac{f_{kp}^{*}}{\rho T} \left[\nabla^{2}p - \frac{\nabla p \cdot \nabla T}{T} - \frac{\nabla p \cdot \nabla \rho}{\rho} \right] = \Lambda_{k}\psi_{k}^{*} - \rho \frac{dc_{k}^{*}}{dt} + \frac{c_{k}^{*}}{h} \left[\rho \frac{d\tilde{u}}{dt} + \bar{\tau} : \operatorname{grad} \bar{V} + p \operatorname{div} \bar{V} \right] \qquad (k = 1, 2, \dots, n).$$
(26)

The Gibbs-Duhem equation expressed in terms of the transformed variables may now be written as

$$h \operatorname{grad} \frac{1}{T} + \frac{1}{\rho T} \operatorname{grad} p + \sum_{k=1}^{n} c_k^* \nabla \psi_k^* = 0.$$
 (27)

We now proceed to demonstrate the applicability of this approximate theory by employing it in the analysis of reacting Couette flows.

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[†]Note that to obtain equation (15) we have assumed that the term (c_i/h) is constant. The limitation of this assumption as well as the assumption of Onsager's reciprocal relations (see below) should be stressed here in spite of the fact that Brown [10] and DeGroot and Mazur [5] made the same assumption (but without a due declaration). For this and other reasons we call this an approximate theory.

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3. MULTICOMPONENT REACTING COUETTE FLOWS

For an incompressible Couette flow the following assumptions apply (see Fig. 1):

$$v = 0 \qquad u = U \cdot \frac{y}{d} \tag{28}$$

$$\nabla p = 0 \tag{29}$$

$$\rho = \text{const.}$$
(30)

$$\frac{\partial}{\partial x} \equiv 0$$
 for every variable. (31)



The Planck-potential equation for Couette flow becomes

$$\frac{\partial^2 \psi_i^*}{\partial y^2} - \Lambda_i^* \psi_i^* = -\frac{c_i^*}{h} \mu \frac{U^2}{d^2} \qquad (i = 1, 2, ..., n)$$
(32)

$$\Lambda_i^* \neq 0 \quad \text{for} \quad i \leq r; \qquad \Lambda_i^* = 0 \quad \text{for} \quad i > r.$$

and the energy equation is

$$\frac{\partial}{\partial y}(J_q) = \frac{\mu U^2}{d^2}.$$
(33)

Upon integration this equation gives

$$J_{q}\left(\pm\frac{d}{2}\right) = J_{q}(0) \pm \frac{\mu U^{2}}{2d}.$$
(34)

The boundary conditions for an impermeable wall are expressed here as

$$J_i^*\left(\pm\frac{d}{2}\right) = \frac{\partial\psi_i^*}{\partial y}\Big|_{\pm d/2} + \frac{c_i^*}{h} J_q\left(\pm\frac{d}{2}\right) = 0 \qquad (i = 1, 2, \dots, n).$$
(35)

The solution of equation (32) with (35) yields

$$\psi_{i}^{*} = -\frac{J_{q}(0)d_{i}c_{i}^{*}}{2h\cosh\frac{d}{d_{i}}}\sinh\frac{2y}{d_{i}} + \frac{c_{i}^{*}\mu U^{2}}{4h} \left(\frac{d_{i}}{d}\right)^{2} - \left(\frac{d_{i}}{d}\right)\frac{\cosh\frac{2y}{d_{i}}}{\sinh\frac{d}{d_{i}}} \qquad (i \le r)$$

$$\psi_{i}^{*} = -\frac{c_{i}^{*}}{h}\left[\frac{\mu}{2}\frac{U^{2}}{d^{2}}y^{2} + J_{q}(0)y\right] \qquad (i > r)$$
(36)

where the characteristic length d_i is defined by

$$d_i = \frac{2}{\left(\Lambda_i\right)^{1/2}}.\tag{37}$$

The temperature profile may now be obtained by integration of the Gibbs-Duhem equation, viz.,

$$-\frac{h}{T^2}\frac{\partial T}{\partial y} + \sum_{i=1}^n c_i^* \frac{\partial \psi_i^*}{\partial y} = 0$$
(38)

$$T(y) - T(0) = \frac{T^2}{h} \sum_{i=1}^{n} c_i^* [\psi_i^*(y) - \psi_i^*(0)].$$
(39)

