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RESEARCH MEMORANDUM

CORRELATION OF TURBULENT HEAT TRANSFER IN A TUBE FOR THE DISSOCIATING SYSTEM $N_2O_4 \rightleftharpoons 2NO_2$

By Richard S. Brokaw

Lewis Flight Propulsion Laboratory
Cleveland, Ohio

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SUMMARY

Experimental data on turbulent heat transfer in a tube for the dissociating system $N_2O_4 \rightleftharpoons 2NO_2$ are in accord with the usual Nusselt-Prandtl-Reynolds number correlation for this type of convective heat transfer.

The thermal conductivities and kinematic viscosities needed to formulate the Nusselt, Reynolds, and Prandtl numbers have been computed using rigorous expressions from the kinetic theory of gases. The thermal conductivity includes a large contribution arising from the diffusional transport of chemical enthalpy.

Three methods of selecting average property values for the boundary layer were tested. All gave good correlation using the equilibrium transport properties.

An approximate method using frozen gas properties gave somewhat poorer results, but should be useful for complex systems where exact computations would be prohibitively tedious. This approximate method should also be applicable to systems in which chemical equilibrium is not attained, provided the enthalpy of the gases in the free stream and at the wall is known.

INTRODUCTION

Currently there is considerable interest in heat transfer in dissociating gases, because such gases may exist in the exhaust nozzles of jet engines or within the boundary layer on hypersonic aircraft. In reacting gases, heat transport may be considerably higher than in "frozen" (nonreacting) mixtures. Large amounts of heat may be carried as chemical enthalpy of molecules which diffuse because of concentration gradients. These gradients arise because the gas composition varies with temperature.

For example, in a gas which absorbs heat by dissociating as the temperature is raised, heat is transferred when a molecule dissociates in the high-temperature region and diffuses toward the low-temperature region (there is a lower concentration of dissociated molecules at low temperature). In the low-temperature region the gas reassociates, releasing the heat absorbed from the high-temperature dissociation.

Heat transfer in practical systems with chemical reaction has been theoretically examined (e.g., refs. 1 and 2). Parallel theoretical studies of the thermal conductivity of reacting gases have been made: in reference 3 a general expression for the thermal conductivity of gas mixtures in chemical equilibrium is developed, while in reference 4 systems with finite reaction rates are considered.

Recently Beal and Lyerly (ref. 5) have experimentally investigated turbulent heat transfer in a tube for the dissociating system $N_2O_4 \rightleftarrows 2NO_2$. Essential features of their apparatus included a 3/4-inch-diameter test section with a rounded entrance. Heat was added at a thermally guarded hot spot 23 diameters from the tube entry. The guard heater was short, so that the length-diameter ratio for the thermal boundary layer was only 2. Tests were carried out with air in the Reynolds number range from 2500 to 34,000 and with $N_2O_4 \rightleftarrows 2NO_2$ mixtures in the range from 18,000 to 104,000.

Beal and Lyerly were unable to analyze their results fully, because of the dearth of experimental conductivity and viscosity data for the $N_2O_4 \rightleftarrows 2NO_2$ system in the literature. This report shows that the data of Beal and Lyerly are in complete accord with the usual Nusselt-Prandtl-Reynolds number correlations for this type of convective heat transfer. Thermal conductivities and viscosities for the $N_2O_4 \rightleftarrows 2NO_2$ system (needed to formulate the Nusselt, Prandtl, and Reynolds numbers) have been computed using rigorous kinetic theory expressions.

SYMBOLS

The following symbols are used in this report:

A dimensionless constant, eq. (1)

c_p specific heat at constant pressure

D binary diffusion coefficient

d tube diameter

G mass velocity

Δh specific enthalpy difference between wall and fluid bulk

h heat-transfer coefficient

k thermal conductivity

Nu Nusselt number, hd/k

Pr Prandtl number, $c_p\mu/k$

Re Reynolds number, Gd/μ or $Gd/\rho\nu$

T absolute temperature

δ $\rho D c_{p,f}/k_f$ (dimensionless)

μ absolute viscosity

ν kinematic viscosity

ρ density

φ generalized temperature dependent property

Subscripts:

B bulk fluid

e chemical equilibrium

f chemically frozen

i contribution due to internal energy

m value for mixture of monatomic gases

r contribution due to chemical reaction

w wall

Superscripts:

