DEGENERATE THERMAL IGNITION OF GASEOUS SUSPENSIONS

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Study has been made of the effect of a reduction of the heat of reaction per unit mass on the critical conditions for ignition of a gaseous suspension. Calculations are presented to show the relation between critical ignition temperature and the particle diameter and concentration.

1. In earlier discussions of ignition in gaseous suspension [1-5], the heat of reaction per unit mass of suspension was assumed to be so high that $\gamma \equiv \gamma_0/B \ll 1$, ($\gamma_0 = CRT_0^2/Eq$, C is the heat capacity of the particle material; q, the heat of reaction per unit particle mass; T_0, the initial temperature; B, the mass particle concentration; E, the activation energy; and R, the molar gas constant). In many of the reported experiments, however, the mass particle concentration was so low that the parameter γ must have been comparable to unity in value.

The parameter γ relates to the heat of reaction of the gaseous suspension and is the analog of the γ_0 appearing in homogeneous thermal explosion theory [6, 7]. It has become customary to refer to thermal explosion at low γ_0 values as "degenerate" [6, 8]. Specifically, a degenerate homogeneous thermal explosion is one in which burn-out during the induction period has become a factor of importance, reaction becoming weakly autocatalytic with rising temperature and the critical ignition limit disappearing with an increase in γ_0.

Thermal ignition of the gaseous suspension at higher γ values will also be designated as degenerate. Ignition degeneracy in such two-phase systems differs quantitatively from ignition degeneracy in homogeneous systems. For example, under conditions such that ignition of the individual isolated particle remains nondegenerate ($\gamma_0 \ll 1$), the critical limit for suspension ignition will not disappear as the particle mass concentration is reduced and the value of the parameter γ brought closer to unity. In view of the role of critical burning in nondegenerate ignition theory [2, 3], component combustion can be expected to have a pronounced effect on degenerate ignition in the gaseous suspension.

Certain aspects of degenerate thermal ignition of gaseous suspensions will be treated in the present paper, the results obtained being checked against the experimental data of [9].

We will consider ignition in an enclosed suspension, assuming the gas temperature uniform throughout and the heat loss proportional to the temperature difference between gas and container walls.

For the case of identical particles uniformly distributed throughout the container, the following equations describe the isolated particle warm-up, the thermal energy balance in the gas, and the rate of particle combustion:

$$B[1+s\eta]\frac{d\theta}{d\tau} = \varphi(\eta)\exp\left(\frac{\theta}{1+\beta\theta}\right) - [1+s_1\eta]^{1/3}\frac{\theta-\theta_g}{\varkappa} \tag{1.1}$$

$$(1-B)\left[1 - s\frac{B}{1-B}\eta\right]\frac{d\theta_g}{d\tau} = [1+s_1\eta]^{1/3}\frac{\theta-\theta_g}{\varkappa} - s\gamma_0(\theta-\theta_g)\varphi(\eta)\exp\left(\frac{\theta}{1+\beta\theta}\right) - \frac{\theta_g-\theta_w}{A\varkappa} \tag{1.2}$$

$$\frac{d\eta}{d\tau} = \gamma\varphi(\eta)\exp\left(\frac{\theta}{1+\beta\theta}\right) \tag{1.3}$$

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$$\varphi(\eta) = (1-\eta)^{2/3}\left(1 - \frac{\nu}{a_0}\frac{B}{1-B}\eta\right)^m \tag{1.4}$$

the initial conditions being

$$\begin{gathered}
\tau \doteq 0, \quad \theta = \theta_0, \quad \theta_g = \theta_{g0}, \quad \eta = 0 \\
\theta = \frac{E}{RT_*^2}(T - T_*), \quad \theta_g = \frac{E}{RT_*^2}(T_g - T_*), \quad \theta_w = \frac{E}{RT_*^2}(T_w - T_*) \\
\theta_{g0} = \frac{E}{RT_*^2}(T_{g0} - T_*), \quad \eta = 1 - (r/r_0)^3, \quad \gamma_0 = CRT_*^2/Eq \\
\gamma = \gamma_0 / B, \quad \tau = 3ta_0^m k \exp(-E/RT_*)/r_0\gamma\rho, \quad \beta = RT_*/E \\
B = \frac{4}{3}\pi r_0^3 \rho CN \Big/ \left(\frac{4}{3}\pi r_0^3\rho CN + \rho_g C_g\right) \\
\varkappa = qka_0^m \exp\left(-\frac{E}{RT_*}\right) \Big/ \mathrm{Nu}\frac{\lambda_g}{r_0}\frac{RT_*^2}{E} \\
A = NV4\pi r_0^2 \mathrm{Nu}\, L / \Sigma \mathrm{Nu}_g r_0 \equiv 3\frac{\rho_g}{\rho}\frac{\mathrm{Nu}}{\mathrm{Nu}_g}\frac{B}{1-B}\left(\frac{VL/\Sigma}{r_0^2}\right)
\end{gathered} \tag{1.5}$$

