

Chemical Nonequilibrium Effects in Hypersonic Pure Air Wakes

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The effect of finite chemical reaction rates on the chemical structure of hypersonic wakes is examined. The technique of incipient nonequilibrium coefficients, based on linearized chemical kinetics for a seven-component (O_2 , N_2 , O , N , NO , NO^+ , e^-), ten-reaction model of high-temperature air, is utilized in the analysis. The turbulent far wake is treated by means of a simple analysis. A range of altitudes (70–200 kft) and flight speeds (15–21 kft/sec) is considered. The results show that chemical nonequilibrium does occur at these conditions, and, except near the wake axis, it is due mainly to diffusive rather than convective effects. The species N and O are identified as the principal contributors to these effects.

Nomenclature

A	= reference area for wake drag
C_i, C_j	= mass fraction of i th, j th species
ΔC_{i2}	= composition deviation for i th species; Eq. (9)
C_D	= drag coefficient, $2D/A (\rho_\infty) u_\infty^2$
D	= total drag
D/Dt	= convective derivative
E_{chem}	= chemical energy in mixture, $\sum_{i=1}^{n+m} E_i C_i$
E_i	= chemical energy for i th species
ΔE	= energy deviation; Eq. (13)
G_i	= w_i/ρ
h	= enthalpy
H	= total enthalpy, $h + u^2/2$
I.N.I.	= incipient nonequilibrium index, $\Delta E/h$
n	= number, independent species in mixture
m	= number, independent conservation laws relating species
p	= pressure
p_c	= $u_1 (dp_1/dx)$; Eq. (10)
Pr	= Prandtl number
q_i	= species diffusional flux velocity
\bar{R}	= far wake radius
Re	= Reynolds number
Sc	= Schmidt number
t_c, t_f	= chemical and fluid mechanic reference times, respectively
T	= temperature
\dot{T}_D, \dot{T}_{CD}	= see Eq. (10)
u, v	= axial, radial velocities in wake
u_D	= velocity defect in far wake, $u_\infty - u$
w_i	= species mass generation rates due to chemical reactions
x, y	= axial, radial coordinates in wake
\bar{Y}	= transformed radial coordinate; Eq. (15)
ϵ	= expansion parameter t_c/t_f
Θ	= transformed wake radius; Eq. (17)
μ	= viscosity
ρ	= density

Subscripts

1, 2	= first, second terms in ϵ expansion
∞	= freestream

C, CD, D = correspond to expressions in Eq. (10)
eq = in chemical equilibrium
 i, j = i th, j th species

Introduction

AERODYNAMICS at hypersonic speeds is greatly complicated by chemical changes occurring in high-temperature air. In addition, the chemical state of air in the wake determines the electrical transmission and radiative properties of the wake, which are important means for the detection, observation, and identification of hypersonic re-entry bodies. Under many circumstances of practical interest in the re-entry problem, chemical-kinetic and fluid-mechanic rates of change in the flow are comparable. Much of current research has been directed toward justifying simplifications that make this problem of coupled chemical-kinetics and fluid-mechanics more tractable in specific situations.

The present investigation is directed toward solving this general problem for the specific case of uncontaminated air wakes behind bodies moving at hypersonic speeds. This type of flow is characterized by the presence of both convective and diffusive effects, but the latter (as in boundary layers) occur only along constant pressure surfaces. An approximation procedure, which is based on expanding the coupled fluid-mechanic and chemical-kinetic equations in powers of a small aerothermochemical parameter, is used to describe these types of flows. Deviations from the chemical equilibrium model are due both to the rapidity of convection of the fluid particles along the wake streamlines and to the interdiffusion of the reacting species across the streamlines, each of which dominates in various parts of the wake.

Investigations of wake fluid mechanics (e.g., Ref. 1) show clearly that the description of wakes is completely different for laminar and turbulent flows. As a re-entry body penetrates into the atmosphere, transition from laminar to turbulent wake flow will occur at a certain point, and the chemical processes in the flow will pass through a complete spectrum from chemically frozen flow to instantaneous chemical equilibrium flow. At the two ends of the spectrum, therefore, the wake flow will be laminar and frozen, and turbulent and in equilibrium, respectively. The expansion procedure of the present investigation, which attempts to justify and improve the chemical equilibrium description of reacting flows, is therefore particularly suitable for investigating properties of turbulent wakes. Although body shape has important bearing on the development of turbulent hypersonic wakes near the body, the wake behavior far downstream can be adequately described without reference to body shape. The present investigation utilizes the turbulent far wake as a basis of estimating chemical nonequilibrium effects in hypersonic wakes.

