JOURNAL OF THERMOPHYSICS AND HEAT TRANSFER
Vol. 20, No. 2, April–June 2006

Technical Notes

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Surface-Absorption Assumption for Radiant Heating and Ignition of Energetic Solids

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Nomenclature

K_a	=	absorption coefficient
k	=	thermal conductivity
q	=	absorbed radiant flux
T	=	temperature
T_o	=	initial temperature
$T_s(t)$	=	surface temperature ($x = 0$)
T_s	=	reference temperature
x	=	coordinate normal to surface, positive into solid
α	=	thermal diffusivity
β_s	=	in-depth absorption parameter $= (T_s - T_o)/(q/kK_a)$

Introduction

IN MODELING and analysis of radiant heating and ignition of energetic solids the assumption of surface absorption (i.e., a surface heat-flux boundary condition) is routinely invoked without justification or discussion of its applicability.[1–4] However, it is well known that radiation absorption is a volumetric phenomenon even in opaque solids. Nevertheless, no analytical assessment of the surface absorption assumption in radiant heating and ignition of solids, either energetic or nonenergetic, has been reported.

In this Note a simple analytical model is described for investigating the surface absorption assumption. The analysis is based on the one-dimensional heat equation with in-depth, volumetric absorption. The results show that in many cases in-depth absorption is significant and that assuming surface absorption can cause ignition times to be severely underestimated. The analysis also sets forth a new nondimensional conduction/radiation parameter for radiant ignition of solids that can be used to evaluate the validity of the surface-absorption assumption.

Analytical Model and Governing Equations

In-Depth Absorption

A collimated, monochromatic radiative flux is imposed on a semi-infinite, homogeneous, nonemitting, nonscattering solid. The solid is characterized by K_a, k, and α. The flux absorbed (i.e., not reflected at the surface) by the solid is q. The initial condition is a uniform temperature T_o (room temperature). The boundary conditions are T_o deep in the solid and a surface gradient of zero, corresponding to no conductive/convective heat flux at the surface (i.e., the solid is adjacent to a poorly conducting fluid, such as a gas). Ignition is assumed to occur when the surface reaches a fixed temperature T_s. This criterion is, of course, an overly simplistic view of ignition of energetic solids, but it is a common assumption, and furthermore it is a useful approximation for the purposes of this Note (i.e., investigation of ignition criteria is not the point of this analysis). All properties are assumed constant (uniform in space and time). In dimensional variables the energy equation and associated boundary and initial conditions are as follows:

$$\frac{1}{\alpha}\frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} + \frac{qK_a}{k}\exp(-K_a x) \tag{1}$$

$$T(x, t=0) = T_o, \qquad \left.\frac{\partial T}{\partial x}\right|_{x=0} = 0, \qquad T(x\to\infty, t) = T_o \tag{2}$$

Even though the radiant flux is treated as monochromatic with the absorption coefficient being the corresponding spectral quantity at that wavelength, this assumption is merely a convenience. Broadband radiation and nongray absorption could be readily handled by spectral integration.

Introducing the following variables

$$\beta = (T - T_o)/(q/kK_a), \qquad X = x/(1/K_a), \qquad \tau = t\Big/\left(1\Big/\alpha K_a^2\right) \tag{3}$$

gives a nondimensional description that is parameter-free:

$$\frac{\partial \beta}{\partial \tau} = \frac{\partial^2 \beta}{\partial X^2} + \exp(-X) \tag{4}$$

$$\beta(X, \tau=0) = 0, \qquad \left.\frac{\partial \beta}{\partial X}\right|_{X=0} = 0, \qquad \beta(X\to\infty, \tau) = 0 \tag{5}$$

Because it is parameter-free this formulation is the most efficient for representing the solution.