Here, T, T_g, and T_W are the temperatures of particle, gas, and container wall, respectively; T_* is a scale temperature, to be defined below; T_0 and T_{g0} are initial temperatures of particle and gas, respectively; t is the time; r_0 and r are the initial and current radii of the particle reacting surface; B is the mass particle concentration in the suspension; λ_g is the coefficient of thermal conduction of the gas; C and C_g are the respective specific heat capacities of particle and gas; ρ, ρ_0, and ρ_g are the densities of particle, oxides, and gas, respectively; q is the heat of reaction per unit mass of condensed particle material; E is the activation energy; a_0 is the oxidizer concentration in the gaseous phase; R is the molar gas constant; m is the reaction order with respect to the oxidizer; ν is the ratio of mass of oxidizer to mass of particle in the stoichiometric suspension; Nu and Nu_g are the respective Nusselt numbers for particle–gas and suspension–container wall heat exchange; L and V are the characteristic dimension and volume of the container; Σ is the heat-transfer surface area of the container; N is the number of particles per unit volume of suspension; and s and s_1 are given by $s = \nu$, $s_1 = [(1+\nu)(\rho/\rho_0) - 1]$ and $s = -1$, $s_1 = -1$ for the condensed and gaseous reaction products cases.

The first term on the right side of Eq. (1.1) describes the rate of liberation of heat on the particle surface; this same term also appears in the expression for the rate of particle burning. The second term on the right of Eq. (1.2) describes the rate at which heat builds up in the gas as a result of mass transfer between particles and gas. The remaining terms of this equation describe particle–gas and suspension–container wall heat exchange. The form of the kinetic function in Eq. (1.4) is such as to allow for oxidizer combustion and alteration in the reacting surface area of the particle.

In setting up the expression for the particle–gas thermal energy flux [second term of Eq. (1.1)], account was taken of the fact that the heat-transfer coefficient is inversely proportional to the particle diameter, and the heat-transfer surface area, directly proportional to the second power of this diameter.

The discussion that follows will center around three limiting forms of kinetic function, namely,

$$\varphi_1 = (1-\eta)^{2/3}, \quad \varphi_2 = (1-\eta)^{m+2/3}, \quad \varphi_3 = \left(1 - \frac{\nu}{a_0}\frac{B'}{1-B}\eta\right)^m \tag{1.6}$$

φ_1 applying when the oxidizer is present in excess and $\nu B / a_0(1-B) \ll 1$, φ_2 applying to the stoichiometric mixture with $\nu B/a_0(1-B) = 1$, and φ_3 applying when there is an oxidizer deficiency and $\nu B/a_0(1-B) \gg 1$. This last case can be formally reduced to that in which the oxidizer is present in excess by making the change in variable $\eta' = \eta\nu B/a_0(1-B)$ and replacing the parameter γ by $\gamma' = \gamma\nu B/a_0(1-B)$.

2. Let us now investigate critical ignition effects in the gaseous suspension. The possibility of such effects arises from the interaction of heat loss from suspension to container walls and burning of the limiting component, particles or oxidizer, as the case may be. Nondegenerate critical ignition ($\gamma \ll 1$) has already been discussed in [2-5].

