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International Journal of Multiphase Flow 25 (1999) 1505–1519

International Journal of
**Multiphase
Flow**

www.elsevier.com/locate/ijmulflow

On the existence of multiphase thermal detonations

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Received 11 January 1999; received in revised form 18 May 1999

With best wishes to Gad Hetsroni on the occasion of his 65th birthday.

Abstract

The classical Board–Hall model is re-examined and shown to imply that, contrary to the model's intention and widely-held interpretation, multiphase thermal detonations are not physically possible. A new key physical concept is introduced, and the results show the existence of supercritical detonations, even in lean (melt content) premixtures, in agreement with recent experimental evidence. This opens the way for a rational approach to predicting the energetics of such explosions, and indicates the kind of constitutive laws needed for this purpose. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Microinteractions; Multiphase explosions; Steam explosions; Thermal detonations; Fuel–coolant interactions

1. Introduction

In 1975, Board et al. (1975) proposed the now-classic model of multiphase thermal detonations (a physical explosion where the thermal energy of a melt drives and sustains the pressure wave). In complete analogy with chemical detonations, it involves a premixed state, a reaction zone over which the constituents react and equilibrate, and seeks to determine conditions under which a pressure wave can be sustained in a steady state as it propagates through the premixture. The solution of the basic conservation equations augmented by the equation of state (and any chemical energy involved in the reaction) also yields the pressure and speed of the shock wave. Early on, the model was extensively criticized on grounds

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varying from the seemingly excessive fragmentation rates required to sustain the wave, to the attainability of the steady state solution in systems of finite dimension, to more esoteric aspects of multiphase interactions (Condiff, 1982). Eventually, however, the Board and Hall concept prevailed, as workers set out to address the above criticisms through the use of more detailed numerical models — that is, accounting for melt fragmentation rates (obtained from experiments or analyses), and finite rates of escalation, as predicted by the multiphase dynamics (Sharon and Bankoff, 1981; Scott and Berthoud, 1978; Fletcher, 1991a; Medhekar et al., 1991). However, the basic frame of melt fragmentation and mixing with the coolant remained the same, as in the original Board and Hall model, and this turned out to be very problematic.

One purpose of this paper, as may be evident already from the title, is to point out this problem. The other purpose is to resolve it, and to provide an initial illustration of the implications of this resolution.

2. The Board and Hall solution

The basic concept is illustrated in Fig. 1. Assuming a sufficiently ‘thin’ reaction zone, the conservation equations across the shock can be written, with reference to Fig. 2, as

$$\frac{u_1}{v_1} = \frac{u_2}{v_2} \quad (1)$$

$$p_1 + \frac{u_1^2}{v_1} = p_2 + \frac{u_2^2}{v_2} \quad (2)$$

$$h_1 + \frac{1}{2}u_1^2 = h_2 + \frac{1}{2}u_2^2 \quad (3)$$

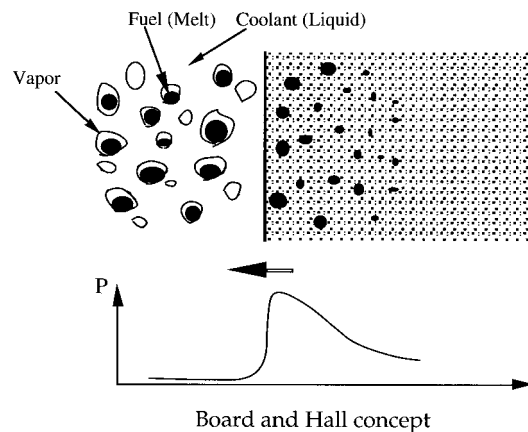


Fig. 1. Illustration of the Board and Hall thermal detonation concept.

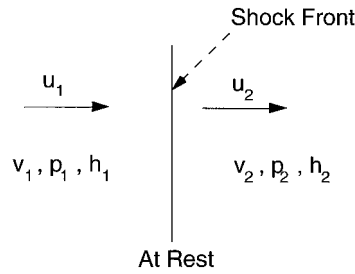


Fig. 2. Definition of quantities in a frame moving with the shock.

where u denotes the velocity, v the specific volume, p the pressure, and h the enthalpy. The subscripts 1 and 2 indicate the state ahead and behind the shock, respectively.

The solution then is obtained by

$$h_1 - h_2 + \frac{1}{2}(p_2 - p_1)(v_1 + v_2) = 0 \tag{4}$$

which with the equation of state, for the coolant,

$$h = h(p, v) \tag{5}$$

leads to the Hugoniot, as illustrated in Fig. 3, for the example given in the original Board and Hall paper. The definition of the Chapman–Jouget (C–J) point, from a given premixture state (p_1, v_1) , is also illustrated in this figure. The shock speed is given by the slope of the tangent to the Hugoniot that defines the C–J point, i.e.,

$$\rho_1 u_1 = j = \sqrt{\frac{p_2 - p_1}{v_1 - v_2}} \tag{6}$$

where ρ is density and j is the mass flux.