The above solution will now be further detailed for a binary system with a single chemical reaction. Equation (39) gives [after inserting ψ_i^* as calculated in (36) and after some manipulations]

$$T(y) - T\left(-\frac{d}{2}\right) = \frac{\Delta T}{d} \frac{(c_1^*)^2 \frac{d_1}{2\cosh d/d_1} \left[\sinh\left(\frac{2y}{d_1}\right) - \sinh\left(-\frac{d}{d_1}\right)\right] + (c_2^*)^2 \left(y + \frac{d}{2}\right)}{(c_1^*)^2 \frac{d_1}{d} tgh \frac{d}{d_1} + (c_2^*)^2} + \frac{\mu U^2}{2d^2} \left(\frac{T}{h}\right)^2 \left\{(c_1^*)^2 \frac{d_1 d\left(\cosh \frac{d}{d_1} - \cosh \frac{2y}{d_1}\right)}{2\sinh \frac{d}{d_1}} + (c_2^*)^2 \left[\frac{d^2}{4} - y^2\right]\right\}.$$
 (40)

The above expression is composed of two terms: the first is contributed by the temperature difference ΔT between the two plates and the second is due to viscous dissipation.

For frozen flows $(d_1 \rightarrow \infty)$, or for equilibrium flows $(d_1 \rightarrow 0)$, the first of these terms is linear and the second is parabolic:[†]

$$\left[T(y) - T\left(-\frac{d}{2}\right)\right]_{d_1 \to \infty} = \frac{\Delta T}{d}\left(y + \frac{d}{2}\right) + \frac{\mu U^2}{2d^2}\left(\frac{T}{h}\right)^2 \left[(c_1^*)^2 + (c_2^*)^2\right]\left(\frac{d^2}{4} - y^2\right)$$
(41)

$$\left[T(y) - T\left(-\frac{d}{2}\right)\right]_{d_1 \to 0} = \frac{\Delta T}{d}\left(y + \frac{d}{2}\right) + \frac{\mu U^2}{2d^2}\left(\frac{T}{h}\right)^2 \left[(c_2^*)^2\left(\frac{d^2}{4} - y^2\right)\right].$$
(42)

4. DIMENSIONLESS REPRESENTATION

The following dimensionless parameters may now be defined:

$$\bar{y} = \frac{y}{d} \tag{43}$$

$$\theta(\vec{y}) = \frac{T(y) - T(-d/2)}{T}$$
(44)

$$\Omega = \frac{\mu U^2 T}{2h^2} (c_2^*)^2 \tag{45}$$

$$\vec{d}_1 = \frac{d_1}{d} \tag{46}$$

$$\Delta \theta = \frac{\Delta T}{T} \tag{47}$$

$$\bar{c}^* = \frac{c_2^*}{c_1^*}.$$
 (48)

Then the dimensionless temperature profile is expressed by

$$\theta(\bar{y}) = \Delta \theta \frac{\frac{\bar{d}_1}{2\cosh(1/\bar{d}_1)} \left[\sinh\left(\frac{2\bar{y}}{\bar{d}_1}\right) - \sinh\left(-\frac{1}{\bar{d}_1}\right) \right] + (\bar{c}^*)^2 (\frac{1}{2} + \bar{y})}{\bar{d}_1 tgh(1/\bar{d}_1) + (\bar{c}^*)^2} + \Omega \left[\frac{1}{\bar{c}^{*2}} \frac{\bar{d}_1 (\cosh 1/\bar{d}_1 - \cosh 2\bar{y}/\bar{d}_1)}{2\sinh(1/\bar{d}_1)} + (\frac{1}{4} - \bar{y}^2) \right].$$
(49)

For a frozen flow $(\tilde{d}_1 \to \infty)$, and for an equilibrium flow $(\tilde{d}_1 \to 0)$, the dimensionless temperature profile is given by equations (50) and (51); namely by

$$\theta(\bar{y})|_{d_1 \to \infty} = \Delta \theta(\bar{y} + \frac{1}{2}) + \Omega \left[1 + \frac{1}{(\bar{c}^*)} \right] (\frac{1}{4} - \bar{y}^2)$$
(50)

$$\theta(\bar{y})|_{d_1 \to 0} = \Delta \theta(\bar{y} + \frac{1}{2}) + \Omega(\frac{1}{4} - \bar{y}^2).$$
(51)

For the more general case, [finite d_1 , as given in equation (49)], $\theta(\bar{y})$ can be expressed as a linear combination of two functions $\tau(\bar{d}_1, \bar{c}^*, \bar{y})$ and $\phi(\bar{d}_1, \bar{c}^*, \bar{y})$ (which represent the contributions due to temperature difference and viscous dissipation), i.e.

$$\bar{\theta}(\bar{y}) = \tau \cdot \Delta \theta + \phi \Omega. \tag{52}$$

T on the right side of equations (40)-(42) represents an average temperature.