Here the effect of the parameter γ on the gaseous suspension ignition limit was first studied through numerical integration of the system of equations (1.1)-(1.5), assuming reaction with an excess oxidizer to form condensed reaction products and identity of initial temperatures of suspension and walls ($\theta_0 = \theta_{g0} = \theta_W = 0$). The results of these calculations are shown in Fig. 1, where the critical value of the parameter $\varkappa$ has been plotted as a function of A and γ with $\beta = 0$: $\varkappa_c = \varkappa_c(A, \gamma)$. Curve 1 applies to the case in which no allowance for the effect of burning was made in calculating the heat loss ($\gamma_0 = 0$) [2]

$$\varkappa_c = 1/(1+A)e \tag{2.1}$$

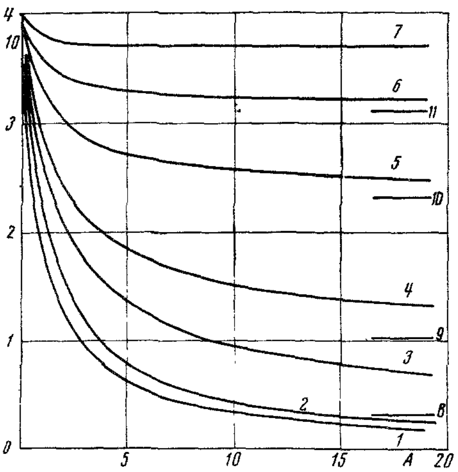

Fig. 1

while curves 2-7 apply to the overall critical effect ($\gamma_0 = 0.016$) at various concentrations: 2) $B = 0.32$, $\gamma = 0.05$; 3) $B = 0.08$, $\gamma = 0.2$; 4) $B = 0.0532$, $\gamma = 0.3$; 5) $B = 0.0318$, $\gamma = 0.503$; 6) $B = 0.0228$, $\gamma = 0.7$; 7) $B = 0.016$, $\gamma = 1$.

The line segments 8-11 show the positions assumed by curves 3-6 in the limit as $A \to \infty$. Curves 2-7 converge to a single point at $A = 0$ thus marking out critical ignition conditions for the isolated particle, account being taken of burning ($\gamma_0 \neq 0$). The effect of burning on isolated particle ignition has been treated in [6], where the shift in critical conditions resulting from this factor has been calculated through the equation

$$\varkappa_0 = \frac{1}{e}\left[1 + 2.7\left(\frac{2}{3}\gamma_0\right)^{2/3}\right] \tag{2.2}$$

An increase in the parameter A reduces $\varkappa_c$, which then asymptotically approaches the value $\varkappa_i$ fixed by γ; in this range of A values, ignition is limited by critical particle burning, a factor independent of the heat transfer out of the system [2, 3]. The ratio $\varkappa_i/\varkappa_0(\gamma_0)$ has been plotted as a function of γ in Fig. 2 (curve 1). With an increase in γ, the critical conditions for gaseous suspension ignition asymptotically approach the critical conditions for isolated particle ignition, remaining, however, clearly expressed at all values of γ. In distinction to the case of the gaseous suspension, passage into the degenerate thermal explosion region leads to a disappearance of critical ignition effects in homogeneous gaseous and condensed systems [8].

The results of calculations on the critical ignition effect $\varkappa_c = \varkappa_c(A, \gamma)$ could be satisfactorily reproduced by the approximation equation

$$\varkappa_c = \varkappa_i(\gamma) + [\varkappa_0 - \varkappa_i(\gamma)]\frac{1}{1+A} \tag{2.3}$$

Using this equation, it was possible to mark out a region of parameter values over which the critical effect and induction period are only weakly dependent on the heat loss. The gaseous suspension can be considered adiabatic when the conditions are such that

$$\varkappa > \varkappa_c \gg [\varkappa_0 - \varkappa_i(\gamma)]/(1 + A) \tag{2.4}$$

In determining the form of the $\varkappa_i(\gamma)$ function, use was made of the fact that preexplosion warm-up of the gaseous suspension is a quasistationary process [3], the particle–gas thermal equilibrium being displaced toward the particle side as a result of gas heating and an alteration in the rate of evolution of thermal energy. If the derivative term in the expression for particle warm-up, (1.1), is neglected, an algebraic equation of the form $F(\theta, \theta_g, \eta) = 0$ results. When the condition of (2.4) is satisfied, heat loss through the container walls has no longer any effect on ignition, and the system of equations (1.1)-(1.3) reduces to a total energy balance equation for the gaseous suspension,

