Table III. Properties of Trinitromethane Compounds

	$(NO_2)_3CH$	$(NO_2)_3CF$	$(NO_2)_3CCl$	$(NO_2)_3CBr$
Density, g/ml	1.59 (24° C)	1.595 (20° C)[a]	1.66 (20° C)	2.034 (20° C)
Melting point, ° C	+27–28	–29	+5.7	+18
Heat of vaporization, kcal per mole	13.1 (25°)	8.171 (25°)	10.86 (25° C)	11.41 (25° C)
Enthalpy of formation, kcal per mole	-16.25 ± 0.75	-51.99 ± 0.48	-5.57 ± 0.18	$+3.22 \pm 0.24$
Estimated halogen-carbon bond energy, kcal	. . .	10.35	73.2	52.63

[a] Through oversight densities of trinitrofluoromethane are reported incorrectly in *(11)*. The first digits (.7 and .6) in the density series should read 5 and 4—i.e., 1.595 instead of 1.795; 1.489 instead of 1.689.

NOMENCLATURE

m = mass of (′)compound, (″)auxiliary material, (‴)polyethylene film, and (⁗)fuse, grams

n = number of moles of (′)compound, (″)auxiliary material, (‴)polyethylene film, and (⁗)fuse

n^i = initial number of moles of water in bomb

t_i = initial temperature of calorimeter, ° C

t_f = final temperature of calorimeter, ° C

t_h = reference temperature to which combustion reaction is referred (25° C)

Δt_{corr} = rise in temperature of calorimeter due to thermal leakage of outer jacket

Δt_c = $t_f - t_i + \Delta t_{corr}$

ξ(calor.) = energy equivalent of calorimeter, cal per deg

ξ(cont.) = energy equivalent of contents, cal per deg [ξ^i (cont.)$(t_i - 25)$ + ξ^f (cont.)$(25 - t_f + \Delta t_{corr})$ where ξ^i (cont.) and ξ^f (cont.) are energy equivalents of contents before and after combustion, respectively]

ΔE_{ign} = energy released due to ignition, cal

ΔE_{corr} st. states = energy reduction to standard states, cal *(5, 7)*

ΔE_{dec} (HNO_3) = energy released from formation of nitric acid, cal

ΔE_{dec} (As_2O_5) = energy released from formation of As_2O_5, cal

ΔE_c° = standard energy of idealized combustion reaction, calories per mole

ΔH_c = heat of reaction between 1 mole of diethyl phthalate + 1 mole of bromotrinitromethane, oxygen, and water as given in Equation 1 at constant pressure, kcal

ΔE_c = energy of reaction between 1 mole of diethyl phthalate + 1 mole of bromotrinitromethane, oxygen, and water as given in Equation 1 at constant volume kcal

ΔH_f° = enthalpy of formation of bromotrinitromethane, kcal per mole

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Natural Convection Heat Transfer from a Horizontal Cylinder to a Reacting Gas

$N_2O_4 \rightleftarrows 2NO_2$ System

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The transfer of heat between a solid surface and a fluid may be significantly affected by the occurrence of a homogeneous chemical reaction in the fluid and/or a heterogeneous reaction on the surface of the solid. The general problem of transport in reacting systems is discussed by Brokaw *(3)*, Brokaw and Butler *(4)*, and Rosner *(8)*. In a reacting medium, temperature gradients induce concentration gradients, causing diffusion of the reacting species. In this diffusional process, the enthalpy of each species is transported, making a major contribution to the thermal conductivity, and thus to heat transfer when the enthalpy of reaction is large.

Values of the equilibrium thermal conductivity of the N_2O_4 system, which is a nonmonotonic function of temperature, are presented by Svehla and Brokaw *(9)*. The rate of dissociation of N_2O_4 is so large that for all subsonic-flow regimes at pressures above 0.1 atm the system is effectively at equilibrium and thus the equilibrium proper-

