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A Unified Theory of Solid Propellant Ignition

Part 3. Computer Solutions

by

H. H. Bradley, Jr.
Research Department

DECEMBER 1975

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R. G. Freeman, III, RAdm., USN Commander

G. L. Hollingsworth Technical Director

FOREWORD

This report was prepared as part of a research program at the Naval Weapons Center on the subject of Ignition of Solid Propellant Rockets, sponsored under Naval Ordnance Systems Command Task Assignment ORD 331-001/200-1/UR024-02-02. Previous reports include an extensive summary of the then current status of various theories of solid propellant ignition (NAVWEPS Report 8987, NOTS TP 3954), and extended analyses of solid phase theory (NWC TP 4618) and heterogeneous theory (NWC TP 4864). The present report, supported in addition by Ballistic Research Laboratories MIPR RN-516-75, is the third in a series of three reports on a Unified Theory of Solid Propellant Ignition. It presents some results of a parameter study obtained through use of the theory and computer program developed in the first two parts.

This report has been prepared for timely presentation of information. Because of the continuing nature of research in this area, refinements may be made in the future.

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Under authority of
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(U) *A Unified Theory of Solid Propellant Ignition.*

Part 3. *Computer Solutions* by H. H. Bradley, Jr.
China Lake, Calif., Naval Weapons Center, December
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(U) Computer solutions are obtained for several ignition models involving either surface or gaseous exothermic chemistry. Surface regression and convection of products from the surface are found to be limiting for the surface reaction of gaseous oxidizer and solid fuel. Fast gas phase reactions cannot in general be distinguished from surface and solid phase reactions by the experimental determination of the dependence of ignition time on igniting flux; a more usable technique may be the effect of concentration on ignition regimes. Slow gas phase reactions would be best studied by photographic observation of the developing gaseous reaction zone. Further analyses, coupled with judiciously chosen numerical solutions, offer a reasonable path to the solution of the remaining unresolved questions.

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INTRODUCTION

Ignition of combustible solid materials depends upon many interrelated phenomena, including chemical kinetics and heat and mass transport. A general ignition theory would deal with all processes simultaneously but is too complicated for tractable solution or presentation of results of calculations. When mass diffusional processes are neglected, the ensuing theory is called the thermal ignition theory. Heat losses to the gas phase may be included but are usually of lesser importance. Several reviews (e.g., Price,¹ and Merzhanov and Averson²) have been published dealing with thermal ignition theory and with various mathematical techniques of solving the equations either numerically or by appropriate approximate methods. The approximate solutions agree exceptionally well with the more exact numerical solutions and encompass a variety of models and boundary conditions, so that further exploration in this field does not seem fruitful. Less restricted models (e.g., Williams,³ and Linán and Crespo⁴), including surface reactions and diffusional processes, have been presented. The broader field, in which diffusion and convection play an important role, has not yet been so exhaustively studied. The importance of the various processes appears in proper perspective if it is recognized that

1. Kinetics and thermal transport govern time to incipient ignition (temperature runaway).
2. Diffusional mass transport establishes limits for incipient ignition.
3. Convection controls the transition from incipient ignition to steady-state combustion.

The success of approximate analytical methods of solutions in the case of thermal ignition models rests entirely with the great sensitivity of the chemical reaction rate to temperature over the range of activation

¹ Price, E. W., and others. "Theory of Ignition of Solid Propellants," AIAA J, Vol. 4, No. 7 (July 1966), pp. 1153-81.

² Merzhanov, A. G., and A. E. Averson. "The Present State of the Thermal Ignition Theory: An Invited Review," COMBUST AND FLAME, Vol. 16 (1971), pp. 89-124.

³ Williams, F. A. "Theory of Propellant Ignition by Heterogeneous Reaction," AIAA J, Vol. 4, No. 8 (August 1966), pp. 1354-57.

⁴ Linán, A., and A. Crespo. "An Asymptotic Analysis of Radiant and Hypergolic Heterogeneous Ignition of Solid Propellants," COMBUST SCI AND TECHNOL, Vol. 6 (December 1972), pp. 223-32.

energy encountered, thereby localizing the chemistry and permitting division of the ignition process into an initial inert heating phase followed by a chemical phase of rapidly increasing intensity. With decreasing activation energy, the chemistry becomes more diffuse in time and position, leading to a reduction in accuracy and ultimate breakdown of approximations. In systems containing large diffusional or convectional effects, a situation similar to low activation energy may prevail. Indeed, it has been noted⁵ that interpretation of experimental results to yield activation energies may be in error when mass transport effects are ignored.

While the characteristics of ignition of systems not containing important diffusional or convectional effects are quite well known, the relative paucity of information regarding the other end of the spectrum leads to complications in the analysis of experimental results and makes difficult the choice between the various ignition mechanisms. Two methods suggest themselves for approaching the problem. The first involves extension of the techniques of approximation used in thermal ignition theory, especially that of dividing the process into stages, each of which may be treated separately. The second would be the development of an entirely different concept of approximation tailored to the convection-diffusion problem. As in the development of thermal ignition theory, accurate numerical solutions are required against which to test approximations.

This report is the third in a series of which the first⁶ presented a unified ignition model containing as many of the diverse ignition situations as feasible; the second⁷ described a computer program for solving the equations developed in Reference 6. The current report presents some preliminary results of computer calculations. It is not the purpose of this report to provide a handbook of solutions spanning a wide range of parameters. An exhaustive or even comprehensive parameter study using numerical methods would be prohibitive in terms of time and money; reasonably accurate approximate analytical methods are needed for such purposes. Future reports in the series will be addressed to the study of such analyses.

The framework for the present study was developed in Reference 6, which contains complete details. To place the current effort in proper perspective, the principal assumptions by which the equations of the ignition model were derived from conservation laws are listed without comment:

⁵ Rosner, D. E. "Convective Diffusion as an Intruder in Kinetic Studies of Surface Catalyzed Reactions," AIAA J, Vol. 2, No. 4 (April 1964), pp. 593-610.

⁶ Naval Weapons Center. *A Unified Theory of Solid Propellant Ignition, Part 1. Development of Mathematical Model*, by H. H. Bradley, Jr. China Lake, Calif., NWC, August 1974. 36 pp. (NWC TP 5618, Part 1.)