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Method of Analysis

For a mixture comprised of $n + m$ species with m independent conservation laws relating the species concentrations to each other (e.g., over-all mass conservation, charge conservation in a neutral plasma, etc.), there will be n independent species conservation laws of the following type:

$$\frac{DC_i}{Dt} + \frac{1}{\rho} \operatorname{div}(\rho C_i \mathbf{q}_i) = G_i(p, T, C_j \dots) \quad (1)$$

where

$$G_i = w_i/\rho$$

In addition to Eq. (1), the usual fluid-mechanic conservation laws for mass, momentum, and energy, the equation of state, and other auxiliary equations relating thermodynamic and transport properties are necessary completely to formulate the general problem. However, Eq. (1) is particularly significant in the analysis due to the appearance of the chemical-kinetic mass-generation-rate terms (right-hand side).

For "fast" chemical reactions it is possible to treat the general problem in terms of an expansion procedure.² The expansion parameter ϵ is the ratio of an appropriate chemical time t_c (which is based on the sensitivity of the chemical mass-generation-rate functions to changes in composition) and a fluid-mechanic reference time t_f . As a result of the expansion procedure, incipient nonequilibrium coefficients can be defined for a reacting mixture, which can be used to predict nonequilibrium effects both qualitatively and quantitatively from properties of the flow solution based on instantaneous local chemical equilibrium. Such solutions are comparatively easy to obtain for wakes and other hypersonic aerodynamic problems of current interest. In what follows, the results of Ref. 2 will be used in conjunction with equilibrium wake solutions. The salient features of the expansion procedure, particularly the application of incipient nonequilibrium coefficients to wakes, will first be reviewed.

Equation (1), which has units of inverse time, can be written in nondimensional form by the use of a flow time (for the convective term), diffusion time (for the diffusive term), and chemical time (for the reaction term) to normalize the equations. These reference times must be appropriately chosen for the application that is being considered. Then, the expansion parameter, $\epsilon \equiv t_c/t_f$, will appear explicitly in the equation as follows:

$$\epsilon \left\{ \frac{DC_i}{Dt} + \frac{1}{ReSc} \frac{1}{\rho} \operatorname{div}(\rho C_i \mathbf{q}_i) \right\} = G_i(p, T, C_j \dots) \quad (2)$$

If now all dependent variables are expanded in powers of ϵ and terms grouped according to powers of ϵ in all the pertinent equations, the over-all mass, momentum, and energy conservation equations for the leading terms are unchanged (since the parameter ϵ does not appear in these equations explicitly), but species conservation Eq. (2) becomes

$$G_i(p_1, T_1, C_{j1} \dots) = 0 \quad (3)$$

where subscript 1 denotes the leading terms in the ϵ expansion. Equation (3) forms an algebraic system which can be solved for the first-order composition

$$C_{i1} = C_{ieq}(p_1, T_1) \quad (4)$$

without reference to any fluid mechanic problem, thereby decoupling the chemical problem from the fluid mechanics. This justifies use of the chemical equilibrium composition for these types of problems. The effect of the flow process on composition can be ascertained by considering the next term in the expansion of Eq. (2). Specializing now to axisymmetric steady wakes, the species conservation equations take on the simplified form

$$u \frac{\partial C_i}{\partial x} + v \frac{\partial C_i}{\partial y} - \frac{1}{y\rho} \frac{\partial}{\partial y} \left(\frac{\mu y}{Sc} \frac{\partial C_i}{\partial y} \right) = G_i(p, T, C_j \dots) \quad (5)$$

