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Ignition of Explosive Gas Mixtures by Electric Sparks. II. Theory of the Propagation of Flame from an Instantaneous Point Source of Ignition.1

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A theory of the propagation of flame from an instantaneous point source of ignition such as a capacitance spark is developed. When a mass element passes through a combustion wave, it at first gains heat by conduction from preceding hotter elements and later loses heat by conduction to succeeding cooler elements. Correspondingly, the sum h of the thermal and chemical energy per unit mass at first increases above the level ho for the unburned or adiabatically burned gas, and later decreases to the same level. It follows that in a unit-area segment of a plane combustion wave the excess energy $\int_{x_0}^{x_0} \rho(h-h_0) dx$ is stored, where x_n and x_b denote reference points before and behind the wave. This energy is acquired at the expense of the energy content of the burned gas as the flame grows from a small sphere to its final size. The energy that is thus lost by the burned gas is insignificant except at the flame origin. Here, the excess energy must be furnished by the ignition source. It is the function of the latter to initiate the reaction by producing a high local concentration of heat and chain carriers, and to furnish at least as much energy as is necessary to satisfy the excess energy requirement of the smallest flame that is capable of selfpropagation. Such a flame may be visualized as a burning sphere of the smallest volume consistent with the condition that the rate of heat production should equal the rate of heat conduction to the surrounding unburned gas, and the minimum ignition energy is the integral $\int_0^\infty 4\pi r^2 \rho (h-h_0) dr$. The calculation of this energy is made possible by assuming a simplified, partly linear temperature gradient in the combustion wave. This approximate gradient closely circumscribes the actual gradient. The resulting temperature and energy distribution can be calculated numerically from data on the flame diameter, the burning velocity, the initial and flame temperature, the heat conductivity, density and specific heat of the unburned gas. In a calculation of this kind, no issue arises concerning a "thermal" or "kinetic" theory of flame propagation. The issue arises only when, conversely, an attempt is made to calculate the burning velocity and the width of the reaction zone from speculative values of the ignition temperature and the rate of the chemical reaction.

No data are available for the diameters of the smallest self-propagating flames; however, they are smaller than the readily measurable flame quenching distances between plane parallel plates by a factor of about 2. Using energy data reported in a previous publication and the other enumerated data, flame diameters have been calculated and compared with quenching distances. A satisfactory agreement is found.

A marked difference is noted between lean and rich mixtures of methane and oxygen with and without inert gas. In the former the chemical reaction begins at a much higher temperature and the temperature gradient is much steeper, than in the latter. An explanation may possibly be found in the suggestion that in the combustion zone of rich mixtures the concentration of free radicals and particularly hydrogen atoms is much larger. If such species diffuse toward the unburned gas in considerable quantity, significant reaction occurs already at low temperature levels. The higher temperature levels are then depleted of chain carriers, and the temperature gradient correspondingly is much less steep than in lean mixtures, where rapid reaction is confined chiefly to high temperature levels.

IN a previous paper4 the ignition of mixtures of methane, oxygen and nitrogen by capacitance sparks has been described. In this paper a theory of the process is developed assuming a very short time of electric discharge and a very small spark volume, i.e., an instantaneous point source of ignition. The theory shows the existence of an absolute minimum energy which can be correlated to the burning velocity, the width of the combustion wave, and the thermal conductivity, specific heat and density of the gas mixture.

A general picture of the distribution of temperature and energy in a combustion wave is shown in Fig. 1. For the case of a plane wave the origin of the x-coordinate is placed at a constant distance from the wave and the gas flows from left to right at the velocity of flame propagation. At any point x the sum of the rates of heat gain or loss in a unit volume of a layer dx due to thermal conduction, mass flow and chemical reaction is zero. This is expressed by the following

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M. V. Blanc, P. G. Guest, G. von Elbe, and B. Lewis,

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equation:

$$\mu d^{2}T/dx^{2} - \rho Sc_{p}dT/dx + q(x) = 0$$
 (1)

where T=temperature, μ =heat conductivity coefficient, ρ =density, S=velocity of the gas flow, c_p =specific heat, and q(x)=rate of heat release by chemical reaction per unit volume.