⁷ Naval Weapons Center. *A Unified Theory of Solid Propellant Ignition, Part 2. Computer Program*, by H. H. Bradley, Jr. China Lake, Calif., NWC, August 1974. 28 pp. (NWC TP 5618, Part 2.)

NWC TP 5618, Part 3

1. One space dimension
2. No cross-coupling of forces and fluxes
3. No radiation feedback from gas phase reaction zone
4. No momentum transport (constant pressure)
5. Temperature-independent properties
6. Semi-infinite gas and solid phases
7. Validity of Chapman-Rubens approximation
8. No subsurface gasification in solid
9. Adequacy of global kinetics

To simplify computer programming, some additional approximations were used in Reference 7, namely:

10. Constant heat flux boundary condition, hence no absorption of incoming stimulus by evolved gases
11. Chemistry far removed from equilibrium, so that rate effects are governing
12. Sensible heat small compared to heats of reaction and phase change
13. Adequacy of the binary diffusion approximation
14. Equal molecular weights of all species

In order to restrict the amount of computational work still further, the general model was specialized to four simple models to be described in subsequent sections. Also, some additional limitations were imposed on the calculations:

15. Completely opaque solid phase--no subsurface absorption of incoming radiant flux
16. No solid phase reactions
17. Uniform initial temperature--equal in the solid and gas

Assumption 16 was made because of the availability of comprehensive studies for the case of solid phase reactions.⁸⁻¹⁰

SELECTION OF CASES FOR STUDY

Ignitable systems may be placed into two categories according to whether all the ingredients participating in the ignition chemistry

⁸ Linán, A., and F. A. Williams. "Radiant Ignition of a Reactive Solid with In-Depth Absorption," COMBUST AND FLAME, Vol. 18 (February 1972), pp. 85-97.

⁹ Bradley, H. H., Jr., "Theory of Ignition of a Reactive Solid by Constant Energy Flux," COMBUST SCI AND TECHNOL, Vol. 2 (August 1970), pp. 11-20.

¹⁰ Linán, A., and F. A. Williams. "Theory of Ignition of a Reactive Solid by Constant Energy Flux," COMBUST SCI AND TECHNOL, Vol. 3 (April 1971), pp. 91-98.

arise from the propellant itself or whether at least one reactant is provided as an element of the igniter in addition to its thermal stimulus. A different breakdown is provided by the site (gas or surface) of the key reaction which initiates steady state combustion. Solid phase bulk reactions are excluded from this study by Assumption 16. We have then the possibility of four subtheories of the unified ignition theory:

1. Case A: Surface chemistry between a solid fraction and an ambient reactive gas.
2. Case B: Surface chemistry between one pyrolyzed and one unpyrolyzed solid fraction. This is Case A with the ambient reactive gas provided by pyrolysis of the solid itself.
3. Case C: Gas phase reaction between a pyrolyzed solid fraction and an ambient reactive gas.
4. Case D: Gas phase reaction between two pyrolyzed ingredients initially present in the solid.

In addition, a brief study, accompanied by computer calculations, was made on endothermic and exothermic surface pyrolysis, since one or both processes are basic to several of the four main subtheories.

In all four cases, the stimulus may be provided by radiant flux, as in arc image experiments, or by an initial temperature difference between phases, as in shock tube experiments. Combinations of one or more of the simplified cases, as well as other possibilities are most likely as ignition mechanisms; nevertheless, it is believed that a thorough understanding and correlation of the characteristics of the simpler cases are basic to the solution of the overall ignition problem.

Comprehensive studies of Case A with both temperature difference and radiant heating have been reported.¹¹ This report will concentrate on only one aspect of Case A, namely, the effect of convective mass flux, engendered by surface reaction, on the character of the solution for a representative range of parameters.

No studies of Case B are currently available, although results on a related model with exothermic surface reaction rather than distributed solid reaction have been reported.¹² The distinction between Case B and Reference 12 is that in the latter, no mechanistic modeling of gas phase processes is attempted, boundary conditions being based on a reasonable application of steady state relationships. A moderate effort was expended on Case B to provide orientation for future studies.

¹¹ Bradley, H. H., Jr., and F. A. Williams. "Theory of Radiant and Hypergolic Ignition of Solid Propellants," COMBUST SCI AND TECHNOL, Vol. 2 (August 1970), pp. 41-52.

¹² Baer, A. D., and N. W. Ryan. "An Approximate but Complete Model for the Ignition Response of Solid Propellants," AIAA J, Vol. 6, No. 5 (May 1968), pp. 872-77.

A presentation of Case C with radiant stimulus has been published,¹³ including the frequently neglected effects of convection and surface regression. For this reason Case C will not be covered here; computer calculations were limited to check runs to establish program capability and execution times.

Studies of Case D are seemingly limited to instances with a temperature difference (relevant to shock tube work) and also are combined with Case C.¹⁴ Thus they apply to the interesting situation in which two oxidizers compete for the fuel vapors in gas phase reactions. The complexity of Case D, owing to its greater number of parameters, has precluded any comprehensive study in this report. Some representative calculations have been made, primarily for comparison with analogous runs of Case B.

STUDIES OF INDIVIDUAL CASES

It is almost universal practice to use dimensionless parameters in the analysis of complicated systems of equations. The required number of calculations or experiments is usually significantly reduced if the optimum set of parameters is chosen. An optimum set is one from which only a few members have an important influence on the outcome of the calculations or experimental results. Often such an optimum set cannot be recognized a priori, especially in studies involving new or little explored fields. In such instances, a sparse coverage of the parameter space renders interpretation of results in terms of actual physical quantities almost impossible. Therefore, the choice was made to deal with dimensional equations directly, although a dimensional analysis was carried out to establish the minimum number of dimensionless parameters required to describe the problem. The single exception was Case A, for which an extensive study had already been completed.¹¹ Otherwise, standard values were assigned to those physical quantities (Table 1) which were considered constant throughout the study, while a reference set was chosen for the other parameters about which to conduct a sensitivity study. No claim is made for the relevance of the reference set to a particular physical system; the choice was made from among published values and from other studies.

¹³ Kashiwagi, T. "A Radiative Ignition Model of a Solid Fuel," COMBUST SCI AND TECHNOL, Vol. 8, No. 5 and 6 (1974), pp. 225-36.

¹⁴ Kumar, R. K., and C. E. Hermance. "Ignition of Homogeneous Solid Propellants under Shock Tube Conditions: Further Theoretical Development," AIAA J, Vol. 9, No. 8 (August 1971), pp. 1615-20.