The expansion procedure just described can now be applied to (5). The first step, which is independent of the fluid-mechanic problem, results in Eqs. (3) and (4). The quantities G_i in Eq. (3) are considered specified functions of the dependent thermodynamic (two variables) and chemical (n variables) quantities, so that Eq. (3) must be considered a mathematical relation between dependent variables rather than a physical statement implying vanishing reaction rates. Thus, the artifice of describing G_i as an indeterminate quantity which approaches a limit as chemical equilibrium is approached is not necessary in this formulation. Instead, the next step in the expansion of (5) indicates clearly that the chemical processes that do take place in equilibrium flow are due to second-order perturbations of the equilibrium thermochemical state; namely,

$$u_1 \frac{\partial C_{i1}}{\partial x} + v_1 \frac{\partial C_{i1}}{\partial y} - \frac{1}{y\rho_1} \frac{\partial}{\partial y} \left(\frac{\mu_1 y}{Sc} \frac{\partial C_{i1}}{\partial y} \right) = \sum_{j=1}^n \left(\frac{\partial G_i}{\partial C_j} \right)_{p, T, C_k} C_{j2} + \left(\frac{\partial G_i}{\partial p} \right)_{C_j, T} p_2 + \left(\frac{\partial G_i}{\partial T} \right)_{C_j, p} T_2 \quad (6)$$

The foregoing equation forms a system of n linear algebraic equations for the unknown second-order terms; formulation of the second-order problem should be completed by considering corresponding expansions of the remaining conservation equations. Before proceeding further, it may be observed that the coefficients on the right-hand side of (6) are not all independent. Equation (4) can be substituted into (3) and the resulting identity differentiated with respect to p_1 and T_1 separately; e.g.,

$$\frac{\partial}{\partial p_1} \left\{ \frac{G_i}{\rho} [p_1, T_1, C_{ieq}(p_1, T_1)] \right\} \Big|_{T_1} = \left(\frac{G_i}{\rho} \right)_{C_j, T} + \sum_{j=1}^n \left(\frac{\partial G_i}{\partial C_j} \right)_{p, T, C_k} \frac{\partial C_{ieq}}{\partial p_1} \Big|_{T_1} = 0 \quad (7)$$

and similarly for T_1 . Equations (4) and (7) can now be substituted into the left- and right-hand sides of (6), respectively, resulting in

$$\frac{\partial C_{ieq}}{\partial p} \Big|_T \dot{p}_c + \frac{\partial C_{ieq}}{\partial T} \Big|_p \dot{T}_{cD} + \frac{\partial^2 C_{ieq}}{\partial T^2} \Big|_p \dot{T}_D = \sum_{j=1}^n \frac{\partial G_i}{\partial C_j} \Delta C_{j2} \quad (8)$$

where the quantities

$$\Delta C_{j2} = C_{j2} - \frac{\partial C_{ieq}}{\partial p_1} \Big|_{T_1} p_2 - \frac{\partial C_{ieq}}{\partial T_1} \Big|_{p_1} T_2 \quad (9)$$

group all second-order terms into one "composition deviation" term, and the terms

$$\begin{aligned} \dot{p}_c &\equiv u_1 (dp_1/dx) \\ \dot{T}_{cD} &\equiv u_1 \frac{\partial T_1}{\partial x} + v_1 \frac{\partial T_1}{\partial y} - \frac{1}{y\rho_1} \frac{\partial}{\partial y} \left(\frac{\mu_1 y}{Sc} \frac{\partial T_1}{\partial y} \right) \\ \dot{T}_D &\equiv \frac{\mu_1}{Sc \rho_1} \left(\frac{\partial T_1}{\partial y} \right)^2 \end{aligned} \quad (10)$$

account for the flow effects based on the first-order (i.e., chemical-equilibrium) solution of the wake. The linear algebraic system (8) is solvable for the ΔC_{j2} , and, due to the linearity of the system, it is possible to write

$$\Delta C_{j2} = \left(\frac{\Delta C_{j2c}}{\dot{p}_c} \right) \dot{p}_c + \left(\frac{\Delta C_{j2cD}}{\dot{T}_{cD}} \right) \dot{T}_{cD} + \left(\frac{\Delta C_{j2D}}{\dot{T}_D} \right) \dot{T}_D \quad (11)$$

As pointed out in an earlier note,³ the coefficients in parentheses of (11) can be calculated once and for all without con-

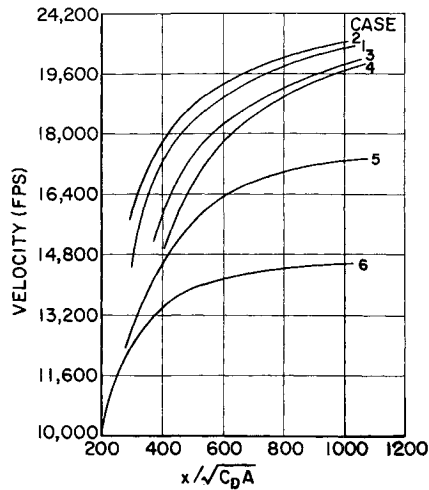


Fig. 1 Velocity along centerline of far wake.