If a characteristic temperature T_s is introduced [where T_s is a constant, not the variable surface temperature $T_s(t)$],

$$\theta = (T - T_o)/(T_s - T_o), \qquad \chi = x/[k(T_s - T_o)/q]$$
$$\eta = t\Big/\left[k^2(T_s - T_o)^2\Big/\alpha q^2\right] \tag{6}$$

a second nondimensional formulation results, one that has a single parameter, β_s:

$$\frac{\partial \theta}{\partial \eta} = \frac{\partial^2 \theta}{\partial \chi^2} + \beta_s \exp(-\beta_s \chi), \qquad \beta_s = \frac{T_s - T_o}{q/kK_a} \tag{7}$$

$$\theta(\chi, \eta=0) = 0, \qquad \left.\frac{\partial \theta}{\partial \chi}\right|_{\chi=0} = 0, \qquad \theta(\chi\to\infty, \eta) = 0 \tag{8}$$

Because of the appearance of the parameter β_s the solution of this formulation cannot be represented as compactly as that for the preceding one. Nevertheless this formulation is useful because it identifies the parameter β_s as a nondimensional conduction–radiation

Received 8 August 2005; revision received 18 August 2005; accepted for publication 18 August 2005.

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parameter for in-depth vs surface absorption. This formulation also shows how the volumetric absorption problem transforms to the surface absorption one in the limit of a very opaque solid or the uniform volumetric absorption problem in the limit of a very transparent solid, as discussed later.

Surface Absorption

In the limit of a very opaque solid ($K_a \to \infty$, $\beta_s \gg 1$) the classical surface absorption limit is approached. The radiation source term of the previous formulation moves from the differential equation to the surface boundary condition and in the process the β_s parameter drops out:

$$\frac{\partial \theta}{\partial \eta} = \frac{\partial^2 \theta}{\partial \chi^2} \tag{9}$$

$$\theta(\chi, \eta = 0) = 0, \qquad \left.\frac{\partial \theta}{\partial \chi}\right|_{\chi = 0} = -1, \qquad \theta(\chi \to \infty, \eta) = 0 \tag{10}$$

This problem, which describes a semi-infinite solid heated by a surface heat-flux boundary condition, has a classical analytic solution that appears in most heat transfer texts:

$$\theta = \frac{2}{\sqrt{\pi}}\sqrt{\eta}\exp\left(-\frac{\chi^2}{4\eta}\right) - \chi\, erfc\left(\frac{\chi}{2\sqrt{\eta}}\right), \qquad \theta_s = 2\sqrt{\frac{\eta}{\pi}} \tag{11}$$

$$\frac{T(x,t) - T_o}{T_s(t) - T_o} = \exp\left(-\frac{x^2}{4\alpha t}\right) - \frac{x}{2}\sqrt{\frac{\pi}{\alpha t}}\, erfc\left(\frac{x}{2\sqrt{\alpha t}}\right) \tag{12}$$

$$T_s - T_o = \frac{2q}{k}\sqrt{\frac{\alpha t}{\pi}}, \qquad t = \frac{\pi}{\alpha}\left[\frac{k(T_s - T_o)}{2q}\right]^2 \tag{13}$$

Uniform Volumetric Absorption

In the limit of a very transparent solid ($K_a \to 0$, $\beta_s \ll 1$) the general problem reduces to a description of uniform volumetric heating with negligible conduction:

$$\frac{\mathrm{d}\theta}{\mathrm{d}\eta} = \beta_s, \qquad \theta(0) = 0 \tag{14}$$

$$\theta = \beta_s \eta, \qquad T(t) = T_o + K_a q\left(\frac{\alpha}{k}\right)t, \qquad t = \frac{k(T_s(t) - T_o)}{\alpha q} \tag{15}$$

For this case there is no spatial variation in temperature: $\theta_s(t) = \theta(t)$ and $T_s(t) = T(t)$.

Results

The parameter-free, nondimensional, volumetric absorption problem was solved numerically using explicit finite-differencing. The results are shown in Fig. 1. The characteristic depth of heating ($X \sim 1$) is the photon mean free path, $1/K_a$, and the characteristic time for heating ($\tau \sim 1$) is $1/\alpha K_a^2$. The characteristic temperature rise ($\beta \sim 1$) is q/kK_a. Near the surface (for $X < 1, x < 1/K_a$) the temperature profiles show the effect of in-depth absorption, regardless of time; that is, slope going to zero. For times less than the characteristic time ($\tau < 1$, $t < 1/\alpha K_a^2$) the temperature profiles show the effect of in-depth absorption (deviations from a surface absorption analysis) not only near the surface but at x-locations deeper in the solid (see discussion of Fig. 2). Although this nondimensional representation of the solution is the most compact, it is not necessarily the most conducive to physical insight.

