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THERMAL EFFECTS WHICH MAY RESULT IN THE UNEXPECTED IGNITION OF GASEOUS MIXTURES

by

James T. Dehn

April 1972

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U.S. ARMY ABERDEEN RESEARCH AND DEVELOPMENT CENTER
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Terminal Ballistics Laboratory

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ABERDEEN PROVING GROUND, MARYLAND

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MEMORANDUM REPORT NO. 2179

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Aberdeen Proving Ground,Md.
April 1972

THERMAL EFFECTS WHICH MAY RESULT IN THE UNEXPECTED IGNITION OF GASEOUS MIXTURES

ABSTRACT

When gases are admitted rapidly to a vessel the temperature may rise several hundred degrees in some circumstances. A simple formula involving the specific heat ratio, thermal conductivity and molecular weight of a gas is derived which successfully describes the maximum value of this temperature pulse as a function of temperature and pressure for a variety of gases. Safety applications and corrections to measurements of ignition limits and ignition delay times are discussed.

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LIST OF SYMBOLS

f = number of degrees of freedom in a molecule

N = number of molecules

k = Boltzmann's constant

T = gas temperature

T_w = vessel wall temperature

m = mass of a molecule

M = molecular weight

v_m = most probable molecular speed

n = number of moles of gas

K = thermal loss coefficient

γ = ratio of specific heats

R = universal gas constant

q = heat per mole flowing into a system

c_v = specific heat at constant volume

c_p = specific heat at constant pressure

p = pressure

v = specific volume = V/n

V = volume

λ = coefficient of thermal conductivity

L = effective heat loss distance

τ = time for gas to enter vessel and reach maximum temperature

I. INTRODUCTION

When gases are admitted rapidly into an evacuated vessel the temperature at the center of the vessel can rise dramatically hundreds of degrees above the temperature of its surroundings. If this is not realized and isothermal conditions are assumed to hold, large experimental errors can be made in the determination of ignition limits. Figure 1 shows the explosion limits of a hydrogen-oxygen mixture with large discrepancies in the upper limits as determined by static heating (dashed line) and rapid admission of gas to a pre-heated vessel through a solenoid valve (solid line)[1*].

If the gases form a combustible mixture and no provision for explosion has been made the consequences could be disastrous. Industrial accidents have occured during the transfer of potentially explosive gases from one vessel to another. Sometimes such accidents are blamed on the buildup of static charge in spite of the fact that adequate electrical grounds have been provided. A more probable cause is the phenomenon described in this report.

In summary, both the combustion chemist and the safety engineer can benefit by a better understanding of what is happening. It is the purpose of the present report to contribute to this understanding.

II. THE PHENOMENON

The phenomenon has been carefully investigated by Fine, Gray and MacKinven[2-5] in the course of their work on spontaneous ignition. Figure 2 gives a sample of their measurements of the maximum temperature increase ΔT above the vessel walls reached at the center of an evacuated spherical vessel (1.222×10^3 cm^3) at 190°C as a function

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[*] *References are listed on page 28.*