sidering any particular flow problem. Associating each of the species with the appropriate amount of chemical energy, which is required to create it from a low-temperature reference state, incipient nonequilibrium coefficients can be defined which account for the combined effect of all the species, e.g.,

$$\Delta E_c / \dot{p}c = \sum_{i=1}^{n+m} E_i (\Delta C_{i2c} / \dot{p}c) \quad (12)$$

and similar equations apply for $\Delta E_{CD} / \dot{T}_{CD}$ and $\Delta E_D / \dot{T}_D$.

For a given equilibrium wake solution, the local value of enthalpy is known and the quantities in (10) can be calculated, so that the ratio

$$\frac{\Delta E}{h} = \frac{1}{h_1} \left(\frac{\Delta E_c}{\dot{p}c} \dot{p}c + \frac{\Delta E_{CD}}{\dot{T}_{CD}} \dot{T}_{CD} + \frac{\Delta E_D}{\dot{T}_D} \dot{T}_D \right) \quad (13)$$

can be calculated everywhere in the flow. This ratio $\Delta E/h$ can be considered the "incipient nonequilibrium index" (I.N.I.) for the flow. If I.N.I. $\ll 1.0$, the equilibrium solution is valid; if I.N.I. ≥ 1.0 , nonequilibrium effects are certainly present. This parameter is analogous to similar criteria developed by Penner,⁴ Boyer et al.,⁵ and others to determine incipient nonequilibrium effects in nozzle flows. The present criterion differs because it is the outcome of a systematic expansion procedure for the general problem of flow of reacting mixtures with both convective and diffusive effects present. Because of the linearity of the pertinent equations, the causes of deviation from equilibrium can be ascertained: the fluid-mechanic effects are seen from the magnitudes of the various terms in (10), and the principal chemical species that cause deviations from equilibrium can be seen from the magnitudes of the various terms in the equations (e.g., 12) for the incipient nonequilibrium coefficients.

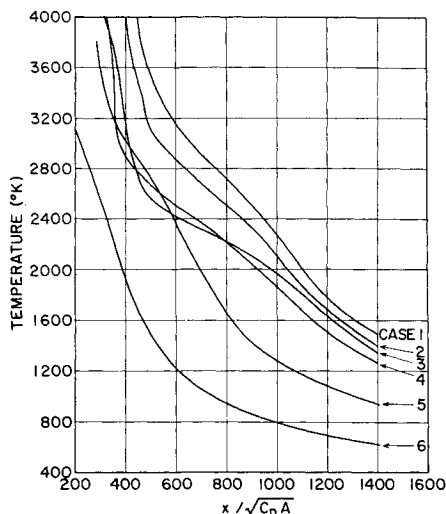


Fig. 2 Temperature along centerline of far wake.

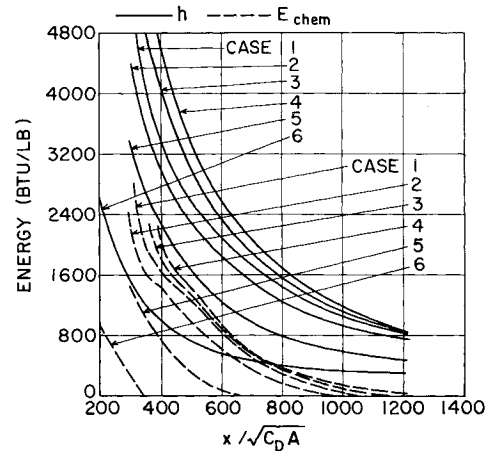
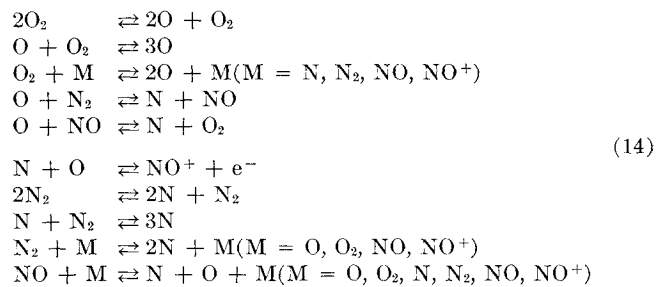


Fig. 3 Enthalpy and chemical energy along centerline in far wake.