At the shock front the pressure is known as the von Neumann ‘spike,’ and it falls towards

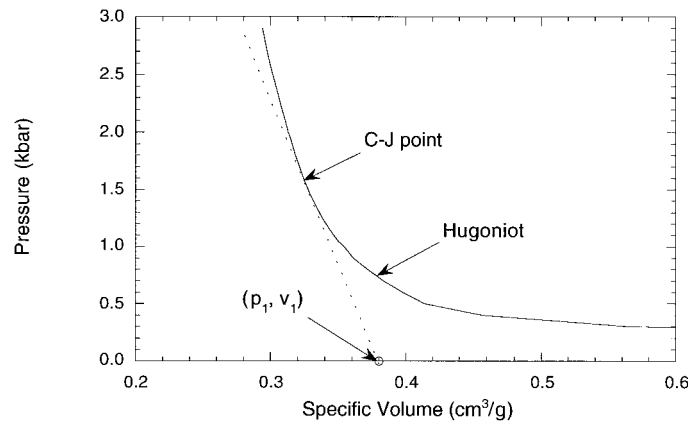


Fig. 3. Illustration of the Board and Hall solution for the specific numerical example given in their original paper.

the Hugoniot, more or less rapidly depending on the reaction rate (fragmentation rate in the present case), to the C–J point.

Applying this procedure for the complete range of premixture void fractions (defined as the fractional volume of the whole coolant space occupied by vapor), we obtain the C–J pressures shown in Fig. 4. In their specific illustration, Board and Hall used an equal volume, three phase premixture; that is, $\sim 33\%$ each, melt, water, and steam, and obtained a C–J pressure of ~ 800 bar. In Figs. 3 and 4, we see a C–J pressure of ~ 1500 bar. The discrepancy probably is due to differences in the equation of state, which was not specified by Board and Hall. In our solution, we use property data generated from numerical interpolation of a standard table (Harr et al., 1984).

In Fig. 4, we also show results for melt volume fractions of 5, 10, 20, and 50%, and the following trends can be discerned.

- ‘Lean’ premixtures cannot detonate, to supercritical pressures (above the thermodynamic critical value), unless the premixture void fraction is very nearly zero (when the compressibility of the mixture decreases rapidly to that of a pure liquid). Such low void fractions are not physically possible in premixtures of any significant size.
- ‘Lean’ premixtures can yield weak propagations at the very high end of the void fraction scale.
- ‘Rich’ premixtures can detonate, under most void fractions, but produce only weak propagations (i.e., not highly supercritical) at the very high end of the void fraction scale. However, this (high end) is precisely where rich premixtures of any significant size would find themselves physically, due to heat transfer (film boiling and radiation) during premixing.

We see in particular that the premixture specification chosen by Board and Hall, i.e., rich in fuel, is on the one hand necessary to produce a supercritical detonation, but on the other hand it is justifiably open to criticism as physically impossible. In fact, this was one reason, we suspect, that subsequently much attention was focused on the phenomenon of ‘water depletion’ in premixtures (Henry and Fauske, 1981). It now has been verified both experimentally

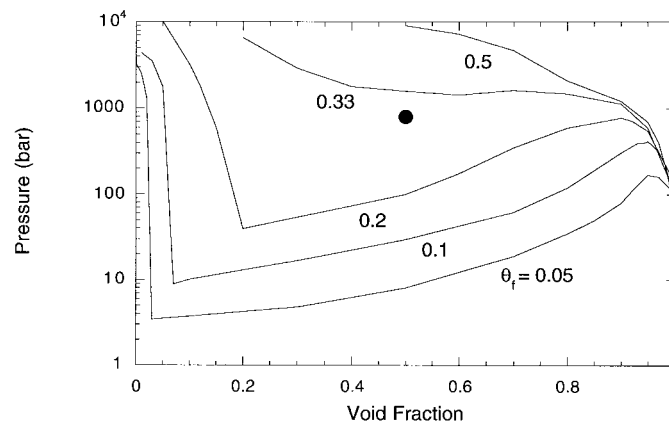


Fig. 4. The C–J pressures obtained according to the Board and Hall model for 1500°C tin premixtures in water. The parameter is tin volume fraction (θ_f). ● Board and Hall example.

(Angelini et al., 1992, 1995) and analytically (Theofanous et al., 1998a; Fletcher and Thyagaraja, 1991).

Similar trends are found for the practically much more important (than tin) nuclear reactor fuel, as illustrated in Fig. 5. It is important to note that in this case radiation heat transfer overwhelms the premixing behavior, so that a melt concentration of only a few percent is more than sufficient to effectively deplete large-scale premixtures from all water (void fraction, $\alpha > 90\%$) (Angelini et al., 1995; Theofanous et al., 1998a).

Thus, the Board and Hall model predicts that steady state supercritical detonations require highly contrived premixtures that cannot be expected in most practical situations.

Actually, experience with transient propagation models shows that the problem is even deeper, in that explosions cannot propagate unless, in **addition** to rich premixtures, **unreasonably high** fragmentation rates are assumed (Fletcher and Anderson, 1990; Fletcher and Theofanous, 1997). Thus, the very existence of supercritical thermal detonations has to be questioned.

