OCT 1 2 1976

MEMORANDUM FOR: Benard C. Rusche, Mirector Office of Nuclear Reactor Regulation

FROM:

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Saul Lovine, Acting Director Office of Nuclear Regulatory Research

SUBJECT: SRI 6: DRAFT REPORT: "A CRITIQUE OF THE BOARD-HALL MODEL FOR THERMAL DETONATIONS IN THE BO,-HA SYSTEM"

Reference: Memorandum of August 19, 1976 from C. N. Kalber to L. Rubenstein entitled "Purdue Research Foundation Proposal for Continuation of the Work Initiated Under Contract No. AT(49-24)-018."

Enclosed for your information is a draft copy of the subject report prepared as part of the effort of the Office of Nuclear Regulatory Research to determine research meeds in this area. The author is Dr. David C. Williams of Sandia Laboratories, and the paper will be presented at the ANS-ENS "International Meeting of Fast Reactor Safety and Related Physics" at Chicago in October. This evaluation is relevant to the DPM-supported experiments at Purdue University on the validity of the Board-Hall model, and is the RES generated evaluation of this model referred to in the reference memorandum.

The Williams report concludes that the Board-Hall model for propagation of a detonation-like thermal explosion in a postulated pre-mixed UO₂ sodium system is not valid for LMFBR accident conditions because of the very high magnitude of the shock pressure required for triggering, because of deficiencies in the fragmentation model, and because of neglect of the dispersive characteristics of the pre-mixed UO₂-sodium system. The report does say, however, that the possibility of the occurrence and propagation of a detonation-like thermal explosion in a pre-mixed UO₂-sodium system by some mechanism other than the Board-Hall fragmentation model cannot at present be ruled out completely.

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Benard C. Rusche

We hope that this report will prove useful to you and your staff in svaluating the Board-Hall model in particular, and also the value of the experimental research proposed by Purdue to BPM. Further discussions on the technical aspects of this problem should be conducted with R. W. Wright of my staff.

> Original Signed by Saul Levine

Saul Levine, Acting Director Office of Nuclear Regulatory Research

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Enclosure: As stated

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A CRITIQUE OF THE BOARD-HALL MODEL FOR DETONATING THERMAL EXPLOSIONS AS APPLIED

TO UO2- Na SYSTEMS

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ABSTRACT

The Board-Hall model for detonating thermal explosions is reviewed and some criticisms are offered in terms of its application to UO₂-Na systems. The basic concept of a detonation-like thermal explosion is probably valid provided certain fundamental conditions can be met; however, Board and Ball's arguments as to just how these conditions can be met in UO₂-Na mixtures appear to contain serious flaws. Even as given, the model itself predicts that a very large triggering event is needed to initiate the process. More importantly, the model for shock-induced fragmentation greatly overestimates the tendency for such fragmentation to occur. The shockdispersive effects of mixtures are ignored. Altogether, the model's deficiencies imply that, as given, it is not applicable to LMFBR accident analysis; nonetheless, one can not completely rule out the possibility of meeting the fundamental conditions for detonation by other mechanisms.

INTRODUCTION

In 1974, the British workers S.J. Board and R.W. Hall[1] proposed a model for the propagation of vapor explosions which was based upon a close analogy with chemical detonations. Since the model predicts that, under certain conditions, very powerful explosions can occur in molten UO.-Na mixtures, it is of obvious interest to the LMFBR safety community. In this paper, we review the model and its theoretical foundation, and then offer some criticisms which suggest that Board and Hall may have overestimated the possibility that such 'explosions could occur in LMFBR accident situations. We conclude with some additional discussion, including some cautionary remarks against over-interpreting the results offered here.