These coefficients are functions of thermodynamic state only and have been calculated² for a seven-component (O_2 , N_2 , O , N , NO , NO^+ , e^-), ten-reaction model of high-temperature air. The ten reactions are:



Wake Analysis

As indicated in the introduction, the incipient nonequilibrium concept outlined in the foregoing can be applied to turbulent far wakes, where instantaneous chemical equilibrium can certainly be justified at some flight conditions. The calculation of the turbulent far wake is based on Long's adaptation to hypersonic wakes⁶ of the classical description¹ of incompressible turbulent axisymmetric wakes. The effects of density variations, which are important in hypersonic wakes due to the large temperature range, are accounted for by means of a Howarth-Dorodnitsin transformation of the radial coordinate

$$YdY = (\rho/\rho_\infty) ydy \quad (15)$$

Based on this simple model,[†] the analysis⁶ can be outlined as follows:

$$C_D A = 4\pi \int_0^R \frac{\rho}{\rho_\infty} \frac{u}{u_\infty} \frac{u_D}{u_\infty} ydy \quad (16)$$

where $u_D = u_\infty - u$, and R is the radius of the turbulent wake. In terms of transformation (15), the turbulent wake radius is denoted by Θ , so that integrating (15) gives

$$Y \Big|_{y=R} \equiv \Theta = R \left[2 \int_0^1 \frac{\rho_\infty}{\rho} \frac{Y}{\Theta} d \left(\frac{Y}{\Theta} \right) \right]^{-1/2} \quad (17)$$

[†] In a more elaborate analysis, Lees and Hromas⁷ account for both the effects of compressibility and the effect of ingesting a highly nonuniform inviscid flow into the turbulent wake. The latter effect is particularly important near the body, where body shape significantly affects the development of the turbulent wake,⁸ but in the present study the description of the far wake is based on the asymptotic behavior of the fluid far from the body, so that a simpler treatment⁶ is adequate (and is in essential agreement with that of Ref. 7 in this region), because it is reasonable to assume that the pressure has reached the ambient value and that the total drag is contained in the turbulent wake.

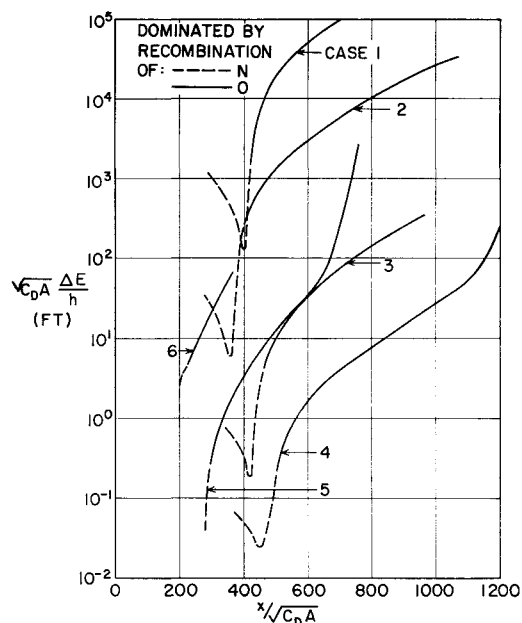


Fig. 4 Incipient nonequilibrium index along wake center-line.

It can now be assumed that the velocity profile has the same shape in terms of the dimensionless transformed variable as the corresponding incompressible wake

$$\frac{u_D}{u_\infty} = \left(\frac{u_D}{u_\infty} \right)_{\max} \left[1 - \left(\frac{Y}{\Theta} \right)^{3/2} \right]^2 \quad (18)$$

The physical justification for this assumption is that it is reasonable to assume that the turbulent exchange of momentum between adjacent streamlines follows the same mechanism for the compressible and incompressible cases based on the mass flux in the respective streamlines.

Far from the body, $u_D \ll u_\infty$, and the growth of the turbulent wake can be described as for the incompressible case

$$R = 0.6 x^{1/3} (C_D A)^{2/3} \quad (19)$$

Now it only remains to relate the density and velocity profiles in the wake by means of the energy equation

$$h + u^2/2 = H_\infty \quad (20)$$

and the equation of state for air in chemical equilibrium, which can be used to find the density $\rho = \rho(h, p_\infty)$.