NWC TP 5618, Part 3

TABLE 1. Standard Physical Properties Used in Report.

Name	Symbol	Definition	Value	Units
Solid density	ρ_1	...	1.7	g/cm ³
Solid specific heat	c_1	...	3(10 ⁻¹)	cal/g °K
Solid conductivity	λ_1	...	1(10 ⁻³)	cal/cm °K sec
Solid responsivity	Γ_1	$\sqrt{\lambda_1 \rho_1 c_1}$	2.258(10 ⁻²)	cal/cm ² °K sec ^{1/2}
Solid permittivity	a_1	$\lambda_1 \rho_1 / c_1$	5.667(10 ⁻³)	g ² /cm ⁴ sec
Gas density (initial)	ρ_{20}	...	1.016(10 ⁻³)	g/cm ³
Gas specific heat	c_2	...	3(10 ⁻¹)	cal/g °K
Gas conductivity	λ_2	...	4(10 ⁻⁵)	cal/cm °K sec
Gas responsivity	Γ_2	$\sqrt{\lambda_2 \rho_{20} c_2}$	1.104(10 ⁻⁶)	cal/cm ² °K sec ^{1/2}
Gas permittivity	a_2	$\lambda_2 \rho_{20} / c_2$	1.354(10 ⁻⁷)	g ² /cm ⁴ sec
Pressure	...	...	1	atm
Gas molecular weight	μ	...	25	...
Initial temperature	T_0	...	300	°K
Lewis number	Le	$\frac{\rho_2 c_2 D}{\lambda_2}$	1	dimensionless

PYROLYSIS

Transient pyrolysis is described by the following set of equations adapted from Reference 6.

Field equations:

Solid enthalpy

$$\frac{\partial h_1}{\partial t} = \frac{a_1}{\rho_{10}^2} \frac{\partial^2 h_1}{\partial \psi_1^2} - \frac{m_s}{\rho_{10}} \frac{\partial h_1}{\partial \psi_1} \quad (1)$$

Gas enthalpy

$$\frac{\partial h_2}{\partial t} = \frac{a_2}{\rho_{20}^2} \frac{\partial^2 h_2}{\partial \psi_2^2} - \frac{m_s}{\rho_{20}} \frac{\partial h_2}{\partial \psi_2} \quad (2)$$

Pyrolyzed ingredient

$$\frac{\partial Y}{\partial t} = Le \frac{a_2}{\rho_{20}^2} \frac{\partial^2 Y}{\partial \psi_2^2} - \frac{m_s}{\rho_{20}} \frac{\partial Y}{\partial \psi_2} \quad (3)$$

Boundary equations:

Energy

$$\left. \frac{a_1}{\rho_{10}} \frac{\partial h_1}{\partial \psi_1} \right|_{0^-} = \left. \frac{a_2}{\rho_{20}} \frac{\partial h_2}{\partial \psi_2} \right|_{0^+} + q_r + m_s Q \quad (4)$$

Pyrolyzed ingredient

$$m_s = m_s Y - Le \frac{a_2}{\rho_{20}} \frac{\partial Y}{\partial \psi_2} \quad (5)$$

where

$$m_s = \rho_1 v \exp(-E/RT_s) \quad (6)$$

In the absence of gas phase reactions it can be shown¹⁵ that the gas phase temperature approaches a spatially constant value equal to the steady state surface temperature (T_1) and given by solving the following steady state heat balance equations for the surface:

$$q_r = \rho_{10} r [-Q + c_1 (T_1 - T_0)] \quad (7)$$

$$z = v \exp(-E/RT_1) \quad (8)$$

In Reference 15, a matched asymptotic expansion was completed on the energy equations alone, with the result that the surface temperature was found to approach its steady state value asymptotically from below. Integration of the current equations, including mass flow effects, reveals a temperature which overshoots the steady state value T_1 by 2 to 20%, becoming most pronounced for exothermic reactions with high activation energy. This is explainable by the action of pyrolyzed mass in carrying away high-temperature surface material in increasing amounts as surface regression increases toward its final steady state value as given by solution of Equation 7 and 8. The calculations do not indicate any effect of gaseous mass diffusivity (manifest in Le) on the shape of the curve of surface temperature versus time, thus implying that convection, not diffusion, is controlling in this instance. An interesting and unexplained phenomenon of the solution of Equations 1 through 6 is the presence of small amplitude, low frequency damped oscillations in surface temperature for endothermic pyrolysis. No oscillations were noted for exothermic pyrolysis.

¹⁵ Kindelan, M., and F. A. Williams. "Theory for Endothermic Gasification of a Solid by a Constant Energy Flux," COMBUST SCI AND TECHNOL, Vol. 10, No. 1 and 2 (1975), pp. 1-19.

CASE A

The following governing equations for Case A may be readily written by reference to Part 1 of this report:⁶

Field equations:

Solid enthalpy

$$\frac{\partial h_1}{\partial t} = \frac{a_1}{\rho_{10}^2} \frac{\partial^2 h_1}{\partial \psi_1^2} - \frac{m_s}{\rho_{10}} \frac{\partial h_1}{\partial \psi_1} \quad (9)$$

Gas enthalpy

$$\frac{\partial h_2}{\partial t} = \frac{a_2}{\rho_{20}^2} \frac{\partial^2 h_2}{\partial \psi_2^2} - \frac{m_s}{\rho_{20}} \frac{\partial h_2}{\partial \psi_2} \quad (10)$$

Oxidizer

$$\frac{\partial Y_4}{\partial t} = Le \frac{a_2}{\rho_{20}^2} \frac{\partial^2 Y_4}{\partial \psi_2^2} - \frac{m_s}{\rho_{20}} \frac{\partial Y_4}{\partial \psi_2} \quad (11)$$

Product

$$\frac{\partial Y_6}{\partial t} = Le \frac{a_2}{\rho_{20}^2} \frac{\partial^2 Y_6}{\partial \psi_2^2} - \frac{m_s}{\rho_{20}} \frac{\partial Y_6}{\partial \psi_2} \quad (12)$$

Boundary conditions:

Energy

$$\left. \frac{a_1}{\rho_{10}} \frac{\partial h_1}{\partial \psi_1} \right|_{0^-} = \left. \frac{a_2}{\rho_{20}} \frac{\partial h_2}{\partial \psi_2} \right|_{0^+} + q_r + m_6 C_6 \quad (13)$$

