THERMAL DIFFUSION EFFECTS IN MASS TRANSFER*

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Abstract-Recent interest in the influence of thermodynamic coupling on mixture boundary layers has prompted the presentation of exact solutions with and without such coupling. Results are presented for stagnation point injection of helium and Freon-13 into an airstream layer. Significant heat transfer rate and recovery temperature effects are discussed in terms of the sign of the thermal diffusion ratio which formed the basis for the injectant choices.

NOMENCLATURE

- mass fraction, *i*th species, ρ_i/ρ ; $c_i,$
- specific heat, constant pressure; c_n
- coefficient of mass diffusion; D_{12}
- D_T coefficient of thermal diffusion;
- modified stream function, $\psi/\sqrt{2s}$; f,

$$
= (2-\beta)^{1/2} \frac{(\rho v)_w}{(\rho u)_o} \sqrt{\left[\frac{(\rho u)_o x}{\mu_o}\right]};
$$

- heat-transfer coefficient ; h_{-}
- thermal diffusion ratio, D_{12}/D_T (from kr . Section 14.71, 2nd Ed., [2]);
- molecular weight ratio, m_i/m_j ; M_{ii}
- $Pr,$ Prandtl number, $\mu c_p/k$;
- heat flux per unit time; $q,$
- R_{\cdot} gas constant;

Reynolds number, $\frac{\rho u x}{u}$; Re.

- *(** Schmidt number, $\mu/\rho D_{12}$; $Sc.$
- Stanton number, $h/(\rho u c_n)$; $St.$
- T. temperature.

Greek symbols

 f_{w}

- β , pressure gradient similarity parameter, 0.5 for hemisphere stagnation point:
- γ , ratio of specific heats;
- η , similarity parameter;
 θ , Sutherland constant;
- Sutherland constant:
- $\rho\mu/(\rho\mu)_o = [(\mu R_2)/(\mu_2 R)][(1 +$ λ. θ)/ $(\bar{T} + \theta)$] $\bar{T}^{1/2}$;
- μ , coefficient of viscosity;
- *P,* density.

Subscripts

- i, ith species;
- 1, injected fluid ;
- \mathfrak{o} either no injection or external edge (stagnation) condition;
- $W_{\rm S}$ condition at surface, $\eta = 0$;
- aw, condition at insulated surface;
- $($)', differentiation with respect to η ;
- \bigcap quantity in units of external stream value, $(\)/(\)$ _o.

1. INTRODUCTION

THERMODYNAMIC coupling is essentially the simultaneous transport of a macroscopic fluid property by virtue of two or more physical property gradients. The concern here will be the interaction of energy and mass transport phenomena.

The vast majority of mixture boundary-layer analyses omit such coupling phenomena in view of the implied complications and the negligible effects thought to be involved. Recent evidence [l] suggests that such phenomena may be of some importance in the interpretation of coolant behavior primarily due to evaluations which are generally conducted under conditions far different from those of free, high-speed, flight. As a consequence, several investigations into thermal diffusion effects by means of exact solutions have been completed and may be of interest. The

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object here is to present the results in relative detail and to indicate the precise effect upon heattransfer rate and recovery temperature.

So-called thermal diffusion and diffusiontherm0 effects relate specifically to the coupling between concentration and temperature gradients. such that each contributes to a mass and energy flux. The latter may both be present in an "equilibrium" body of fluid, their relative magnitudes depending upon the thermal diffusion ratio, k_T , which is the ratio of the phenomenological transport coefficients associated with each gradient. For a gas, this ratio derives from a consideration of the particle description of the fluid, e.g. the molecular weight, interparticle force field behavior, size and geometry. There is available from kinetic theory [2] adequate theoretical specifications of the magnitude of k_T and there exists experimental evidence 131 for corrections to the idealized particle descriptions used in analysis.