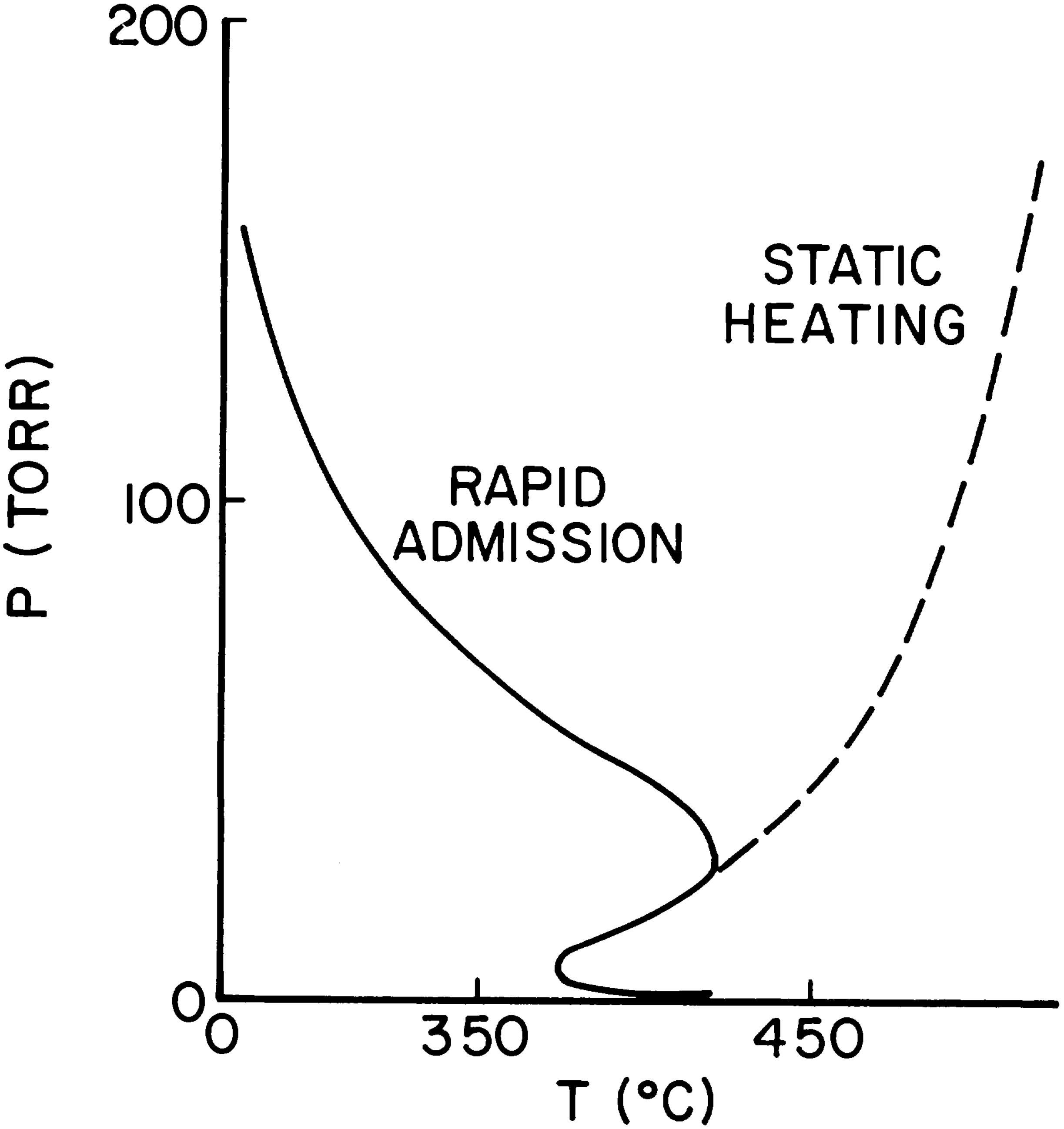

Figure 1. Explosion Limits of a Hydrogen-oxgyen Mixture as Determined by Static Heating (dashed line) and Rapid Admission of Gas to a Pre-Heated Vessel (Solid Line)

of the final pressure in the vessel for various gases. Smaller temperature rises occur closer to the vessel walls with a parabolic profile along a vessel diameter. Generally speaking, the greater the pressure rise the greater the maximum of the transient temperature pulse with a larger effect for monatomic gases than for diatomic gases and with small changes for heavy polyatomic gases. Within a monatomic or diatomic series however the lighter atoms usually have a smaller effect. Figure 3 shows the same relations for a vessel at room temperature[2]. Relations within a series are brought out more clearly by Figure 4 where ΔT is plotted against the ratio of specific heats, for the same experimental conditions as Figure 2 but with the final pressure only equal to 10 torr[2]. Again it is clear that the heavier atoms show a larger effect in the monatomic series, however in the diatomic series oxygen shows a smaller effect than nitrogen in spite of the fact that it is more massive. Still both show a larger effect than hydrogen. The fact that there are other factors at work besides the molecular weight and γ of a molecule is made clear by the observation that ΔT for argon increases with an increase in vessel temperature while ΔT for neon and helium decrease. Figure 5 shows the same relations as Figure 4 under the same conditions except that the vessel is at room temperature instead of 190°C.

All of these gases were studied under identical conditions and were admitted to the vessel through a solenoid valve with the temperature pulses for a final pressure of 10 torr reaching their maximum values in about 0.1 or 0.2 seconds and decaying almost as rapidly. The temperature changes took place throughout the volume but the changes at the center were always greatest. Cooling effects were also observed on expansion of gases from a vessel at higher than atmospheric pressure[2]. In summary, the size of the effect is a function of the properties of the gases themselves, the temperature and pressure employed and the speed of transfer. In the next section