Oxidizer

$$0 = m_s Y_4 - Le \frac{a_2}{\rho_{20}} \frac{\partial Y_4}{\partial \psi_2} + \frac{\mu_h}{\mu_{10}} b'_4 m_6 \quad (14)$$

Product

$$0 = m_s Y_6 - Le \frac{a_2}{\rho_{20}} \frac{\partial Y_6}{\partial \psi_2} - \frac{\mu_6}{\mu_{10}} b_6 m_6 \quad (15)$$

where

$$m_s = m_6 = \rho_1 Y_{10} v_b (\rho_2 Y_4 / \mu_4)^{s_6} \exp(-E_6 / RT_s) \quad (16)$$

Taking constant specific heats and defining

$$\theta_i = (T_i - T_0) / T_0 \quad (17)$$

it may be shown by dimensional analysis that the solutions of Equations 9 through 16 are expressible in the functional form

$$(\theta_i, \eta_k) = \bar{f}_A(\tau, \xi_1, \xi_2; \bar{P}_A) \quad (18)$$

where

$$\eta_k = (\mu_{10}/\mu_k b'_k) Y_k \quad (19)$$

$$\tau = (q_r^2/\Gamma_1^2 T_0^2) t \quad (20)$$

$$\xi_1 = (\rho_{10} q_r / a_1 c_1 T_0) \psi_1 \quad (21)$$

$$\xi_2 = \xi_1 (a_1/a_2)^{1/2} (\rho_{20}/\rho_{10}) \psi_2 \quad (22)$$

$\bar{P}_A$ = the parameter set:

$a_1/a_2, c_1/c_2, Le, Q_6/c_1 T_0, E_6/RT_0$, and

$$[\rho_1 Y_{10} Q_6 \nu_6 (\rho_{20} b'_k / \mu_{10})^{S_6} \exp(-E_6/RT_0)] / q_r$$

In Reference 11, advantage is taken of an integral solution to represent the surface conditions as a function of time with no regression in terms of three parameters:

$$\alpha = \Gamma c_1 T_0 / \Gamma_1 \cdot e^{1/2} Q_6 Y_{60} \quad (23)$$

$$\beta = E_6 / RT_0 \quad (24)$$

$$\gamma = q_r / Q_6 B Y_{60} \exp(-\beta) \quad (25)$$

where dimensionless time τ is defined as

$$\tau = [(\gamma + 1) / \gamma T_0]^2 q_r^2 t \quad (26)$$

and B replaces the expression $\rho_1 Y_{10} (\rho_{20} b'_k / \mu_{10})^{S_6}$. In the present study of Case A, the same parameters are used for convenience in comparing results. The additional parameter b'_k is introduced and all the Equations 9 through 16 are solved in arriving at results. Again, only results for the surface are presented, otherwise the full six parameters would be required to describe temperature and concentration distributions. The choice $b'_k = 1$ represents reaction of one mass unit each of oxidizer and fuel, leading to evolution of two units of product and to a net convective flow from the surface. Assignment of smaller values of b'_k would correspond to a more severe convective effect since b'_k is the ratio of oxidizer mass to fuel mass reacted.

Before describing the results of the parameter study it is convenient to define three characteristic temperatures of the process. First, the steady state temperature in the absence of external heating ($q_r = 0$) and ignoring convection is shown in Reference 11 to be given by

$$\theta_{ss} = 1/\alpha \quad (27)$$

This is the dimensionless temperature at which the rate of chemical reaction of oxidizer just balances its rate of diffusion to the surface. Second, we have the temperature at which, in the absence of diffusional or convectional effects, the applied flux, q_r , equals the chemically generated surface heat

$$\theta_Y = \ln \gamma / (3 - \ln \gamma) \quad (28)$$

This condition obviously cannot exist if $\beta < \ln \gamma$. Third, there is the steady state pyrolysis temperature θ_1 , obtained by solving the pair of Equations 7 and 8. This dimensionless temperature θ_1 to indicate exothermicity, is generally greater than θ_Y , although the trend reverses at higher values of θ_Y . We also note here that the steady state adiabatic ($q_r = 0$) surface temperature with regression is given by Equation 7 because at steady state, as noted previously, the gas phase temperature approaches a space-uniform value. Hence no sensible heat is lost to the gas phase and all the surface chemical heating goes into heating the solid. Under these conditions

$$T_{ss \text{ conv}} = T_0 + Q_6/c_1 \quad (29)$$

or in dimensionless terms

$$\theta_{ss \text{ conv}} = (T_{ss} - T_0)/T_0 = Q_6/c_1 T_0 \quad (30)$$

Only two steady state solutions exist for the distribution of Y_k , namely, $Y_k = 0$ and $Y_k = u_k b_k / u_{1k}$. Corresponding to $Y_k = 0$ is zero concentration gradient, mass flux, and chemical heating. In a number of numerical calculations, external heating was interrupted (to simulate go/no-go experiments) at various times during heating. In all cases, regardless of the state of the system at the time of interruption, the ensuing transient led ultimately to an extinguished state in which Y_k and $dY_k/d\psi_k = 0$ at the surface. The go/no-go criterion of ignition is thus seen to be inapplicable to ignition corresponding to Case A.

Turning now to Figure 1, we see the results of calculations for continuous heating over a broad range of values of parameters α , β , and γ . Numbered points on the figure are keyed to run numbers of Table 2, which presents other quantitative results. Classical ignition curves characterized by a rapid temperature rise (Figure 2a) occurred only in Runs 1 and 2. Even these exhibited the transition to zero surface chemical heating rate described above for interrupted heating. For this reason an alternative definition of ignition time, used in Reference 11 and represented by the solid curves in Figure 1, was employed. This was the time of occurrence of equal applied and chemical heating rates, which, without convection, was often close to the time of rapid temperature rise or the go/no-go time. With the single criterion chosen, convective and nonconvective times were nearly equal, but the range of parameters for ignition to occur was reduced. The inequality