The functions τ and ϕ are given by

$$\tau(\bar{d}_1, \bar{c}^*, \bar{y}) = 0.5 + \frac{(\bar{c}^*)^2 \bar{y} + \frac{d_1}{2\cosh 1/\bar{d}_1} \sinh\left(\frac{2\bar{y}}{d_1}\right)}{d_1 t g h 1/\bar{d}_1 + (\bar{c}^*)^2}$$
(53)

$$\phi(\bar{d}_1, \bar{c}^*, \bar{y}) = \left[0.25 + \frac{\bar{d}_1}{2(\bar{c}^*)^2 tgh(1/\bar{d}_1)}\right] - \left[\bar{y}^2 + \frac{\bar{d}_1 \cosh 2\bar{y}/\bar{d}_1}{2(\bar{c}^*)^2 \sinh(1/\bar{d}_1)}\right].$$
(54)

 $\tau(\bar{y})$ is plotted in Fig. 2 for $\bar{d}_1 = 0.1$ and $\bar{c}^* = 0.1, 1, 10, \phi(\bar{y})$ is plotted in Fig. 3 for $\bar{d}_1 = 0.1$ and $\bar{c}^* = 0.1, 1, 10$.

The actual temperature profile for any specific situation is a linear combination of $\tau(\bar{y})$, $\phi(\bar{y})$, the weighing functions being $\Delta\theta$ and Ω .







4. CRITICAL CONCLUSIONS

In trying to evaluate the merits of this theory we first encounter two fundamental problems, namely:

(1) To what extent are analytical procedures, such as the one developed here, useful in comparison with current computer procedures? This question is especially pressing when we deal with chemical kinetics differing from those which can be approximated by first-order kinetics. Obviously, there is no simple answer to this question. Hence, the minimum that one can say in this respect is that analytical procedures like the one presented here are useful in obtaining approximate parametric trends in complicated systems on which there is no *a priori* experimental and/or exact (computerized) information. Such analytical procedures may also be useful in the evaluation of the validity of a numerical solution.

(2) We stress the fact that the approximate theory presented here remains valid and necessary even when the assumed thermodynamic coupling between the chemical reaction rates is neglected. This can be seen from the definition of b_{ik} in equation (14), i.e. when the coupling coefficients $l_{jj'}$ vanish for $j \neq j'$, b_{ik} remains a non-diagonal matrix requiring the mathematical procedures presented here. For the specific application presented here, this means that without neglecting the thermodynamic coupling between energy and species fluxes, the neglect of the assumed "thermodynamic couplings between the chemical reactions" affects only the values of d_1 and \bar{c}^* in the parametric solutions presented in equations (53) and (54) (i.e. it does not change the form of the solution). Hence, the approximate theory presented here is sufficiently flexible to be regarded as a general procedure for describing the behavior of reacting flow systems.

(3) Some other simplified applications of the approximate theory presented here can be obtained by analytical procedures. These include the case of a multicomponent reacting flow in a heated two-dimensional channel. The solution of this case is available elsewhere [11].

(4) Finally, it should be noted that our solution is in good agreement with the numerical results presented by Broadwell [14] for the specific case of uncoupled, reacting Couette flows in binary systems.

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TRANSFERT DE CHALEUR ET DE MASSE DANS DES SYSTEMES D'ECOULEMENT AVEC REACTION: UNE THEORIE APPROCHEE

Résumé—On développe une théorie approchée pour décrire les effets combinés du transfert de la chaleur et de la matière à plusieurs composants dans des systèmes d'écoulement avec réaction chimique. Les potentiels chimiques des composants sont exprimés en fonction des potentiels de Planck. On démontre l'applicabilité de la théorie à l'écoulement de Couette avec réaction et ses limitations sont discutées.

WÄRME- UND STOFFÜBERGANG IN REAGIERENDEN STRÖMUNGSSYSTEMEN —EINE NÄHERUNGSTHEORIE

Zusammenfassung—Eine Näherungstheorie wird entwickelt, um die kombinierten Effekte von Wärmeund Stoffübergang mehrerer Komponenten in chemisch reagierenden Strömungen zu beschreiben. Die chemischen Potentiale der Komponenten werden in der Form von Planck-Potentialen ausgedrückt. Die Anwendbarkeit der Theorie wird für reagierende Couette-Strömung gezeigt, und ihre Grenzen werden ebenfalls diskutiert.

ТЕПЛО- И МАССОПЕРЕНОС В РЕАГИРУЮЩИХ ПРОТОЧНЫХ СИСТЕМАХ. ПРИБЛИЖЕННАЯ ТЕОРИЯ

Аннотация — Разработана приближенная теория для описания одновременных процессов теплои массопереноса многокомпонентных химически реагирующих проточных систем. Химические потенциалы компонентов выражаются через потенциалы Планка. На примере течения Куэтта реагирующей жидкости показана применимость теории и рассмотрены её ограничения.