3. The microinteractions model

The above reveal a very fundamental difficulty with the whole concept, and it is interesting that due to this difficulty, **all** subsequent, more detailed, transient multiphase models have actually taken significant steps **backwards**. The reason for this is rather straightforward, when one thinks about how the feedback supporting the detonation wave is generated. It really involves the heating and consequent expansion of the coolant, in single or two-phase, and this is a strong function of mixing mass fractions of the fragmented debris and of the coolant mixing with it, immediately, behind the shock. Board and Hall assume that all fuel (one could just as easily take a fraction, but this is not essential here) will become debris, and all of it will equilibrate with all the coolant at the C–J point. This **eventually** (that is, at a sufficient distance behind the shock) would be correct, provided the C–J point exists, and that it can be reached

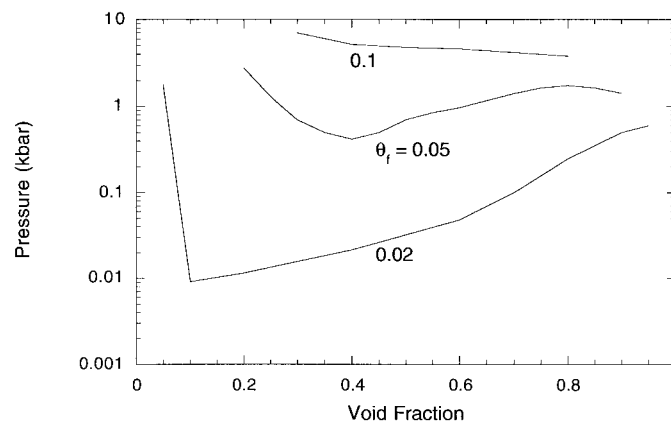


Fig. 5. The C–J pressures obtainable according to the Board and Hall model for 3300°C uranium dioxide premixtures in water. The parameter is melt volume fraction (θ_f).

with physically reasonable rates of fragmentation and mixing. But these are the ‘ifs’ that beg the question. On the other hand, the transient multifield models pursued as an extension to Board and Hall, using finite fragmentation rates, as they should to properly represent the physics of fragmentation, are bound to produce an essentially infinite dilution to begin with (by mixing the debris with all available coolant in the volume swept by the front in the time increment); hence, they would **always** be incorrect. This is why they cannot produce sufficient feedback to sustain a given trigger wave, unless unreasonably high melt concentrations **and** fragmentation rates are assumed.

This ‘dilution’ problem was first identified and addressed by Yuen et al. (1992). They made use of the two-dimensional (2D) feature of their propagation code to restrict the amount of coolant initially available to mix with the debris generated behind the shock. They also showed physical evidence of a limited mixing, with a melt drop exploding under simulated large scale explosion conditions (sustained pressure wave, obtained at UCSB’s SIGMA facility). This in turn led to the formulation of the microinteractions model (Yuen and Theofanous, 1994), its numerical implementation in a code known as ESPROSE.m (Theofanous and Yuen, 1994; Yuen and Theofanous, 1995; Theofanous et al., 1998c), and a related experimental program (on the SIGMA again) aimed at the constitutive laws of these microinteractions (Chen et al., 1995; Theofanous et al., 1998c). Here, we make use of the microinteractions concept to address the question raised in the previous section. The solutions obtained have also been useful to test the numerics and other salient features of the ESPROSE.m code. More importantly, we provide here the first interpretation of the steady state theory in terms of fully dynamic propagations computed numerically.

A sample of the physical evidence for microinteractions is shown in Fig. 6(a)–(c). They contain high-speed movie (Fig. 6(a)) and flash X-ray images (Fig. 6(b)) of a molten (1650°C) steel drop forced to explode with water under a sustained pressure wave of 265 bar (4000 psi). Respective images of the steel drop just prior to the explosion are shown in Fig. 6(c). The key observations are that the fragmentation develops rapidly, in 10 s of microseconds, and that the resulting debris mixes with water (to produce what we call the m-fluid) in a confined region within the immediate vicinity of the drop. For example, at 0.24 ms, the ratio of the mixing volume to the initial volume of the drop is ~ 4 .

The physical concept of microinteractions is illustrated in Fig. 7. For the mathematical model, we will assume, again, that the reaction zone is very thin and that velocity equilibration is obtained immediately behind it. The continuity and momentum equations are thus identical to the original Board and Hall model and are given by Eqs. (1) and (2). However, now thermal equilibrium is restricted to the m-fluid (a mixture of the fragmented debris and entrained coolant), while the non-participating coolant is simply compressed (adiabatically), and the m-fluid expands against it towards a common pressure. The final state is now characterized by the two temperatures, one of the m-fluid ($T_{m,2}$) and one of the non-participating coolant ($T_{c,2}$), and the common pressure p_2 . Accordingly, two separate energy equations are required, one for the non-participating coolant and one for the constituents (fuel and coolant) of the m-fluid. Supposing that the coolant-to-fuel mass ratio in it is expressed by \hat{f}_c (mass of entrained coolant per unit mass of fuel), we have

