STRUCTURE AND STABILITY OF DEFLAGRATIONS IN POROUS ENERGETIC MATERIALS*

Stephen B. Margolis[†] **and Forman A. Williams**[‡]

[†]*Combustion Research Facility, Sandia National Laboratories, Livermore, California 94551-0969 USA*
E-mail:margoli@sandia.gov

[‡]*Applied Mechanics and Engineering Sciences, University of California, La Jolla, CA 92093-0411 USA*
E-mail:faw@ames.ucsd.edu

Extended Abstract

The combustion behavior of porous energetic materials is of increasing interest due to the realization that even supposedly nonporous materials may develop significant porosities over time due either to aging or to other types of degradation that may arise from exposure to abnormal environments. In such materials, two-phase-flow effects are especially significant due to the presence of gas flow relative to the condensed material, both within the unburned porous solid as well as in the exothermic liquid/gas layers that typically form on the surfaces of many types propellants, such as nitramines. In the presence of confinement, the significance of the convective transport effects due to two-phase flow are enhanced, leading, through gas permeation into the unburned solid, to a preheating of the solid and, consequently, to a strong enhancement of the burning rate relative to the unconfined case. Indeed, this type of preheating associated with gas permeation into the unburned solid is generally associated with the onset of specially identified modes of combustion, such as convective burning.[1] However, even in unconfined problems, two-phase-flow effects play an important role, affecting not only fundamental thermodynamic characteristics such as the burned temperature, but also the burning rate and the stability of steady, planar burning.

In the present work, we synthesize and extend the results of several recent analyses that, by means of asymptotic methods, have been successful in predicting the effects of two-phase flow on the structure, burning rate and linear stability of a propagating deflagration wave.[2–5] For the case of a confined or partially confined geometry, a quasi-steady propagation regime can be identified such that the structure and other features of the deflagration wave collapses to that of an unconfined deflagration in the limit that the pressure difference, or overpressure, between the burned and unburned regions approaches zero. Indeed, a sketch of the geometry for the simplified chemistry adopted here is shown in Fig. 1, which indicates an unburned solid/gas region, a melting surface, a liquid-gas region within which is embedded a thin reaction zone in which burning takes place, and finally a burned gaseous product region. The primary difference between the unconfined and confined problems is that in the case of confinement, the direction of gas flow in the unburned region is likely to be negative, resulting in a preheating effect that results in a rapid increase in the burning rate as a function of overpressure. This permeation-enhanced burning is generally referred to, for obvious reasons, as convective burning, and one of the successes of the present model formulation and analysis is the ability to predict the transition from ordinary conduction-controlled burning that is characteristic of unconfined deflagrations to one in which convection plays a significant role.

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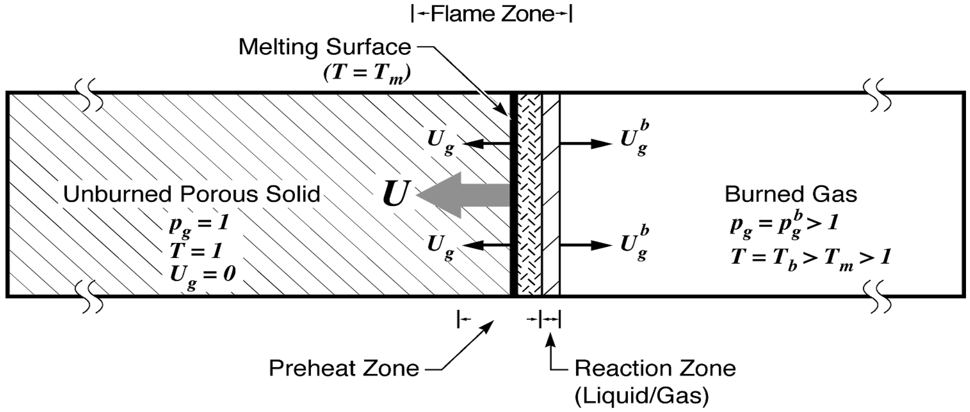
Figure 1



The model used to investigate the wave structure described above is essentially a simplified version of more general models of two-phase reacting flow. In particular, the model essentially consists of continuity and energy equations for each coexisting phase, a simplified accounting for momentum conservation appropriate for deflagrations, an equation of state for the gas, and an appropriate set of boundary and melting surface conditions. Although it is critical to the focus of our study to allow for velocity differences between coexisting phases, for simplicity we assume good thermal contact and adopt the single-temperature approximation that the temperature at a given spatial location is the same for each phase. Also, in keeping with our goal of focusing on two-phase-flow effects, we deliberately simplify the chemistry by postulating the overall process $R(s) \longrightarrow R(l) \longrightarrow P(g)$, where the first step denotes the melting (assumed to be slightly endothermic) of the solid material, and the second represents a one-step exothermic process in which liquid-phase reactants are directly converted to burned gaseous products. Extensions of the analysis to more complicated global mechanisms have been given,[6,7] but such extensions are not critical to the examination of the primary two-phase-flow effects of interest here.