The classical description of heat transfer in compressible fluids involves a specification of a rate and a temperature "potential". Thermodynamic coupling affects both of these but to different extents. The potential uses as a reference the adiabatic surface temperature defined by a balance between conduction (temperature gradient) and diffusion-thermo (concentration gradient) effects [4]. For negligible k_T , i.e. the ratio of thermal to mass diffusion coefficients. the resulting Fourier's law defines the usual reference temperature corresponding to a vanishing temperature gradient at the surface. For finite (although small) k_T a rough assessment of the change **in** reference temperature has been shown to be possible but not extremely accurate [I]. However. the change does prove to be appreciable. Depending then upon the accompanying effects upon the heat-transfer coefficient a choice of "coolant" in high-speed applications does involve a consideration of *kr.*

An earlier study of transpiration cooling with helium-air boundary layers has explained some differences between experiment and analysis on the basis of thermal diffusion [I]. Later experiments [5, 6. 7] have supplied additional confirmation. Whereas larger thermal capacities tend to reduce adiabatic surface temperatures. thermal diffusion (specifically negative k_T),

increases such temperatures to levels higher than without the injection process. A **reversal** of the k_T sign favors a reduction but is usually associated with material of small thermal capacity. Since only helium-air solutions (with $k_T \neq 0$) were available heretofore some additional computations have been completed for a coolant with $k_T > 0$ (Freon-13).

The specific model chosen for study was the hemispherical stagnation point in a stream of stagnation temperature equal to $1360^{\circ}R$. This conforms to a Mach number 8.0 flow considered for a related experimental investigation.

2. **PROBLEM DESCRIPTION AND METHOD**

The detailed mathematical formulation will not be explicitly presented since it is available elsewhere [4, 81. Baron and Scott [X] considered the possible similarity solutions \vith pressure gradients, omitting k_T terms. The results to be discussed use the same description with the inclusion of thermodynamic terms as derived for boundary layers **in** [4]. As demonstrated in $[1]$ a complete description of the thermal diffusion ratio is necessary and no effort was made here towards simplification by either the introduction of a constant thermal diffusion factor in place of k_T , or by assuming Prandtl and Schmidt numbers to be constant.

Similarity solutions were completed for $\beta =$ 0.5 in the terminology of $[8]$, i.e. the nondimensional pressure gradient parameter appropriate to the stagnation region on a hemisphere. Five surface temperatures were considered including the adiabatic and four levels relative to the stream stagnation (i.e. boundary layer external edge) temperature $(1.0, 0.8, 0.6 \text{ and } 0.4)$. Injectants were chosen on the basis of anticipated sign effects of thermodynamic coupling. although it was realized other parameters were involved. Fig. 1 indicates the variation of k_T with mass concentration for Freon-13 and helium mixing with air. Both mixtures exhibit peak (absolute) \alucs of about the same magn.itude and **skewed** distributions such that the peaks occur for approximately a I :3 proportion by mass for the components. The signs, however. differ and indicate that helium tends to migrate to warmer regions while Freon-13 tends toward cooler regions.

FIG. 1. Thermal diffusion ratio dependence upon mass concentration of injectant. Helium-air, $k_T \leq 0,$ ---; Freon-air, $k_T > 0,$ - - - -.

Although the k_T expressions are cumbersome the numerical procedure for the boundary layer solutions remains the same as in their absence. One assumes that at the surface there is no slip, a known injection rate and temperature (or adiabatic restriction on the temperature gradient), and the required coupling between surface concentration and its gradient to ensure zero net flow of the external stream gas into the surface. In order to integrate away from the surface additional assumptions must be made for the velocity, concentration and temperature gradients (or adiabatic temperature) and are then modified repeatedly until proper conditions result far from the surface.

An appendix summarizes the system of relations that were actually employed for this study (see also $[4]$, $[8]$). A summary of surface values is given in Tables 1 and 2.

3. RESULTS AND DISCUSSION

Recovery temperature

In the absence of thermodynamic coupling

the recovery temperature at a stagnation point is simply the total temperature of the oncoming stream. Departures from such levels were shown in [I] to be both expected and measured for mixture layers. Fig. 2 illustrates the dependence

FIG. 2. Recovery temperature at stagnation point on hemisphere as a function of injection rate. Helium-Air, $O = 1360^{\circ}R$, $\Box = 570^{\circ}R$; Freon-air, $O = 1360^{\circ}R$. (For $k_T = 0$, $T_{av}/T_0 = 1.0$.)

of *Taw* upon injection rate, temperature level, and the sign of k_T . Helium injection into air $(k_T < 0)$ results in appreciable increases in T_{aw} above T_o , the more so with increasing total temperature level. This conforms to a conclusion [9] that tunnel test conditions generally yield lower recovery factors than free flight for low density injectants on a $k_T \equiv 0$ basis. Fig. 2 also shows that the injection of Freon $(k_T > 0)$ resulted in depressed *Taw* values but to a relatively much lesser degree.