This completes the system of equations necessary to describe the wake. The momentum integral can be integrated by using (15), (17), and (18) in (16):

$$C_D A = 0.5143 \pi (u_D/u_\infty)_{\max} \Theta^2 \quad (21)$$

The actual calculation of the wake can now proceed in the following sequence of steps. First, one assumes (u_D/u_∞) , from which Θ and the velocity profile can be determined via (21) and (18). (All distances are nondimensionalized by $(C_D A)^{1/2}$.) The density profile can now be determined and used to relate the transformed and physical coordinates by means of (15). The radius of the wake R can now be used to find the distance behind the body by means of (19). The description of the wake flow field is now complete, because all quantities are determined in terms of the physical coordinates x and y .

Nonequilibrium Effects

The results of the wake analysis can now be combined with the incipient nonequilibrium coefficients to estimate the effects of finite chemical rates in the wake. The terms described by Eqs. (10) must be calculated from the wake solution. In the absence of pressure gradients, the first of these terms is zero. The second term combines the effects of axial and radial convection and a term that is due to diffusion and

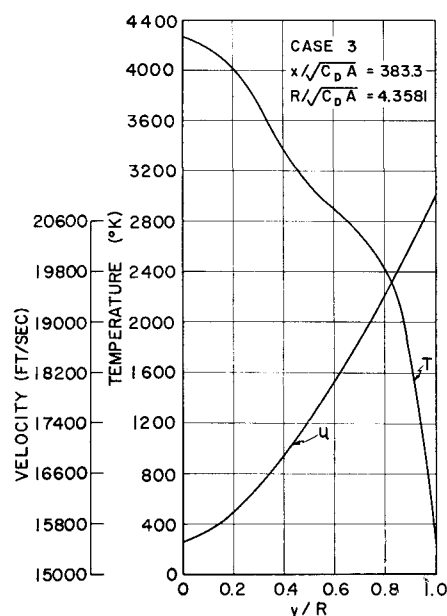


Fig. 5 Velocity and temperature profile in far wake.

is essentially proportional to the second derivative of the temperature profile. The axial convection term can be calculated directly from the known profiles. The radial velocity v must be calculated by integrating the continuity equation

$$v(x, y) = \frac{1}{\rho y} \int_0^y \frac{\partial(\rho u)}{\partial x} dy \quad (22)$$

For turbulent flow, $Sc \sim 1.0$, and the eddy viscosity from Prandtl's mixing length theory is

$$\mu = \rho(0.18R)^2 |\partial u / \partial y| \quad (23)$$

This expression is in agreement with the assumed velocity profile.¹ The third expression in (10) is due to diffusion and is proportional to the square of the temperature gradient; it can be calculated by combining some of the previously determined quantities.

Before proceeding with a presentation of numerical results, it is worthwhile to discuss the potential significance of these calculations. The expansion procedure that forms the basis of the incipient nonequilibrium analysis is not valid unless succeeding terms in the expansion are small throughout the flow field. This implies that if deviations from chemical equilibrium occurred in the flow field surrounding the re-entry

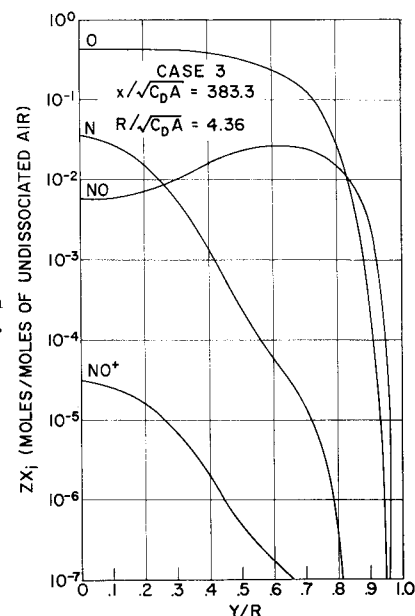


Fig. 6 Composition profile in far wake.