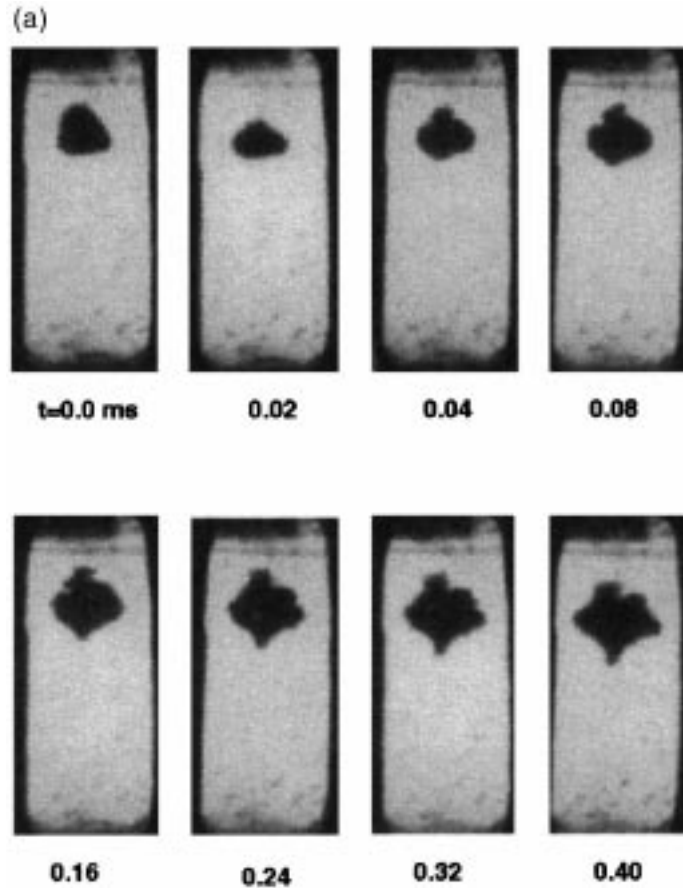


Fig. 6. (a) High-speed movie images of an exploding steel drop in run S-4-15.5. (b) Top: high-speed movie and flash X-ray images of an exploding steel drop, at 0.32 ms after forcing the interaction, SIGMA run S-4-15.5. Bottom: quantitative rendering of the X-ray image (projected mass distribution). (c) Images corresponding to those in (b), but just prior to the interaction (prior to the arrival of the pressure wave).

$$h_{c,1} + \frac{1}{2}u_1^2 = h_{c,2} + \frac{1}{2}u_2^2 \tag{7}$$

and

$$h_f(T_{f,1}) + \frac{1}{2}u_1^2 + \hat{f}_e \left(h_{c,1} + \frac{1}{2}u_1^2 \right) = h_f(T_{m,2}) + \frac{1}{2}u_2^2 + \hat{f}_e \left(h_{m,2} + \frac{1}{2}u_2^2 \right) \tag{8}$$

In these, $h_{m,2}$ is computed from the equation of state of the coolant at temperature and pressure of $T_{m,2}$ and p_2 , respectively, while the $h_{c,2}$ and $T_{m,2}$ are obtained from adiabatic compression and compression/equilibration of the non-participating coolant, and of the m-fluid constituents, respectively.

The solution of Eqs. (1), (2), (5), (7) and (8) produces a whole family of ‘microinteraction