$-$ mean value

$*$ evaluated at reference condition

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METHODS OF ANALYSIS

Transport Properties

The computed thermal conductivities, kinematic viscosities, and Prandtl numbers for the $N_2O_4 \rightleftarrows 2NO_2$ system at 1 atmosphere are shown in figure 1. The methods of computation are detailed in the appendix. Calculated conductivities agree with recent experimental determinations (ref. 6), which are also plotted in figure 1. Frozen thermal conductivities and Prandtl numbers are shown by the dashed lines. (The frozen values refer to equilibrium gas composition without chemical reaction.) The horizontal bars across the top of the figure indicate the temperature differences between the wall and bulk fluid for the individual $N_2O_4 \rightleftarrows 2NO_2$ heat-transfer tests of Beal and Lyerly; approximate Reynolds numbers are given adjacent to the bars. (The exact values of the Reynolds numbers depend on the way mean property values are chosen.) Over the temperature range of the heat-transfer experiments, the conductivity varies by a factor of 2.2, the kinematic viscosity by a factor of 2.3, and the Prandtl number by a factor of 1.4. These variations are much larger than those encountered with nonreacting gases; over a comparable temperature range, conductivity might increase by 10 to 15 percent and kinematic viscosity by 30 to 40 percent, while Prandtl number would remain substantially constant. Accordingly, for the $N_2O_4 \rightleftarrows 2NO_2$ system, suitable mean property values for correlation must be selected.

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Selection of Mean Property Values

In order to formulate the Reynolds, Nusselt, and Prandtl numbers, mean values of viscosity, thermal conductivity, and Prandtl number are required. In actual fact, a mean value of kinematic viscosity is required in the Reynolds number, which may be written $Gd/\bar{\mu} = Gd/\rho_B\bar{\nu}$; hence,

$$\bar{\mu} = \rho_B\bar{\nu}$$

Mean properties obtained by the following three methods were tested:

(1) Temperature-integrated properties. If a property is designated as φ, then the integrated average value is

$$\bar{\varphi} = \frac{\int_{T_B}^{T_w} \varphi(T)\,dT}{T_w - T_B}$$

This type of average suggests itself naturally for thermal conductivity because it is the proper average for heat transfer in the absence of convection (e.g., thermal-conductivity cells).

(2) Reference-temperature properties: Properties are taken at a reference temperature, as suggested in reference 7.

$$T^* = T_B + 0.58\ (T_w - T_B)$$

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(3) Reference-enthalpy properties: Properties are evaluated at a temperature corresponding to a reference enthalpy h^*, defined as

$$h^* = h_B + 0.58\ (h_w - h_B)$$

COMPARISON OF CORRELATIONS

Correlations Using Equilibrium Transport Properties

All three methods of averaging properties give good correlation of the air and $N_2O_4 \rightleftharpoons 2NO_2$ heat-transfer data of Beal and Lyerly. Figure 2 shows a log-log plot of $Nu/Pr^{1/3}$ against Reynolds number for temperature-integrated average properties. A mean kinematic viscosity is required in formulating the Reynolds number. If a mean value of the absolute viscosity is used, correlation is very poor, with errors of up to fifty percent in the Reynolds number. This is shown in figure 3, a reference-enthalpy equilibrium-property correlation, in which a mean absolute viscosity was used. The experimental data for each of the three correlations using mean kinematic viscosity were fitted by an equation of the form

$$Nu/Pr^{1/3} = ARe^{0.79} \tag{1}$$

or

$$\log Nu - \frac{1}{3} \log Pr = \log A + 0.79 \log Re \tag{1a}$$

For each correlation, the constant A was chosen to give a least-squares fit of all the data (air and N_2O_4) at Reynolds numbers greater than 9000 to equation (1a). (The air points at $Re < 9000$ were excluded because they lie in the transition region.) Results of the correlations are summarized in the first three columns of table I[1].

[1]It might be noted that in the Reynolds number range under consideration (9000 to 110,000) equation (1), with the A values of table I, agrees to within 4 percent with Colburn's correlation (ref. 8): $Nu/Pr^{1/3} = 0.023\ Re^{0.8}$. This is somewhat surprising, in view of the short length-diameter ratio for the thermal boundary layer in Beal and Lyerly's apparatus.

From table I it is seen that all three correlations represent the $N_2O_4 \rightleftharpoons 2NO_2$ data very well. (The standard deviations for the air data and over-all correlations merely indicate the consistency of the correlations.) For engineering heat-transfer calculations the reference-enthalpy or reference-temperature methods would ordinarily be used, because with these methods, transport properties need be calculated at only one condition.

However, table I shows that the integrated property correlation is slightly better. This is merely a tentative conclusion, however, since the differences between the correlations are largely the result of a single data point (albeit the point with the largest temperature difference). More experimental measurements with larger temperature differences are required to firmly establish the most reliable method for computing mean properties. In any event, for all three correlation methods, deviations for N_2O_4 and air data points are similar. This fact strongly supports the notion that heat-transfer correlations for nonreacting systems may be applied to dissociating systems, provided that transport properties are properly computed and suitable mean values are chosen. 4760