To facilitate physical insight and the interpretation of Fig. 1 with respect to the surface-absorption assumption for radiant ignition, a baseline set of dimensional parameters was selected as follows: $K_a = 333\ \mathrm{cm}^{-1}$, $k = 0.001$ W/cm · K, $\alpha = 6.4\text{e–}4\ \mathrm{cm}^2/\mathrm{s}$, and $T_o = 300$ K ($1/K_a = 30\ \mu$m and $1/\alpha K_a^2 = 14$ ms). These parameters were selected so as to correspond nominally to a polymeric hydrocarbon, such as rubber, plastic, or solid propellant binder, subject to CO_2 laser radiation; they are not meant to simulate any particular material precisely, however.

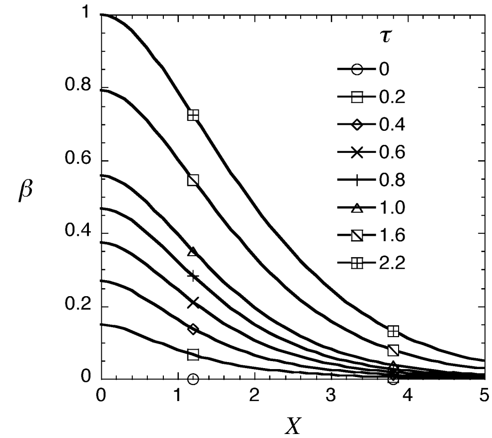

Fig. 1 Nondimensional temperature profile for radiatively heated solid with in-depth absorption (parameter-free solution).

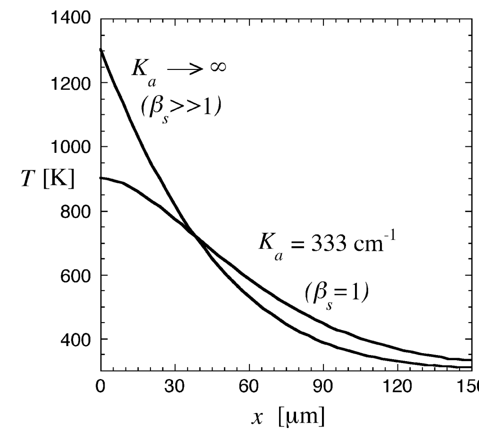

Fig. 2 Temperature profile at 31 ms for $k = 0.001$ W/cm · K, $\alpha = 6.4\text{e–}4\ \mathrm{cm}^2/\mathrm{s}$, $T_o = 300$ K, and $q = 200\ \mathrm{W/cm}^2$ for both surface ($K_a \to \infty$) and in-depth ($K_a = 333\ \mathrm{cm}^{-1}$) absorption.

The ignition problem can be posed by prescribing an ignition temperature, setting the maximum (i.e., surface) temperature to this value and solving for the time necessary to reach this T_s for a given absorbed flux. For $T_s = 900$ K and $q = 200\ \mathrm{W/cm}^2$ the surface absorption parameter (from its definition) is $\beta_s = \beta(X = 0) = 1.0$ (the characteristic temperature rise is $q/kK_a = 600$ K or $q/kK_a + T_o = 900$ K). From the numerical solution, Fig. 1, the nondimensional time needed to reach this nondimensional surface temperature is $\tau = 2.2$, which corresponds to a dimensional time of $t = (2.2)(14\ \mathrm{ms}) = 31$ ms.

Figure 2 shows the dimensional temperature profile in the solid for these conditions ($q = 200\ \mathrm{W/cm}^2$ and $T_s = 900$ K). Figure 2 also shows the temperature profile for the surface-absorption case. For surface absorption, the surface would reach 1305 K in 31 ms (if the solid remained inert with constant properties) or the ignition temperature of 900 K in only 11 ms. Thus Fig. 2 shows how the surface absorption analysis overpredicts surface temperature or underpredicts ignition time; in this case, the latter is underpredicted by a factor of three.