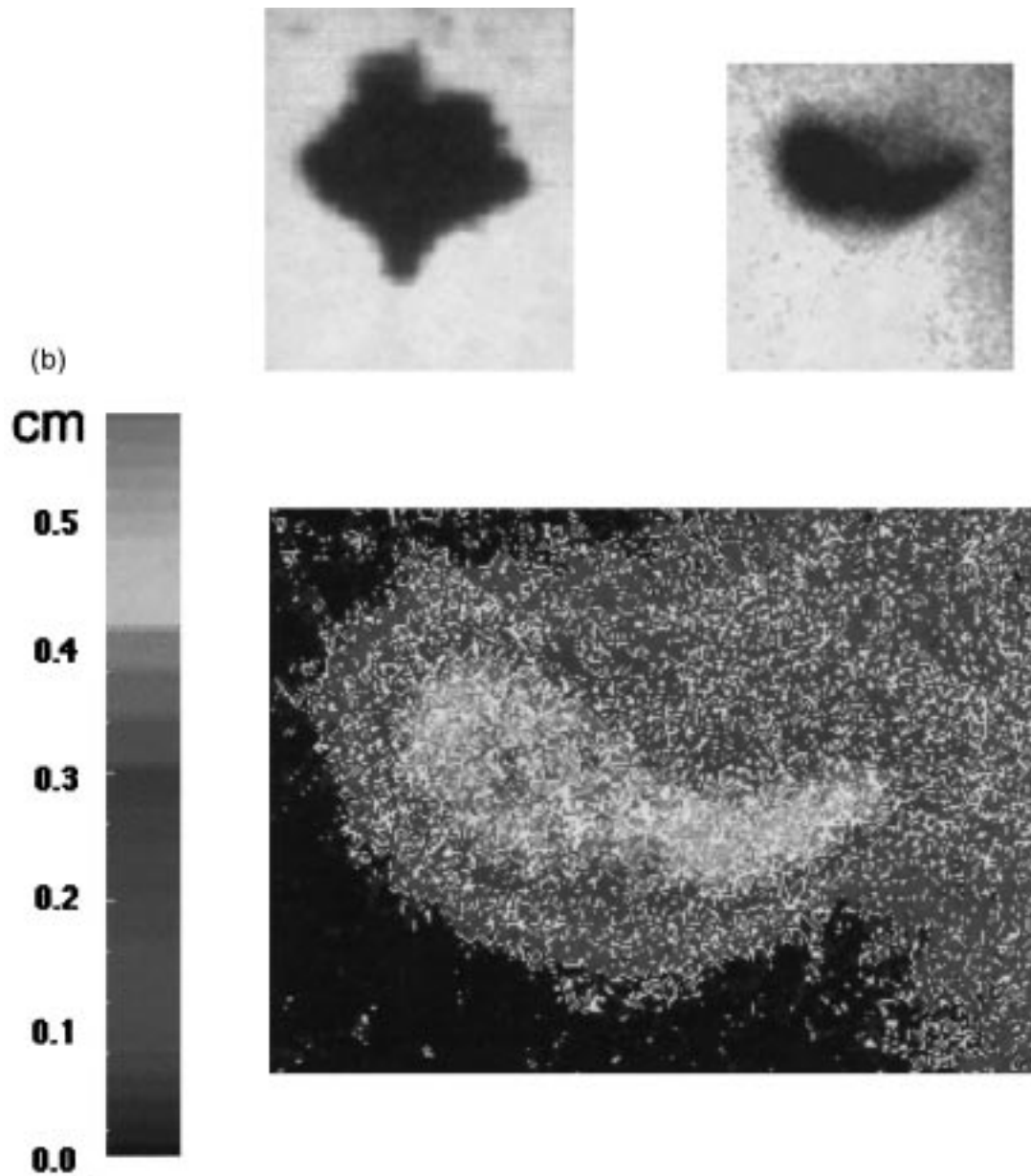


Fig. 6. (continued).

Hugoniots,' with the entrainment factor as the parameter. For a given premixture condition, we can obtain the C–J point, by tangency, as previously, to the appropriate Hugoniot — that is, the results now also depend on the entrainment factor. Sample results for two relatively 'lean' mixtures (volume fraction, $\theta_f = 0.05$ and 0.02 for tin/water and UO_2 /water, respectively) are shown in Figs. 8 and 9, and they are discussed below.