Approximate Correlation Using Frozen Transport Properties

In reference 3 it is shown that the equilibrium and frozen thermal conductivities may be related to the equilibrium and frozen specific heats by the equation

$$\frac{k_e}{k_f} = 1 + \delta\left(\frac{c_{p,e}}{c_{p,f}} - 1\right)$$

If the dimensionless group δ is assumed to be unity, then $k_e \cong k_f c_{p,e}/c_{p,f}$. (Aerodynamicists sometimes refer to δ as the "Lewis number." However, this usage is not generally accepted.) Hence, for a system with a finite temperature difference, it seems reasonable to define an approximate mean thermal conductivity by

$$\bar{k}_e \cong \left(\frac{\bar{k}_f}{\bar{c}_{p,f}}\right) \frac{\int_{T_B}^{T_w} c_{p,e}\,dT}{T_w - T_B} = \bar{k}_f \frac{\Delta h}{\bar{c}_{p,f}\Delta T} \tag{2}$$

with the frozen specific heat and conductivity evaluated at a reference temperature. Subject to this approximation, the Nusselt and Prandtl numbers are

$$Nu_e \cong Nu_f \frac{\bar{c}_{p,f}\Delta T}{\Delta h}$$

$$Pr_e \cong Pr_f$$

A reference-temperature correlation using frozen properties is shown in figure 4. (If the correction term $\bar{c}_{p,f}\Delta T/\Delta h$ were not applied to the frozen Nusselt number, Nusselt numbers for the $N_2O_4 \rightleftharpoons 2NO_2$ system would be eight- to ninefold too high.) As expected, the correlation of figure 4 is poorer than the correlation of figure 2; this is reflected in a higher standard deviation, shown in the fourth column of table I. In reality the deviations are systematic with δ and might amount to 10 percent or more under some conditions. For this system δ actually ranges from 1.13 at 310° K to 0.64 at 375° K. The frozen property correlation can be improved by introducing a correction term involving δ and plotting

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$$\left(Nu_f/Pr_f^{1/3}\right)\left(\bar{c}_{p,f}\Delta T/\Delta h\right)\left[1 + (\delta - 1)\left(1 - \frac{\bar{c}_{p,f}\Delta T}{\Delta h}\right)\right]^{-2/3}$$

against Reynolds number. The correction term is reminiscent of similar corrections used in treatments of hypersonic stagnation-point heat transfer (refs. 2 and 9). However, heat transfer in multicomponent mixtures cannot, in general, be characterized by a single diffusion coefficient, and thus the correction term is not generally useful[2].

The approximate correlation without the correction should be applicable to many complex systems where an exact computation would be prohibitively tedious (e.g., jet-engine and rocket exhausts). Indeed, it should be applicable to systems in which chemical equilibrium is not attained, provided the chemical compositions (and hence, enthalpy) of the gases in the free stream and at the wall are known.

CONCLUSIONS

Experimental data for turbulent heat transfer in a tube have been analyzed and correlated. The following conclusions may be drawn:

1. Experimental heat-transfer data in a tube for the dissociating system $N_2O_4 \rightleftharpoons 2NO_2$ correlate well with air data using transport properties computed on the basis of chemical equilibrium.

2. In computing Reynolds number a mean kinematic viscosity is required.

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[2]For multicomponent mixtures δ may be defined as $[(k_e/k_f) - 1]/[(c_{p,e}/c_{p,f}) - 1]$, and the correction term is valid. This is not useful, however, since the equilibrium properties calculated to obtain δ might better be applied directly in the correlations of the preceding section.

3. The best correlation was obtained using temperature-integrated properties. Reference-temperature and reference-enthalpy properties were almost as good.

4. Errors are somewhat larger for an approximate correlation which uses frozen properties. In this method the frozen Nusselt number is corrected by the factor $\bar{c}_{p,f}\Delta T/\Delta h$ and it is assumed that $\rho D c_{p,f}/k_f = 1$. (Mean value of the specific heat at constant pressure and chemically frozen conditions, $\bar{c}_{p,f}$; temperature difference between wall and fluid bulk, ΔT; specific enthalpy difference between wall and fluid bulk, Δh; density, ρ; binary diffusion coefficient, D; specific heat at constant pressure and chemically frozen conditions, $c_{p,f}$; chemically frozen thermal conductivity, k_f.)

Lewis Flight Propulsion Laboratory
National Advisory Committee for Aeronautics
Cleveland, Ohio, December 6, 1957

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APPENDIX - CALCULATION OF EQUILIBRIUM PROPERTIES

FOR THE $N_2O_4 \rightleftharpoons 2NO_2$ SYSTEM

Transport-property calculations were carried out assuming a Lennard-Jones (6-12) potential for the molecular interactions (ref. 10). Gas compositions were obtained from the equilibrium data of Bodenstein (ref. 11). The means by which various properties were computed are as follows:

Force constants. - In order to compute the transport properties, force constants σ and ε/k characteristic of molecular interactions must be estimated. For the N_2O_4 molecule, the force constants were estimated from critical temperature and density (ref. 10, eqs. (4.1-17) and (4.1-18)). The critical temperature and density for pure N_2O_4 were in turn estimated from the boiling point and density of liquid N_2O_4 by the methods of references 12 and 13. (It was assumed that the boiling point and liquid density of $N_2O_4 \rightleftharpoons 2NO_2$ equilibrium mixtures is close to the value which would be expected for pure N_2O_4.) Force constants chosen for N_2O_4 were: $\sigma = 4.74$ Å and $\varepsilon/k = 383^\circ$ K.