$$B\theta + (1 - B)\theta_g = \eta/\gamma - sB\eta(\theta - \theta_g)$$

The nonlinear term $sB\eta(\theta - \theta_g)$ in the total energy balance equation can be neglected, since it will be small in comparison with the other terms if the conditions are such that $sB\gamma \equiv s\gamma_0 \ll 1$; and this is generally the case, γ_0 being much less than unity in systems capable of ignition. We will now set up the $F(\theta, \theta_g, \eta) = 0$ equation for the case of condensed reaction products, neglecting the alteration in particle diameter resulting from oxidation and the difference in particle and oxide densities (e.g., for magnesium particles with $s_1 = -0.2$, complete oxidation reduces the particle diameter by some 7%).

By solving the two simplified transcendental algebraic equations, a quasistationary relation between the extent of particle burning η and the particle temperature is obtained:

$$\eta = 1 - \gamma \mid \gamma^{-1} - \theta \mid \Psi^3(U, K) \tag{2.5}$$

The K parameter of this last equation describes the reaction conditions: $K = 2/3$, for the case of excess of oxidizer and condensed reaction products; $K = 1/3$, for the case of excess of oxidizer and gaseous reaction products; $K = 2/3 + m$, for the case of stoichiometric proportions, particles to oxidizer, and condensed products; $K = 1/3 + m$, for the case of stoichiometric proportions, particles to oxidizer, and gaseous products; and $K = m$, for the case of oxidizer insufficiency.

$$U = \exp(z)\exp[-(\gamma^{-1}-\theta)] / |\gamma^{-1}-\theta|^{1-k}$$
$$z = \gamma^{-1} - K\ln\gamma^{-1} + \ln[\varkappa(1-B)]$$

The function $\Psi(U, K)$ is to satisfy the equation

$$\Psi^3 - U\Psi^{3k} \mp 1 = 0 \qquad (2.6)$$

the upper sign applying when $\theta < \gamma^{-1}$ and the lower, when $\theta > \gamma^{-1}$.

Calculations based on the equation system (1.1)-(1.5) show the integral curve $\eta[\theta(\tau)]$ to be essentially identical with the curve of Eq. (2.5) as long as it is possible for a quasistationary particle–gas thermal equilibrium to be maintained; beyond this point the difference between the temperatures of the two phases changes discontinuously and the particles in the suspension ignite. This point is reached in the neighborhood of that temperature θ_2 at which Eq. (2.5) no longer can be solved for θ. The value of θ_2 is obtained from the equation $d\theta/d\eta = \infty$, or its equivalent $d\eta/d\theta = 0$.

$$\gamma^{-1} - \theta_2 = \Phi(z, K) \qquad (2.7)$$

The function $\Phi(z, K)$ satisfies the transcendental equation

$$z = \Phi - K\ln\left[\left(K + \frac{1}{3}\right) + \Phi\right] + (1-K)\ln\left[1 - \frac{K - {}^2/_3}{\Phi + (K - {}^2/_3)}\right] \qquad (2.8)$$

Particle ignition at temperatures in excess of θ_2 will initiate an autocatalytic reaction, regardless of how small the heat of reaction per unit mass of suspension may be. The situation here is markedly different from that met in homogeneous systems, where a reduction in the heat of reaction abruptly quenches the autocatalytic reaction [8].

Gaseous suspension particle ignition occurs at those values of the parameters z and K for which Eq. (2.8) has a solution, a condition which cannot be met at critical values of the burning rate.

It is also true that (2.8) does not have a solution when the conditions are such that $d\Phi/dz = \infty$, the function Φ then satisfying an equation of the form

$$[\Phi_i + {}^1/_3] / [\Phi_i + K + {}^1/_3] = (1-K)({}^2/_3 - K) / \Phi_i[\Phi_i + (K - {}^2/_3)] \qquad (2.9)$$

Φ_i being the value of Φ at the critical burning rate.