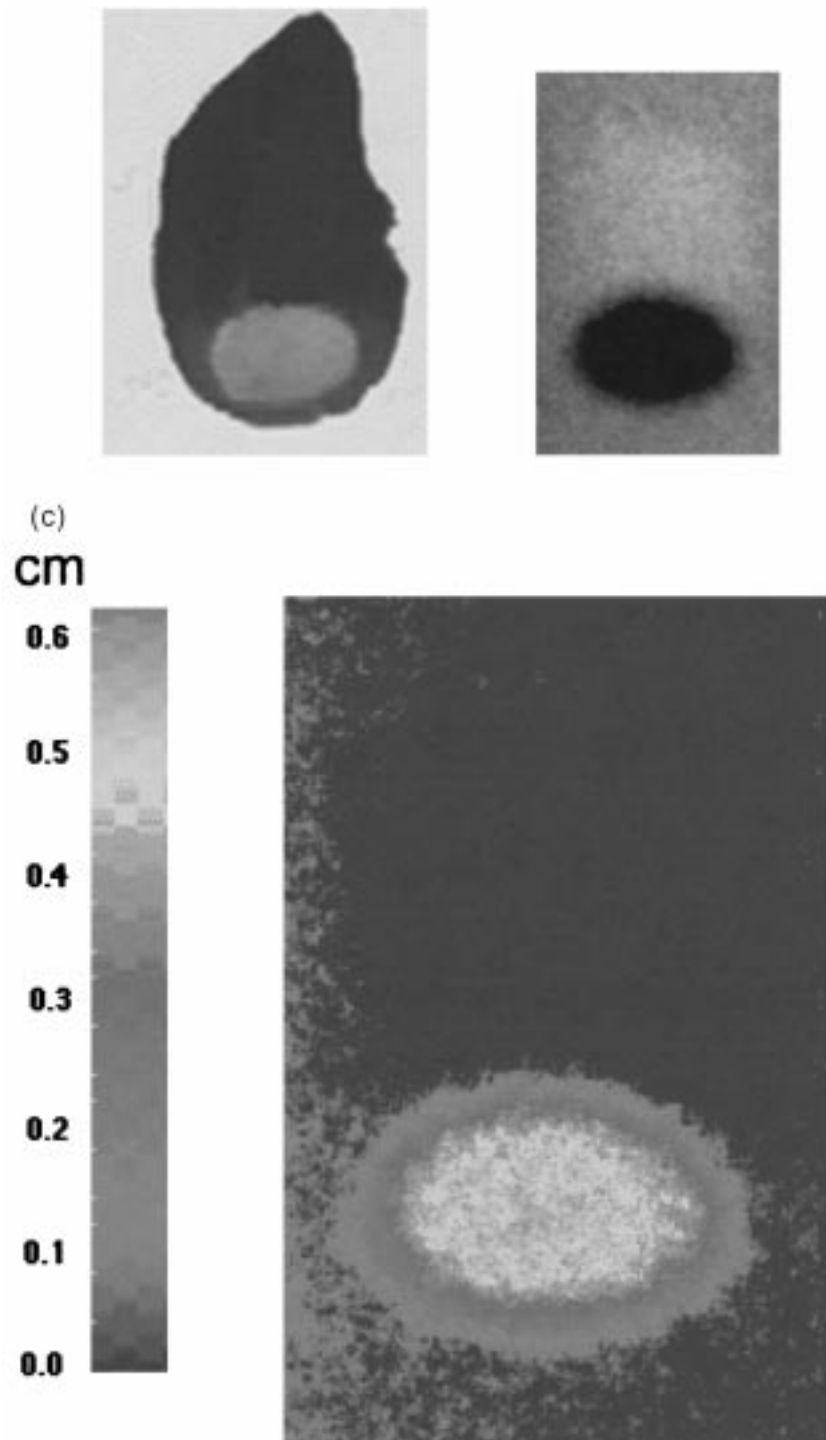


Fig 6 (continued).

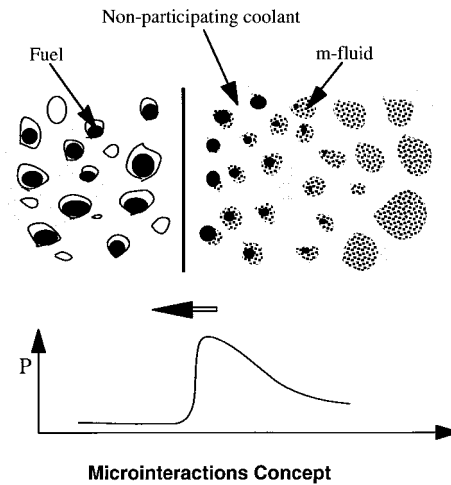


Fig. 7. The microinteractions thermal detonation concept (Theofanous and Yuen, 1993). The ‘non-participating’ coolant is compressed behind the shock, but it does not react thermally with the debris.

4. Discussion

Figs. 8 and 9 demonstrate that thermal detonations can be obtained under much less restrictive premixture conditions than deduced from the Board and Hall model. The constitutive law for the entrainment factor, f_e (volume of entrained coolant per unit volume of fuel) can be obtained experimentally, under conditions (pressure level, fluid velocities) relevant to the particular detonation that pertains (Chen et al., 1995), in an iterative fashion if necessary, so that mutual consistency is clearly established. This then guarantees the existence of steady self-sustaining, detonations, at least in 1D, provided:

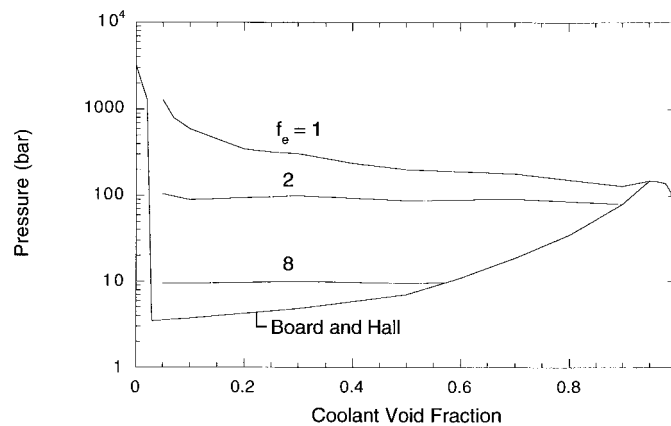


Fig. 8. Solutions of the microinteractions thermal detonation model, for the conditions of Fig. 4 and the case with $\theta_f = 0.05$. Here, $f_e = \hat{f}_e(\rho_f/\rho_l)$.

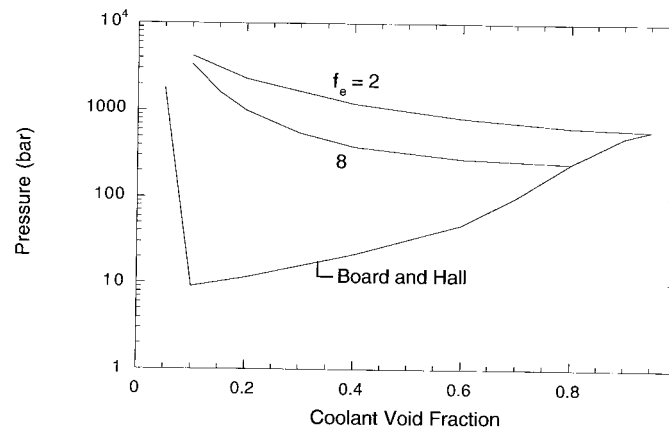


Fig. 9. Solutions of the microinteractions thermal detonation model, for the conditions of Fig. 5 and the case with $\theta_f = 0.02$. Here, $f_e = \hat{f}_e(\rho_f/\rho_l)$.

- a sufficient mechanism for escalation, to these conditions, exists, and **in addition**
- the finite fragmentation rates expected physically do not invalidate in a basic way the thin reaction zone assumption used to obtain the steady solutions.

These provisions can be examined by applying the microinteractions idea by means of a transient, multifield simulation, supplemented by constitutive laws for fragmentation rates and microinteractions.