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Natural convection heat-transfer rates from horizontal electrically heated cylinders were measured in the equilibrium, homogeneously reacting gaseous $N_2O_4 \rightleftarrows 2NO_2$ system from 0.1 to 1.0 atm and at a bulk temperature of approximately 25° C. The maximum cylinder surface temperature was 150° C. Values of the Nusselt number ranged from about 0.5 to 1.5, corresponding to Grashof numbers of about 10^{-2} to 10^2. These Grashof number values are below those for the laminar flow regime. Because of the marked sensitivity of properties to temperature in reacting systems, ways of handling properties appearing in the Nusselt and Grashof numbers had to be developed in order to obtain a single empirical correlation between the Nusselt number and the Grashof number that was applicable to both reacting or nonreacting systems. Heat-transfer measurements on nonreacting nitrogen and air were made over the same range of Nusselt and Grashof numbers for comparison with the N_2O_4 data.

ties tabulated by Svehla and Brokaw may be utilized in treating experimental data. Because of the existence of molecular weight gradients when heat is transferred to reacting systems, density gradients and thus buoyancy forces in natural convection heat transfer can be markedly affected, causing velocity, temperature, and concentration distributions to be interrelated. Thus, a numerical or analytical solution of the equations of change under these conditions becomes extremely complicated, if not impossible, and experimentation must be resorted to.

There have been few experimental studies in the field of natural convection heat transfer in reacting systems since the work of Langmuir (*6*). Beer (*1*) and Beer and Deubel (*2*) measured natural convection heat-transfer rates in the N_2O_4 system for a few Grashof numbers in the range 10^2 to 10^5. The bulk and surface temperatures were similar to those employed in this study. Experimental data were compared with relationships obtained from laminar boundary layer theory instead of with correlations obtained with other experimental data on nonreacting systems.

Applied problems where natural convection from small horizontal cylinders is of importance occur in chemical regeneration of metal surfaces such as incandescent light filaments, discussed by Tower (*11*) and Fast (*5*); and manufacture of metal fibers, such as boron, which are used as composite structures in the aerospace industry, described by Talley and Lasday (*10*). In the first application, a light bulb is filled with a gaseous halide compound of tungsten which dissociates on the electrically heated tungsten filament to regenerate tungsten. In the second case, a mixture of gaseous boron trichloride and hydrogen reacts on an electrically heated substrate such as tungsten to deposit solid boron on the substrate. In both cases, the reaction takes place at steady state in the presence of free-convection motion.

Since the Prandtl number in reacting gases is relatively constant at about 0.7, similar to nonreacting gases (cf. Svehla), correlations may be obtained directly between the Nusselt number and the Grashof number without the Prandtl number appearing. There is, however, a complication in correlating data in reacting systems due to the fact that all physical properties are sensitive to temperature, particularly thermal properties such as thermal conductivity. Density and viscosity are unusual, in that they are affected not only by temperature but by the change in molecular weight with temperature peculiar to reacting systems. Thus the main problem in obtaining a good engineering correlation is finding the appropriate way of handling the variable properties. In reacting gaseous systems the buoyancy driving force in the Grashof number is expressed correctly as $(\rho_\infty/\rho_w - 1)$ and not as $(T_w/T_\infty - 1)$. These terms are not identical even for a perfect reacting gas where

$$\left(\frac{\rho_\infty}{\rho_w} = \frac{T_w}{T_\infty}\,\frac{M_\infty}{M_w}\right)$$

since the molecular weight varies with temperature.