SUMMARY OF THE MODEL

Since the Board-Hall model is based upon shock and detonation physics, we briefly review this topic, omitting many refinements and qualifications that are required when dealing with solid materials [2].

consider a severe stress wave propagating through a medium for which the sound speed C tends to increase as the material is compressed; most materials satisfy this condition. The high-pressure portion of the wave then tends to overtake the leading edge, so that the compression phase of the wave steepens into a neardiscontinuous jump in the pressure called a shock. By applying the equations of mass, momentum, and energy conservation, it may be shown that the material properties before and after shock passage must be related by the well-known Rankine-Bugoniot jump relations:

$$E_{1} = E_{1} + \frac{1}{2}(P_{2} + P_{1})(\nabla_{1} - \nabla_{2}) \qquad (1c)$$

Here U, u, E, V and P are the shock propagation velocity, the shock-induced material velocity, specific energy, specific volume, and pressure, respectively; subscripts 1 and 2 refer to conditions before and after shock passage, respectively. In addition, the material must obey its own equations of state, P = P(E,V)For a given set of initial conditions, we have four equations in five unknowns (U, u, P₂, V₂, E₂). Specifying any one of the latter in effect specifies the other four. In particular, for any two of the quantities (Q₁ and Q₁, say), a plot of the states which may be obtained (from given initial conditions) by shocks of various strengths lies along a curve in the Q₁ - Q₂ plane called the Eugoniot. For many fully-dense (non-porous) materials, it has been found experimentally that the U-u Hugoniot approximately follows an especially simple form,

$$\mathbf{U} = \mathbf{C}_{\mathbf{A}} + \mathbf{s}\mathbf{u}, \qquad (2\mathbf{a})$$

where s is an empirical, non-dimensional constant which usually lies between 1 and 2; we have also assumed that the initial state is the uncompressed reference state. By combining Equation (2a) with Equations (1a) and (1b), it may be shown that the P - V Hugoniot will then be of the form

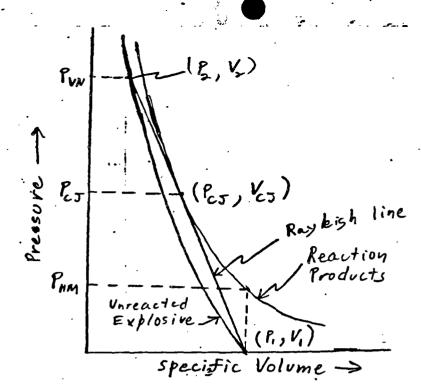
$$P = c_0^2 \varepsilon / [V_0 (1 - \varepsilon)^2] = K_0 \varepsilon / (1 - \varepsilon)^2, \qquad (2b)$$

where the subscript o refers to the uncompressed reference state, ε is in the volumetric strain (1 - V/V) and K is the bulk modulus. It is also worth noting that the difference between the Hugoniots and isentropes varies as ε^3 , and is therefore slight for small ε , in some of what follows, this difference is ignored.

The detonation of chemical high explosives is more complicated. It involves the following sequence of events:

- (1) The detonation subjects the unreacted explosive to a severe shock.
- (2) As a result, the explosive undergoes energetic decomposition on a time scale,t, so short that the reaction zone thickness Ut is < 1 mm, often << 1 mm.</p>
- (3) The hot reaction gases expand with a substantial conversion of heat energy to work, which supplies further energy to continue driving the detonation wave.

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The system is diagramed in terms of the P-V Hugoniots in Fig. 1. The lower curve represents the Hugoniot for the unreacted explosive initially in the state (P_1,V_1) . The upper curve represents the Hugoniot for the high-temperature, gaseous reaction products. It can be shown that the detonation wave will propagate steadily, without attenuation or growth in amplitude, if and only if the line connecting the initial state with the state existing at the completion of the reaction is tangent to the reaction product Hugoniot. The point of tangency, (P_{CJ}, V_{CJ}) , gives the conditions at the end of the reaction and is called the Chapman - Jouget point or C.J. point, and is a characteristic constant for a given explosive at a given (P_1, V_1) .

From Eq. (3.1a), the slope of the line connecting (P_1, V_1) and the C.J. point, called the Rayleigh line, is equal to U^2/V^2 . Since the shock initially propagates through unreacted explosive, the initial pressure must be given by the intersection of this line with the corresponding Hugoniot at P_2V_2 . This still higher pressure, P_{VN} , is called the Von Neuman spike; it is so narrow, however, that it attenuates very rapidly in a non-explosive material and the CJ pressure is normally the quantity of interest.

The basis of the Board-Hall model was its authors' observation that thermal explosions can proceed with an essentially identical structure if the following three fundamental assumptions are valid:

A. The liquid - liquid system is initially in the form of a mixture that is too coarse to permit significant heat transfer on a time scale comparable to the time required for the detonation wave to traverse the system.