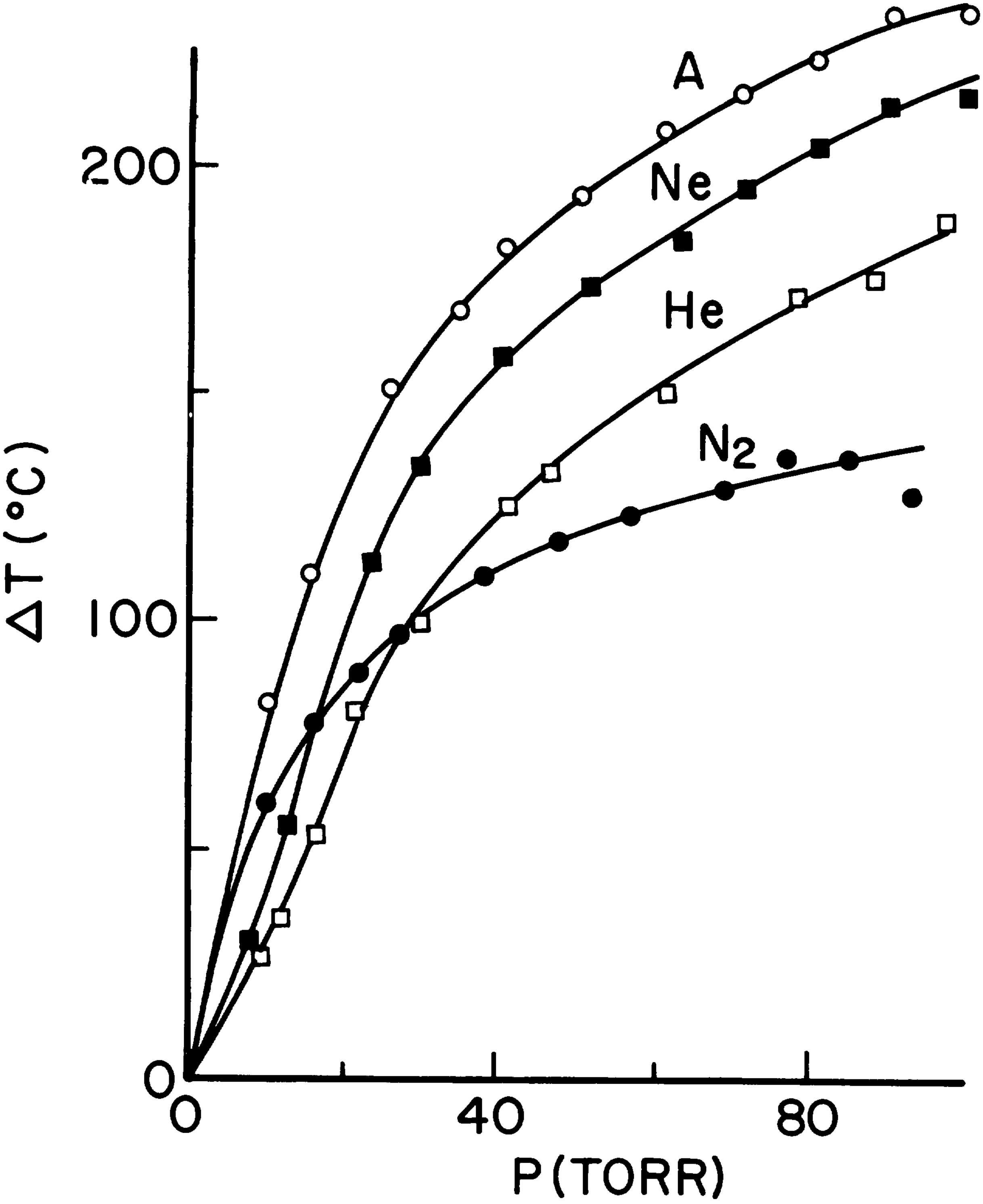

Figure 2. Maximum Temperature Increase ΔT at the Center of an Evacuated Spherical Vessel (Wall Temperature 190°C) as a Function of Final Pressure for Various Gases)