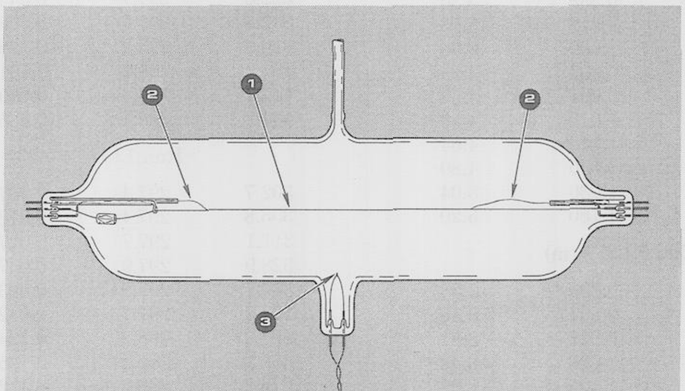

Figure 1. Natural convection heat transfer cell

1. Electrically heated cylinder
2. Voltage taps
3. Thermocouple for bulk temperature measurement

Table I. Natural Convection Heat Transfer to N_2O_4

T_w, °K	T_∞, °K	q, Watts/Cm^2	N_{Nu}	N_{Gr}
Run 1 (D = 0.0127 Cm, 1 Atm)				
307.7	299.0	0.554	0.647	0.0861
322.6	299.0	1.875	0.721	0.204
322.9	299.4	1.871	0.719	0.201
325.5	299.2	2.528	0.854	0.219
346.7	299.4	4.114	0.749	0.288
355.6	299.5	4.764	0.751	0.296
366.4	299.9	5.338	0.749	0.294
382.2	300.0	5.998	0.753	0.287
415.0	300.4	6.870	0.765	0.265
Run 2 (D = 0.0127 Cm, 1 Atm)				
303.8	297.9	0.305	0.554	0.0607
314.2	297.9	1.212	0.723	0.155
336.6	298.2	3.165	0.712	0.278
350.6	298.2	4.305	0.718	0.304
369.3	299.0	5.520	0.747	0.301
388.4	298.7	6.292	0.755	0.293
413.5	298.5	6.993	0.767	0.278
Run 3 (D = 0.0127 Cm, 1 Atm)				
304.5	298.2	0.317	0.533	0.0643
316.1	298.2	1.236	0.656	0.167
336.1	297.7	3.116	0.704	0.282
350.8	297.7	4.405	0.727	0.309
360.7	297.7	5.035	0.728	0.313
373.7	297.7	5.816	0.749	0.310
388.2	298.2	6.207	0.742	0.297
Run 4 (D = 0.200 Cm, 0.090 Atm)				
315.9	298.9	0.109	0.844	2.64
315.0	298.7	0.104	0.830	2.59
322.0	298.9	0.147	0.899	3.16
337.3	298.6	0.201	0.896	3.95
348.3	298.7	0.225	0.901	4.16
350.3	298.8	0.229	0.905	4.17
357.0	299.4	0.243	0.937	4.13
371.2	298.4	0.273	0.947	4.42
374.9	298.9	0.275	0.953	4.34
379.2	298.9	0.287	0.977	4.35
387.2	298.9	0.294	0.969	4.36
Run 5 (D = 0.200 Cm, 0.100 Atm)				
305.6	300.0	0.044	0.919	1.34
313.0	300.3	0.097	0.962	2.60
314.0	300.2	0.098	0.903	2.78
316.8	300.3	0.118	0.936	3.13
317.3	300.2	0.124	0.953	3.22
318.2	300.3	0.121	0.898	3.30
322.7	300.4	0.145	0.912	3.73
325.5	300.4	0.173	1.000	3.96
326.3	300.5	0.174	0.992	3.99
327.2	300.6	0.177	0.991	4.03
338.3	300.7	0.234	1.070	4.58
340.1	300.7	0.248	1.110	4.64
346.9	300.8	0.250	1.050	4.80
369.9	301.1	0.297	1.080	5.04
397.2	300.7	0.357	1.150	5.20
Run 6 (D = 0.200 Cm, 0.100 Atm)				
280.1	274.2	0.034	0.800	3.58
286.3	275.0	0.076	0.867	6.40
291.4	274.9	0.122	0.927	8.81
298.0	279.2	0.162	1.020	8.49
300.0	280.5	0.184	1.100	8.32
298.7	276.7	0.169	0.920	10.30
306.3	280.7	0.221	1.010	9.76
309.2	279.9	0.262	1.060	10.80
314.9	280.2	0.269	0.943	11.40
314.2	278.8	0.311	1.070	12.10
313.7	276.6	0.305	1.000	13.30
327.4	279.9	0.378	1.060	12.30
336.7	280.3	0.410	1.060	12.20

