Ignition Analysis of Adiabatic, Homogeneous Systems Including Reactant Consumption

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Ignition of reactants initially at some moderate temperature, with reactant consumption effects included by introducing the adiabatic flame temperature and an effective specific heat into a First Law analysis, is analyzed using a thermal model, for first- and second-order reactions. After the normal Zel'dovich expansion of the Arrhenius term, the approximate equation so obtained is analytically solvable using a Poincaré expansion method. The resulting solution, in the form of a truncated series, is in excellent agreement with numerical integration of the approximate equation. Applying the Poincaré method to an extended Zel'dovich type expansion yields a solution for first-order reactions that is acceptably close to that obtained by numerical integration of the governing differential equation before transformation. A method is developed for obtaining approximate thermal theory activation energy and pre-exponential factor values for a system of reactants, using experimental data and this solution.

Nomenclature

A $= \gamma^2+3\gamma+3$, constant in Eq. (10)
B $= \gamma+1$, constant in Eq. (10)
$\bar{c}$ = mean specific heat of mixture, kcal/m^3 °K, and defined as equalling $QC_{f,o}/(T_f-T_o)$
$C_{f,o}$ = initial mass concentration of fuel, kg/m^3
E = activation energy of chemical reaction, kcal/mole
F_i, f_i = dimensionless functions
$g_m(u)$ = general term of series, see Eqs. (5–7)
k = mass oxidizer/fuel stoichiometric ratio, or integration constant
n = order of reaction
Q = heat of combustion of fuel, kcal/kg
R = universal gas constant 1.986 kcal/mole °K
T, T_o, T_f = local, initial, and adiabatic flame temperature of gas mixture, °K
t = real time, sec
$\hat{t}$ $= t(k\bar{c}T_o/Q)^{n-1}Z$, scaled time variable, dimensionless
u = running dimensionless time variable
Z = pre-exponential factor of reaction, sec^{-1} or m^3 kg^{-1} sec^{-1}
β $= \varepsilon\gamma = RT_o^2/E(T_f-T_o)$, a small parameter
γ $= T_o/(T_f-T_o)$, a constant
ε $= RT_o/E$, dimensionless small parameter
θ $= E(T-T_o)/RT_o^2$, dimensionless temperature
τ $= \hat{t}/\varepsilon\gamma^n \exp(1/\varepsilon)$, dimensionless time

Introduction

IT is well-known[1,2] that thermal models of the combustion of premixed gases (and solids) can adequately describe the reaction process as long as branched-chain reactions do not occur under the prescribed conditions of pressure and temperature. As reviewed by Kondrat'ev,[2] thermal models accurately predict the critical conditions for explosion in such systems, and have the form of the "Stationary State" model described by Frank-Kamenetski.[1] The Stationary State model inquires into the possible, steady-state, temperature distributions a system may exhibit when the maximum temperature in the system is not very much larger than the initial temperature. When the system can no longer support a low maximum temperature, explosion occurs. The value of the Stationary State theory is that the question of critical conditions for explosion is answered in terms of the initial pressure, temperature, container size, and effective chemistry of the reactants concerned.

Thermal modeling has been used to predict laminar flame speeds for premixed gases[3,4] and in models for various diffusion flames such as Burke and Schumann's flowing gas system[5] and the liquid droplet burning models of Godsave[6] and later authors. Similarly, thermal modeling of reactant chemistry has also been used in examining the diffusive ignition of polymeric fuels[7] and in homogeneous and composite solid propellants,[8,9] with seemingly reasonable success.

In spite of all these applications, even in ignition processes of initially unmixed systems, the basically elementary thermal model of the "Nonstationary State" of a premixed, reactable system[1] has not undergone the same development of understanding and usefulness as has been done for the Stationary State case. This is unfortunate both from the point of view of fundamental, theoretical understanding of combustion processes, and the subsequent possible difficulty in interpreting data from ignition experiments involving premixed gases. In this regard, it would be useful to be able to supply data obtained from ignition experiments to questions of critical explosion conditions, for the same reactants, using Stationary State thermal theory. The reason for this lack of development appears to be the difficulty in finding a general method of solving the mathematical form of the thermal model.

One of the simplest of such problems is that encountered in the adiabatic ignition of premixed gases, either in a shock tube or a mechanical, adiabatic compression device. The solution to this problem is described in this paper; the solution of the Nonstationary State will be the subject of another paper.