The value of Φ_i obtained from Eq. (2.9) can be substituted into (2.8), and the parameter z given the value z_i, to obtain a relation between the parameters values at the critical burning rate, namely,

$$\ln\frac{\varkappa_0(0)}{\varkappa_i} = (\gamma^{-1} - K) + K\ln(\gamma K) - \ln[f(K)] + \ln(1-B) \qquad (2.10)$$

$$f(K) = \exp[\Phi_i + 1 - K]\frac{\Phi_i}{\Phi_i + (K - {}^2/_3)}\left[\frac{\Phi_i - (K - {}^2/_3)}{\Psi_i} + \frac{(1-K)(K - {}^2/_3)}{\Phi_i^2}\right]^K$$

$\varkappa_0(0)$ being the critical value of $\varkappa$ for the isolated particle with no allowance for burning effects ($\gamma_0 = 0$), and $\varkappa_0(0) = e^{-1}$. With $\gamma \ll 1$, the critical conditions of (2.10) for burning pass over the critical conditions for nondegenerate thermal ignition, the relation then becoming identical with that obtained earlier in [2, 3].

Computations show that preexplosion warm-up begins with an essentially nonstationary particle heating, and then passes over into a quasistationary stage, where the greater part of the particle burning is concentrated. The derivation of Eq. (2.10) considered only this second, quasistationary stage, neglecting particle burning in the first stage, and it is for this reason that the equation itself contains the parameter $\varkappa_0(\gamma_0)$ at $\gamma_0 = 0$.

Let us now consider in more detail the condensed reaction products case with $s = \nu$, $K = {}^2/_3$. For this value of K, Eqs. (2.7) and (2.9) have the solutions $\theta_2 = \gamma^{-1} + {}^1/_3$, $\Phi_i = -{}^1/_3$, and $f(K)$ is equal to unity. Curve 2 of Fig. 2 shows the $\varkappa_i/\varkappa_0(0)$ ratio plotted as a function of the parameter γ at $K = {}^2/_3$. Although the approximation to critical ignition conditions for the gaseous suspension given by Eq. (2.10) for $\gamma \geq {}^3/_2$ is identical with the critical ignition conditions for the isolated particle, numerical computations (curve 1, Fig. 2) indicate that there should be an asymtotic approach of the one set of conditions to the other. To a considerable degree of accuracy it could be considered that that there is essentially no departure from $\varkappa_0(\gamma_0)$ in calculations at $\gamma \geq {}^3/_2\,\varkappa_i$.

We will now attempt to find the effect on the extent of burning of departures from the critical burning rate. Equations (2.5) and (2.6) can be drawn on to show that at θ_2, the temperature of cut-off from

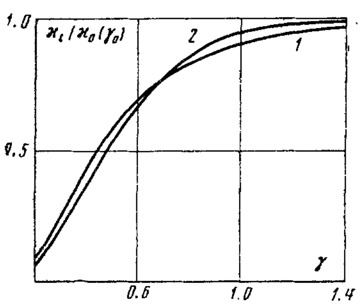

Fig. 2

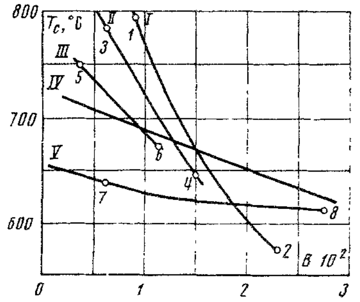

Fig. 3

TABLE 1

No.	1	2	3	4	5	6	7	8
r_0, μ	7	7	12	12	15	15	55	55
$B\cdot10^2$	0.94	2.3	0.64	1.5	0.33	1.13	0.64	2.78
$\gamma_0\cdot10^3$	4.1	2.3	3.9	2.7	3.6	3.1	3.5	2.9
γ	0.43	0.10	0.60	0.18	1.12	0.27	0.54	0.10
A	5.55	13.8	1.28	3.12	0.42	1.45	0.06	0.34
$\varkappa_i/\varkappa_0(0)$	0.45	e^{-7}	0.67	0.04	0.95	0.16	0.6	e^{-7}
T°_c, °C	875	875	810	810	775	775	650	650
T_c, °C	787	572	772	647	757	677	641	622
$T_c(\gamma=0)$	672	572	717	637	732	672	637	622
$T_c(A=\infty)$	767	377	752	427	757	602	607	302

the quasistationary state, ψ should satisfy the equation

$$\Psi^3 \mp 1 = 1/|\gamma^{-1} - \theta_2| \tag{2.11}$$

By substituting (2.11) into (2.5), one obtains an expression for the extent of particle burning at the instant of ignition (η_2),