$$\theta_Y(\text{numerical}) < 1/\alpha \quad (31)$$

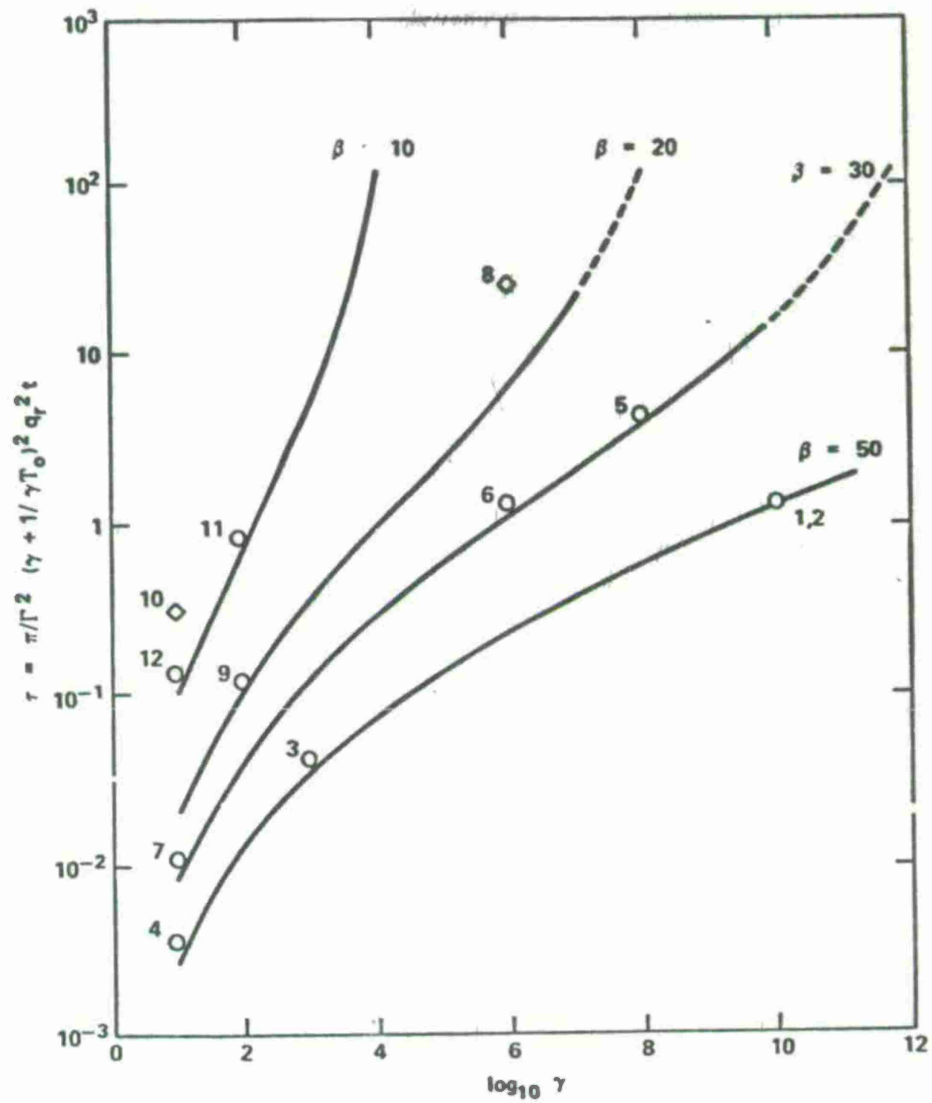


FIGURE 1. Comparison of Ignition Times for Case A With Surface Regression (Points) and Without Surface Regression (Solid Lines).

TABLE 2. Results of Case Calculations.

Run no.	α	β	$\log_{10} \gamma$	θ_Y^a	θ_Y^b	θ_{1+}^c	$1/\alpha$
1	0.1	50	10	0.855	0.86	1.063	10
2	1	50	10	0.855	0.86	1.063	1
3	5	50	3	0.16	0.17	1.000	0.2
4	10	50	1	0.0482	0.058	1.000	0.1
5	0.1	30	8	1.591	1.59	1.680	10
6	1	30	6	0.855	0.92	1.126	1
7	5	30	1	0.083	0.1	1.000	0.2
8	0.1	20	6	2.234	...	2.160	10
9	1	20	2	0.299	0.32	1.000	1
10	5	20	1	0.130	0.32	1.100	0.2
11	0.1	10	2	0.855	0.86	1.330	10
12	1	10	1	0.299	0.35	1.050	1

^a Calculated by Equation 28.

^b From numerical solution.

^c Calculated from Equation 7,8.

^d Not reached.

is still required for ignition as in the nonconvective case. A new characteristic associated with convection is the presence of oscillations of the relaxation type (Figure 2b) superimposed on the rising portion of the curve of surface temperature versus time and occurring after the temperature has risen beyond the value corresponding to θ_Y . Each example of oscillatory behavior is associated with the inequality

$$\theta_Y < 1/\alpha < \theta_{1+} \quad (32)$$

including Runs 3, 4, 6, 7, and 9. Runs 1, 2, 5, 11, and 12, which are near the periphery of the parameter space in Figure 1, show little or no tendency toward oscillation; temperature-time traces resemble the lower portion of the curve of Figure 2a. No ignition occurs in Run 8 where θ_Y is never reached or in Run 10 for which $\theta_Y > 1/\alpha$. While the correlations are not perfect, they suggest that the interaction between chemical reaction and convection alternately consumes the gas phase reactant at the surface and blocks it from entering until provided by the slower diffusional process. The implication of this convective blocking, unique to Case A, is that some other transport mechanism, not included in the present model, must be responsible for ignition or combustion processes. The mechanism is probably turbulent mixing, generated in real situations by cross flow of gases or by the combustion itself. The result of such mixing would be to reduce the effective value of α , thereby extending the range of other parameters for which ignition occurs.