Assuming a quasi-steady gas phase and weak nonplanarity of the combustion front, it is possible, in the limit of large activation energy, to derive a closed asymptotic model given by

$$(1-\alpha_s)\left\{\begin{matrix}1\\ rb\\ 0\end{matrix}\right\}\frac{\partial T}{\partial t}-\left\{\begin{matrix}1+\alpha_s(\hat{r}\hat{b}-1)\\ rb+\alpha_s(\hat{r}\hat{b}-rb)\\ \hat{r}\hat{b}\end{matrix}\right\}\frac{\partial x_r}{\partial t}\frac{\partial T}{\partial \zeta}-(1-\alpha_s)\left[(1-r)\left\{\begin{matrix}0\\ b\\ \hat{b}\end{matrix}\right\}\frac{\partial x_m}{\partial t}+(r-\hat{r})\left\{\begin{matrix}0\\ 0\\ \hat{b}\end{matrix}\right\}\frac{\partial x_r}{\partial t}\right]\frac{\partial T}{\partial \zeta}$$
$$=\left\{\begin{matrix}1+\alpha_s(\hat{l}-1)\\ l+\alpha_s(\hat{l}-l)\\ \hat{l}\end{matrix}\right\}\nabla_r^2 T+\hat{r}\hat{b}(1-\gamma^{-1})\alpha_s\left\{\begin{matrix}1\\ 0\\ 0\end{matrix}\right\}\frac{\partial x_r}{\partial t}\frac{\partial p_g}{\partial \zeta}\,,\quad \begin{matrix}\zeta<-(x_r-x_m)\\ -(x_r-x_m)<\zeta<0\,,\\ \zeta>0\end{matrix} \tag{1}$$

$$T=p_g\left[1-\frac{\kappa}{\alpha_s}\frac{\partial p_g}{\partial \zeta}\left(\frac{\partial x_r}{\partial t}\right)^{-1}\right],\quad \zeta<-(x_r-x_m)\,, \tag{2}$$

$$-r\frac{\partial x_r}{\partial t}-(1-r)\frac{\partial x_m}{\partial t}=G_r\exp\left(-\frac{\beta}{2}\cdot\frac{T_b-T\big|_{\zeta=0}}{T_b-1}\right), \tag{3}$$

subject to

$$T\to p_g\to 1\ \text{ as }\ \zeta\to-\infty\,,\quad T\to T_b\ \text{ as }\ \zeta\to+\infty\,, \tag{4}$$

$$T=T_m\ \text{ at }\ \zeta=-(x_r-x_m)\,,\quad p_g=p_g^b\ \text{ for }\ \zeta\geq-(x_r-x_m)\,, \tag{5}$$

$$\left(-\frac{\partial x_m}{\partial x_1}, -\frac{\partial x_m}{\partial x_2}, 1\right) \cdot \left\{ \left[l + \alpha_s(\hat{l} - l)\right] \nabla_r T\Big|_{\zeta=-(x_r-x_m)^+} - \left[1 + \alpha_s(\hat{l} - 1)\right] \nabla_r T\Big|_{\zeta=-(x_r-x_m)^-} \right\} = \frac{\partial x_m}{\partial t}(1-\alpha_s)\left[\gamma_s + (1-b)T_m\right] , \tag{6}$$

$$T\Big|_{\zeta=0^-} = T\Big|_{\zeta=0^+} , \tag{7}$$

$$\hat{l}\frac{\partial T}{\partial \zeta}\bigg|_{\zeta=0^+} - \left[l + (\hat{l} - l)\alpha_s\right] \frac{\partial T}{\partial \zeta}\bigg|_{\zeta=0^-} + (1-\alpha_s)(l-\hat{l})G_r^{-2}\left(\frac{\partial x_r}{\partial x_1}\frac{\partial T}{\partial x_1} + \frac{\partial x_r}{\partial x_2}\frac{\partial T}{\partial x_2}\right)\bigg|_{\zeta=0} = -(1-\alpha_s)\left[Q + (b-\hat{b})T\Big|_{\zeta=0}\right] G_r^{-1} \exp\left(-\frac{\beta}{2}\cdot\frac{T_b - T\big|_{\zeta=0}}{T_b - 1}\right) , \tag{8}$$