It is interesting to consider simultaneously the concentrations achieved by the injection process as shown in Fig. 3. Peak values of $|k_T|$ occur at approximately $c_{1_w} \approx 0.18, 0.70$ for helium and Freon respectively (Fig. 1). These correspond to $f_w \approx -0.1$, -0.65 from

Table 1. Summary of surface values (helium injection, $\beta = 0.5$, $T_o = 1360^\circ R$)

f_w	T_{w}	f_w "	c_{1w}	T_{w}	S_{I} \vee Re	Pr_{w}	Sc_v	$k_{T\,\rm u}$
θ	$*1.0$	0.9227	$\mathbf{0}$	Ω	θ	0.6870	0.2381	Θ
-0.2	$*1.0$	2 1229	0.2521	θ		0.6070	0.6478	θ
-0.2	0.8	1.8000	0.2512	0.0585	0.3460	0.5900	0.6439	$\{\}$
-0.2	0.4	1.1427	0.2512	0.1355	0.3477	0.5534	0.6524	()
-0.4	$*1.0$	3.8591	0.6626	θ		0.6648	$1 - 2957$	Θ
-0.4	0.8	3.2151	0.6646	0.0465	0.2273	0.6476	1.2910	0
-0.4	0.4	1.8876	0.6793	0.1046	0.2250	0.6106	1.3423	$^{()}$
-0.6	$*1.0$	4.4969	0.9215	θ	$\mathcal{L}_{\mathrm{eff}}$	0.7278	1.6948	$\langle \rangle$
-0.6	0.8	3.6909	0.9243	0.0298	0.1290	0.7098	1.6876	Ω
-0.6	0 ₄	2.0296	0.9383	0.0664	0.1269	0.6711	1.7508	0
-0.01	$*1.0044$	0.9677	0 0076	0.0027		0.6843	0.2461	-0.0098
-0.01	$1-0$	0.9651	0.0076	-0.0007	0.5355	0.6841	0.2461	-0.0098
-0.2	$*1.1050$	2.3012	0.2555	0.0387	\sim \sim	0.6153	0.6551	-0.0763
-0.2	$1-0$	2.1201	0.2526	-0.0020	0.3661	0.6070	0.6485	-0.0765
-0.2	0.8	1.7799	0.2471	-0.0591	0.3261	0.5901	0.6374	0.0768
-0.2	0.6	1.4303	0.2386	$+0.1049$	0.3210	0.5726	0.6240	-0.0772
-0.2	0.4	1.0813	0.2274	$+0.1354$	0.3190	0.5549	0.6133	0.0776
-0.4	$*1.2048$	4.5368	0.6668	-0.0352	~ 10	0.6821	1.3138	0.0330
-0.4	$1-0$	3.8605	0.6631	-0.0183	0.1970	0.6649	1.2965	0.0334
-0.4	0.8	3.1890	0.6583	0.0607	0.1956	0.6463	$1 - 2813$	-0.0339
-0.4	0.6	2.5095	0.6525	0.0932	0.1962	0.6257	$1 - 2732$	-0.0345
-0.4	0.4	1.8178	0.6467	0.1104	0.1925	0.6037	$1-2904$	-0.0352
-0.6	$*1.2698$	5.5977	0.9222	0.0129		0.7504	$1 - 7187$	-0.0070
-0.6	$1-0$	4.5045	0.9212	$+0.0289$	0.111	0.7277	1.6944	-0.0071
-0.6	0.8	3.6920	0.9221	0.0528	0.1096	0.7093	1.6842	-0.0070
-0.6	0.6	2.8621	0.9238	0.0686	0.1067	0.6892	1.6880	0.0068
-0.6	0.4	2.0162	0.9284	0.0764	0.1042	0.6686	1.7352	0.0064
-0.8	$*1.3087$	5.3154	0.9887	-0.0026		0.7713	1.8263	0.0010
-0.8	$1-0$	4.1229	0.9892	-0.0195	0.0523	0.7455	1.7984	-0.0009
-0.8	0.8	3.3411	0.9898	0.0299	0.0513	0.7267	1.7868	-0.0009
-0.8	0.6	2.5498	0.9908	0.0364	0.0498	0.7060	1.7897	-0.0008
-0.8	0.4	1.7461	0.9926	0.0383	0.0475	0.6846	1.8359	-0.0006
$\mathbf 0$	0.9	0.8708	$\mathbf{0}$	0.0453	0.5534	0.6830	0.2340	Ω
θ	0.8	0.8139	θ	0.0877	0.5514	0.6803	0.2340	θ
θ	0.6	0.6996	$\bf{0}$	0.1637	0.5465	0.6830	0.2340	0
θ	0.4	0.5864	θ	0.2287	0.5402	0.7065	0.2340	θ

