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On Excess Enthalpy, Flame Extinction and Minimum Ignition Energies[1]

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Excess Enthalpy of Ozone Decomposition Flames

USING Hirschfelder's numerical solution for the ozone decomposition flame (1),[4] the total excess enthalpy per gram of mixture has been calculated as a function of temperature for a mixture containing 75 per cent oxygen and 25 per cent ozone and an assumed Lewis number *Le* of 0.94.

The thermal (Δh_T), chemical (Δh_{ch}), and total enthalpies $\Delta h = \Delta h_T + \Delta h_{ch}$ per gram of mixture are plotted as a function of temperature in Fig. 1. Reference to Fig. 1 shows that Δh is everywhere positive but small because the chemical and thermal enthalpy changes nearly cancel for *Le* close to unity.

Lewis and von Elbe (2) have defined the excess enthalpy as

$$\Delta h_{Le} = \frac{\lambda}{m}\frac{dT}{dx} \quad [1]$$

where λ is the thermal conductivity and m equals the mass flow rate per unit area. The quantity Δh_{Le} is also shown in Fig. 1 and is seen to attain a maximum value nearly ten times

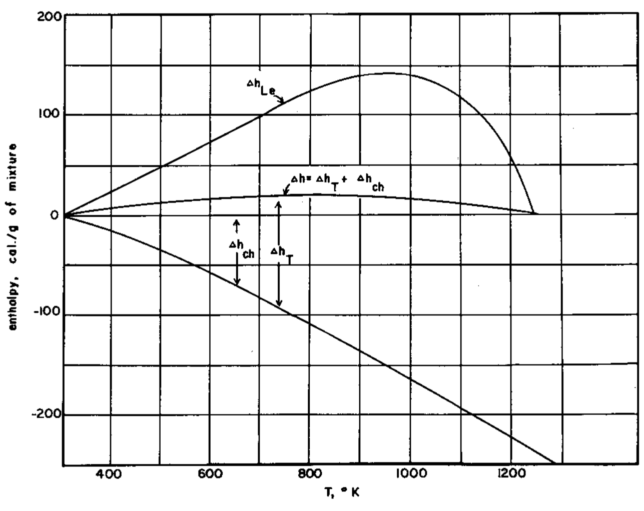

Fig. 1 The enthalpy terms Δh_T, Δh_{ch}, Δh_{Le} and Δh as a function of temperature for the ozone decomposition flame

as large as Δh. Also Δh_{Le} is linear with T and equal to Δh_T in the colder parts of the flame, because here reaction proceeds so slowly that the change in $\bar{c}_pT$ is produced largely by heat conduction. In the hotter parts of the flame, chemical reactions occur rapidly and the change in $\bar{c}_pT$ is now produced mostly by chemical heat release.

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In calculating minimum ignition energies, it is customary to consider the excess enthalpy h_a per unit area of flame front referred to the unburnt gas of density ρ_u, viz.

$$h_a = \rho_u \int_{-\infty}^{\infty} \Delta h \, dx \quad [2]$$

If diffusion is neglected in comparison with heat conduction, then h_a is replaced by

$$h_{Le} = \rho_u \int_{-\infty}^{\infty} \Delta h_{Le} \, dx = \frac{\bar{\lambda}}{Su}(T_f - T_0) \quad [3]$$

where T_f is the adiabatic flame temperature, *Su* equals the laminar burning velocity, and the bar indicates that a suitable average has been used for the entire flame width. Lewis' estimate of the minimum ignition energy, $h_{\min}$, is

$$h_{\min} = \pi d_q^2 h_{Le} \quad [4]$$

where d_q is the quenching distance. Performing an approximate integration on our graphs we find that, for the ozone decomposition flame, h_{Le} is larger than h_a by about a factor of 5.

In view of this large discrepancy it therefore appears likely that the demonstrated empirical utility of Equation [4] reflects the fact that enthalpy changes produced by heat conduction, rather than total enthalpy changes, are of dominant importance in ignition processes. This last statement is in accord with the conclusions reached by J. W. Linnett (3) after a critical examination of the arguments in favor of the excess enthalpy concept advanced by Lewis and von Elbe (2) and by Burgoyne and Weinberg (4, 5, 6), and after a critical examination of the relevant objections raised by Spalding (7).