The temperature rises from T_u of the unburned gas to T_b of the burned gas. The chemical term q(x) is initially very small; hence, according to Eq. (1), $d^2T/dx^2 \sim dT/dx$, which means that the temperature gradient and also the temperature increase exponentially with x, until at some temperature T the chemical term becomes significantly large. d^2T/dx^2 now decreases and finally becomes negative, corresponding to the dotted temperature curve in Fig. 1. Accordingly, as shown in the lower part of the figure, a mass element entering the combustion wave first acquires some excess energy by heat conduction from preceding elements and then returns this "borrowed" energy to succeeding elements; so that, as demanded by thermodynamics, its thermal energy after passage through the combustion wave is equal to the sum of its thermal and chemical energy before it entered the combustion wave. In the aggregate, the mass elements inside a plane wave segment of unit area carry an excess energy, $\int_0^{+\infty} \rho(h-h_0) dx$, where h represents the sum of thermal and chemical energy per unit mass and the subscript zero refers to the state before and after passage through the combustion zone.

The initial small flame which is formed around a spark derives its excess energy from the spark

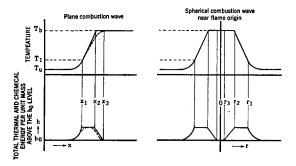


Fig. 1. Distribution of temperature and energy per unit mass in the combustion wave. $h_0 = \text{sum of thermal}$ and chemical energy per unit mass before and after combustion.

---- Actual distribution Approximate distribution

itself. It is the function of the latter to initiate the reaction by producing a high local concentration of heat and chain carriers, and to furnish at least as much energy as is necessary to satisfy the excess energy requirement of the smallest flame sphere that is capable of self-propagation. Such a flame may be visualized as a burning sphere of the smallest volume consistent with the condition that the rate of heat production should equal the rate of heat conduction to the surrounding unburned gas. As the flame grows the additional energy required by the growing flame surface is taken from the burned gas; that is, the combustion temperature is slightly lower than the temperature T_b of the plane wave. However, the spark must furnish at least the energy corresponding to the integral $\int_0^\infty 4\pi r^2 \rho (h-h_0) dr$ of the smallest self-propagating flame, r being the distance from the center of the flame.

The calculation of this energy from other data, notably data on the burning velocity and the diameter of the smallest flame, is made possible by substituting for the actual temperature gradient the simple approximate gradient illustrated in Fig. 1. The term q(x), Eq. (1), is taken as zero up to the temperature T_1 , and thus the gradient initially rises exponentially. Between T_1 and T_b , q(x) is taken as constant and d^2T/dx^2 as zero, and the gradient thus remains constant. A mass element within this part of the gradient neither loses nor gains h = thermal + chemicalenergy. However, heat is continually being furnished to the fresh gas at x_1 ; this heat is being produced at the constant temperature level T_b from residual chemical energy between x_2 and x_3 , and transported down the gradient by a fictitious mechanism. Although the resulting temperature and energy distributions are not physically possible, they closely circumscribe the actual distributions. They can be calculated numerically from the data on the width of the reaction zone x_3-x_1 , the burning velocity S_u , the temperatures T_u and T_b , and the heat conductivity, density and specific heat of the unburned gas.

In a calculation of this kind no issue arises concerning a "thermal" or "kinetic" theory of flame propagation. The issue arises only when, conversely, an attempt is made to calculate S_u and x_3-x_1 from speculative values of T_1 and the rate of the chemical reaction. These quanti-

ties would have to be determined from comprehensive knowledge of reaction kinetics and the diffusion of chain carriers in the reaction zone. However, if such knowledge were available, one would certainly not attempt to calculate the burning velocity and width of the reaction zone from a system of equations based on the heat balance Eq. (1) and the concept of ignition temperature. Rather, since the values of S_u and x_3-x_1 are mainly determined by chemical and diffusion processes within the zone of active chemical reaction, a suitable approximation may consist in the elimination of the heat balance Eq. (1) by an assumption regarding the energy content of any layer dx and giving detailed consideration to reaction and diffusion processes only. This principle was used in the calculation of burning velocity and width of the reaction zone of ozone flames by Lewis and von Elbe.5

For the present problem of calculating the minimum ignition energy the heat balance equation for a spherical combustion wave must be developed, using the above approximation.