* Denotes adiabatic surface case.

Fig. 3 and thus to 3-4 per cent changes in T_{aw} from T_0 for either injectant on Fig. 2.

In view of this Fig. 4 has been prepared to illustrate the variation of T_{aw}/T_o with $\vert k_T \vert_w$, although it is realized that one should not anticipate a sole dependence of the T_{aw} effect upon surface values. It is suggested that an evaluation of the effect corresponding to (k_T) $(k_T)_{w, \text{max}}$ for the given mixture defines the coupling effect on T_{aw} over the rather larger range of injection rates. Despite a need for additional confirmation of the "universality" of Fig. 4 it should serve as a reasonable approximation for other mixtures at this time.

Heat transfer

Despite interest in recovery temperature the major concern remains with the heat-transfer rate. The classical concept of a temperature potential—or driving "force" is of value when heat transfer proves to be proportional to some ΔT . Modern development and needs result in variable proportionality "constants" (heat-transfer coefficients) and also, as we have seen,

* Denotes adiabatic surface case.

variable reference temperature levels depending respectively. When $T_0 < T_w < T_{aw}$, q/q_0 is upon injection rate and mixture constituents. It negative. Allowing for k_T , at a fixed rate of upon injection rate and mixture constituents. It negative. Allowing for k_T , at a fixed rate of thus is proper to dispense with heat-transfer injection q is reduced with increasing rates of thus is proper to dispense with heat-transfer injection *q* is reduced with increasing rates of coefficient and T_{aw} concepts and first present heat transfer $(T_w/T_o \ll 1)$ in the helium case coefficient and T_{aw} concepts and first present heat transfer $(T_w/T_o \ll 1)$ in the helium case actual heat-transfer rates (Figs. 5, 6) for the while the Freon case behaves oppositely. In the actual heat-transfer rates (Figs. 5, 6) for the while the Freon case behaves oppositely. In the mixtures considered. Regardless of the coolant absence of thermal diffusion $(k_T \equiv 0)$ the mixtures considered. Regardless of the coolant absence of thermal diffusion $(k_T \equiv 0)$ the choice q decreases markedly with increasing surface temperature level has little effect but choice q decreases markedly with increasing surface temperature level has little effect but injection for the surface temperature levels does tend to agree more closely with very cold reported. For each injection rate there exists wall results for $k_T \neq 0$. In essence this amounts a T_w such that $q/q_0 = 1.0$. Lesser and greater to a small diffusion-thermo effect relative to the a T_w such that $q/q_0 = 1.0$. Lesser and greater to a small diffusion-thermo effect relative to the injection rates for such T_w loci imply $q/q_0 \ge 1.0$ conductive (Fourier) effect for very large rates injection rates for such T_w loci imply $q/q_o \geq 1.0$

does tend to agree more closely with very cold wall results for $k_T \neq 0$. In essence this amounts

FIG. 3. Injectant mass concentration at surface as a function of injection rate. \bigcirc = Helium-air, \bigcirc = Freonair. Temperature and k_T effects negligible.