B. Astrong triggering shock is supplied.

Figure 1

Schematic P-V Diagram For

Detonating Explosive

C. A shock having the CJ amplitude will fragment the coarse initial mixid re into a much finer mixture permitting extremely rapid heat transfer, with the total time required for fragmentation and thermal equilibration being much less than that required for the detonation wave to traverse the system.

Given these three assumptions, Board and Hall show that the explosion can propagate with a structure identical to that of the chemical explosion, with the zone of rapid fragmentation and heat transfer corresponding to the reaction zone for the chemical case. They also show that such a detonation can actually generate

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pressures considerably higher, and work potentials comewhat higher, than those calculated by Hicks and Menzies. The reason is that, here, the mixture is first compressed by the shock and heat is then transferred to the volatile componentat above normal densities; Hicks and Menzies assumed heat would be transferred at a constant volume corresponding to the normal density thereby leading to P_{HM} in Fig. 1.

The author knows of no flaw in the basic argument, but it is much more debatable as to whether the model's three fundamental assumptions are valid in practice. The third assumption is the crucial one in terms of the model's internal structure; the first two are actually assumptions about the presumed initial conditions.

Board and Hall argue that this third assumption will be met by noting that, because of the large UO_2 -Na density difference, the shock will accelerate the two phases to guite different velocities, and the resulting velocity differential V will tend to induce UO_2 fragmentation due to Taylor instabilities. Drawing an analogy with the data Simpkins and Bales [37 obtained for shock-induced breakup of liquid droplets in gases, they concluded that the time t required for fragmentation of a UO_2 drop of radius r_1 is given by

$$(\rho/\rho')^{\frac{1}{2}} vt/r_1 = 44B_{\rho}^{-\frac{1}{2}}$$
, (3a)

where ρ and ρ' are the Na and UO₂ densities and the Bond number, B₂, is defined by

$$B_{0} = \rho' gr_{1}^{2} / \sigma = 3\rho v^{2} r_{1} C_{D} / (8\sigma)$$
(3b)

(4)

where g is the acceleration imparted by drag forces, σ the surface tension, and C the drag coefficient. It is then assumed that the drop fragments down to a final size r₂ governed by a Weber-type criterion, $\rho v^2 r_2 g/\sigma^R$ 8; r₂ turns out to be so small (microns or less) that heat transfer is essentially instantaneous compared with the time required for fragmentation.

On the other hand, Board and Hall indicate that the relative velocities of the two fluids should tend to equilibrate during a characteristic time t' given by

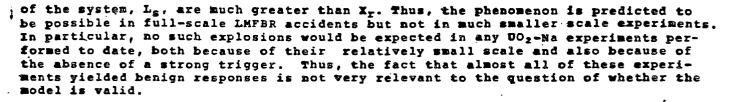
 $t' \approx 8p' r_1 / (3pvC_p)$.

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This estimate was evidently obtained simply by taking t' = v/g, where the acceleration, g, is given by the drag force, $F_D = \frac{1}{2}\rho v^2 C_D \lambda$ divided by the droplet mass, $4\pi\rho' r_1/3$, and λ is the cross sectional area πr_1^2 . Based upon the results for liquid droplets, the drag coefficient, C_D , was taken to be about 2. Board and Hall then argue that the third fundamental assumption will be satisfied if t is greater than the fragmentation time, t.

Board and Hall consider the case of UO₂ at 3550 K, Na at 700 K, a UO₂/Na ratio of 10 by weight, and a 50% void (vapor) fraction by volume. They find that the fragmentation criterion is indeed met, and calculate $U = 1.9 \times 10^5$ cm/sec, P = 15 kbar, and $u = 2.3 \times 10^4$ cm/sec at the C.J. point; if vapor is absent, they state that P is approximately doubled.

The authors also note that the reaction zone thickness, $X_r = Ut$, is very much : greater than for chemical explosives, of the order of 10 cm or more if $r_1 = 0.5$ cm, as they assumed. The model is one-dimensional and cannot apply unless the dimensions



Indeed, these facts point to a singularly vexatious aspect of the Board-Hall model from the point of view of LMFBR safety analysis: the model predicts that extremely powerful explosions are possible in full-scale LMFBR accidents yet the model also predicts that it will be virtually impossible to give the model any rigorous experimental test in an actual UO_2 -Na experiment of reasonable size. Before attempting an experimental test, it is therefore worthwhile to review some features of the model a little more carefully.