For $r \ge r_1$ (cf. Fig. 1) the chemical term is zero; hence, the increase of heat in a unit volume of any shell $4\pi r^2 dr$ during the time dt is given by

$$\mu \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} \right) = c_p \rho \frac{\partial T}{\partial t}. \tag{2}$$

Here $\partial T/\partial r$ is a negative quantity since the temperature decreases along the radius r, but $\partial^2 T/\partial r^2$ is positive.

As the flame spreads the radii r_1 and r_2 increase at the rate $\partial r/\partial t = S_b$, which is the velocity of the combustion wave relative to the gas at the temperature T_b . This burned gas is at rest relative to the flame center, and the sphere with radius r_2 gains the mass $\rho_b S_b$ per unit surface in unit time. In the outer colder shells a flow of gas away from the center is induced by the thermal expansion, and each shell expands relative to the moving gas at a velocity $S < S_b$. It appears consistent with the assumptions of this treatment to specify that the shells expand at such a rate that the mass crossing unit area is the same for all shells, i.e., $\rho_b S_b = \rho S = \rho_u S_u$. Thus during unit time the mass

TABLE I.

$$\frac{ar_1}{\int_{r_1}^{\infty} a^3 r^2 [-Ei(-ar)] dr} = 0.514 \quad 0.296 \quad 0.142 \quad 0.058 \quad 0.018$$

 $\rho_u S_u$ passes through the volume dr times unit area of a shell of total volume $4\pi r^2 dr$, and receives the heat $c_p \rho_u S_u dT$; hence $|c_p \rho_u S_u \partial T/\partial r|$ is the rate of increase of heat in a unit volume of the shell. Equation (2) may then be written

$$\mu \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} \right) = -c_p \rho_u S_u \frac{\partial T}{\partial r}$$
 (3)

or

$$\partial^2 T/\partial r^2 + (a+2/r)\partial T/\partial r = 0,$$
 (3a)

where $a = c_p \rho_u S_u / \mu$. Writing $\partial T / \partial r = u$, Eq. (3a) is integrated to

$$u = u_1(r_1/r)^2 e^{-a(r-r_1)}$$
 (4)

and further,

$$\frac{T - T_u}{|u_1| r_1} = \frac{r_1}{r} e^{-a(r-r_1)} - ar_1 e^{ar_1} [-Ei(-ar)]$$
 (5)

where

$$-Ei(-ar) = \int_{r}^{\infty} \frac{e^{-ar}}{r} dr.6$$

The temperature T_1 and the gradient $|u_1|$ at r_1 for the spherical wave and x_1 for the plane wave, respectively (Fig. 1), are considered to be identical. For the plane wave the heat balance at x_1 is

$$\mu u_1 = c_p \rho_u S_u(r_1 = \infty) (T_1 - T_u)$$
 (6)

or

$$(T_1 - T_u)/u_1 = 1/a_\infty$$
 (7)

where a_{∞} is the value of a for a very large flame radius, i.e., plane wave. From (5) for $r=r_1$ and $T=T_1$, and (7),

$$\frac{1}{a_{\infty}r_1} = 1 - ar_1e^{ar_1} \left[-Ei(-ar_1) \right]. \tag{8}$$

The coefficient a is thus determined from values of r_1 and the coefficient a_{∞} . The latter is

⁶ B. Lewis, and G. von Elbe, J. Chem. Phys. 2, 537 (1934).

⁶ This function is tabulated in "Tables of sine, cosine and exponential integrals," Vol. 1, Federal Works Agency, WPA, 1940; sponsored by National Bureau of Standards.

ar_1	3	5	10	15	20	- 30	40	50
ar_a	3.90	5.82	10.77	15.74	20.73	30.72	40.69	50.68
$\frac{(T_a-T_u)}{(T_b-T_u)}$	0.263	0.166	0.0816	0.0537	0.0418	0.0254	0.0191	0.0153

TABLE II. Average temperature T_a at radius r_a between T_1 and T_u .

larger than a corresponding to an increase of the burning velocity S_u as the flame expands from a sphere to a plane wave.