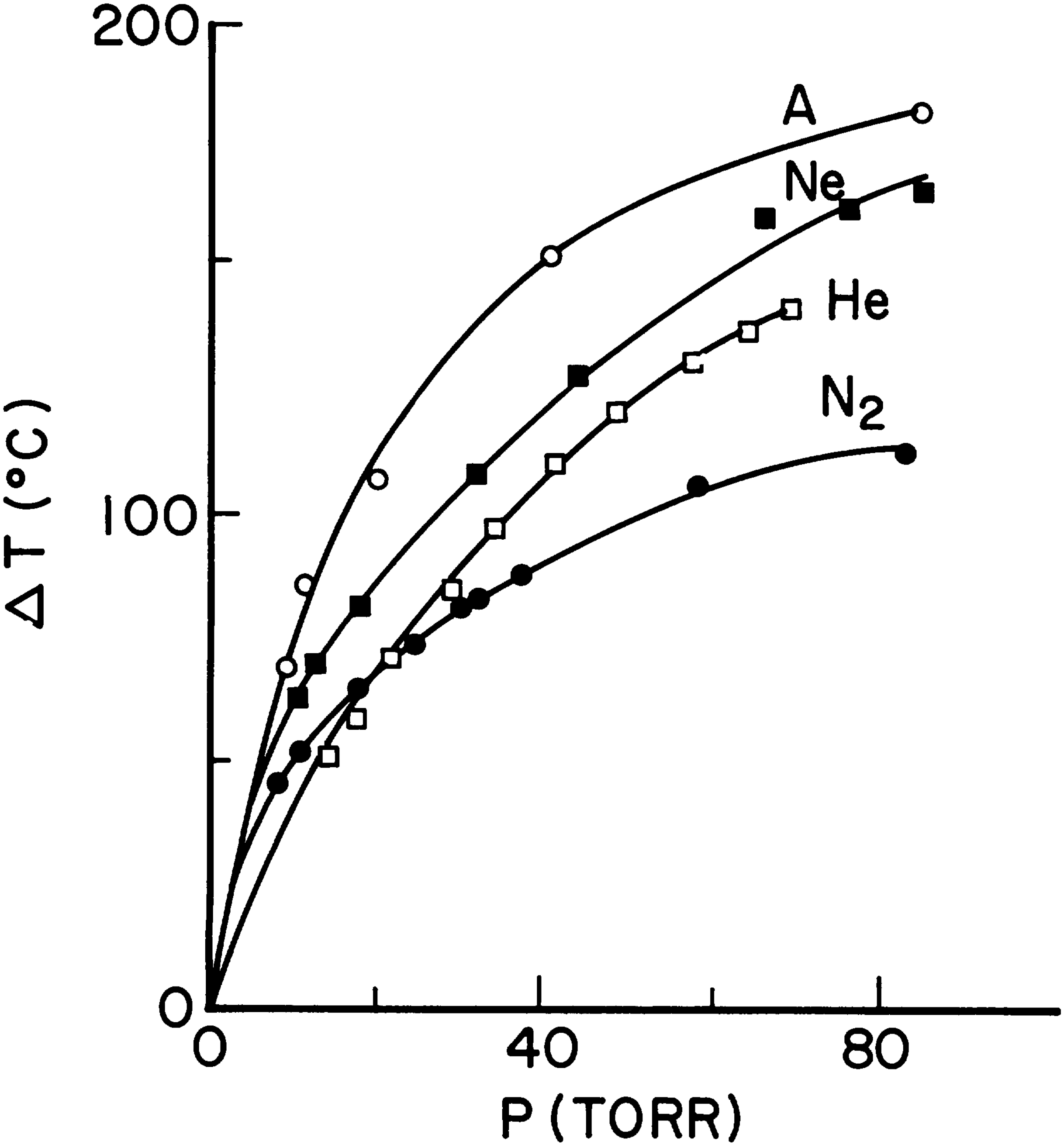

Figure 3. Maximum Temperature Increases ΔT at the Center of an Evacuated Spherical Vessel at Room Temperature as a Function of Final Pressure for Various Gases.