where the above equations have been written in a nonorthogonal moving coordinate system (x_1, x_2, ζ) such that $\zeta = 0$ coincides with the infinitesimally thin reaction zone. The dependent variables are thus the (single) temperature T, the gas pressure p_g, and the locations $x_3 = x_r(x_1, x_2, t)$ and $x_3 = x_m(x_1, x_2, t)$ of the melting and reaction sheets, respectively, in a fixed frame of reference. The remaining quantities (parameters) of interest are the porosity α_s, permeability κ, heat release Q, ratio of specific heats γ, heat of melting γ_s, Zel'dovich number β, liquid-to-solid density, conductivity and heat-capacity ratios r, l and $\hat{b}$, respectively, and the corresponding (upstream) gas-to-solid ratios $\hat{r}$, $\hat{l}$ and $\hat{b}$. The operators ∇_r and ∇_r^2 are the gradient and Laplacian in the moving coordinate system, and $G_r = 1 + (\partial x_r/\partial x_1)^2 + (\partial x_r/\partial x_2)^2$. Other quantities of interest, such as the gas velocity u_g, are expressed in terms of the four dependent variables x_r, x_m, T and p_g, where a single equation for p_g is obtained by combining Eqs. (1) and (2) in the region $\zeta < -(x_r - x_m)$, the latter being derived from gas-phase continuity, Darcy's law and the (ideal) equation of state.

A basic solution corresponding to steady, planar burning is obtained from the above formulation by retaining only derivatives with respect to the normal coordinate ζ. The resulting reaction-zone analysis leading to Eq. (3) and the jump condition (8) also determines the propagation speed $\tilde{U}$ for this solution, as well as the nontrivial velocity and pressure profiles in the solid/gas region. The first of these is determined from the ratio $U^* = \tilde{U}(p_g^b)/\tilde{U}(1) = U_n[\tilde{A}(p_g^b)/\tilde{A}(1)]^{1/2}$, where $\tilde{A}(p_g)$ is the rate coefficient and U_n is given by

$$U_n = \frac{T_b(p_g^b)}{T_b(1)}\left[\frac{(b-\hat{b})T_b(1) + Q}{(b-\hat{b})T_b(p_g^b) + Q}\right]^{1/2} \exp\left[\frac{N_u}{2}\left(\frac{1}{T_b(1)} - \frac{1}{T_b(p_g^b)}\right)\right] , \tag{9}$$

with T_b a function of p_g^b according to

$$T_b = \frac{(1-\alpha_s)(Q + 1 + \gamma_s) + \hat{r}\hat{b}\alpha_s\left[1 + \chi(p_g^b - 1)\right]}{\hat{b}(1 - \alpha_s + \hat{r}\alpha_s)} . \tag{10}$$

In this form, it is readily seen, since T_b is a linearly increasing function of overpressure and the nondimensional activation temperature N_u (expressed as a multiple of the unburned temperature) is typically very large, that U_n is exponentially sensitive to T_b and hence p_g^b as the overpressure $p_g^b - 1$ increases from zero. Thus, as the overpressure increases, the burning rate increases exponentially (Fig. 2), reflecting its sensitivity to the corresponding increase in the rate of gas permeation into the solid/gas region. In the limit of large overpressures, T_b^{-1} becomes small and the exponential factor in Eq. (9) approaches a constant value. Consequently, in the range of large overpressures, the dependence of U_n on p_g^b becomes algebraic. This is also illustrated in Fig. 2, in which case (since $b = \hat{b}$) the saturated dependence of U_n on p_g^b is linear. We note that this feature (exponential transition to an algebraic pressure-dependent burning rate) is qualitatively consistent with most experiments in Crawford-type (large volume) bombs that indicate a rapid increase in the burning rate frequently associated with the onset of convective burning,[1] followed by a less dramatic pressure dependence that is typically represented in the form Ap^n. The gas velocities at the melting surface, $u_g(0) = T_m/p_g^b - 1$, and in the burned region, u_g^b, are especially of interest and are shown in Fig. 3. It is seen that gas permeation into the solid, as indicated by negative values $u_g(0)$, begins for extremely modest overpressures, and that for sufficiently large overpressures, even the gas velocity in the burned region is directed upstream, in the direction of the solid.