This was done in the ESPROSE.m code, as noted above, and consistent interpretations of 1D experiments in the KROTOS facility were possible, for both weak propagations obtained with tin, as well as with strong supercritical detonations with aluminum oxide melt (Hohmann et al., 1993). Together with proper treatment of wave dynamics, this opens the way to a priori predictions of 2D and 3D explosions (Theofanous et al., 1998b), as definitely needed in practical geometries.

Spontaneous triggering and escalation can also be addressed in the same frame, but here we would need much more extensive efforts in determining the microinteractions constitutive laws (under a much wider range of pressures, from very low to rather high), and perhaps even extending some aspects of the formulation. Such work is currently pursued by UCSB's program, including the use of an upgraded SIGMA facility.

Finally, it is interesting to examine this steady state theory in the context of the above mentioned full simulation of the wave-dynamics/microinteractions process. We used ESPROSE.m for this purpose. For a tin/water mixture with a fuel fraction of 0.05 and a void fraction of 0.05, calculations were carried out with high enough fragmentation rates to approach a behavior similar to that assumed in the steady state theory. In addition, accounting for the fact that due to the still-finite rates of fragmentation used in the simulation, the major portion of the m-fluid is created from a compressed state, we modified Eqs. (7) and (8) so that a fraction γ of the \hat{f}_e mixes in at conditions corresponding to the pressure behind the shock (the remaining mixing at the conditions ahead). Specifically, we use $\gamma = 0.9$, a fuel volumetric fragmentation rate of 10^5 g/s cm^3 , and an entrainment factor of $f_e = 1$. Illustrative results are summarized in Figs. 10–12.

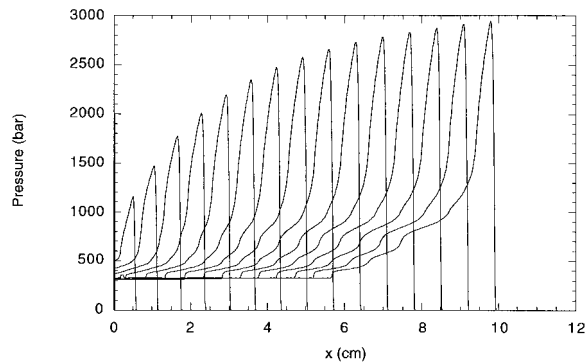


Fig. 10. Illustration of escalation and approach to steady state propagation in an ESPROSE.m simulation of a tin (1500°C)–water explosion, with a premixture void fraction of 5% and fuel fraction of 5%. Print interval is 5 μ s.

In Fig. 10, we see the escalation and approach to a steady-state propagation with a self-similar character. In Fig. 11, we see the wave evolution from the shock adiabat towards the Hugoniot, which is approached tangentially and is followed thereafter over the whole expansion branch.

Certain other key features of the numerical solution (ESPROSE.m) and the exact results (steady-state theory) are illustrated in Fig. 12. In this figure, u_1 , the velocity of the shock in the laboratory frame, has a minimum (~ 1.38 km/s) at a pressure of ~ 1.34 kbar. On the p – v plane (as in Fig. 3), this condition corresponds to the tangent to the Hugoniot drawn from the initial state of the premixture, and it is the C–J point as discussed above. This tangency condition can also be interpreted (Landau and Lifshitz, 1959) as a state at which the speed of the reaction products relative to the shock front (u_2) becomes equal to the local speed of sound. Indeed, the local sound speeds (the tangents) along the Hugoniot are shown in Fig. 12, to intersect the u_2 line (speed of the reaction products relative to the shock) at a pressure of ~ 1.33 kbar, i.e., at the C–J plane.

The wave speeds from ESPROSE.m shown in Fig. 12 indicate the speed of propagation of each pressure amplitude of the detonation wave (see Fig. 10). Note that the wave is subsonic in the front (high pressure) position of the wave, and that it becomes supersonic somewhere ahead of the C–J plane. Also note that the propagation velocity of the wave front, from ESPROSE.m, agrees quantitatively with the steady state result (u_1). Finally, the choking condition at the C–J plane discussed above for the steady-state solution is found in the ESPROSE.m results as well (not shown in the figure, to maintain clarity). In fact, the exact u_2 result shown in the figure is perfectly matched.

5. Conclusions

- The Board and Hall model leaves significant questions about the existence of supercritical thermal detonations in physically meaningful premixtures.
- In attempting to address criticisms by including finite rate phenomena, such as melt