The force constants for the NO_2 molecule were assumed to be similar to those of CO_2 and N_2O (ref. 10, p. 1111); values used were $\sigma = 3.9$ Å and $\varepsilon/k = 230^\circ$ K.

For the N_2O_4-NO_2 interaction, σ was taken as the arithmetic mean of NO_2 and N_2O_4 values; ε/k was taken as the geometric mean (ref. 10, eqs. (8.4-8) and (8.4-9)).

These force constants differ somewhat from those used in reference 3; they are believed to be more realistic estimates.

Viscosities. - Viscosities were calculated by equation (8.2-22) of reference 10.

Thermal conductivities. - The thermal conductivity of a reacting gas mixture may be broken into three portions:

$$k_e = k_m + k_i + k_r \tag{3}$$

The first term on the right side of equation (3) represents collisional kinetic energy transport; it was computed using equation (8.2-36) of reference 10. The second term (in the nature of an Eucken correction) represents a diffusional transport of internal energy (rotation and vibration) and was calculated using equation (75) of reference 14. The third

and dominant term for these dissociating mixtures is the increase in conductivity due to chemical reaction. It was computed from equation (14) of reference 3. The frozen thermal conductivity is simply the first two terms of equation (3).

Diffusion coefficients. - Diffusion coefficients were computed according to equation (8.2-44) of reference 10.

Specific heats. - The specific heat may be broken up into a frozen part and a contribution due to chemical reaction. The frozen specific heats were computed from the known vibrational frequencies of the NO_2 and N_2O_4 molecules. The contribution due to chemical reaction was computed according to reference 3.

Heat of reaction. - The heat of reaction at 298.16° K was computed from the heats of formation of the NO_2 and N_2O_4 molecules presented in reference 15. Values at other temperatures were obtained by correcting the 298.16° K value for the integrated heat capacity difference between products and reactant.

Calculated values of viscosity, density divided by pressure, frozen and equilibrium thermal conductivity, and frozen and equilibrium specific heat are contained in table II. Values were computed at 10° intervals from 290° to 490° K and for pressures of 1.0, 0.74, and 0.33 atmospheres. (Additional thermal conductivities computed for pressures of 0.5, 0.2, 0.1, 0.05, and 0.02 atmospheres may be found in reference 6.) Only the 1.0-atmosphere values were used in the correlations of this report.

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TABLE I. - COMPARISON OF HEAT-TRANSFER CORRELATIONS

	Integrated properties	Reference-temperature properties	Reference-enthalpy properties	Reference-temperature frozen properties, $\delta = 1$
A (eq. 1(a))	0.0261	0.0262	0.0257	0.0262
Standard deviation				
$N_2O_4 \rightleftharpoons 2NO_2$ data	2.4 percent	3.3 percent	3.5 percent	5.3 percent
Air (Re > 9000)	3.0 percent	3.1 percent	3.2 percent	----------
Over-all (Re > 9000)	2.9 percent	3.1 percent	3.3 percent	----------

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TABLE II. - COMPUTED PROPERTIES FOR $N_2O_4 \rightleftharpoons 2NO_2$ EQUILIBRIUM

(a) Pressure, 1 atmosphere.

Temperature, °K	Viscosity, poises	Equilibrium thermal conductivity, cal/(cm)(sec)(°K)	Frozen thermal conductivity, cal/(cm)(sec)(°K)	Density + pressure, gm/(cm³)(atm)	Equilibrium specific heat, cal/gm	Frozen specific heat, cal/gm
290	1.124×10^{-4}	20.083×10^{-5}	3.110×10^{-5}	3.414×10^{-3}	1.0168	0.2017
300	1.192	25.668	3.337	3.130	1.2973	.2042
310	1.270	31.176	3.587	2.832	1.6051	.2060
320	1.356	35.233	3.860	2.540	1.8730	.2075
330	1.445	36.366	4.143	2.272	2.0067	.2083
340	1.534	33.977	4.427	2.043	1.9395	.2089
350	1.615	29.086	4.693	1.864	1.6893	.2094
360	1.689	23.308	4.938	1.726	1.3483	.2099
370	1.754	18.037	5.163	1.621	1.0158	.2106
380	1.811	14.087	5.366	1.542	.7556	.2115
390	1.865	11.262	5.561	1.479	.5650	.2125
400	1.915	9.478	5.745	1.428	.4415	.2138
410	1.961	8.375	5.923	1.385	.3622	.2151
420	2.005	7.742	6.098	1.346	.3134	.2166
430	2.048	7.397	6.272	1.312	.2831	.2181
440	2.091	7.219	6.446	1.280	.2634	.2196
450	2.133	7.143	6.617	1.250	.2503	.2211
460	2.174	7.163	6.791	1.221	.2429	.2226
470	2.215	7.223	6.958	1.195	.2382	.2240
480	2.254	7.315	7.127	1.169	.2353	.2255
490	2.294	7.436	7.300	1.145	.2341	.2270

(b) Pressure, 0.74 atmosphere.