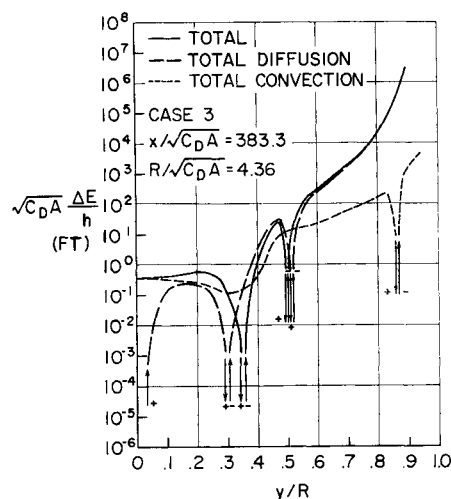
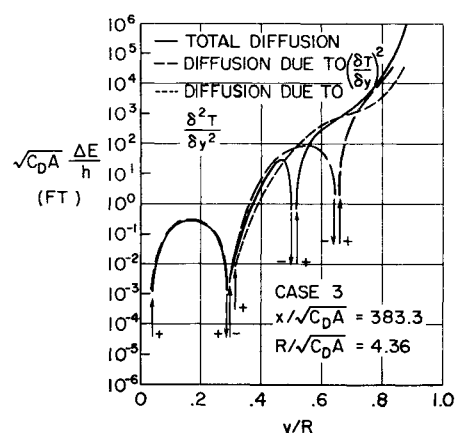
Table 1 Flight conditions considered (freestream)

Case	Alt, kft	Speed, fps	Static pressure, atm $\times 10^5$	Stagnation temp., $^{\circ}\text{K}$
1	200	21,400	23	4630
2	150	21,400	145	4885
3	100	21,000	1090	5380
4	70	21,000	4430	5710
5	70	18,000	4430	5320
6	70	15,000	4430	4680

body, application of this method to the wake could not be justified rigorously. It may be argued that the sensitivity of the chemical rate effects to pressure and temperature indicates that upstream effects play a minor role compared to local properties of the flow in bringing about nonequilibrium effects. This will tend to justify the use of incipient nonequilibrium coefficients on a local basis. On the other hand, sudden changes in flow properties around the body, particularly expansions around the base or the nose region of bodies, or incomplete relaxation behind the bow shock, may cause a lack of equilibrium in the fluid that empties into the wake despite the relatively higher pressure and temperature upstream from the wake. These considerations indicate that the present method can only give necessary, but not sufficient, conditions for the validity of instantaneous local equilibrium wake flow. Therefore the subsequent results simply indicate limits of the possible usefulness of local equilibrium wake flow. Because of the linearity of all pertinent equations, the results do permit conclusions as to the particular chemical and fluid-mechanic phenomena that are the principal causes of nonequilibrium deviations.

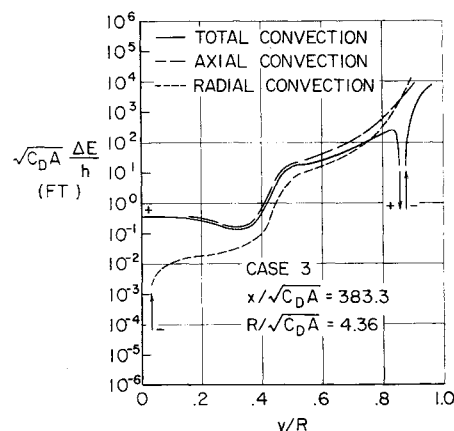
Numerical Results

The flight conditions considered are listed in Table 1; they were chosen to represent various altitudes and speeds of practical interest. Velocities, temperatures, enthalpies, and chemical energies along the centerline are given in Figs. 1, 2 and 3, respectively. The chemical energy E_{chem} is the energy absorbed by dissociation and ionization; when compared to h , it is an indicator of the potential significance of chemical effects. Figure 4 shows $\Delta E/h$, the incipient nonequilibrium index (I.N.I.), for the same six cases along the

**Fig 7 Incipient nonequilibrium index in far wake.****Fig. 8 Incipient nonequilibrium index in far wake (diffusion only).**

far wake centerline. Since the temperatures are the highest near the centerline, for a 1-ft body $[(C_D A)^{1/2} = 1.0]$, this figure indicates the existence of an equilibrium region in the far wake (where the I.N.I. < 1.0) in only three of the six cases studied. Near the body, N is the largest contributing species to the I.N.I., whereas further downstream, it is O. As the temperature drops (Fig. 2), the I.N.I. in Fig. 4 passes through a minimum for many of the cases; despite the lower rates at the lower temperatures, the tendency to deviate from equilibrium becomes smaller due to the diminished supply of N atoms that are "required" to recombine, and the terms in Eqs. (10) become smaller. However, as soon as O recombination becomes significant, the tendency to deviate from equilibrium becomes several orders of magnitude larger. One may surmise from these results that for the high-altitude condition (case 1), both the N and O recombinations will lag far behind, whereas at lower altitudes (e.g., cases 3 and 4) most of the N atoms will have recombined along the centerline; the excess atoms in the far wake will be mostly O atoms. This might have important consequences regarding electron densities, due to the increased occurrence of NO^+ ions in regions where both types of atoms are present.