Flame Extinction

Equation [4], as well as Spalding's critical size for a burnt gas slab which will just support combustion (8), are consistent with the following basic postulate: Thermal ignition will occur if the rate of energy transport per unit area by thermal conduction equals the rate of enthalpy change per unit area for a steadily propagating flame when energy transport by diffusion is neglected.

For one-dimensional, steady, inviscid, constant-pressure flow problems without external forces, the equation for conservation of energy reduces to

$$m\Delta h = \Delta q \quad [5]$$

where Δh denotes the change in the specific enthalpy for the mixture with constant mass flow rate m to which the total heat transfer by conduction and diffusion is Δq. The preceding relation may be applied to a stationary combustible mixture through which a laminar flame is propagated. In this case we identify $v = m/\rho$ with the mass-weighted average velocity of the burning gas relative to the flame front.

Consider a slab of burnt gas of finite width at the adiabatic flame temperature T_f in contact with a semi-infinite slab of combustible gas at the temperature T_0. According to our basic postulate, we may use for Δq in Equation [5] a suitable average value for $\lambda(dT/dx)$ which we approximate by $\bar{\lambda}(T_f - T_0)/\delta$ where δ is the effective width of the laminar flame front which is propagated at the constant speed *Su*. Equation [5] now becomes

$$\rho_u \delta \Delta h = \frac{\bar{\lambda}}{Su}(T_f - T_0) \quad [6]$$

[4] Numbers in parentheses indicate References at end of paper

since $m = \rho_u Su$. Furthermore, for gases with constant average specific heat $\bar{c}_p$

$$\Delta h = \bar{c}_p(T_f - T_0)(\rho_b/\rho_u) \quad [7]$$

in order to make the required heat conduction rate per unit area equal to $\rho_b Su\, \bar{c}_p(T_f - T_0)$. From Equations [6, 7] it is apparent that

$$\frac{\delta Su}{\bar{\lambda}/\rho_b \bar{c}_p} = 1 \quad [8]$$

where δ may now be identified with the smallest width of a heated burnt gas slab of density ρ_b which will prevent flame extinction and produce a steadily propagating one-dimensional laminar flame. For slabs of width smaller than δ, the energy per unit area in the flame front is presumably insufficient to propagate the combustion wave by thermal ignition. Equation [8] differs from Spalding's result (8) for the critical minimum size $d_{crit} = \delta$ of a uniform burnt gas slab at the adiabatic flame temperature T_f only in so far as the number 1 replaces 4 on the right-hand side. Spalding obtains the number 4 because he considers flame propagation from the hot slab in two directions (which accounts for a factor of two) and because he uses a maximum temperature gradient evaluated at the mean temperature (which accounts for another factor of two).

By an argument similar to that used in the derivation of Equation [8], we find that for a spherical source of burnt gas at the adiabatic flame temperature T_f, in an infinite combustible medium, the minimum radius r_{crit} of the sphere is determined by the relation

$$\frac{r_{crit} Su}{\bar{\lambda}/\rho_b \bar{c}_p} = 3 \quad [9]$$

Minimum Ignition Energies

The interpretation of Equation [6] which led to our identification of δ as d_{crit} for one-dimensional wave propagation may be restated as

$$\frac{h_{min}}{A} = \bar{\lambda}\left(\frac{T_f - T_0}{\delta}\right)\left(\frac{\delta}{Su}\right) \quad [10]$$

with h_{min}/A representing the minimum thermal enthalpy per unit area required to produce a self-propagating flame; the right-hand side of Equation [10] identifies the total conduction heat transfer per unit area during the life of a width δ when the flame is propagating at the speed Su.

In spark ignition measurements, h_{min} corresponds to the minimum ignition energy, and $A = \pi d_q^2$ is the effective spherical area bounding the gas volume into which the energy h_{min} is introduced. It is customary to choose d_q equal to the quenching distance in order to make certain that all of the energy introduced by the spark remains in the combustible gas. Equation [10] now reduces to Equation [4].

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