290	1.134×10^{-4}	22.332×10^{-5}	3.136×10^{-5}	3.352×10^{-3}	1.1407	0.2015
300	1.206	28.201	3.373	3.053	1.4500	.2039
310	1.288	33.498	3.633	2.745	1.7685	.2056
320	1.377	36.578	3.916	2.449	2.0051	.2069
330	1.468	36.073	4.203	2.189	2.0543	.2076
340	1.554	32.025	4.482	1.974	1.8776	.2081
350	1.633	26.197	4.740	1.811	1.5482	.2086
360	1.702	20.304	4.975	1.688	1.1803	.2092
370	1.763	15.542	5.188	1.596	.8677	.2101
380	1.817	12.201	5.384	1.526	.6410	.2111
390	1.870	9.939	5.573	1.469	.4841	.2123
400	1.918	8.576	5.753	1.422	.3866	.2136
410	1.963	7.769	5.929	1.380	.3258	.2150
420	2.006	7.331	6.102	1.343	.2890	.2164
430	2.049	7.114	6.275	1.310	.2666	.2180
440	2.092	7.024	6.448	1.278	.2522	.2196
450	2.133	7.008	6.618	1.249	.2427	.2210
460	2.175	7.069	6.792	1.221	.2377	.2226
470	2.215	7.154	6.959	1.194	.2344	.2240
480	2.254	7.267	7.128	1.169	.2328	.2255
490	2.294	7.401	7.300	1.145	.2322	.2270

(c) Pressure, 0.33 atmosphere.

290	1.166×10^{-4}	29.106×10^{-5}	3.221×10^{-5}	3.152×10^{-3}	1.5476	0.2010
300	1.250	34.829	3.488	2.819	1.9064	.2030
310	1.342	37.844	3.776	2.497	2.1643	.2043
320	1.435	36.481	4.072	2.216	2.1774	.2051
330	1.523	31.189	4.352	1.994	1.9129	.2057
340	1.600	24.217	4.608	1.828	1.4900	.2063
350	1.667	18.109	4.835	1.709	1.0850	.2070
360	1.727	13.554	5.042	1.621	.7694	.2080
370	1.780	10.557	5.234	1.554	.5566	.2092
380	1.828	8.752	5.414	1.499	.4248	.2105
390	1.877	7.663	5.593	1.452	.3422	.2118
400	1.922	7.078	5.766	1.411	.2943	.2133
410	1.966	6.781	5.937	1.373	.2658	.2148
420	2.009	6.667	6.108	1.339	.2494	.2163
430	2.051	6.657	6.279	1.307	.2398	.2179
440	2.093	6.711	6.451	1.276	.2342	.2195
450	2.134	6.796	6.620	1.247	.2307	.2210
460	2.175	6.918	6.794	1.220	.2293	.2225
470	2.215	7.048	6.960	1.194	.2287	.2239
480	2.255	7.191	7.129	1.169	.2287	.2254
490	2.294	7.346	7.301	1.145	.2293	.2270