Model Formulation

Assume that at zero time, a specified mass of premixed, combustible gas is uniformly brought to a specified temperature and pressure, and that the total ignition delay is sufficiently short to neglect heat losses from the bulk of the system. The system can then be regarded as adiabatic. It is assumed that the chemistry can be represented by an over-all reaction using an Arrhenius rate expression. Then, after using the first law of thermodynamics to relate temperature to reactant concentrations,[2] one obtains the following equation which describes the

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Presented as Paper 73-215 at the AIAA 11th Aerospace Sciences Meeting, Washington, D.C., January 10–12, 1973; submitted February 6, 1973; revision received May 31, 1973. Research supported by the Royal Institute of Technology, Institute for Gasdynamics, Stockholm, Sweden.

Index categories: Combustion Stability, Ignition, and Detonation; Combustion in Gases.

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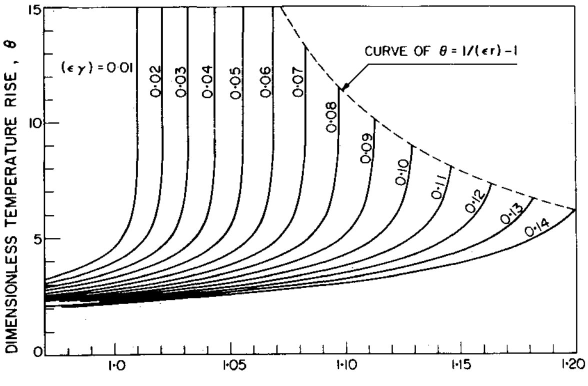

Fig. 1 Solution plant plot, θ vs τ, for adiabatic ignition involving a first-order reaction and classical expansion of exponential argument.

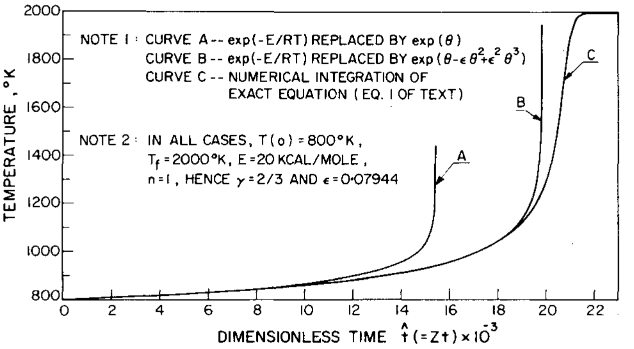

Fig. 2 Comparison of temperature T vs dimensionless real time t behavior predicted by exact and approximate equations for adiabatic ignition and first-order reaction with $E = 20$ kcal/mole.

development of the explosion for a reaction of over-all order n and a mass oxidizer/fuel stoichiometric ratio k:

$$dT/d\hat{t} = (T_f - T)^n \exp(-E/RT); \quad T(0) = T_o \tag{1}$$

Here $\hat{t} = Z(k\bar{c}T_o/Q)^{n-1}t$ where Z is the pre-exponential factor of the reaction. Q is the heat of combustion per unit mass of fuel, T_f is the real adiabatic combustion temperature, E is the over-all activation energy, and k is the reaction stoichiometry constant (A/F or $0/F$ ratio). The effective specific heat $\bar{c}$ of the mixture, per unit volume, is defined as

$$\bar{c} = QC_{f,o}/(T_f - T_o)$$

where $C_{f,o}$ is the initial fuel mass concentration.

To solve Eq. (1), one expands the exponential argument in terms of the deviation of T from T_o as demonstrated in Refs. 1 and 2. In this case, however, we carry the expansion somewhat further, obtaining

$$d\theta/d\tau = [1-(\varepsilon\gamma)\theta]^n \exp(\theta - \varepsilon\theta^2 + \varepsilon^2\theta^3); \quad \theta(0) = 0 \tag{2}$$

where $\theta = E(T-T_o)/RT_o^2$, $\tau = \hat{t}/\varepsilon\gamma^n \exp(1/\varepsilon)$, $\varepsilon = RT_o/E$, and $\gamma = T_o/(T_f - T_o)$. This problem will be referred to as the "extended" problem.