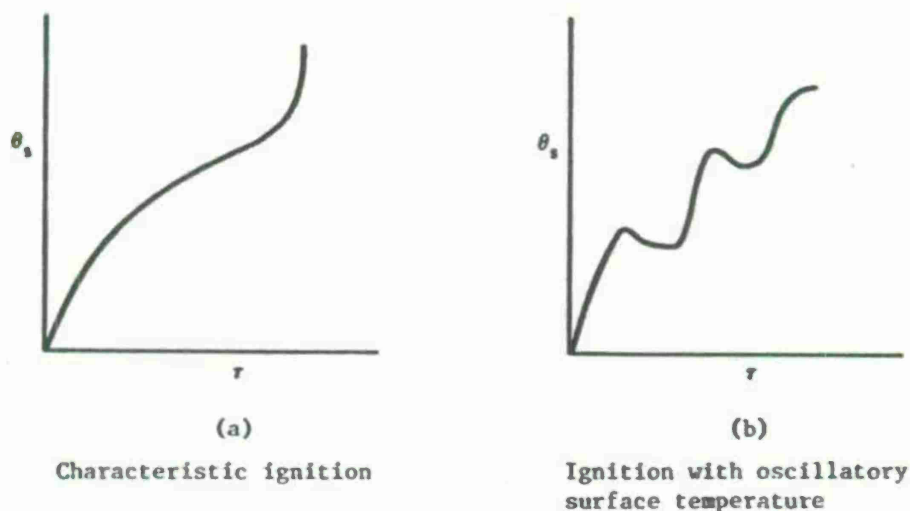


FIGURE 2. Typical Dimensionless Curves of Surface Temperature History During Heating by Constant Radiant Flux (Case A).

CASE B

There are two rate processes in Case B: (1) endothermic pyrolysis of one constituent of the solid propellant to produce an active gaseous species, and (2) exothermic reaction of this active gas in a surface process with the unpyrolyzed fraction. The equations solved were:

Field equations:

Enthalpy--same as Equations 9 and 10

Oxidizer

$$\frac{\partial Y_2}{\partial t} = Le \frac{a_2}{\rho_{20}^2} \frac{\partial^2 Y_2}{\partial \psi_2^2} - \frac{m_s}{\rho_{10}} \frac{\partial Y_2}{\partial \psi_2} \quad (33)$$

Product

$$\frac{\partial Y_5}{\partial t} = Le \frac{a_2}{\rho_{20}^2} \frac{\partial^2 Y_5}{\partial \psi_2^2} - \frac{m_s}{\rho_{10}} \frac{\partial Y_5}{\partial \psi_2} \quad (34)$$

Boundary conditions:

Energy

$$\frac{a_1}{\rho_{10}} \frac{\partial h_1}{\partial \psi_1} = \frac{a_2}{\rho_{20}} \frac{\partial h_2}{\partial \psi_2} + m_5 Q_5 + m_{11} Q_{11} + q_r \quad (35)$$

Oxidizer

$$m_{11} = m_s Y_2 - Le \frac{a_2}{\rho_{20}} \frac{\partial Y_2}{\partial \psi_2} + \frac{\mu_2}{\mu_{10}} b'_2 m_s \quad (36)$$

Product

$$0 = m_s Y_5 - Le \frac{a_2}{\rho_{20}} \frac{\partial Y_5}{\partial \psi_2} - \frac{\mu_s}{\mu_{10}} b'_5 m_s \quad (37)$$

where

$$m_s = \rho_1 Y_{10} v_5 (\rho_2 Y_2 / \mu_2)^{ss} \exp(-E_5 / RT_s) \quad (38)$$

$$m_{11} = \rho_1 Y_{11} v_{11} \exp(-E_{11} / RT_5) \quad (39)$$

$$m_s = m_5 + m_{11} \quad (40)$$

When the same temperature, time, and distance scales are used (see Equations 17, 20, 21, 22), dimensional analysis yields the following functional solution:

$$(\theta_1, \eta_2) = \bar{F}_B(\tau, \xi_1, \xi_2; \bar{P}_B) \quad (41)$$

where

$$\eta_2 = (\mu_{10} / \mu_2 b'_2) Y_2$$

$\bar{P}_B$ = the parameter set:

$$a_1/a_2, c_1/c_2, Le, Q_5/c_1 T_0, E_5/RT_0, Q_{11}/c_1 T_0, E_{11}/RT_0,$$

$$[\rho_1 Y_{10} Q_5 v_5 (\rho_2 b'_2 / \mu_{10})^{ss} \exp(-E_5/RT_0)]/q_r, \text{ and}$$

$$[\rho_1 Y_{11} Q_{11} v_{11} \exp(-E_{11}/RT_0)]/q_r$$

The last two measure the relative intensities of the two rate processes and the stimulating flux at the initial temperature T_0 .

The rapid proliferation of parameters, even in the nondimensional representation, suggests the prohibitive number of calculations which would be associated with a complete parameter study. Even the more modest approach of conducting a sensitivity study, which ignores interactions among parameters, requires the choice of a reference set of parameters for variation. The decision was to make a limited study of

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the effect of varying the physical quantities E_5 , E_{11} , v_5 , v_{11} , q_r , p , Q_5 , Q_{11} , and Le with values of the other quantities as shown in Table 3. The variation of E_5 was carried out in two ways: (1) independently, and (2) with v_5 to provide a constant reference value of the exothermic reaction rate between fuel and pyrolyzed oxidizer. Likewise E_{11} was varied independently and with v_{11} to yield a constant reference value of oxidizer pyrolysis rate. The radiation flux level was varied from 3 to 30 cal/cm²sec, pressure effect was evaluated from 0.1 to 10 atmospheres, and Lewis number was varied from 0.1 to 10. The effect of Q_5 and Q_{11} was found by keeping their algebraic sum equal to 2000 cal/g, thereby allowing both endothermic and exothermic pyrolysis.

TABLE 3. Parameters Used in Case B Calculations.

Parameter	Reference value	Range
E_5	30 kcal/mole	25 - 35
v_5	$7.26(10^{19})$	$1.35(10^{17}) - 3.91(10^{22})$
E_{11}	30 kcal/mole	25 - 35
v_{11}	$2.91(10^{18})$	$5.40(10^{11}) - 1.56(10^{17})$
q_r	10 cal/cm ² sec	3 - 30
Q_5	2150 cal/g	1850 - 2000
Q_{11}	-150 cal/g	0 - 150
Le	1	0.1 - 10
p	1 atm	0.1 - 10

Three characteristics of the solution were examined: (1) the time (t_1) to attain zero surface chemical heating (endothermic heat of pyrolysis equal to exothermic heat of reaction), (2) time (t_2) at which total surface chemical heating equals incident flux, and (3) time (t_3) of occurrence of the inflection point in the curve of surface temperature versus time when such a point exists. For the parameter values examined, t_2 and t_3 agree within 5%.