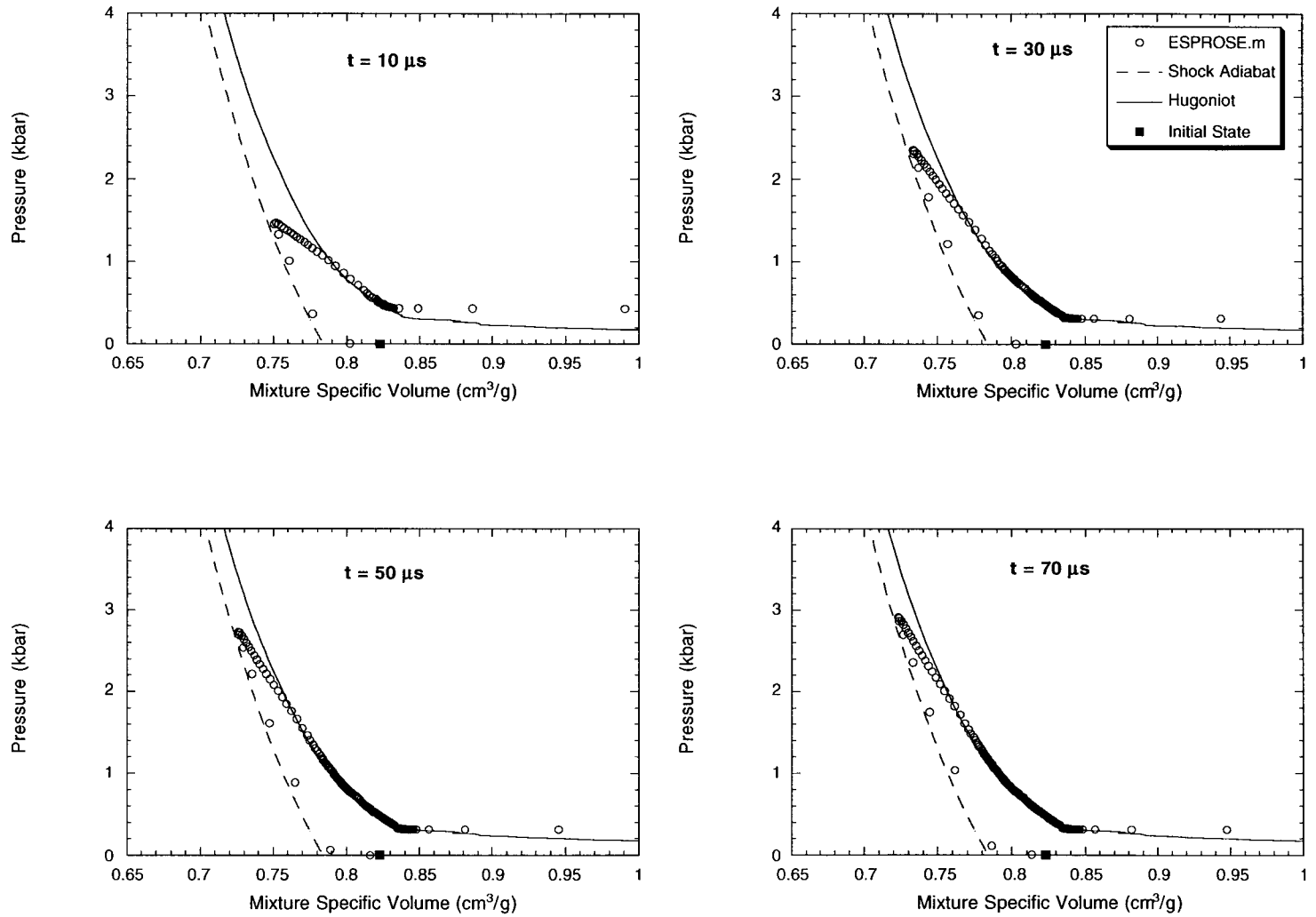


Fig. 11. The ESPROSE.m result at $20 \mu\text{s}$ intervals in the $p-v$ plane, in relation to the shock adiabat and the Hugoniot.

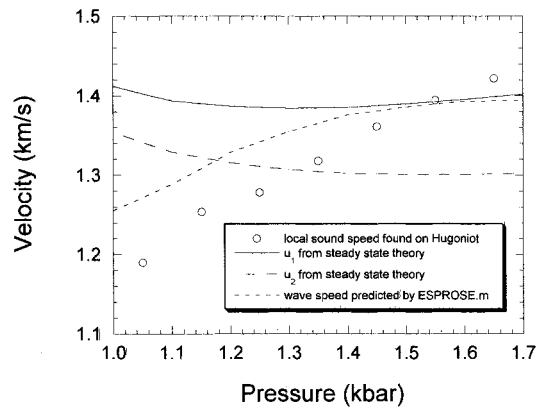


Fig. 12. Demonstration of choking at the C–J point as predicted by the steady state theory and the wave speed of the ESPROSE.m solution.

fragmentation, all transient multifield formulations have misused the Board and Hall concept, and produced the misleading result that supercritical thermal detonations are not possible in physically meaningful premixtures.

- The microinteractions idea allows a physically consistent bridge between Board and Hall and finite rate behaviors. The microinteractions model applied in the steady detonation frame (i.e., assuming infinite fragmentation rates) provides a useful tool to properly discern conditions of the existence of such detonations. It is also useful for checking the numerics of the microinteractions model applied in the transient (finite rates) multifield frame; that is, in rendering detonations in terms of physically meaningful terms. Key aspects in this are the inclusion of constitutive laws for microinteractions from experiments simulating large scale explosions, and of multidimensional wave dynamics in the formulation. A rational approach to addressing the energetics of large scale explosions is thus possible, without the pitfalls of extrapolating small experiments, or the need for large ones.

Acknowledgements

This work was supported by DOE's ARSAP program at UCSB, with Mr. Steven Sorrell (DOE Idaho Operation's Office) as the program manager.

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