Solution Description and Discussion

It can be shown that a regular perturbation solution of (2) is incorrect; it is found that τ can never exceed unity, regardless of the value of $(\varepsilon\gamma)$. This indicates that perhaps both θ and τ are functions of (ε), leading naturally to the idea of applying a Poincaré expansion of the form

$$\begin{aligned} \theta &= F_o(u) + \varepsilon F_1(u) + \varepsilon^2 F_2(u) + \cdots \\ \tau &= u + \varepsilon f_1(u) + \varepsilon^2 f_2(u) + \cdots \end{aligned} \tag{3}$$

where both $F_i(u)$ and $f_i(u)$ are functions to be determined.

Insertion of Eq. (3) into Eq. (2), and applying Lighthill's principle in a somewhat altered form yields useful solutions to the problem. Details of this calculation are shown in the Appendix for the case when $n = 1$ and the higher-order terms, θ^2 and θ^3, are dropped in the exponential argument of Eq. (2). Thus, for an nth order reaction we wish to solve the equation

$$d\theta/d\tau = [1-(\varepsilon\gamma)\theta]^n \exp(\theta); \quad \theta(0) = 0 \tag{2a}$$

The solution, using the Poincaré method, is given by the following two equations for θ and τ as functions of the new time variable "u"

$$\theta = -\ln(1-u) \tag{4}$$

and

$$\tau = u + \sum_{m=1}^{\infty} (\varepsilon\gamma)^m g_m(u) \tag{5}$$

where, for $n = 1$

$$g_m(u) = [mg_{m-1} - (-1)^m(1-u)\ln^m(1-u)]; \quad g_o = u \tag{6}$$

and, for $n = 2$

$$g_m(u) = (m+1)[g_{m-1} - (-1)^m(1-u)\ln^m(1-u)]; \quad g_o = u \tag{7}$$

The series resulting from inserting Eqs. (6) or (7) into Eq. (5) are, in principle, only semiconvergent. However, for reasonable values of $(\varepsilon\gamma)$, using the first three to six terms produces results which, when plotted, are indistinguishable from Runge-Kutta integration of Eq. (2a). This indicates acceptable accuracy in using solutions (4, 5, and 6 or 7). Assuming $n = 1$ and six terms in the series, Eqs (3, and 4 and 5) are plotted in Fig. 1 for a range of $(\varepsilon\gamma)$ values. Calculations were terminated, where applicable for the scale chosen, at $\theta = (1/\varepsilon\gamma) - 1$, where $d\theta/d\tau$ is a maximum. The τ values associated with this maximum value of θ can be acceptably calculated by setting $u = 1$ in Eqs. (5) and (6) or (7) with the result that, for $n = 1$

$$\tau_{\infty_1} = 1 + (\varepsilon\gamma) + 2(\varepsilon\gamma)^2 + 6(\varepsilon\gamma)^3 \tag{8}$$

and, for $n = 2$

$$\tau_{\infty_2} = 1 + 2(\varepsilon\gamma) + 6(\varepsilon\gamma)^2 + 24(\varepsilon\gamma)^3 \tag{9}$$

Comparison—for $n = 1$—of the preceding solution with numerical integration of Eq. (1)—curves "C"— is shown in Figs. 2 and 3, assuming $E = 20$ kcal/mole and 40 kcal/mole, respectively, with $T_o = 800$°K, and $T_f = 2000$°K. In each case T vs $\hat{t}(=Zt)$ is plotted, using three terms of the series (5) and (6); the corresponding curves are labelled "A" in the figures.

However, a much better approximation to Eq. (1) is obtained using Eq. (2) and applying the same type of Poincaré expansion. For first-order reactions ($n = 1$), the result—to order ε^2—is

$$\theta = -\ln(1-u) \tag{10}$$

$$\begin{aligned} \tau = u &+ \varepsilon\{(\gamma+2)[u + (1-u)\ln(1-u) - \ln^2(1-u)]\} + \\ &\varepsilon^2\{2Au + (1-u)[2A\ln(-u) - A\ln^2(1-u) + \\ &B\ln^3(1-u) - (\tfrac{1}{2})\ln^4(1-u)]\} \end{aligned} \tag{11}$$