The effect of radiant flux level on t_1 and t_2 is displayed in Figure 3 for the reference set of physical parameters. The relationship is similar to results obtained for solid phase bulk reactions and for pure exothermic surface reaction. An essential difference from Case A is the absence of convective blocking of reactants by products, although the loss of energy from the surface would have a marked effect during

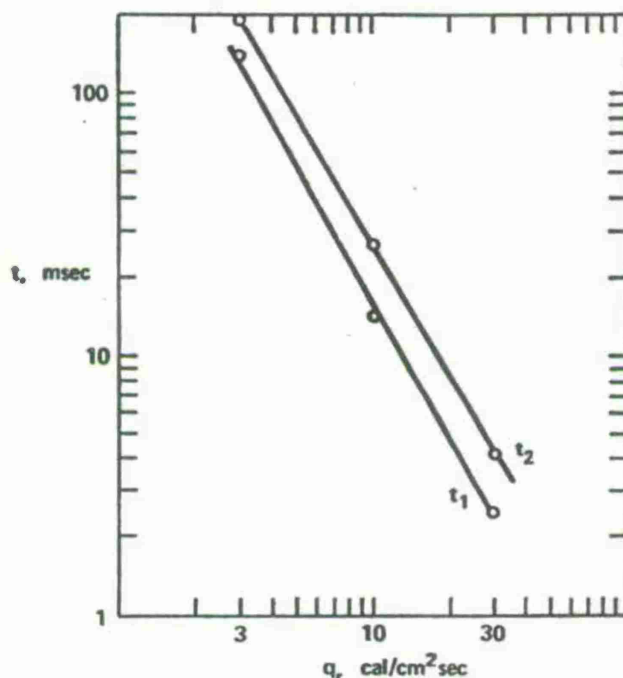


FIGURE 3. Effect of Flux Level on Ignition Times t_1 and t_2 for the Reference Set of Parameters (Case B).

transition to steady state. No quantitative test of this phenomenon has been made, although either suppression of surface regression or interrupted stimulus runs would be important.

The influence of propellant kinetic parameters is shown in Figure 4. The effect of varying either activation energy with its pre-exponential factor is nearly undetectable in terms of characteristics (1) and (2); however, there is no inflection point unless at least one activation energy exceeds an approximate value of 30 kcal/mole. When either activation energy is varied independently, the results are much more dramatic, ranging from very short ignition times to no ignition at all. When the pyrolysis activation energy is lower than the reaction activation energy, no ignition of type (1) or (2) was observed to occur. Physically, this is consistent with the temperature-limiting effect of the surface endotherm, which prevents the attainment of a temperature high enough to promote the exothermic reaction within practical time limits. Exposure of the solid fuel to constant temperature reactive pyrolysis products might lead to a situation similar to an "adiabatic" ignition were it not for the effect of convection in removing any developing thermal zone from the surface. When pyrolysis activation energy is higher, the potential for reaction occurs prior to the availability of both species; as the pyrolysis temperature is approached, exothermic reaction occurs rapidly

at the onset of pyrolysis. Ignition times within the ignition regimes increase with either activation energy when such energy is varied independently of its associated pre-exponential factor.

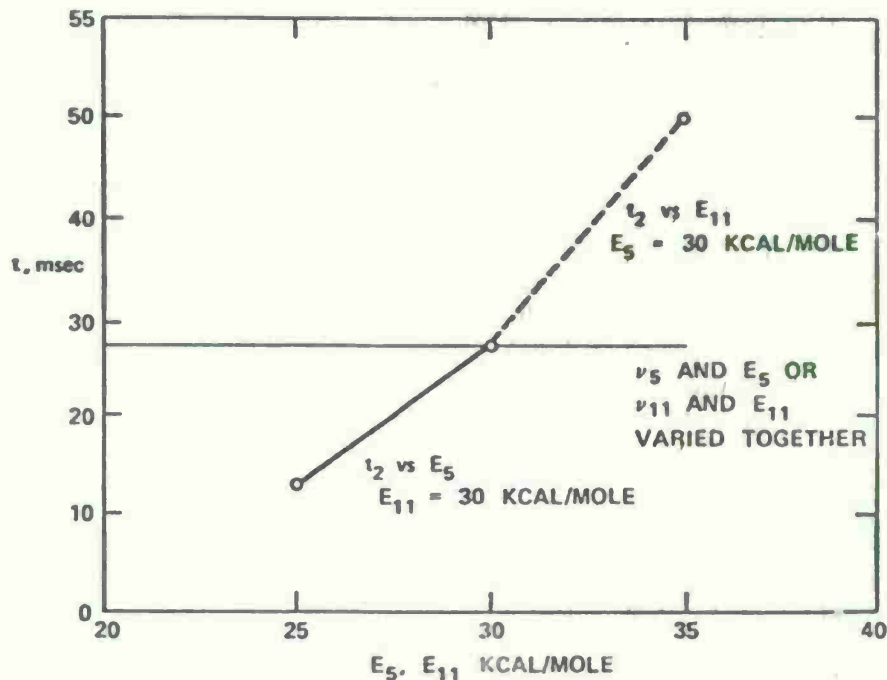


FIGURE 4. Effect of Activation Energy on Ignition Time t_2 for the Reference Set of Parameters (Case B).

The effect of pressure is similar to the effect of transport properties in imposing an ignition limit but not otherwise affecting ignition time significantly. At a pressure of 1/10 atmosphere, characteristic (2) was not established so that ignition would not occur. At 10 atmospheres the ignition time was decreased approximately 5%. The result is anticipated since, by the Chapman-Rubesin approximation, $\rho^2 D$ is constant, and diffusional processes with their induced convection and surface cooling are more pronounced at low pressures.

Ignition occurred over the entire range of Lewis numbers investigated. For $Le = 0.1$, corresponding to low mass-diffusion rates, the ignition time was reduced from 27 to 24 milliseconds, while for $Le = 10$ an increase to 28 milliseconds was noted based again on characteristic (2).

Finally, it was observed that changes in Q_5 and Q_{11} , keeping the sum equal to 2000 cal/g, resulted in no perceptible change in ignition time.