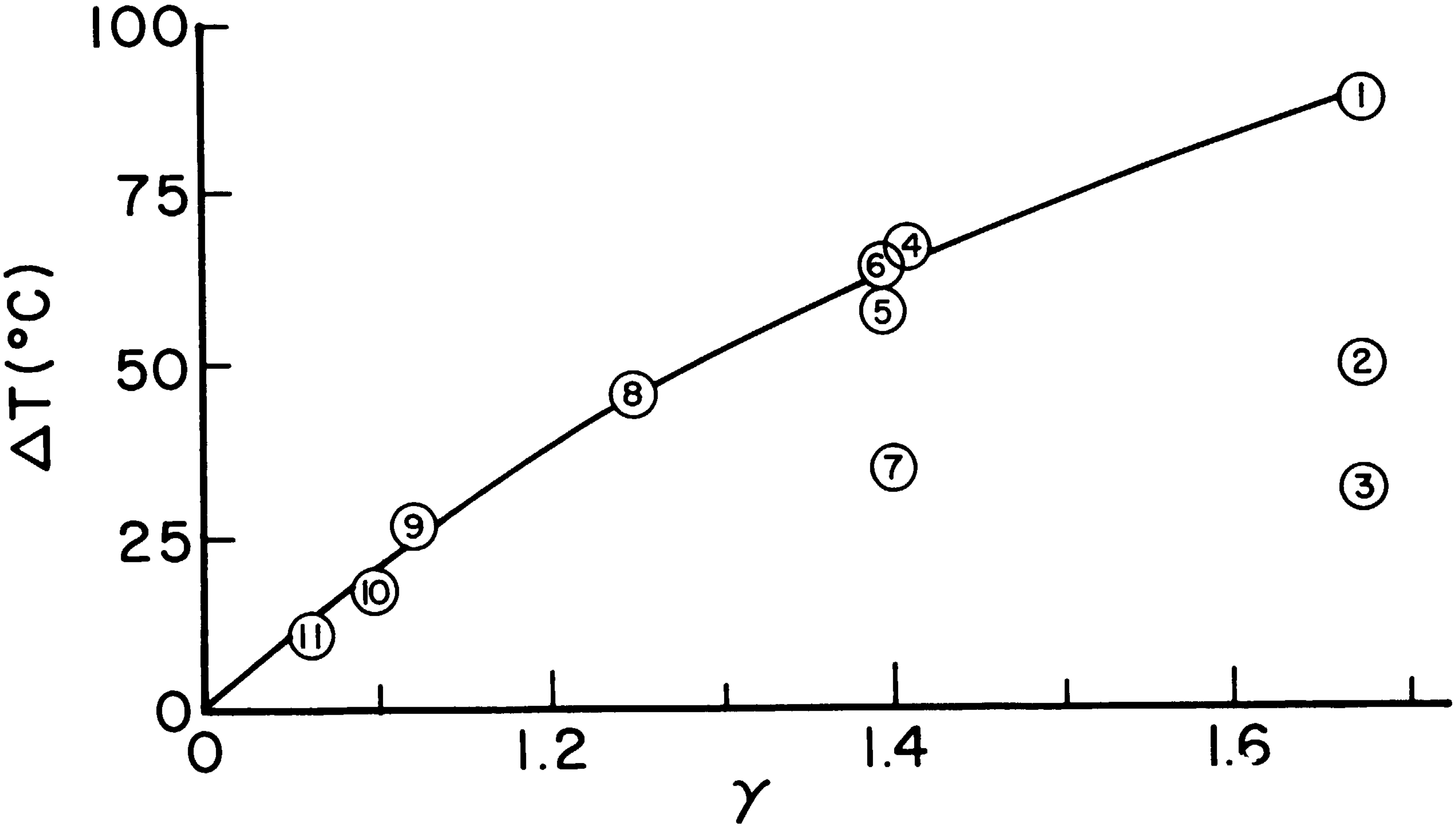

Figure 4. Maximum Temperature Increase ΔT at the Center of an Evacuated Spherical Vessel (wall temperature 190°C) for a Final Pressure of 10 Torr as a Function of the Ratio of Specific Heats, for Various Gases: 1, argon; 2, neon; 3, helium; 4, nitrogen; 5, oxygen; 6, carbon monoxide; 7, hydrogen; 8, methane; 9, ethane; 10, propane; 11, diethyl ether.