where $A = \gamma^2 + 3\gamma + 3$ and $B = (\gamma + 1)$. Equations (10) are

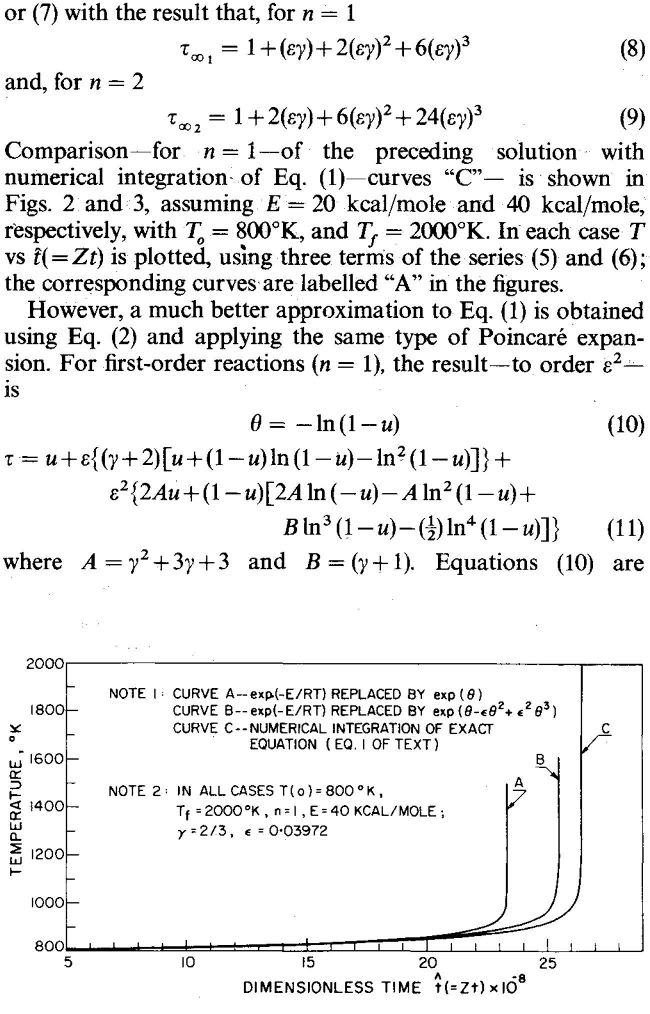

Fig. 3 Comparison of temperature T vs dimensionless real time t behavior predicted by exact and approximate equations for adiabatic ignition and first-order reaction with $E = 40$ kcal/mole.

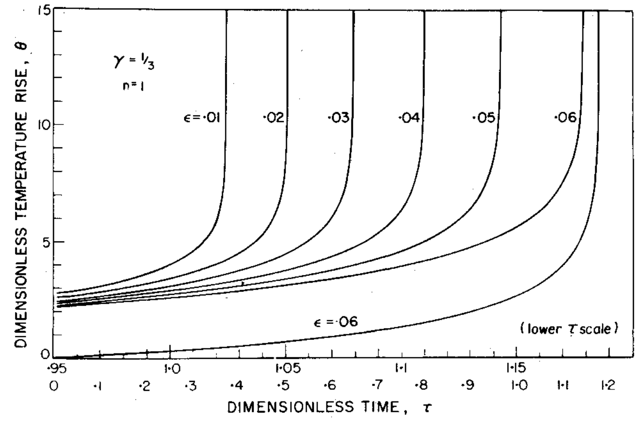

Fig. 4 Dimensionless temperature rise θ vs dimensionless time τ, parametric with ε for $\gamma = 1/3$, for the extended problem and a first-order reaction, Eqs. (10) of text.

plotted for a range of ε values in Figs. 4 and 5 for $\gamma = \frac{1}{3}$ and $\gamma = \frac{2}{3}$, respectively. As before, setting $u = 1$, the τ asymptote is acceptably calculable from

$$\tau_\infty = 1 + (\gamma + 2)\varepsilon + (2\gamma^2 + 6\gamma + 6)\varepsilon^2 \tag{12}$$

which is plotted in Fig. 6 for a range of ε, γ values. T vs $\hat{t}$, using Eq. (11) are shown as curves "B" in Figs. 2 and 3; a marked improvement compared to using Eqs. (5) and (6) is clearly evident.