Ignition of energetic solids is often studied experimentally by varying radiant heat flux and observing its effect on ignition time. Figure 3 shows the effect of absorbed flux on ignition time (time to reach $T_s = 900$ K) for both surface absorption ($K_a \to \infty$, $\beta_s \gg 1$) and in-depth absorption ($K_a = 333\ \mathrm{cm}^{-1}$). For the latter case when the in-depth absorption parameter β_s is of order one, the general numerical results are used in the plot (recall that the value

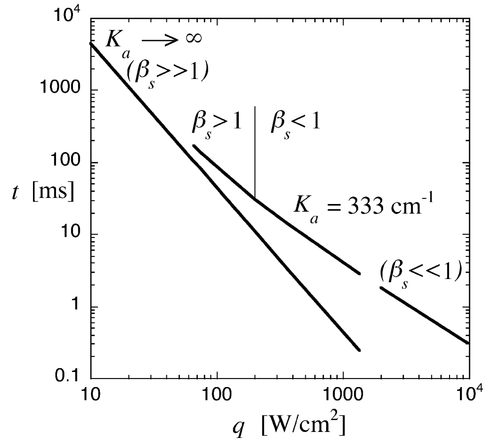

Fig. 3 Effect of absorbed flux on time to reach $T_s = 900$ K (ignition time) for $k = 0.001$ W/cm · K, $\alpha = 6.4\mathrm{e}{-4}$ cm^2/s, and $T_o = 300$ K for surface absorption ($K_a \rightarrow \infty, \beta_s \gg 1$) and in-depth absorption ($K_a = 333$ cm^{-1}). For intermediate values of β_s (order one), the general numerical results are plotted; for $\beta_s < 0.1$, the uniform volumetric absorption solution is used (labeled $\beta_s \ll 1$).

$\beta_s = 1$ occurs at $q = 200$ W/cm^2); for $\beta_s \leq 0.1$ (which corresponds to $q \geq 2000$ W/cm^2) the uniform volumetric absorption solution is appropriate and is thus used in the plot (labeled $\beta_s \ll 1$ in Fig. 3). The surface absorption case has the classical slope of -2 on a log-log plot, corresponding to the analytical solution noted previously. The uniform, volumetric absorption case has a slope of -1, corresponding to its analytical solution, also noted previously. The in-depth absorption case exhibits longer ignition times for a given absorbed flux than the surface absorption case, and the slope of its ignition curve is smaller in magnitude and decreases in magnitude with increasing flux. This is noteworthy because such a deviation in the slope from the classical -2 surface heat-flux solution is often observed in experimental data and yet rarely is in-depth absorption cited as a possible factor.[5]

Discussion

This simple analysis shows that assuming surface absorption, when it is in fact not valid, leads to underestimating radiant ignition times of solids. It also shows that in-depth absorption can be a factor in the deviation of experimental data for ignition time vs radiant flux from the classical slope of -2 on a log-log plot, which is often observed. This is not to suggest that in-depth absorption is always the cause for deviation from a slope of -2 or the only cause; clearly there have been situations reported (e.g., CO_2 laser ignition of cyclo-tetramethylene-tetranitramine[6]) where other factors contributed. However, this analysis and these results do suggest that in-depth absorption should always be considered in interpreting experimental data of radiant ignition of both energetic and nonenergetic solids (e.g., Refs. 7 and 8).

The way to estimate the importance of in-depth absorption has also been elucidated by this analysis; it is to evaluate the in-depth absorption parameter β_s, which comes out of the nondimensional formulations. This parameter can be thought of as a conduction–radiation parameter, or more specifically a ratio of characteristic length scales for conduction to radiation, as follows:

$$\beta_s = \frac{k(T_s - T_o)/q}{1/K_a} = \frac{x_c}{x_r} \tag{16}$$

The denominator x_r is the characteristic length scale for in-depth radiation absorption, the photon mean free path, and it is essentially constant in time. The numerator x_c is the characteristic length scale for conduction. It is the thickness of a layer of solid heated by conduction when the heat flux is applied at the surface. It grows as the square root of time from the analytic solution. Early in the radiant heating process, at sufficiently small times, T_s is low and x_c is small relative to $x_r(\beta_s < 1)$, indicating that in-depth absorption is important. In the limit $\beta_s \ll 1$ conduction in the solid is negligible, and the volumetric heating is nearly uniform spatially near the surface. At long times, as T_s rises, x_c becomes large relative to $x_r(\beta_s \gg 1)$ indicating that in-depth absorption is not as important and the surface absorption approximation is more appropriate. Thus the lower the ignition temperature of a material, the less valid the surface-absorption assumption in interpreting its ignition time data.