$$\eta_2 = \gamma(\theta_2 - 1) \tag{2.12}$$

θ_2 being calculated by substituting the solution of (2.8) $\Phi(z, \,^2\!/_3)$ into Eq. (2.7). The extent of particle burning at the critical burning rate, η_i, is obtained from Eq. (2.12) by setting

$$\eta_i = 1 - {}^2\!/_3\,\gamma \tag{2.13}$$

This last relation shows that the gaseous suspension can be ignited only if the degree of burning is less than $\eta_i(\gamma)$. If the degree of burning in preexplosion warm-up exceeds $\eta_i(\gamma)$, the gaseous suspension will not ignite under further heating. Equation (2.13) is valid for cases such that $\gamma < {}^3\!/_2$, i.e., for cases in which there is still a difference between critical ignition conditions for gaseous suspension and isolated particle; when $\gamma > {}^3\!/_2$, one should consider that $\eta_i = 0$ in the quasistationary approximation. Computations show that η_i approaches the degree of burning of the isolated particle under critical conditions $\eta_0(\gamma_0)$ [6]

$$\eta_0(\gamma_0) = 2.7\,(\gamma_0^2 \,/\, {}^2\!/_3)^{1/3} \tag{2.14}$$

the latter value proving to be low under any realistic assumptions concerning the parameter γ_0 (when $\gamma_0 = 0.016$, $\eta_0(\gamma_0) \approx 0.2$).

A relation between the degree of particle burning and departure from the ignition limit measured by $\varkappa$ developed from the approximation equation (2.12), gave results which were in satisfactory agreement with values obtained from computations based on the system of equations (1.1)-(1.5). Increasing the value of the parameter $\varkappa$ from $\varkappa_i = 0.0025$ to 0.0425 at $\gamma = 0.117$ reduced the degree of particle burning, the reduction being from 0.922 to 0.4, according to the computations based on (1.1)-(1.5), and from 0.922 to 0.32, according to the approximation equation.

3. We will now illustrate the type of relation between the critical ignition temperature T_c, and the particle size and concentration, which follows from the equations presented above. The heat of reaction and the particle heat capacity will be set at 6 kcal/g and 0.25 cal/g · deg, respectively. Application of Eq. (2.3) requires a knowledge of the functions $\varkappa(r_0, T_c)$ and $A(r_0, B)$, (which reflect the experimental conditions) and $\varkappa(r_0, T_c)$; the latter can be obtained from the experimentally developed relation between the critical ignition temperature (T_c^0) and the isolated particle diameter [9], namely,

$$\varkappa = 10^{9.3}\,(r_0/T_c^2)\exp[-23\,000\,/\,RT_c] \tag{3.1}$$

The $A(r_0B)$ function was so chosen that the value of A for particles 55 μ in diameter would be less than unity at all concentrations

$$A = 3\cdot10^4\frac{B}{r_0^2(1-B)} \tag{3.2}$$

Figure 3 shows the relation between particle concentration and the critical ignition temperature of the gaseous suspension for particles of various diameters. Curves I-V correspond to the particle diameters I) 7, II) 12, III) 15, IV) 35, V) 55 μ. The numbers 1-8 designate points at which values of T_c ($\gamma = 0$) and T_c ($A = \infty$), critical suspension ignition temperatures uncorrected, respectively, for particle burning and thermal loss, were calculated; these values are shown in Table 1.

It is seen from the table that the critical ignition temperature was determined by burning at points 1, 3, and 5 and by heat loss to the reactor walls at points 2, 4, 6, 7, and 8. On curves I, II, and III a reduction in concentration at fixed particle diameter results in passage into the region of degenerate ignition; γ increases, T_c approaches $T^0{}_c$, and the effect of burning on the critical ignition temperature begins to predominate over that of heat loss (cf. Table 1).

Comparison shows general agreement between the theoretical T_c (r_0, B) relation of Fig. 3 and the experimental results reported in [9], though the experimentally developed curves are displaced with respect to the theoretical by a 1:6 scale factor on the concentration axis. The fact that the working conditions for the experiments of [9] were not fully specified makes for difficulty in comparing theoretical and experimental results.

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