Table II. Natural Convection Heat Transfer to Nitrogen and Air

T_w, °K	T_∞, °K	q, Watts/Cm^2	N_{Nu}	N_{Gr}
Run 7 (D = 0.0127 Cm, 3.05 Atm Nitrogen)				
332.1	296.5	0.406	0.530	0.0756
351.6	299.5	0.646	0.560	0.0975
380.2	298.8	1.141	0.612	0.133
402.5	298.5	1.576	0.645	0.153
454.1	299.2	2.555	0.662	0.178
529.7	300.0	4.222	0.683	0.190
602.4	300.8	6.008	0.692	0.187
659.6	299.0	7.592	0.698	0.184
664.9	302.3	7.639	0.694	0.178
699.0	302.0	8.592	0.694	0.174
860.0	302.9	13.739	0.705	0.147
Run 8 (D = 0.200 Cm, 1.95 Atm Nitrogen)				
528.9	494.2	0.067	0.959	13.8
554.5	494.5	0.137	1.11	21.9
576.2	495.3	0.236	1.40	27.4
610.2	495.9	0.342	1.40	34.8
655.7	496.6	0.495	1.41	42.2
Run 9 (D = 0.0127 Cm, 1 Atm Air)				
311.6	298.5	0.134	0.485	0.0033
327.4	298.5	0.301	0.483	0.00667
332.3	298.8	0.377	0.519	0.00751
346.6	298.4	0.572	0.537	0.0101
361.0	298.5	0.750	0.533	0.0121
371.6	298.8	0.893	0.537	0.0133
385.2	299.5	1.100	0.552	0.0146
403.2	299.2	1.380	0.559	0.0162
425.2	299.2	1.722	0.560	0.0177
446.2	298.8	2.079	0.565	0.0189
Run 10 (D = 0.200 Cm, 0.08 Atm Air)				
299.5	297.8	0.001	0.448	0.0130
302.6	298.2	0.003	0.516	0.0329
303.7	298.2	0.004	0.550	0.0409
310.9	298.2	0.010	0.589	0.0907
324.4	298.2	0.023	0.645	0.174
349.0	298.2	0.051	0.713	0.296
388.2	298.2	0.118	0.886	0.430
373.9	298.2	0.082	0.745	0.388
463.2	298.2	0.222	0.833	0.554
Run 11 (D = 0.200 Cm, 1.0 Atm Air)				
305.8	297.7	0.012	1.12	8.28
313.5	297.7	0.026	1.23	15.50
322.6	297.7	0.043	1.27	23.20
332.4	297.7	0.063	1.32	30.70
360.2	298.2	0.128	1.45	47.10
381.2	298.2	0.191	1.57	56.80
412.7	298.4	0.280	1.61	66.90
433.2	298.4	0.335	1.59	71.70
Run 12 (D = 0.200 Cm, 1.0 Atm Air)				
302.7	297.4	0.007	0.100	5.53
305.8	297.4	0.012	0.108	8.61
311.1	297.7	0.027	0.151	13.3
328.9	297.9	0.057	0.134	27.9
340.7	297.9	0.083	0.139	36.1
322.0	297.7	0.042	0.127	22.7
357.9	298.2	0.124	0.146	45.9
358.7	298.2	0.125	0.145	54.7
376.2	298.2	0.172	0.151	64.0
402.4	298.4	0.246	0.157	
Run 13 (D = 0.200 Cm, 1.0 Atm Air)				
529.5	490.9	0.065	0.820	4.05
566.9	496.3	0.142	0.948	6.38
587.2	493.2	0.243	0.120	8.10
591.7	496.0	0.245	0.119	8.02
572.2	492.0	0.206	0.121	7.29

EXPERIMENTAL

The natural convection cell used is shown in Figure 1. A platinum or gold cylinder was suspended horizontally in the borosilicate glass cell 30 cm long and 7.6 cm in i.d. The electrically heated cylinder served as a nearly isothermal heat source. In the case of the N_2O_4 system, the runs with platinum gave results identical to those with gold. However, at elevated cylinder temperatures (ca. 800° C) platinum behaved as a catalyst for the $2NO_2 \rightleftharpoons 2NO + O_2$ nonequilibrium system, whereas gold proved to be noncatalytic. The results of an investigation of natural convection heat transfer to nonequilibrium dissociating NO_2 reacting homogeneously on gold and catalytically on platinum is being published.

The cylindrical elements were electrically heated in a direct current circuit and the heat loss from the surface was calculated from measurements of the voltage drop along and the current through the element. From these voltage-current measurements the resistance of the wire was established, which was related to wire temperature by calibration. Small wires (0.0025 cm in o.d.), used as voltage taps, were attached about 2 cm from the ends of the cylinder to minimize axial conduction effects.