The effect of several pertinent variables in addition to ignition temperature (i.e., thermal conductivity, heat flux, and opacity) on the in-depth absorption parameter can be estimated from its definition. The following trends are conducive to surface-absorption treatment: long time and/or high ignition temperature, low heat flux, high thermal conductivity, and high opacity. The following conditions on β_s summarize the heating of a solid by a radiant heat flux in the vicinity of the heated surface: $\beta_s \gg 1$, surface absorption (surface heat-flux B.C.) valid; $\beta_s \sim O(1)$, exponentially distributed, in-depth absorption; and $\beta_s \ll 1$, uniform volumetric absorption with negligible conduction.

Summary

A simple numerical analysis is described for investigating the surface absorption assumption in radiant ignition of energetic solids. A new, nondimensional parameter is defined that quantifies the importance of in-depth (volumetric) absorption compared to surface absorption: $\beta_s = (T_s - T_o)/(q/kK_a)$. For $\beta_s \gg 1$ (high ignition temperature T_s, low absorbed radiant flux q, high thermal conductivity k, and high absorption coefficient K_a), the surface absorption assumption is valid; for $\beta_s \geq 1$ (low ignition temperature T_s, high absorbed radiant flux q, etc.), in-depth absorption is important. Applying realistic material properties to these results shows that in many cases in-depth absorption is significant and that assuming surface absorption can cause ignition times to be severely underestimated. At sufficiently short heating times ($t < 1/\alpha K_a^2$), surface absorption analysis substantially overestimates surface temperature.

Acknowledgments

The author appreciates support from the Hermia G. Soo Professorship and the Center for Simulation of Advanced Rockets.

References

[1] Ritchie, S. J., Thynell, S. T., and Kuo, K. K., "Modeling and Experiments of Laser Induced Ignition of Nitramine Propellants," *Journal of Propulsion and Power*, Vol. 13, No. 2, 1997, p. 367.

[2] Baer, A. D., and Ryan, N. W., "An Approximate but Complete Model for the Ignition Response of Solid Propellants," *AIAA Journal*, Vol. 6, No. 5, 1968, p. 872.

[3] Bradley, H. H., Jr., "Theory of Ignition of a Reactive Solid by Constant Energy Flux," *Combustion Science and Technology*, Vol. 2, No. 1, 1970, p. 11.

[4] Linan, A., and Williams, F. A., "Theory of Ignition of a Reactive Solid by Constant Energy Flux," *Combustion Science and Technology*, Vol. 3, No. 1, 1971, p. 91.

[5] Hermance, C. E., "Solid-Propellant Ignition Theories and Experiments," *Fundamentals of Solid Propellant Combustion*, edited by K. K. Kuo and M. Summerfield, AIAA Progress in Astronautics and Aeronautics, Vol. 84, AIAA New York, 1984, Chap. 5.

[6] Ali, A. N., Son, S. F., Asay, B. W., Decroix, M. E., and Brewster, M. Q., "High-Irradiance Laser Ignition of Explosives," *Combustion Science and Technology*, Vol. 175, 2003, p. 1551.

[7] Weber, J. W., Brewster, M. Q., and Tang, K. C., "Radiative Ignition and Extinction Dynamics of Energetic Solids," *Journal of Thermophysics and Heat Transfer*, Vol. 19, No. 3, 2005, p. 1.

[8] Cain, J., and Brewster, M. Q., "Radiative Ignition of Fine-Ammonium Perchlorate Composite Propellants," *Propellants, Explosives, and Pyrotechnics* (submitted for publication).