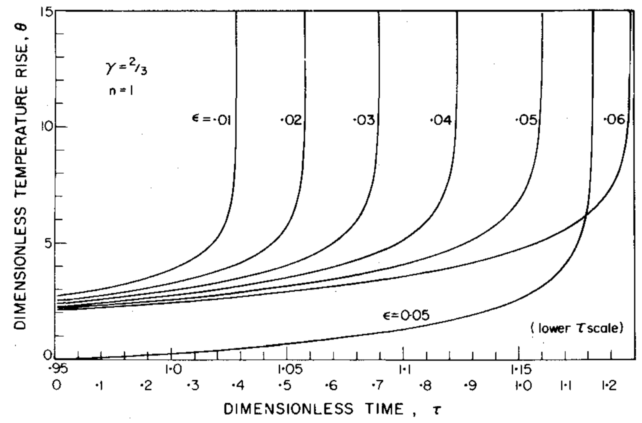

Fig. 5 Dimensionless temperature rise θ vs dimensionless time τ, parametric with ε for $\gamma = 2/3$, for the extended problem and a first-order reaction, Eqs. (10) of text.

Application

The method of extracting the values of E and Z from experimental data is as follows for the first-order ($n = 1$) reactions.† For the same reactant concentrations, and two different initial temperatures, determine the experimental times, $t_i{}^*$, required to each (dT/dt) maximum, and the adiabatic flame temperature for each; thus, two values of t^*, T_o, T_f are known. Theoretically, then, assuming τ_∞ adequately represents the dimensionless time to reach ($d\theta/d\tau$) maximum, and that ($d\theta/d\tau$) maximum and (dT/dt) maximum occur at the same temperature values,‡ we can show that

$$t_i = \varepsilon_i \gamma_i \exp(1/\varepsilon_i) \cdot \tau_{\infty_i}(\varepsilon, \gamma)/Z \tag{13}$$

where τ_{∞_i} is

$$\tau_{\infty_i}(\varepsilon_i, \gamma_i) = 1 + (\gamma_i + 2)\varepsilon_i + 2A_i \varepsilon_i{}^2; \quad A_i = 2\gamma^2 + 6\gamma + 6$$

Since for given reactants, E and Z are presumably constant, the ratio of t_i values is independent of Z and *a* unique function of activation energy alone. Select a trial E value, calculate ε, γ, and τ, for each T_o value, and form the ratio of $t_i{}^*$ values such that

$$\frac{t_1{}^*}{t_2{}^*} = \frac{\varepsilon_1 \gamma_1 \, e^{1/\varepsilon_1} \, \tau_\infty(\varepsilon_1, \gamma_1)}{\varepsilon_2 \gamma_2 \, e^{1/\varepsilon_2} \, \tau_\infty(\varepsilon_2, \gamma_2)} \tag{14}$$

Vary E in (14) until equality is obtained. Once E, and hence ε, is known, Z can be determined by rewriting Eq. (13) such that

$$Z = [\varepsilon_i \gamma_i e^{1/\varepsilon_i} \tau_\infty(\varepsilon_i, \gamma_i)/t_i{}^*] \tag{15}$$

In order to estimate the potential accuracy of using the data reduction procedure just described, suppose that curves "C" in Figs. 2 and 3 represent experimental temperature-time traces of two different (first-order) reactions having known E and Z values. Thus $T(\hat{t})$, $t(\hat{t})$, and $\hat{t}^*$—when $dT/d\hat{t}$ is a maximum—are known; the known E values are 20 and 40 kcal/mole, respectively. Since T_f and T_o are also known, we can compute the quantities γ and ε—for a selected E value—as needed. Using Eq. (12) to calculate τ_∞, we search for an E value such that the value of $\hat{t}$, derived from Eq. (16), equals $\hat{t}^*$, from

$$\hat{t} = \tau_\infty(\varepsilon, \gamma) \cdot \varepsilon\gamma \exp(1/\varepsilon) \tag{16}$$

When this is done for the case at hand, E values of 20.076 and 40.058 kcal/mole are required, respectively, indicating acceptable accuracy may be achieved using the solutions and methods presented herein. Note that use of these derived E values in determining critical Stationary State explosion conditions would imply critical size dimensions only ~2% greater than those calculated using the "correct" E values. This is probably as good as one can expect a thermal theory to be. A higher-order solution —such as that for the external problem but carrid out to $0(\varepsilon^3)$— would still further improve the potential accuracy. But there would be a considerable increase in complexity, making such an extension of doubtful real value.