The excess energy H in the exponential part of the temperature gradient is

$$\left| H \right|_{\tau_1}^{\infty} = \int_{\tau_1}^{\infty} 4\pi r^2 \rho (h - h_0) dr$$

$$= \int_{\tau_1}^{\infty} 4\pi r^2 \rho c_p (T - T_u) dr. \quad (9)$$

Substituting $T-T_u$ from Eq. (5), and integrating,

$$\left|H\right|_{r_{1}}^{\infty} = 4\pi \bar{\rho} \bar{c}_{p} |u_{1}| (r_{1}/a)^{2}$$

$$\times \left(1 + ar_{1} - \int_{r_{1}}^{\infty} a^{3} r^{2} \left[-Ei(-ar)\right] dr\right) \quad (10)$$

where $\bar{\rho}$ and \bar{c}_p now represent average values over the shell volume from r_1 to ∞ . Values of the integral term inside the bracket have been determined and are given in Table I. Since in numerical examples the term ar_1 is generally found to be considerably larger than two, it appears that the integral term may be safely neglected.

For the linear part of the gradient between r_1 and r_2 one obtains

$$\left| H \right|_{r_2}^{r_1} = \int_{r_2}^{r_1} 4\pi r^2 \rho \bar{c}_p(T_1 - T_u) dr
= (4/3)\pi \bar{\rho}_1 \bar{c}_p(T_1 - T_u) (r_1^3 - r_2^3). \tag{11}$$

TABLE III. Thermal conductivities, densities and specific heats.

	cal./cm sec. °C	C g/cm ³				
T,°K	300	300	300	500	600	700
O_2	0.000062	0.00131	0.218	0.232	0.240	0.246
N_2	0.000061	0.00115	0.250	0.253	0.258	0.263
CH₄	0.000075	0.00065	0.518	0.697	0.781	0.868

The average density $\bar{\rho}_1$ is smaller than $\bar{\rho}$ because the temperature rises from T_1 to T_b between r_1 and r_2 . After combining with Eq. (7), Eq. (11) may be written in a form directly comparable with Eq. (10), namely,

$$|H|_{r_2}^{r_1} = 4\pi \bar{\rho}_1 \bar{c}_p |u_1| (r_1^2/aa_\infty) \times [(ar_1 - ar_2^3/r_1^2)/3]. \quad (11a)$$

This shows that $|H|_{r_2}^{r_1}$ has somewhat less than one third the value of $|H|_{r_1}^{\infty}$. The term $|H|_{0}^{r_2}$ is smaller than the second term in Eq. (11a) and hence may be neglected.

The gradient $|u_1|$ is stable, i.e., the flame can propagate, when the rate of heat production in the volume $(4/3)\pi r_2^3$ equals the heat flow across the area $4\pi r_1^2$. In the smallest flame that can propagate the reaction zone is considered to extend to the center, and r_3 (Fig. 1) is zero. In unit time the heat q is liberated per unit volume. This quantity is taken as constant between r=0 and $r=r_1$, and zero at $r>r_1$. Thus, the flame can propagate when

$$(4/3)\pi r_2^3 q = 4\pi r_1^2 \mu |u_1|. \tag{12}$$

Between r_1 and r_2 the heat balance equation becomes

$$q = c_p \rho_u S_u |u_1|. \tag{13}$$

Combining (12) and (13),

$$r_2^3 = 3r_1^2 \mu / c_p \rho_u S_u = 3r_1^2 / a. \tag{14}$$

Since

$$|u_1| = (T_b - T_1)/(r_1 - r_2),$$

$$|u_1| = (T_b - T_1)/r_1 [1 - (3/ar_1)^{\frac{1}{6}}].$$
(15)

Eliminating T_1 by Eq. (7).