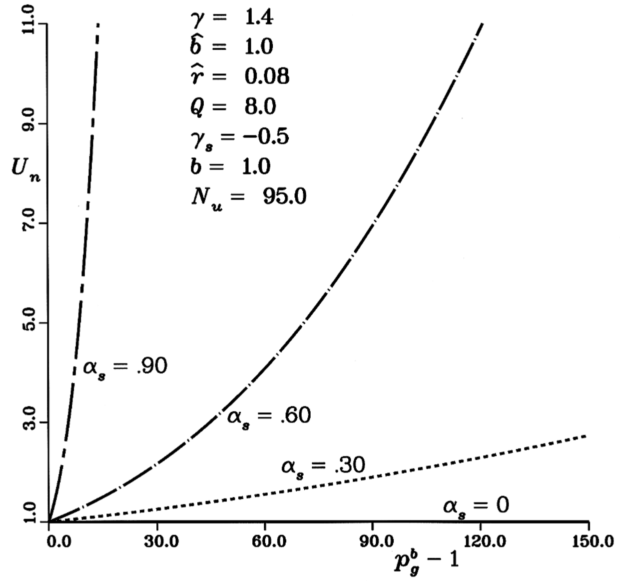


Figure 2

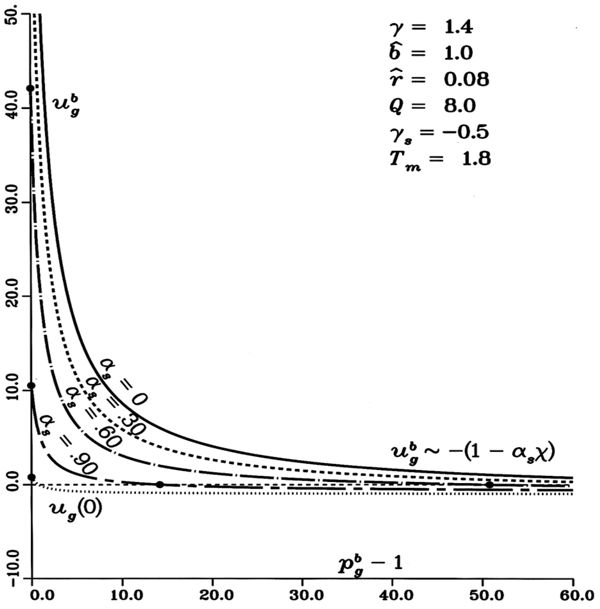


Figure 3

A stability analysis of this basic solution can be undertaken, using the asymptotic model (1) – (8), in a standard fashion. Results for the limiting (unconfined) case $p_g^b \to 1$ indicate the existence of a pulsating stability boundary similar to that predicted for nonporous propellants.[8–12] The effect of porosity is then shown to be destabilizing with respect to the stability threshold predicted in the limit $\alpha_s \to 1$. Corresponding results for the unconfined problem ($p_g^b > 1$) indicate a strong dependence of the stability boundary on $p_g^b - 1$ arising from the dependence of the burned temperature on this parameter, as described above.

References

1. Asay, B. W., Son, S. F., and Bdzil, J. B., *Int. J. Multiphase Flow*, Vol. 22, 1996, pp. 923–952.
2. Margolis, S. B., and Williams, F. A., *J. Propulsion Power*, Vol. 11, 1995, pp. 759–768.
3. Margolis, S. B., and Williams, F. A., *Combust. Sci.Tech.*, Vol. 106, 1995, pp. 41–68.
4. Margolis, S. B., and Williams, F. A., *Int. J. Multiphase Flow*, Vol. 22, 1996, pp. 69–91.
5. Margolis, S. B., *Combust. Theory Modelling*, Vol. 2, pp. 95–113.
6. Li, S. C., Williams, F. A., and Margolis, S. B., *Combustion and Flame*, Vol. 80, 1990, pp. 329–349.
7. Ilincic, N., and Margolis, S. B., *Combust. Sci. Tech.*, Vol. 125, pp. 201–241.
8. Denison, M. R., and Baum, E., *ARS J.*, Vol. 31, 1961, pp. 1112–1122.
9. Krier, H., T'ien, J. S., Sirignano, W. A., and Summerfield, M., *AIAA J.*, Vol. 6, 1968, p. 278.
10. Higuera, F. J., and Liñán, A., *Progress in Astronautics and Aeronautics*, Vol. 95, 1984, pp. 248–256.
11. Margolis, S. B., and Armstrong, R. C., *Combust. Sci. Tech.*, Vol. 47, 1986, pp. 1–38.
12. Margolis, S. B., and Williams, F. A., *Combust. Sci. Tech.*, Vol. 59, 1988, pp. 27–84.