Consequently the solution derived in this paper can be used to extract values of E and Z from adiabatic ignition experiments, with reasonable accuracy. The values thus obtained can then be used for other situations when thermal modeling is desired.

Summary

A method of analyzing the problem of ignition of adiabatic, homogeneous mixtures of reactants, including the effects of reactant consumption, has been described, and several solutions shown. Although the derived solutions are of series rather than

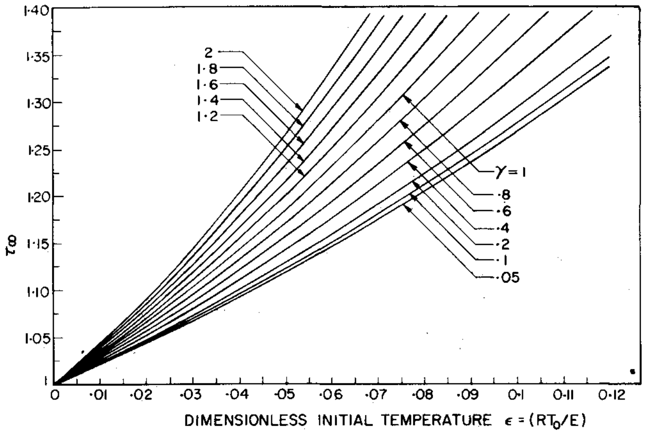

Fig. 6 Asymptotic dimensionless time τ_∞ for the extended problem [from Eq. (11)] vs dimensionless initial temperature ε for a first-order reaction.

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† First-order reactions are stipulated because the solution of the extended problem has only been determined in this case. If one is satisfied with using solutions for Eq. (2a) then Eqs. (13) and (15) may be multiplied by the factor $(\gamma_i k \bar{c}_i T_{o_i}/Q)^{n-1}$ and Eq. (14) by $(\gamma_1 \bar{c}_1 T_{o_1}/\gamma_2 \bar{c}_2 T_{o_2})^{n-1}$ to make them hold for $n = 1, 2$.

‡ Comparison of the plots resulting from using Eqs. (10), and Runge-Kutta integration of Eq. (1), indicate this is a reasonably accurate assumption for E values of some 30 kcal/mole or more.

closed form, only a few terms of the series need be calculated to obtain acceptable accuracy. In the case of first-order reactions with extended expansion of the Arrhenius term, use of only three terms of the resulting series is sufficiently accurate, in comparison with numerical solution of the exact equation describing the problem, to deduce effective activation energy and pre-exponential factor values for the reaction involved. A method of determining these values from experimental data is given. Thus it is shown how the results of adiabatic ignition experiments can be treated to obtain reliable over-all chemical parameters for use in other combustion situations involving the same reactants.

Appendix: The Method of Solution

Essentially, the method used to solve Eq. (2) of the text is a Poincaré, two variable expansion method described by Cole,[10] Ames,[11] and others. Apparently use of this method has been primarily associated with oscillatory, nonlinear systems where the oscillation frequency, and possibly the amplitude, depend upon the size of a small parameter multiplying the nonlinear term. The analysis of the present problem proceeds along the same lines as those of Refs. 10 and 11, but with an important difference. In the oscillatory problems, solutions leading to stable oscillations are sought, and the unknown functions forming the complete solution are determined, in part, by the necessity of removing "secular" terms from the solution. However, the problem of adiabatic ignition inherently consists of a solution exhibiting an exponential growth in the size of the dependent variable (temperature). It is the explosion time which depends on the small parameter in the systems; when this parameter is zero, the differential equation is still nonlinear but solvable in closed form. Therefore a different criterion for determining the unknown functions assumed in the solution must be used as the concept of "secular" terms no longer applies. The criterion used in this case was a form of Lighthill's criterion discussed by van Dyke,[12] to be described subsequently.