$$|u_1| = a_{\infty}(T_b - T_u)/\{1 + a_{\infty}r_1[1 - (3/ar_1)^{\frac{1}{2}}]\}.$$
 (16)

The final equation for the minimum ignition energy is obtained by adding Eqs. (10) and (11a) and eliminating $|u_1|$ by Eq. (16). On the basis

of previous discussions of Eqs. (10) and (11a), numerical calculations will not be significantly affected if the integral term in (10) is neglected and if the contribution of (11a) is simply expressed by changing the remaining numerical factor $1+ar_1$ in Eq. (10) to $1+1.3ar_1$. The equation may then be written

$$H = 4\pi r_1^2 (\bar{\rho}\bar{c}_p/a_\infty) (T_b - T_u)$$

$$\times \left(\frac{a_{\infty}}{a}\right)^{2} \frac{1 + 1.3ar_{1}}{1 + a_{\infty}r_{1}[1 - (3/ar_{1})^{\frac{1}{4}}]}. \quad (17)$$

Values of T_b may be calculated from thermodynamic data. Since the specific heat, heat conductivity and density change with temperature, values of these quantities at an average temperature T_a between T_1 and T_u are used. The following method is used to calculate T_a : A radius $r_a > r_1$ is determined so that

$$\frac{1}{2} \left| H \right|_{r_1}^{\infty} = \left| H \right|_{r_1}^{r_a} = \left| H \right|_{r_a}^{\infty}. \tag{18}$$

Analogous to Eq. (9) for $|H|_{r_1}^{\infty}$, and taking ρc_p as constant for the purpose of this calculation, one may write

$$\int_{r_1}^{r_a} r^2 (T - T_u) dr = \int_{r_a}^{\infty} r^2 (T - T_u) dr. \quad (19)$$

Substituting $T - T_u$ by Eq. (5), and integrating, one obtains

$$(ar_a+1)e^{-a(r_a-r_1)}=0.5(ar_1+1).$$
 (20)

In this equation the terms with factors

(-Ei(-ar)) have been neglected because, as was shown earlier in Eq. (10), they are very small for the values of ar_1 and ar_a which are of interest here.

Values of ar_a calculated from Eq. (20) are used to calculate $(T_a-T_u)/(T_b-T_u)$ from Eqs. (5) and (16). Results are shown in Table II.

Values of μ and ρ at 300°K, and c_p at 300, 500, 600, and 700°K are given in Table III. μ increases approximately proportional to the square root of the temperature and ρ is inversely proportional to the temperature.

The heat conductivity of the mixture is taken as the molar average of the heat conductivities of the component gases. Values of the burning velocity S_u for a plane wave are taken from burner-method measurements.⁷ The coefficient a_{∞} is thus calculated from the equation

$$a_{\infty} = (\bar{c}_p \rho_u S_u / \mu_u) (T_u / T_a)^{\frac{1}{2}}.$$
 (21)

On partial substitution of Eq. (21) in Eq. (17) and since $\bar{\rho} = \rho_u T_u / T_a$, Eq. (17) may be rewritten in the form

$$H = 4\pi r_1^2 \frac{\mu_u}{S_u} \left(\frac{T_u}{T_a}\right)^{\frac{1}{2}} (T_b - T_u) \left(\frac{a_\infty}{a}\right)^2 \times \frac{1 + 1.3ar_1}{1 + a_\infty r_1 [1 - (3/ar_1)^{\frac{1}{2}}]}. \quad (22)$$

The remaining unknown quantity in this equation is the flame radius r_1 . An upper limit is given by the readily measurable flame-quenching distance d between plane-parallel plates,

Table IV. Calculation of flame diameters $2r_1$, temperatures T_b , T_1 , and T_a , and gradients $(T_b - T_1)/(r_1 - r_2)$ for smallest igniting flames. Comparison of flame diameters with flame quenching distances d between parallel plates. $T_u = 300^{\circ}$ K.