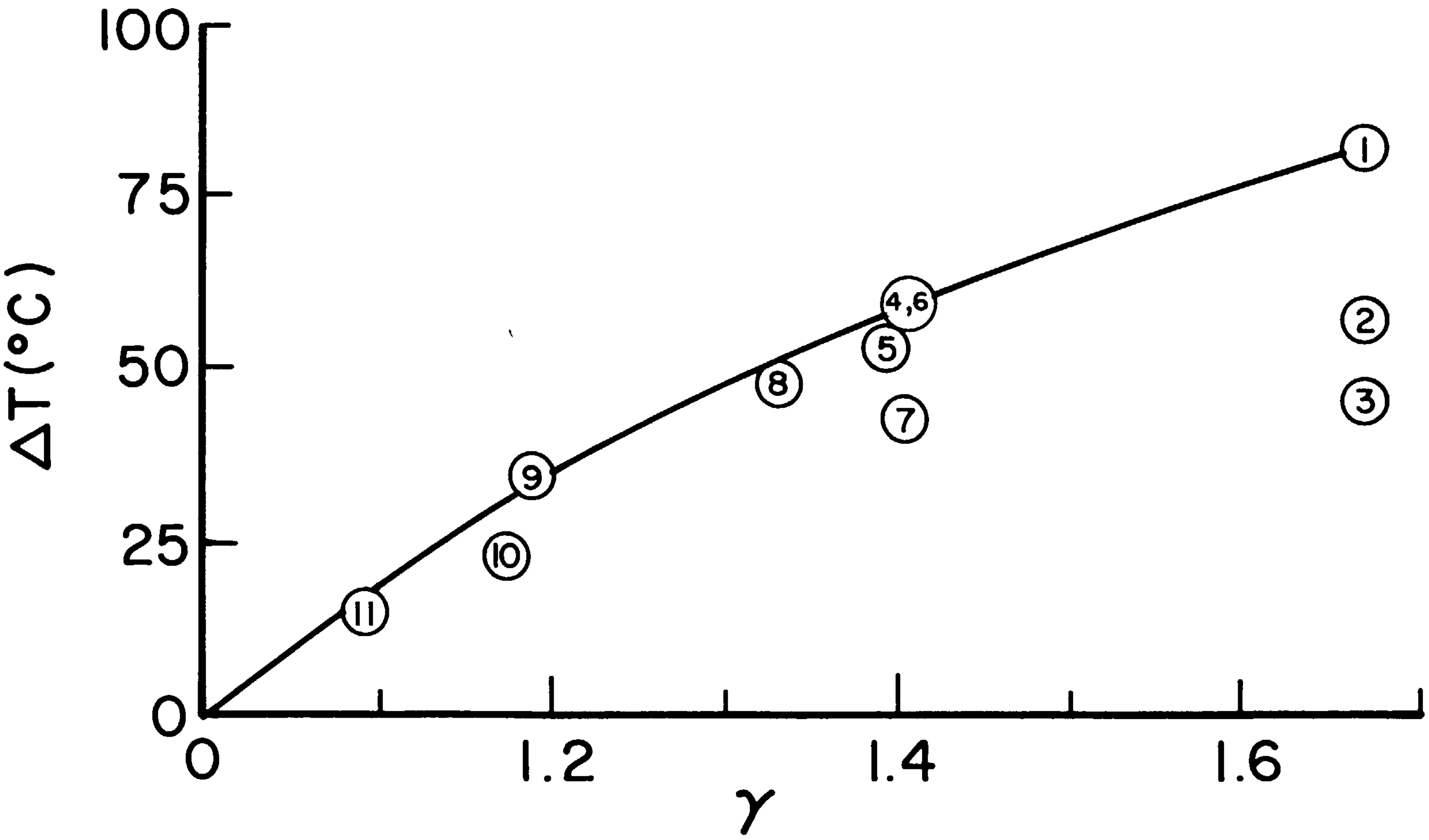

Figure 5. Maximum Temperature Increase ΔT at the Center of an Evacuated Spherical Vessel at Room Temperature for a Final Pressure of 10 Torr as a Function of the Ratio of Specific Heats for Various Gases: 1, argon; 2, neon; 3, helium; 4, nitrogen; 5, oxygen; 6, carbon monoxide; 7, hydrogen; 8, methane; 9, ethane; 10, propane; 11, diethyl ether.

of this report we shall develop a simple formula to describe the observed behavior.

III. THEORY OF THE PHENOMENON

A. A General Formula

When a vessel of gas at higher pressure is connected to one at lower pressure we might think of the gas in the first vessel as having a potential energy relative to the final state in which both are at the same pressure. As soon as the valve connecting the two vessels is opened the gas in the first vessel converts its potential energy into kinetic energy and rushes into the second vessel in the form of a jet. The kinetic energy of this jet is then converted into internal energy of the gas which has been admitted by collisions with the vessel walls and with other molecules. This manifests itself as a temperature rise which reaches its maximum value ΔT at the center of the vessel where heat exchange with the surroundings is slowest.

Let us write this increase in internal energy as $(1/2)\ fNk\Delta T$ where f is the number of degrees of freedom of the molecule, N is the number of molecules, k is Boltzmann's constant and ΔT is the maximum size of the thermal pulse at the center. The energy which has been admitted into the vessel is in the form of n moles of gas with kinetic energy $(1/2)Nmv_m^2$, some of which is lost to the walls by heat transfer with coefficient K(erg per mole-deg). We may then write

$$(1/2)fNk\ \Delta T = (1/2)Nmv_m^2 - nK\ \Delta T \qquad (1)$$

as the average energy balance. Here we are using the most probable speed of the molecules $v_m = (2RT_w/M)^{1/2}$ instead of the root mean square speed $(3RT_w/M)^{1/2}$ in terms of the molecular weight M and the temperature of the vessel wall T_w with $\Delta T = T - T_w$ where T is the maximum temperature reached at the center of the vessel. All except the heavy gases are heated to the temperature of the vessel walls in the entry arm. However, propane and diethyl ether show slight cooling before pulse heating since they are not fully warmed on entry because of their high heat capacities. This results in a lower ΔT than expected. Since $(1/2)f = 1/(\gamma-1)$, $Nm = nM$ and $NK = nR$, we obtain

$$\Delta T = (\gamma-1)T_w/(1 + (K/R)(\gamma-1)). \qquad (2)$$

Alternatively we may consider the gas admitted first to be compressed by the gas admitted later and use the first law of thermodynamics for an ideal gas in the form

$$\Delta q = c_v \Delta T + p\Delta v = c_v \Delta T + R\Delta T - RT\Delta p/p \qquad (3)$$