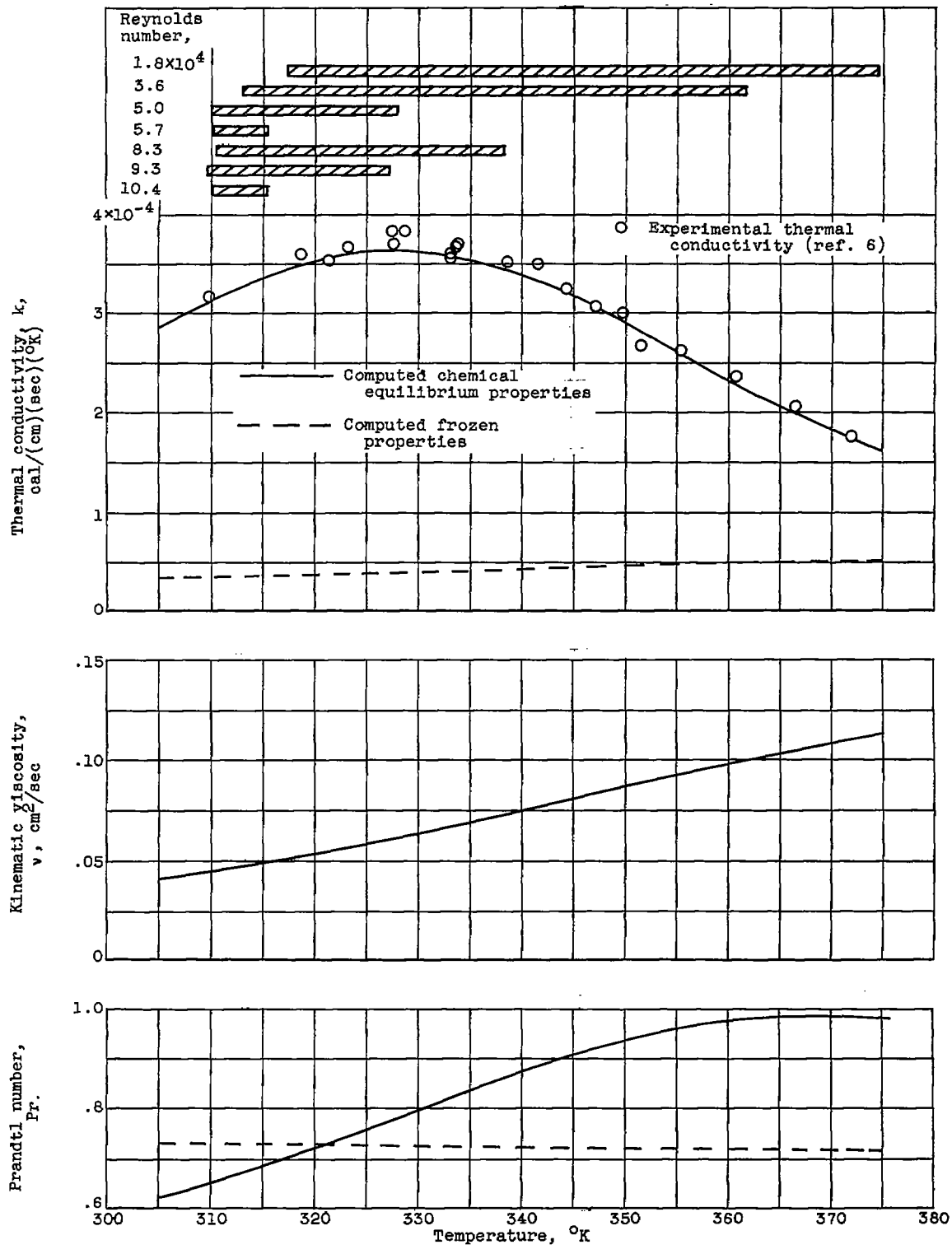

Figure 1. - Equilibrium transport properties for $N_2O_4 \rightleftharpoons 2NO_2$ system at one atmosphere.

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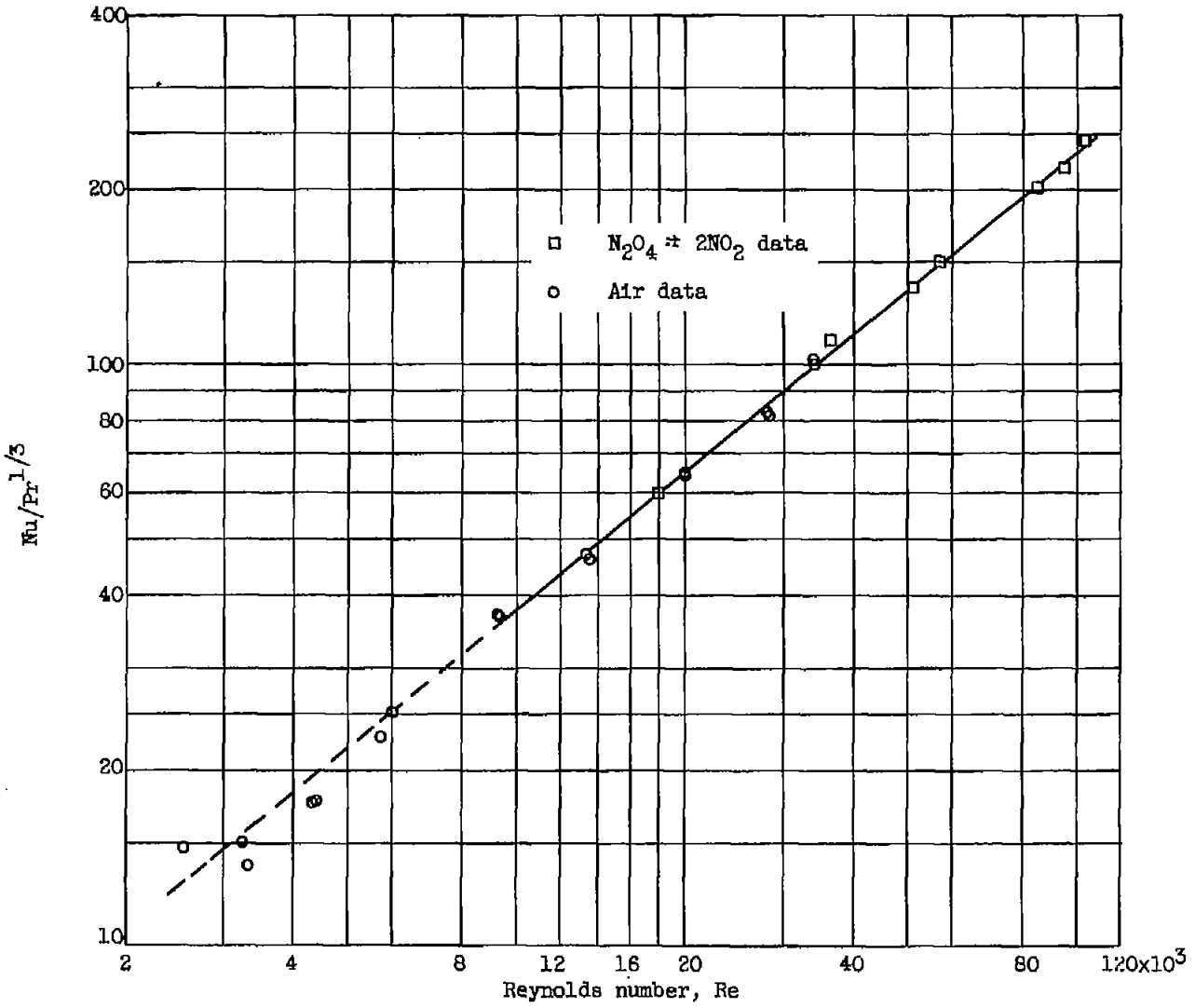