Figures 5 and 6 show velocity, temperature, and composition of a particular equilibrium far-wake profile. A station $383 (C_D A)^{1/2}$ downstream from the body for case 3 was chosen as the example. Figure 7 shows the corresponding I.N.I. profile, showing also the separate effects of convective and diffusive terms. It is clear that, except at the wake center, interdiffusion of the reacting species is the dominant reason for deviating from chemical equilibrium in the far wake. This interdiffusion is due both to the curvature of the tem-

**Fig. 9 Incipient nonequilibrium index in far wake (convection only).**

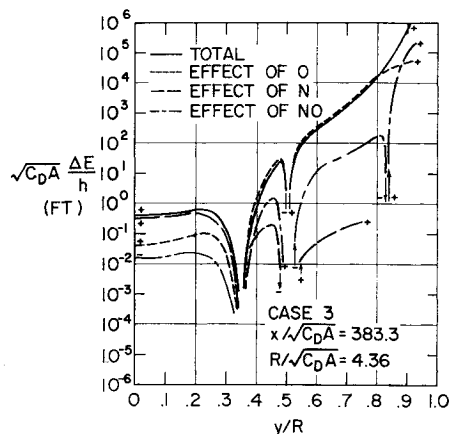


Fig. 10 Incipient nonequilibrium index in far wake (effect of species).

perature profile ($\partial^2 T / \partial y^2$) and to the curvature of the equilibrium composition vs temperature variation in conjunction with the temperature gradients ($\partial T / \partial y$), as shown in Fig. 8. [Curvatures of the equilibrium composition profiles (Fig. 6) also indicate the regions where there will be no diffusion.] The breakdown of the convective effect into axial and radial components (Fig. 9) further demonstrates the utility of the method. Figure 10 indicates contribution of the various species to the I.N.I., atomic N and O dominating near the center and in the outer region of the far wake, respectively; the contribution of NO is not significant. [Note: in the foregoing results only the absolute value of the I.N.I. is significant; the sign merely indicates whether the dominant reaction that is lagging behind is energy releasing (+) or energy absorbing (-).]

Figure 11 compares contours of $|\Delta E/h| = 1.0$ in the far wake for three different cases. Comparison of cases 3 and 4 shows the effect of altitude, whereas comparison of cases 4 and 5 can ascertain the effect of flight speed at the same altitude. As expected, low altitudes and high speeds (and the ensuing high temperatures) will extend the regions of equilibrium flow both laterally and axially. The effect of body size on the contours for one of the cases is also shown, indicating that the equilibrium flow regions will vary with re-entry body size and shape.

Summary

Incipient nonequilibrium coefficients were found to be a practical and versatile research tool in estimating the effects

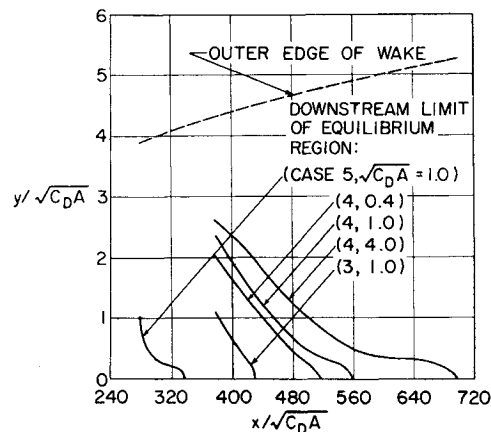


Fig. 11 Equilibrium regions in far wake.

of finite rate chemical kinetics in turbulent hypersonic wake flows. It was possible to account for both convective and diffusive effects in a multicomponent reacting mixture. The results indicate the limits of the possible usefulness of hypersonic wake calculations based on local chemical equilibrium.

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