where v is the specific volume V/n and T is the temperature after a given amount of work has been done. If we take $\Delta q = -K\Delta T$, $c_v + R = c_p = \gamma R/(\gamma-1)$ and $\Delta p = p$ since the initial pressure in the vessel is practically zero, we find

$$\Delta T = T - T_w = (\gamma-1)T/(\gamma + (K/R)(\gamma-1)) \qquad (4)$$

or in terms of T_w

$$\Delta T = (\gamma-1)T_w/(1 + (K/R)(\gamma-1)) \qquad (5)$$

as in equation (2) above. If the vessel already contains some gas, $\Delta p/p$ would be an appropriate fraction r instead of unity. Equation (5) would then be

$$\Delta T = r(\gamma-1)T_w/(\gamma + (K/R - r)(\gamma-1)). \qquad (6)$$

This can also be derived from the first model if we multiply $(1/2)Nmv_m^2$ by the fractional contribution r to the total energy in the vessel. In other words, the added energy must be shared between the molecules added to the vessel and those already inside. The effect of r less than unity is to lower ΔT.

Both derivations are crude energy balances which take no account of the complicated changes which are taking place in time and space. However our only aim is to predict the maximum temperature rise at the center of the vessel. In addition we will treat the timing factor in an empirical fashion leading simply to useful results.

B. The Heat-loss Factor

Heat losses occur to the vessel walls by a combination of convection and conduction. Studies[2] have been made which suggest that conduction is dominant at pressures up to 100 torr at least during the cooling portion of the temperature pulse. Let us write our thermal loss coefficient $K = \lambda L\tau/n$ where λ is the coefficient of thermal conductivity (erg/cm-sec-deg), L is the effective loss distance (cm) and τ(sec) is the time for the gas to enter and the temperature pulse to reach its maximum at the center. If the gas is admitted very slowly, K becomes very large and ΔT approaches zero, a practical way to avoid unwanted temperature pulses, but a poor way to determine delay times or ignition limits in a reaction. Since $n = pV/(RT_w)$ we have $K/R = (L/V)\lambda\tau T_w/p$, showing that losses are smaller at higher pressures giving larger ΔT. If we use the experimental value $\tau = .21$ seconds for nitrogen at 10 torr final pressure in a 1.222×10^3 cm^3 spherical vessel at room temperature which produces a pulse $\Delta T = 57^oC$, we find that K/R must be 2.86 which gives a value of L = 280 cm, much larger than the vessel diameter of about 10 cm. In the context of the semi-empirical approach we have adopted we might attribute this to the effect of convection on the heating portion of the temperature pulse. Forced convection must make a considerable contribution when a jet of gas rushes into an evacuated vessel although density differences and so free convection may be unimportant. In the simple model we have developed we will represent convection through the factor L which we will fix at the value above in all our calculations. Where experimental values of τ are not available we will calculate them from the appropriate values for nitrogen by the formula $\tau = \tau_{N_2} (M/28)^{1/2}$ since it has been observed[2] that τ is approximately proportional to the square root of the molecular weight. It is likely that τ is inversely proportional to the molecular speed which would make it proportional to $(M/T_W)^{1/2}$. This agrees approximately with the observed values (Cf.Table I) and enables us to calculate τ as a function of temperature also.

IV. COMPARISON WITH EXPERIMENT

Table I gives the required parameters for calculating ΔT for a final pressure of 10 torr for a variety of gases and compares these calculated values with experimental values taken from Figures 4 and 5. As we see, if we omit propane and diethyl ether which show an initial cooling before heating the average deviation is about 1.33°C with a maximum deviation of 7°C. If we had used a calculated value for Helium at 300°K instead of the quoted experimental value, the 7°C deviation would have been much reduced. This is remarkably close agreement considering the crudeness of the model and the accuracy of the experiment and encourages us to compare the predictions of the model with experiments as a function of pressure (Figures 2 and 3).

Table I. Required Parameters M (gm/mole),λ(erg/cm-sec-deg),τ (sec)γ , and Experimental and Calculated Values of ΔT (^{o}K) for Admission of Gas to a Final Pressure of 10 Torr at Two Temperatures of the Initially Evacuated Spherical Reaction Vessel.