FIG. 4. Recovery temperature dependence on (k_T) relative to maximum k_T effects. Helium-air, \odot para s 1360°R, \Box 0.570°R; Freon-air, \Box 1360°R.

FIG. 5. Heat-transfer dependence on injection rate for helium into air. $T_w/T_o = 0.8$ (\triangle), 0.6 (\Box), 0.4 (\Diamond). Open symbol, $k_T \neq 0$; solid symbol, $k_T \equiv 0$.

of heat transfer, as has been suggested earlier [I].

Although Figs. 5 and 6 indicate larger proportionate reductions in q for helium than for Freon (if $T_w/T_a \ll 1$) the benefits appear exaggerated when comparison is made in terms of nondimensional heat-transfer coefficients, i.e. Stanton numbers. For $k_T \equiv 0$ the ordinates in Figs. 5 and 6 may be interpreted immediately as St/St_0 . When $k_T \neq 0$, T_{aw} variations shown in Fig. 2 imply the variations shown in Fig. 7. Of note are the rather smaller Stanton numbers for helium, and the relative independence of such coefficients from a surface temperature dependence. In fact such $St = St(f_w)$ curves are in fairly good agreement with the variations obtained upon neglecting k_T . This is shown explicitly in Fig. 8, on a rather large scale, to emphasize the $St = St(k_T)$ influence.

FIG. 6. Heat-transfer dependence on injection rate for Freon into air. $T_w/T_0 = 0.8$ (\triangle), 0.6 (\Box), 0.4 (\Diamond). Open symbol, $k_T \neq 0$; solid symbol, $k_T = 0$.

4. SUMMARY AND CONCLUSIONS

Coupling arising from thermal diffusion and diffusion-therm0 terms introduced into the injection boundary-layer formulation result in measurable differences in both recovery temperatures and heating rates. Of these, the most dramatic is the large increase in stagnation point temperature under zero heat-transfer conditions in the case of helium injection into air. For the unique case of a stagnation point the classical independence of recovery temperature from the influence of surface blowing or mixture properties has permitted a check on the sign reversal of the incremental temperature effect due to a corresponding sign reversal for the thermal diffusion ratio. The results show that negative and positive k_T imply increases and reductions, respectively, of the equilibrium surface temperature.

A consequence of the recovery temperature dependence upon k_T is the possible misleading comparison of Stanton numbers for given coolants at specified injection rates. Helium appears quite superior to Freon-13 whereas, in fact, a comparison of the actual heat-transfer rates (q) strongly depends upon the surface

FIG. **7.** Stanton number dependence upon injection rate, $k_T \neq 0$. Helium-air, unflagged; Freon-air, flagged. $T_w \neq T_{aw}$ (\bigcirc). $T_w = 0.4 T_o$ (\Diamond).

temperature. For a very cold bcundary the departures of *Taw* from the stagnation temperature value are of lesser importance and the helium benefits prevail.

Provisionally, it appears possible to estimate the coupling effect on recovery temperature on the basis of results obtained with the maximum possible value of the thermal diffusion ratio established at the surface.

Fig. 8. Explicit thermal diffusion effect on Stanton number as a function of injection. Helium-air, unfiagged : Freon--air, flagged. $T_w/T_o = 0.8$ (\wedge), 0.4 (\wedge).

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The descriptive relations are of the similarity $[\overline{RT}k_T/c_1c_2]'$ $[\overline{RT}k_T/c_1c_2]'$

$$
(\lambda f^{\prime\prime})^{\prime} + ff^{\prime\prime} - \beta (f^{\prime 2} - \overline{R}\overline{T}) = 0
$$

$$
c_1^{\prime\prime} + Ac_1^{\prime} + Bk_T = 0
$$

$$
a\overline{T}^{\prime\prime} + b\overline{T}^{\prime} + ck_T + d = 0
$$

and which represent, respectively, momentum, injectant species, and energy conservation. The coefficients are explicitly

$$
A = (Sc/\lambda)(f + [\lambda/Sc]^{\prime}) + (M_{12}RT^{\prime}/T)
$$

\n
$$
\left(2[M_{21} - 1]k_T + \bar{R} \frac{\partial k_T}{\partial c_1}\right)
$$