The dissociating N_2O_4 system was chosen for study because it contributes a large reacting effect to the heat transfer rate and because thermodynamic, reaction kinetics, and transport data are readily available from Svehla (*9*). The N_2O_4 used in the experiments was obtained from the Matheson Co. and had a composition of 99.9 mole % N_2O_4. In each experiment to remove any traces of moisture or HNO_3 the gas was passed through a P_2O_5 drying tube prior to entering the evacuated natural convection cell. The purity of the gas was confirmed by freezing a sample and noting the color of the solid, which for pure N_2O_4 is white, even traces of NO giving the solid a bluish tinge.

RESULTS AND DISCUSSION

The detailed experimental natural convection data for N_2O_4, N_2, and air are shown in Tables I and II and are plotted in Figure 2. Values of the Nusselt number for air and N_2 deviate from the solid line by about ±3%, whereas the data points for N_2O_4 deviate about ±7%. To obtain a reasonable correlation, the Nusselt number had to be based on an integrated average of the thermal conductivity between T_∞ and T_w. If, as is commonly done in correlating heat transfer data in nonreacting systems, the value of the thermal conductivity employed in the Nusselt number is taken at a linear average of the bulk and surface temperature, the so-called film temperature, deviation in the Nusselt number as large as 300% can occur under the conditions of the present experiments. It is imperative in reacting systems to use the density driving force in the Grashof number and not temperature driving force times some average thermal expansion coefficient, as is presented by McAdams (*7*). Difficulty arises in the latter case because the thermal expansion coefficient is, like the thermal conductivity, a nonmonotonic function of temperature. The best correlation was obtained by using in the Grashof number values of kinematic viscosity based on the linear averaged bulk and surface temperature. Employing wall values of the kinematic viscosity leads to a maximum in the Grashof number, since the buoyancy term in the numerator and the kinematic viscosity term in the denominator increase with temperature at different rates, causing the Grashof number first to increase at low temperature driving forces and to decrease at sufficiently large temperature driving forces. This behavior occurs because under the condition of these experiments the surface temperature is varied with the bulk temperature maintained constant.

Values of the Nusselt number in the correlation represented by the curve in Figure 2 lay, for a given Grashof number, very close to the lower curve shown by McAdams.

CONCLUSIONS

Natural convection heat-transfer data in equilibrium reacting gases (where physical properties are not influenced by kinetics) may be correlated in the same fashion as for nonreacting systems, using the appropriate average values of the equilibrium properties. Because of the nonmonotonic dependence of thermal conductivity on temperature over certain temperature ranges, an integrated value of the thermal conductivity between the cylinder and bulk temperature has to be used in the Nusselt number. In the Grashof number, a linearly averaged value of kinematic viscosity suffices, since this property is a monotonic function of temperature over a wide temperature range.

In nonequilibrium systems, the processes of chemical kinetics and diffusion influence the over-all rate of heat transfer and the relative magnitude of the rate of reaction and diffusion must be taken into account, with the result that the simple treatment herein is not applicable.