Figure 2. - Correlation of data with temperature-integrated equilibrium properties. (Mean kinematic viscosity used to formulate Reynolds number.)

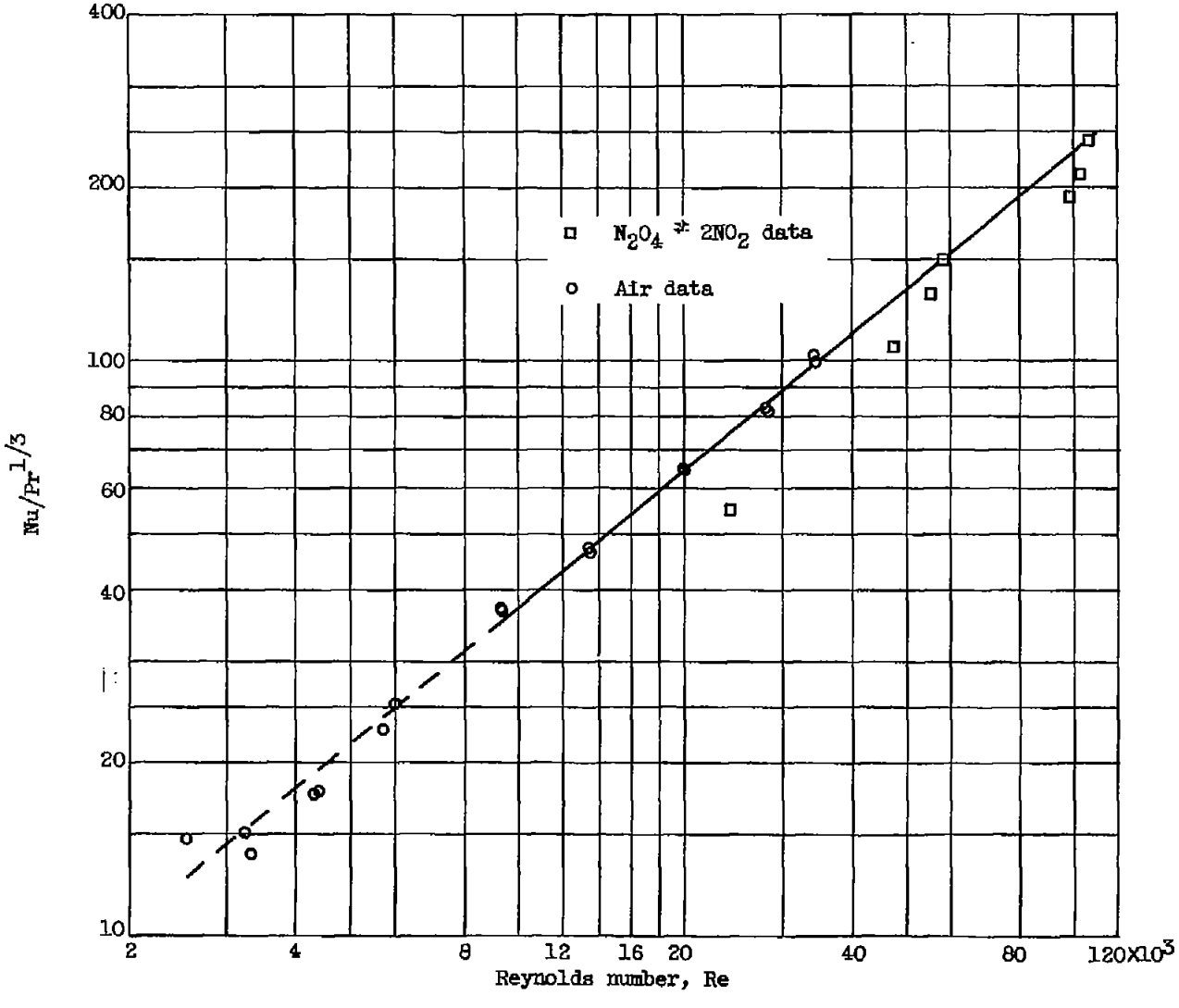

Figure 3. - Correlation of data with reference-enthalpy