SOME CRITICISMS OF THE MODEL

<u>Initiating Event</u>. It is instructive to estimate the magnitude of the triggering event required to initiate the Board-Hall process. Initiation requires a shock above some minimum value P_m of duration at least t. The magnitude of P_m is set by the need to meet the fragmentation criterion. By inserting numerical values for UO₂ and Na material properties [4] into Equations (3) and (4), we obtain

$$t \approx 710 r_1^{3/4} / v^{3/2}, t' \approx 13.1 r_1 / v$$

(5)

For shocks below some limiting amplitude, v will be low enough so that the fragmentation criterion, t < t', will not be met; if $r_1 = 0.5$ cm, for example, Equation (5) implies v > 4000 cm/sec is required.

To estimate P, we assume v $\frac{R_{i}}{2}$ u and assume that, for Na and UO₂, the Hugoniot can be expressed by Eq (2) with s = 1.27 and 1.5, respectively (results that follow are insensitive to s). For the composite, we assume the P - ε curve can be constructed by evaluating ε individually for the two constituents at a given P and taking a volume-weighted average. For the important case where void space (i.e., vapor or gas) is present, we let V₀ represent the mean specific volume of the mixture without void space and represent the specific volume icluding voids as V₁ = α V₀; thus α = 1 and α = 2 correspond to no voids and 50% void fraction respectively. We assume any reasonable final pressure completely collapses the voids and that the final volume V₂ is independent of α . There are several approximations involved here, but refining them would not affect the basic conclusions to be given.

A computer code based upon these assumptions was written to estimate the value of P_{p_1} sufficient to give a velocity u, as calculated from Equation (1b), to meet the fragmentation condition. Equations (1a) and (1c) were then used to estimate the corresponding values of U and E. Since the initiating pressure must be applied for at least a time t, the initiation zone must be of a thickness $X_r = Ut$. If the initiation region must be an order of magnitude greater in lateral extent than the thickness in order to preserve one-dimensionality, the volume of the initiation region is of the order 100 X_r^3 and the total energy E_{tot} imparted to it is ~100 $X_r^3 E/(av_0)$. The latter may be an over-estimate if the lateral extent of the initiation region need not be as great as assumed; on the other hand; we have only attempted to estimate the energy imparted to the initiation region by virtue of its being subjected to the triggering.shock. The total energy available to the triggering event itself must be considerably larger.

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Results are summarized in Table I, where P_{rr} , X_r , and E_{tot} are given for various values of α and r_1 . For $r_1 = 0.5$ and $\alpha = 1$, the "trigger" needed is extremely massive, hardly less destructive than the fully-developed reactorwide explosion. The more realistic cases with $\alpha > 1$ have lesser triggering requirements but they are still very large. If $r_1 = 0.05$, total trigger energy needed is considerably less, but still substantial and P_{rr} is actually increased considerably.

TABLE I

Nagnitude of the Initiating Event Required for Various Parameter Values

G		$r_1 = 0.5 \mathrm{cm}$			$r_1 = 0.05 \text{ cm}$		
	X (cm)	P _m (bars)	E _{tot} (J)	X _r (cm)	P (bars)	Etot ^(J)	
1.0	180	3×10^3	3'x 10"	7	104	1.5×10^6	
1.11	60	700	8 x 10 ⁷	4		3×10^5	
1.5	18	200	2 x 10 ⁶	1.7	2×10^{3}	1.9 x 10 ⁴	
2.0	13	80	4×10^{5}	1.3	800	4×10^{3}	

Thus, even when taken at face value, the model itself predicts that the initial conditions required involve a combination of a rather idealized mixture and a strong initiating event that seems unlikely to be realized in practice.

UO2 /Na Mass Ratio*

Even more serious to the model, Equations (3) and (4) would at best be valid for an isolated drop of UO_2 in an infinite sea of sodium, yet they were applied to a situation with a UO_2/Na mass ratio of about 10. The interfluid drag force applies equally to each component, and the acceleration of the sodium is therefore about ten times that of the UO_2 . Both velocity changes are, of course, in the direction to decrease ∇ , so that the rate of velocity equilibration was seriously underestimated, and t'is correspondingly overestimated by Equation (4).