\n
$$
B = M_{12}\bar{R}^{2}Sc/\lambda)(\lambda \bar{T}^{\prime}/Sc\bar{T})^{\prime}
$$

\n
$$
a = \bar{c}_p \lambda/Pr
$$

\n
$$
b = \bar{c}_p f + (\bar{c}_p \lambda/Pr)^{\prime} + (\lambda \bar{c}_p^{\prime}/Sc)
$$

\n
$$
c = \bar{R} \left\{ \bar{c}_{p2}(c_{p12} - 1)M_{12}\bar{R}\lambda \bar{T}^{\prime 2}/Sc\bar{T} + \frac{\gamma_o - 1}{\gamma_o} \right\}
$$

\n
$$
\frac{\bar{T}}{\bar{c}_1\bar{c}_2} [(\lambda/Sc)(c_1^{\prime} + M_{12}\bar{R}^{2}\bar{T}^{\prime}k_T/\bar{T})]^{\prime} \right\}
$$

APPENDIX
relations are of the similarity

$$
d = [(\gamma_o - 1)\lambda/\gamma_o Sc][c'_1 + M_{12}\overline{R}^2T'k_T/T]
$$

$$
[\overline{R}\overline{T}k_T/c_1c_2]
$$

and the *B, c,* and *d* terms represent the coupling effects. *f* is the familiar modified stream function and all parameters are functions of the similarity variable η defined in an $(x, y) \rightarrow (s, \eta)$ transformation by

$$
\eta = \frac{(\rho u)_o}{\sqrt{2s}} \int_0^y \frac{\rho}{\rho_o} \, \mathrm{d}y
$$

$$
s = \int_0^x (\rho \mu u)_o \, \mathrm{d}x.
$$

The imposed boundary conditions were, at the surface $(\eta = 0)$:

$$
f' = 0, f = \text{constant}, c_1' + (M_{12}\overline{R}^2\overline{T}^{\prime}/\overline{T})k_{\mathcal{V}} = (c_2fSc/\lambda)
$$

and either $\bar{T} = \text{constant or}(\bar{T}'/T)_{aw} = -(\gamma_o - 1)$ γ_0)($\sqrt{RPr/c_1} \lambda \bar{c}_p$) k_T and at the outer edge of the layer $(\eta \rightarrow \infty)$: $f'=\overline{T}=1, c_1=0$.

Résumé—L'intérêt que l'on porte actuellement à l'influence de l'interaction thermodynamique sur le mélange des couches-limites a accéléré la présentation de solutions exactes, compte tenu ou non d'une telle interaction. Les resultats sent presentes pour une injection d'helium et de Freon 13 au point d'arret d'un ecoulement. Les effets du taux d'echange thermique et de la temperature parietale sont etudies en fonction du signe du rapport de diffusion thermique, qui determine les choix du fluide inject&

Zusammenfassung-Das gegenwärtige Interesse für den Einfluss der thermodynamischen Kopplung auf Gemischgrenzschichten veranlasste die Ermittlung exakter Losungen mit und ohne eine derartige Kopplung. Ergebnisse werden gebracht ftir Staupunktseinblasung von Helium und Frigen 13 in eine Luftschicht. Kennzeichnende Einflüsse des Wärmeübergangs und der Rückgewinntemperatur werden im Hinblick auf das Vorzeichen des thermischen Diffusionsverhältnisses, das die Grundlage zur Auswahl der Einblasmedien bildete, diskutiert.

Аннотация—Возникший в последнее время интерес к влиянию термодинамического взаимодействия на пограничные слои, состоящие из смесей газов, способствовал нахождению точных решений для случаев при наличии и при отсутствии такого взаимолействия. Результаты этого решения представлены для случая подачи гелия и фреона-13 в слой воздуха в критической точке. Значительная интенсивность теплообмена и эффекты температуры восстановления находятся в зависимости от знака отношения термодиф-**@y3"", IiOTOpOe FIBJIReTCH OCIIOBOt AJIH BbI60pa BeII(eCTB AJIH l3JQyBa.**