equilibrium properties. (Mean absolute viscosity used to formulate Reynolds number.)

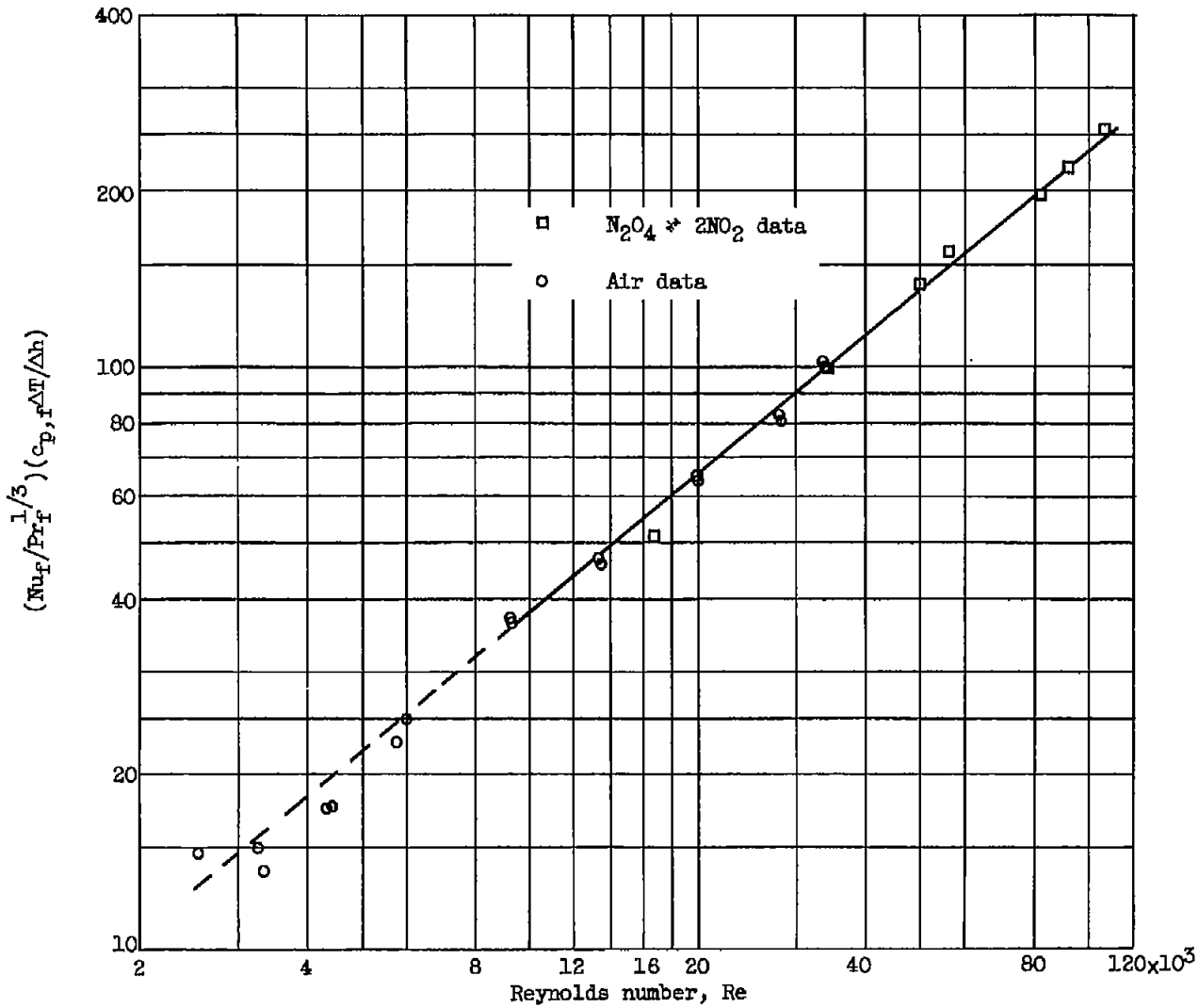

Figure 4. - Approximate correlation of data with reference-temperature frozen properties.

NACA - Langley Field, Va.