For simplicity, we shall only consider the first-order chemical reaction case, when $n = 1$, and the classical Zel'dovich expansion for E/RT in the argument of the exponential to illustrate the method of solution. In this case the energy equation takes the form

$$d\theta/d\tau = (1-\beta\theta)\exp(\theta); \quad \theta(0) = 0 \tag{A1}$$

where $\beta = \varepsilon\gamma$ and normally is much less than unity. Incidentally, it is easily shown that $(d\theta/d\tau) = 1$ when $\theta = 0$ $(d\theta/d\tau)$ is a maximum at $\theta = [(1/\beta)-1]$, and $(d\theta/d\tau) = 0$ at $\theta = 1/\beta$; note that for small β, there should be little difference in τ values between $(d\theta/d\tau)$ and θ reaching their respective maximum values.

The solution of (A1), using the Poincaré method, proceeds by expanding both θ and τ in terms of β and a new time variable u such that

$$\theta = F_o(u)+\beta F_1(u)+\beta^2 F_2(u)+\cdots \tag{A2}$$

$$\tau = u+\beta f_1(u)+\beta^2 f_2(u)+\cdots \tag{A3}$$

where $F_i(u)$ and $f_i(u)$ are functions to be determined.

Then, observing that $(d\theta/d\tau) = (d\theta/du)(du/d\tau) = (d\theta/du)/(d\tau/du)$, we differentiate (A2) and (A3) with respect to (u) and perform the necessary division to obtain, with primes referring to differentiation with respect to u,

$$d\theta/d\tau = F_o'+\beta(F_1-F_o'f_1')+{}_{\beta}^{2}(F_2'-F_1'f_1'- F_o'f_1'^2+F_o'f_2')+\cdots \tag{A4}$$

Insertion of (A2) into the right-hand side of (A1) and series expansion of the exponential function in terms of powers of β yields

$$(1-\beta\theta)\exp(\theta) = \exp(F_o)[1+\beta(F_1-F_o)+ \beta^2(F_2-F_1F_o+F_1^2/2-F_1)+\cdots \tag{A5}$$

Equating (A4) and (A5), and matching according to powers of β gives the following set of differential equations in terms of $F_i(u)$ and $f_i(u)$:

$$F_o' = \exp(F_o) \tag{A6}$$

$$F_1'-F_o'f_1' = (F_1-F_o)\exp(F_o) \tag{A7}$$

$$F_2'-F_1'f_1'-F_o'f_1'^2+F_o'f_2' = (F_2-F_1F_o+F_1^2/2-F_1)\exp(F_o) \tag{A8}$$

to be solved with the boundary conditions $F_i(0) = f_i(0) = 0$, $i = 0, 1, 2 \ldots.$

The solution of (A6) is clearly

$$F_o = -\ln(1-u) \tag{A9}$$

Substitution of (A9) into (A7) and regrouping produces

$$F_1'-F_1/(1-u) = [f_1'+\ln(1-u)]/(1-u) \tag{A10}$$

Then, applying a form of Lighthill's criterion by asserting that "the higher-order solutions, F_i, $i \neq 0$ should be more complicated than F_o," we set

$$f_1' = -\ln(1-u) = F_o \quad \text{and} \quad F_1'-F_1/(1-u) = 0$$

and integrate to obtain

$$f_1(u) = -\int_o^u \ln(1-u)\,du = u+(1-u)\ln(1-u) \tag{A11}$$

$$F_1(u) = k(1-u)^{-1} = 0 \tag{A12}$$

In (A12) the integration constant k must equal zero in order that $F_1(0) = 0$ as required by the initial conditions. In the same manner (A8) yields the differential equations

$$f_2'F_o'-F_o'f_1'^2 = 0; \quad F_2'-F_2/(1-u) = 0$$

Consequently

$$F_2(u) = 0 \tag{A13}$$

and

$$f_2' = (f_1')^2 = \ln^2(1-u)$$

or

$$f_2 = \int_o^u \ln^2(1-u)\,du = 2u+2(1-u)\ln(1-u)-(1-u)\ln^2(1-u) \tag{A14}$$

It can be shown, then, that all $F_k(u) = 0$ for $k \neq 0$, and all

$$f_k(u) = (-1)^k \int_o^u \ln^k(1-u)\,du$$

and hence the solution to (A1) is given by

$$\theta = -\ln(1-u) \tag{A15}$$

$$\tau = u+\sum_{m=1}^{\infty} \beta^m f_m(u) \tag{A16}$$

where $f_m(u) = [mf_{m-1}(u)-(-1)^m(1-u)\ln^m(1-u)]$; $f_o(u) = u$.

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