Mixture composition, percent		Su.	T_b	T_1	T_a	Min. ignition energy H, cal. 10 ⁸		$2r_{1}$,	d.		$\frac{(T_b-T_1)}{(r_1-r_2)},$	
CH ₄	O ₂	N ₂	cm/sec.		°K		Exp.	Calc.a	cm	cm	$d/2r_1$	°C/cm 10
10	90		80	2200	1330	587	19	36	0.038	0.072	1.9	3.5
15	85		175	2650	1570	658	3.1	3.9	.017	.050	2.9	9.5
25	75		304	3000	. 822	515	1.4		.016	.028	1.8	8.0
40	60		305	3000	605	435	5.0		.032	.051	1.6	3.5
50	50		122	2650	446	379	110		.106	.165	1.6	0.8
52.5	47.5		87	2500	388	343	430		.192	.38	2.0	0.38
10	18.8	71.2	36	2200	1115	548	170	382	.090	.28	3.1	1.3
16.3	29.3	54.4	110	2620	1100	567	20		.035	.079	2.3	3.8
21.5	39.75	39.75	170	2810	791	501	9		.031	.053	1.7	3.9
26.4	49.1	24.5	240	2940	592	429	5.3		.030	.045	1.5	3.6

Values obtained by minimizing Eq. (22).

⁷ G. Jahn, Der Zündvorgang in Gasgemischen (Oldenbourg, Berlin, 1934).

which evidently must be larger than $2r_1$. It appears difficult at present to develop a quantitative theoretical relationship between d and $2r_1$; however, the two diameters cannot be very greatly different, and a factor $d/2r_1 \simeq 2$ appears reasonable. Equation (22) has therefore been used to calculate values of $2r_1$ from measured minimum ignition energies, and these values are compared with the corresponding values of d. The calculations are shown in Table IV. The examples chosen include a wide range of mixtures of methane, oxygen and nitrogen, and demonstrate that a reasonable correspondence between d and $2r_1$ indeed exists.

Equation (22) is of such a form that H, as a function of the radius r_1 , passes through a minimum. This is a consequence of the approximating assumptions embodied in the treatment and cannot be considered physically significant. In some cases this calculated minimum value is larger than the experimental value of H and it becomes impossible to satisfy the experimental value by any choice of r_1 . It then appears proper to solve the equation for the minimum value as this is the closest approach to the experimental value of H. It is noted in Table IV that this was done in three cases corresponding to low methane percentages. The discrepancies are not serious.

The values of T_1 and of the temperature gradient $(T_b-T_1)/(r_1-r_2)$ in Table IV indicate a marked difference between lean and rich mixtures of comparable burning velocities and combustion temperatures. In the former the chemical reaction begins at a much higher temperature and the temperature gradient is much steeper, than in the latter. An explanation may possibly be found in the suggestion that in the combustion zone of rich mixtures the concentration of free

radicals and particularly hydrogen atoms is much larger. If such species diffuse toward the unburned gas in considerable quantity, significant reaction occurs already at low temperature levels. The higher temperature levels are then depleted of chain carriers, and the temperature gradient correspondingly is much less steep than in lean mixtures, where rapid reaction is confined chiefly to high temperature levels.

The treatment thus establishes a theoretical concept of the minimum ignition energy of sparks in agreement with experimental data, and also suggests further investigations of the temperature gradient in combustion waves as a means of analyzing the physical-chemical processes governing flame propagation.

In conclusion, it is noted that the treatment is applicable to sparks only when (1) the dimensions of the flame over which the spark energy is distributed during the time of discharge are smaller than the flame radius r_1 , and (2) the time of discharge is small compared to the time required for a substantial growth of the flame radius r_1 . The second condition is easily fulfilled. For example, in the case of a fast burning mixture of 25 percent methane and 75 percent oxygen, a 50-percent increase of flame radius requires approximately 10-5 seconds according to the values of S_u and r_1 in Table IV, while the time of discharge is of the order of 10⁻⁷ seconds. The first condition cannot remain fulfilled indefinitely when the spark electrodes are pulled apart, and thus it is understandable that at large gap lengths the minimum ignition energies increase as shown in the preceding paper. Similarly, future experiments with greatly increased duration of electric discharge should demonstrate the dependence of the minimum ignition energy on the discharge time.