We estimate the importance of this effect by assuming the interfluid drag force per unit volume, $F_{\rm D}$, for a mixture of two fluids, a and b, to be

$$\mathbf{r}_{\mathrm{D}} = \bar{\rho} \mathbf{v}^2 \mathbf{C}_{\mathrm{D}} \mathbf{A} / 2 , \qquad (6)$$

where A is the perpendicular fluid-fluid interfacial area per unit volume, and $\bar{\rho}$ is the average density $f_a \rho_a + f_b \rho_b$, where the f's and ρ 's are the volume fractions and densities of the two fluids, respectively; for the moment, we let Equation (6) be the effective definition of C_D. The relative velocity decays at a rate given by

$$\frac{dv}{dt} = -\left(\frac{1}{f_{a}\rho} + \frac{1}{f_{b}\rho_{b}}\right)F_{D} = -\frac{1}{2}\frac{p^{2}}{f_{a}\rho_{a}f_{b}\rho_{b}} \nabla^{2}C_{D}A$$
(7)

If we let f_m be the volume fraction of the less abundant fluid (i.e., $f_m = Min (f_a, f_b)$) and assume this is in the form of spherical drops of radius r_1

*S. George Bankoff, of Northwestern University, has independently made points similar to those to be discussed here.

dispersed in the more-abundant fluid, $A = 3f_4r_1$, and

$$\frac{dv}{dt} = -\frac{3}{8} \frac{\overline{\rho}^2}{\rho_a f_a \rho_b f_b} v^2 \mathcal{L}_D f_a / r_d$$
(8)

The revised value of the characteristic equilibration time, t⁴, becomes

$$t' = v/(dv/dt) = 8 \rho_{a} f_{a} \rho_{b} f_{b} r_{1} / (3p^{2} v C_{D} f_{m})$$
 (9)

If one fluid overwhelmingly predominates, Equation (9) is easily shown to reduce to the same as Equation (4); hence the value of C_D defined by Equation (6) reduces to the value appropriate for an isolated spherical drop of either fluid moving in a sea of the other fluid, as it should; of course, C may also depend upon the mixing ratio for intermediate cases as well as upon the other usual parameters.

Inserting numerical values for the case of interest, with $f_a = f_b = 0.5$ gives

$$t \approx v/(dv/dt) = 0.45 r_{0}/(vC_{D})$$
, (10)

which implies t'' << t' as given by Equation (5) unless C << 1. As a cross check, we may estimate t'' by approximating the flow of sodium relative to UO₂ as a flow through a packed bed. Starting with a correlation due to Ergun [5] for the resistance to such a flow, and omitting terms that are small in the present case, we obtain

$$t \approx v/(dv/dt) = 0.57 r / v$$
 (11)

which is very close to (10) with $C_D \approx 1$. Though we are applying Ergun's relation to values of v considerably higher than those for which it was established, this result suggests our estimate for t'' is of the right order of magnitude.

Evaluating t from Equation (10) with $C_D = 1$ and still evaluating t from Equation (3) shows that, with a shock amplitude of 30 kbar, t⁴/t ranges from about 0.05 to 0.16 for all cases considered in Table I. That is, even for an initiating shock with an amplitude equal to that of the fully-developed Board-Hall detonation, the fragmentation criterion fails to met by about one order of magnitude. Furthermore, if a still more powerful shock is applied, detonation theory itself tells us it will die down to the CJ amplitude even if fragmentation does occur. Hence, there appears to be no way that a detonation-

like explosion can propagate in just the way proposed by Board and Hall unless t is also an order of magnitude or more shorter than those workers proposed. This is possible, but it is worth noting that either Equation (10) or (11) implies that the relative velocity will decay to less than 10% of its initial value before the total relative motion reaches the order of the mixture scale, ~2r1. Since fragmentation implies liquid-liquid interpenetration, which presumably requires liquid relative motion at least of the order of the mixture scale, it is legitimate to raise the question as to whether the shock-induced velocity differential can cause complete fragmentation by any mechanism.