CASE C

Case C is similar to Case B in having two rate processes: (1) an endothermic surface pyrolysis supplying fuel for the (2) exothermic gas phase reaction with ambient oxidizer. The same number of parameters is required for a complete description. Qualitatively, the surface pyrolytic process limits the surface temperature, but here, the similarity with Case B ends. Even though the surface temperature is restricted, two extremes of ignition characteristics may be noted. First, a fast gas phase reaction occurs almost immediately after pyrolysis commences, and second, a slow gas phase reaction develops into ignition only after a time delay for diffusional mixing and accompanying reactive heating. Both aspects are discussed by Kindelan and Williams¹⁶ and will not be elaborated upon here. In Reference 15, some computer results are presented for the Case C model. An interesting conclusion is that for a given pyrolysis activation energy there is an upper and a lower reaction activation energy outside of which ignition does not occur. Likewise, a given reaction energy has upper and lower limiting values of pyrolysis activation energy. In the current study the only effect noted (qualitatively) is that for given activation energies of the two reactions, there appear to be limits on the relative values of pre-exponential factors for which ignition occurs. The ignition criterion was taken as the appearance of a temperature maximum (spatial) in the gas phase.

CASE D

Case D, with three chemical reactions, is the most resistant to a comprehensive parameter study. It also may represent the most practical model, that of gas phase ignition of two products of surface pyrolysis. Even a breakdown and study of the various subcases according to the relative values of the three activation energies would present a formidable task. In the special subcase where equal pyrolysis energies prevail, ignition certainly occurs near the surface at the onset of pyrolysis for a fast gas phase reaction. A slow gas phase reaction would allow the pyrolyzed reactants, already mixed by simultaneity of pyrolysis, to proceed some distance and time into the gas phase before reacting in a manner approaching that of a bulk explosion. If one pyrolysis product has a much lower activation energy than the other, it will fill the gas phase, simultaneously restricting the surface temperature so that the second ingredient will never pyrolyze. There will be no ignition in this event. Just how close together the two pyrolysis activation energies can be and still permit ignition when both pyrolyses are endothermic has not been determined. The only systematic variation of parameters in this study has been the gas phase activation energy. Reference values are shown in Table 4, including equal activation energies of 20 kcal/mole for all three reactions. Ignition is taken as the development of a

¹⁶ Kindelan, M., and F. A. Williams. "Radiant Ignition of a Combustible Solid with Gas-Phase Exothermicity." (To be published.)

temperature maximum in the gas phase. With a gas phase activation energy of 15 kcal/mole, ignition occurs in 10.5 milliseconds at 65 micrometers from the surface. This maximum develops into a steady state temperature distribution after 1.2 additional milliseconds, with the maximum moving to 33 micrometers from the surface. At 20 kcal/mole, a runaway gas phase temperature occurs in 31.6 milliseconds at 70 micrometers from the surface. At 25 kcal/mole, no ignition occurred before termination of the run, steady state pyrolysis having been established at 60 milliseconds.

TABLE 4. Reference Values for Case D.

Parameters	Reference value
Pyrolysis reactions:	
Activation energy	20 kcal/mole
Heat of pyrolysis	-150 cal/g
Pre-exponential factor	3×10^9
Gas phase reactions:	
Activation energy	20 kcal/mole
Heat of reaction	2300 cal/g
Pre-exponential factor	3×10^{16}
External heat flux	10 cal/cm ² sec

SUMMARY AND DISCUSSION

Four simplifications of the unified ignition model have been considered for numerical study, two each involving exothermic chemical reaction at the surface and in the gas phase. The complexity of the particular case has determined the degree of completeness of the results.

Case A has been treated by an extension of a previous analysis to include gas phase convection and surface regression, both of which were found to be important in restricting the transition to steady state. A unique feature of Case A is the convective blocking of oxidizer from reaching the surface so that other processes must augment a direct reaction between solid and gas phases to achieve ignition.

The sensitivity study on Case B is restricted to a single reference point but is complete otherwise. While interactions may modify the trends for other reference points, activation energy is shown to be the most important single parameter in the model. However, when the kinetics is varied so as to maintain constant reaction rate, the effect of activation energy vanishes. Variation of pressure and transport properties has little effect on ignition time but restricts the ignition regime. The relation between ignition time and external heat flux appears similar to relations obtained for solid phase bulk reactions and simple surface exothermic reactions so that such relations cannot be used as a diagnostic tool for the purpose of selecting the appropriate model.

When we consider gas phase exothermic processes, we find that under certain conditions, namely, those leading to rapid gas phase reaction, the effect of external stimulus level on ignition time is again similar to results obtained for surface exotherms. A unifying statement can thus be made on the basis of the sparse studies now available: The nature and site of an exothermic process cannot be distinguished by q_r versus t relationships if that process occurs near the surface. The effect of varying the concentration seems to offer no solution because of the small effect of concentration versus temperature on reaction rate. Concentration, as well as transport effects, do tend to impose limitations on ignition regimes so that calculations and experiments involving interrupted heating may offer a usable diagnostic tool.

For a slow gas phase reaction, ignition times are significantly increased above the thermal ignition time by diffusional effects. Gas phase reactions in this category should be distinguishable experimentally from any surface reaction by the use of high-speed photographic techniques.

Additional analytical work is needed to settle the unresolved questions remaining from the limited coverage at this time. All answers will not be obtained by the use of numerical techniques because of the great number of parameters involved. Rather more rewarding should be the development of ignition criteria based on reasonable physical arguments and mathematical approximations and tested by spot checking, using a numerical method such as the unified ignition theory.

NOMENCLATURE

a	Thermal permittivity, $\rho\lambda/c$
b, b'	Stoichiometric constants
c	Specific heat
D	Mass diffusivity
E	Activation energy
h	Thermal enthalpy
Le	Lewis number, $\rho c D / \lambda$
m	Surface mass flux
Q	Heats of reaction or pyrolysis
q_r	External heat flux
R	Universal gas constant
r	Linear burning rate
T	Temperature
t	Time
Y	Mass fraction
α	Diffusion parameter (Equation 23)
β	Activation energy parameter (Equation 24)
Γ_1, Γ_2	Solid and gas phase responsivities, $(\lambda_i \rho_i c_i)^{1/2}$
Γ	$\Gamma_1 + \Gamma_2$
γ	External heating parameter (Equation 25).
η	Reduced mass fraction
θ	Reduced temperature, $(T - T_0)/T_0$
λ	Thermal conductivity
μ	Molecular weight
ν	Pre-exponential factor
ξ	Dimensionless length
ρ	Density

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τ Dimensionless time (different for Case A and the other cases)
 ψ Howarth transformed length ($\int \rho dx / \rho_0$)

Subscripts

0 Initial value
1 Solid phase
2 Gas phase
s Surface
ss Steady state
conv With convection

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