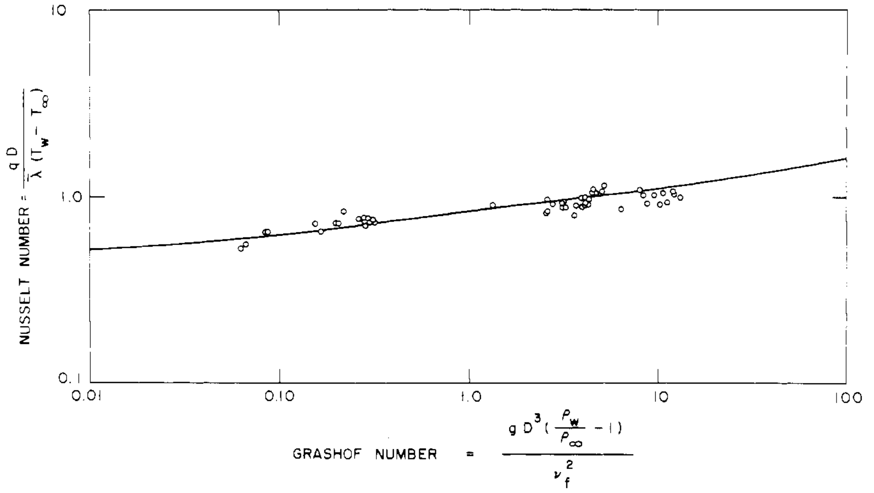

Figure 2. Natural convection heat-transfer correlation

— Air, N_2

O N_2O_4

NOMENCLATURE

D = diameter of cylindrical heating element, cm
g = local acceleration due to gravity, cm/sec^2
M = molecular weight of gas, g/g mole
T = temperature, ° K
q = *wall heat flux density, cal/cm^2 sec*
N_{Gr} = Grashof number, $gD^3(\rho_\infty/\rho_w - 1)/\nu_f^2$, dimensionless
N_{Nu} = Nusselt number, $qD/\lambda(T_w - T_\infty)$, dimensionless
λ = thermal conductivity, cal/cm sec ° K
$\bar{\lambda}$ = integrated temperature average of thermal conductivity

$$= \frac{\int_{T_\infty}^{T_w} \lambda dt}{T_w - T_\infty}, \text{ cal/cm sec °K}$$

ν_f = kinematic viscosity at linear average temperature, $\frac{1}{2}(T_w + T_\infty)$, cm^2/sec
ρ = density, g/cm^3

Subscripts

∞ = state of bulk gas
w = state of cylinder wall surface

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Enthalpy of Formation of Ammonium Hydrogen Monoamidophosphate, $NH_4HPO_3NH_2$

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The enthalpy of formation from the elements in their standard states at 25° C of crystalline (monoclinic) ammonium hydrogen monoamidophosphate, $NH_4HPO_3NH_2$, is −292.42 ± 0.22 kcal per mole. It was determined from its enthalpy of solution in 4m HCl and from published values for the enthalpies of formation of H_3PO_4, $NH_4H_2PO_4$, and H_2O.

In a continuing study of the thermal properties of compounds with potential application in fertilizer technology, measurements were made of the enthalpy of formation of ammonium hydrogen monoamidophosphate, $NH_4HPO_3NH_2$. This compound has a high plant nutrient content and good physical properties and was an excellent source of both nitrogen and phosphorus in greenhouse tests.

MATERIALS AND APPARATUS

Ammonium Hydrogen Monoamidophosphate. $NH_4HPO_3NH_2$ was prepared by adding 27.5 grams of phosphoryl trichloride (0.2 mole) slowly to 250 ml of a stirred, ice-cold 10% aqueous ammonia solution (1.5 moles of NH_3). There was some fuming and evolution of heat, and the reaction was complete within several minutes to form a clear solution.

Paper chromatography with a neutral solvent showed that the phosphate in the solution was distributed as 63.4% monoamidophosphate, 33.1% diamidophosphate, and 3.5% phosphoryl triamide. On dilution of the solution with 1 liter of acetone, two liquid phases were formed; the lower phase contained all the monoamidophosphate and the upper contained all the other species. The bottom layer was separated, neutralized with acetic acid, and cooled to induce crystallization. Additional product was obtained by dilution of the filtrate with an equal volume of ethanol.

The product was identified as $NH_4HPO_3NH_2$ by x-ray diffraction; the four strongest lines and their relative intensities were 5.77 (100), 4.22 (57), 4.30 (50), and 4.26 (51), corresponding, respectively, to 5.77 (100), 4.22 (46), 4.30 (35), and 4.26 (35) reported by Herzog (*3*). Similar agreement was found for the weaker reflections.

The calorimeter sample was a composite of four batches of about equal weight that were mixed thoroughly. Petrographic examination showed that all the individual batches had the same optical properties (monoclinic, $N_\alpha = 1.495$, $N_\beta = 1.522$, $N_\gamma = 1.541$)(*4*) as reference preparations whose identification was established by x-ray and chemical analysis. The composition of the composite was 24.6% total N, 12.4% ammoniacal N, and 27.15% P (stoichiometric: 24.56% total N, 12.28% ammoniacal N,

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