As the Na/UO₂ ratio increases toward infinity, t', as given by Equation (9), increases toward t' as given by Equation (3), but the amplitude of the detonation wave decreases. It seems very questionable as to whether one could find any mixture ratio such that, for a shock of the CJ amplitude, the necessary condition t'' > t would be satisfied; however, this question has not been investigated in any detail,

Throughout this discussion, we have assumed that the sevent shock amplitudes for evaluating fragmentatic should be P_{CJ} , not the higher live P_{VN} . Actually, P_{VN} is not mearly high enough to reverse our conclusions. Note fundamentally, the Von NeuMann spike would be narrow compared with the fragmentation zone and the pressure wave causing fragmentation will have to be of at least the width of the latter.

Shock-Dispersive characteristics

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Board and Hall did not take into account the shock-dispersive characteristics of composites. Speaking roughly, a sharply-defined pressure wave undergoes multiple, partial reflections at the interfaces between the two constituents. The wave profile therefore spreads and becomes rounded; it also attenuates unless backed by a sustained driving pressure. These effects become very important when there is a large acoustic impedance mismatch between the two constituents, as is the case for UO_2 -Na mixtures.

L. M. Barker [6] has analyzed composite response to stress waves. He showed that composites, to a good approximation, could be modeled as a stress-relaxing material. Details cannot be given here, but the key point is that such materials cannot support a steadily-propagating, sharply defined shock at all unless the amplitude exceeds a certain minimum value; lesser-amplitude steady waves must have a rounded profile. Above the minimum value, part (but not all) of the pressure rise may appear as a near-discontinuous jump or shock.

The author applied Barker's model to UO_2 -Na mixtures, using simple stressstrain relationships based upon Equation (2), and it was found that the minimum value of the pressure permitting partial shock formation probably lies between 25 and 50 kbars. This is at least as high as the CJ pressure suggested by Board and Hall, and it is therefore questionable as to whether even the fully-developed detonation could propagate as a sharply defined shock. Failure to achieve a sharp shock would reduce still further the driving force for fragmentation, which already appeared to be inadequate. It would also require careful re-examination of the entire analogy with chemical detonations.

The analysis just summarized would apply directly only when there is little or no void space. With a substantial void fraction, the situation is more complicated and a relatively sharp pressure front cannot be ruled out, though it is not clear how it can be much more sharply defined than the mixture scale. In any case, shock-dispersive effects are still expected and they must be considered. If nothing else, they probably rule out formation of a clearly-defined Von Neumann spike, supporting still further the use of the CJ pressure in the fragmentation analysis as was done here.

SUMMARY AND CONCLUSIONS

If the criticisms offered here are valid, the Board-Hall model, in its present form, cannot be treated as a significant factor in LMFBR safety analysis. When the Board-Hall approach is refined along the lines indicated, the third of the three fundamental conditions for detonation-like behavior (shock-induced fragmentation) fails to be satisfied by rather wide margins, and the effects of shockdispersiveness and the need for very large triggering events cast further doubt upon the model's practical utility in safety analysis even if the idealized mixture considered could be achieved in practice, which is itself questionable.

On the other hand, this rather negative conclusion should not be over-interpreted. The present work was basically limited to refining certain aspects of the original study and showing that, with these refinements, some of the conditions required for internal self-consistency may no longer be satisfied. It is conceivable that quite different mechanisms could cause the rapid fragmentation required to generate detonation-like behavior.

Since fragmentation is a purely mechanical effect in the Board-Hall approach, there is no need to study it with hot-cold liquid pairs. Thus, it could prove

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useful to perform experiments subjecting mixtures of highly dissimilar liquids (e.g., mercury and water) to strong shocks, either with or without vapor present. Care must be taken to ensure that such experiments are consistent with the model's requirements. For example, the input pressure pulse must be relatively long in duration, not only because the Board-Hall mechanism requires such pulses, but also because short pulses could induce fragmentation by mechanisms that would not be present in the long-duration pulses of interest here. If fragmentation is observed, it would then be necessary to establish that it was a prompt effect rather than a delayed effect.

Finally, even if the Board-Hall approach could be shown to be totally invalid, this would not necessarily mean that the possiblility of large-scale, coherent interactions between molten UO_2 and Na can be laid to rest. There is considerable experimental evidence [7] that both triggering and scaling effects are indeed important in vapor explosions, whatever the underlying reason. Unless major advances in the theoretical understanding of vapor explosions are made in the near future, it may eventually be desirable to conduct largescale UO_2 -Na experiments with strong triggering pulses provided.

ACKNOWLEDGEMENT

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