	M	γ_{300}	γ_{463}	$\lambda_{300} \times 10^{-4}$	$\lambda_{463} \times 10^{-4}$	τ_{300}	τ_{463}	ΔT_{300}	ΔT_{463}
A	40	1.67	1.67	0.18	0.25	.225	.185	77(82)	87 (88)
Ne	20	1.67	1.67	0.49	0.65	.147	.130	56(57)	48 (55)
He	4	1.67	1.67	1.50	2.01	.085	.075	43(36)	32 (33)
N_2	28	1.40	1.40	0.26	0.37	.210	.165	57(57)	64 (62)
O_2	32	1.40	1.37	0.27	0.39	(.220)	(.177)	50(53)	58 (57)
CO	28	1.40	1.40	0.25	0.36	(.210)	(.165)	57(57)	63 (63)
H_2	2	1.40	1.40	1.82	2.46	(.056)	(.044)	40(41)	35 (40)
CH_4	16	1.32	1.23	0.34	0.60	(.159)	(.125)	45(50)	45 (44)
C_2H_6	30	1.18	1.12	0.22	0.45	(.217)	(.171)	35(37)	27 (32)
C_3H_8	44	1.16	1.09	0.18	0.37	(.260)	(.206)	21(35)	16 (27)
$(C_2H_5)_2O$	74	1.09	1.06	0.15*	0.32	(.335)	(.270)	14(22)	7 (20)

*Extrapolated ignoring boiling point = 308^{o}K. Values in parentheses were calculated.

Table II. Experimental and Calculated Values of $\Delta T(^{o}K)$ as a Function of Pressure for an Initially Evacuated Reaction Vessel at Two Temperatures.

$T_w = 300^{o}K$

pressure (torr)	10	20	30	40	50	60	80	100
argon	77	110	130	150	155	160	175	---
	(82)	(117)	(135)	(147)	(156)	(163)	(171)	(175)
neon	56	80	105	125	135	145	155	---
	(57)	(88)	(109)	(122)	(132)	(141)	(152)	(160)
helium	43	65	80	105	115	125	---	---
	(36)	(62)	(80)	(94)	(105)	(114)	(128)	(138)
nitrogen	57	75	80	85	95	100	105	---
	(57)	(76)	(87)	(93)	(98)	(101)	(105)	(108)

$T_w = 463^{o}K$

pressure (torr)	10	20	30	40	50	60	80	100
argon	87	140	165	180	200	210	230	245
	(88)	(137)	(168)	(190)	(207)	(218)	(237)	(248)
neon	48	100	125	150	165	180	200	210
	(55)	(94)	(123)	(144)	(161)	(176)	(196)	(212)
helium	32	70	100	125	130	150	170	185
	(33)	(61)	(83)	(102)	(118)	(131)	(150)	(170)
nitrogen	64	85	100	110	120	130	135	---
	(62)	(93)	(111)	(123)	(132)	(139)	(148)	(154)

Calculated values are in parentheses. Experimental values have been estimated from Figures 2 through 5.

Table II compares calculated and experimental values of ΔT as a function of pressure for two temperatures. The agreement is generally good considering the model and the experiment and might have been better if experimental values of τ were available as a function of pressure. However, the values of τ measured or calculated for a final pressure of 10 torr were used throughout (as was a constant L value) and the K/R values of 10 torr were taken simply as inversely proportional to the pressure, probably accounting for much of the variance observed.

V. DISCUSSION

The model is too crude to warrant many refinements, yet it successfully predicts trends including the observed crossover of the nitrogen curve with the neon and helium curves as a function of pressure (Figures 2 and 3). It also predicts an upper limit to the temperature increase of $\Delta T_{max} = (\gamma-1)T_w$ when $K = 0$. When $T_w = 300^oK$, $\Delta T_{max} = 200^oK$ for monatomic gases, 120^oK for diatomic gases, and so forth. When $T_w = 463^oK$ (190^oC), $\Delta T_{max} = 310^oK$ for monatomic gases and 185^oK for diatomic gases, etc. These predictions are of special interest from a safety viewpoint and slow gas admission can be used to eliminate danger. When the measurement of ignition delays and ignition limits is of interest we can do several things to reduce error. If a heavy fuel plus oxygen system is being studied the oxygen might be admitted to the reaction vessel before it is heated and the fuel admitted rapidly after the desired temperature has been reached since a heavy molecule (unlike oxygen) will only give a small temperature pulse. Alternatively, if we desire to admit the oxygen rapidly into a vessel containing the fuel we can get an estimate of the temperature pulse by using an inert gas such as nitrogen (or an inert model of the fuel if the fuel were to be transferred). Or in some circumstances we might adopt the idea of two vessels connected by a wide-bore stopcock with diffusion mixing after pressure equilibration. Finally, if premixed gases are admitted to a vessel effective average values of M, γ, and λ can be used to calculate ΔT and make corrections.

VI. CONCLUSION

We have derived a simple formula for calculating the temperature increase which results when a gas is admitted rapidly to a vessel. In spite of its simplicity the predictions of this formula agree well with experimentally measured values as a function of temperature and pressure for a variety of gases. This enables us to understand the phenomenon more clearly and to take precautions or make corrections as required.

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