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Discrete effects in energetic materials

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Abstract. The classical theory of detonation wave propagation has been highly successful in prediction of detonation dynamics based on a macroscopic, continuum-based approach, wherein the heterogeneity of the energetic material only enters the model via the reaction rate term (e.g., hot-spot based reaction mechanisms). The effects of spatial heterogeneity are rarely treated explicitly in detonation models. However, considerable evidence can be found that mesoscale phenomenon can influence the dynamics of the detonation front on scales larger than the heterogeneities. Two scenarios are identified that may result in spatial non-uniformity of the detonation front influencing the macroscopic dynamics of the wave propagation: (1) large-scale heterogeneities and (2) transverse or longitudinal instability of the front. This evidence is critically reviewed, and possible directions for modeling approaches that incorporate the spatial granularity (discreteness) of the energetic material are suggested.

1. Introduction

The behavior of detonation waves propagating in condensed-phase explosives is entirely dominated by processes occurring at the mesoscale. This fact is most easily appreciated by examining the relative shock insensitivity of monocrystalline samples as compared to polycrystalline samples of the same explosive. Upon loading by the leading shock front of the detonation wave, the friction of contact between grains, work done in viscoplastic collapse of pores, impact of microjets into voids, and shock and shear interactions occurring at impedance interfaces have all been suggested as mechanisms for localizing the energy deposited by the shock into the explosive, creating so-called "hot spots" from which reaction fronts can propagate outward [1–3]. Some of these mechanisms are illustrated schematically in figure 1(a) and are only a subset of all the possible reaction initiation mechanisms that have been suggested.

For engineering applications of explosives, it is usually the macroscopic behavior of detonation waves that is of interest, such as how the detonation wave propagates through an explosive charge of a particular geometry. Some of the more commonly encountered problems are detonation velocity in a cylindrical rate stick or slab and the existence of a critical diameter or thickness necessary for propagation, propagation into a wedge of decreasing thickness, or the corner turning problem, wherein the detonation must emerge from a rectangular prism of explosive into a larger charge of the same explosive, as shown in figure 1(d). The usual practice in modeling these problems is to use a continuum-level model, where the mesoscale processes described above (figure 1(a)) are treated via a source term in the continuum conservation equations that describes the consumption of unreacted explosive, the production of products, and the exothermic heat release of reaction. The reaction initiation and rate are typically described by phenomenological models that account for mesoscale processes using semi-empirically derived

and calibrated formulas; the Forest Fire [4] and Ignition and Growth [1] are two examples of the more popular models, which include terms to describe the formation of hot spots via the processes described in figure 1(a) and the propagation of reaction fronts outward from these localized centers. The resulting continuum equations can then be solved numerically to predict the propagation of the detonation wave. Such an approach does not explicitly consider the spatial inhomogeneity of the polycrystalline media and assumes that the energy release is spatially uniform. For many explosives, this approach can be justified, since the scale of the explosive grains (typically, 1–100 μm) is at least two orders of magnitude smaller than the minimum dimension of the explosive charge (diameters or thicknesses on the order of cm). These disparate scales allow for a clearly defined hierarchy of models to separate mesoscale processes from continuum-level behavior. In fact, the finer the scale of heterogeneity, the more valid this approach is anticipated to be. Fine-scale heterogeneities generate a sufficient density of hot spots to result in an effectively homogeneous energy release that washes out the inherent instabilities that occur in homogeneous media governed by activated reactions (i.e., Arrhenius kinetics). For this reason, explosives with fine-scale heterogeneities are expected to be "ideal" explosives.

If certain additional assumptions are satisfied, such as the detonation front having a large radius of curvature in comparison to its reaction zone thickness (i.e., the front is weakly curved), then it is possible that the propagation of the detonation front through different geometries can be solved for via greatly simplified calculations. This approach is the basis of Detonation Shock Dynamics [5]. The equations describing the reaction zone of a weakly curved detonation front, for which the DSD approximation is valid, makes specific predictions of the scaling of the propagation velocity between the different geometries. In particular, if the detonation wave is confined by a high-impedance inert material such that the flow in the reaction zone is only slightly divergent, then the velocity of a detonation propagating in a two-dimensional slab with wide aspect ratio and thickness t should correspond to the velocity of a detonation in a cylindrical charge with a diameter equal to *twice* the thickness of the slab, i.e., $d = 2t$ [6, 7]. If the explosive is only weakly confined, such that there is a larger divergent flow in the reaction zone, then the scaling between geometries is expected to be less than two, i.e., $d/t < 2$ [6]. Examination of this scaling can also be extended to the scaling of the critical diameter d_c or critical thickness t_c at which the detonation is no longer able to propagate. Near the critical diameter or thickness, the assumption of a detonation front that has a large radius of curvature in comparison to the reaction zone thickness is unlikely to be valid, since the front is highly curved and the reaction zone has significantly thickened near failure. Thus, the diameter/thickness scaling will likely deviate from 2:1 near failure. While only a few measurements of critical diameter and critical thickness have appeared, the measurements previously reported for the critical diameter to critical thickness scaling were approximately two [8, 9]. As discussed below, more recent measurements in nonideal explosives have found large deviations from this value. This scaling parameter may be a useful metric in describing the dynamics of detonation propagation. While there have been many attempts to classify explosives (see, for example, a comprehensive classification [10]), these usually consist in comparative studies between different explosives or a given explosive at different densities. The geometric scaling parameter has the advantage that it is defined by examining the *same* explosive (at the *same* density) in two different geometries and can be compared to a theoretically predicted value.

This paper will review both historical and recent evidence for the influence of the spatial discreteness on the global (or macroscopic) propagation of detonation. Specifically, scenarios under which a continuum-level description of the propagation dynamics may not be valid due to spatial inhomogeneity of the media or multidimensional instability of the wave. Examination of the diameter to thickness scaling ratio (both for propagation velocity and critical dimension) will be used as a "litmus test" of whether the detonation propagation is amenable to a continuum-level model and can be accurately described by the global curvature of the detonation front.

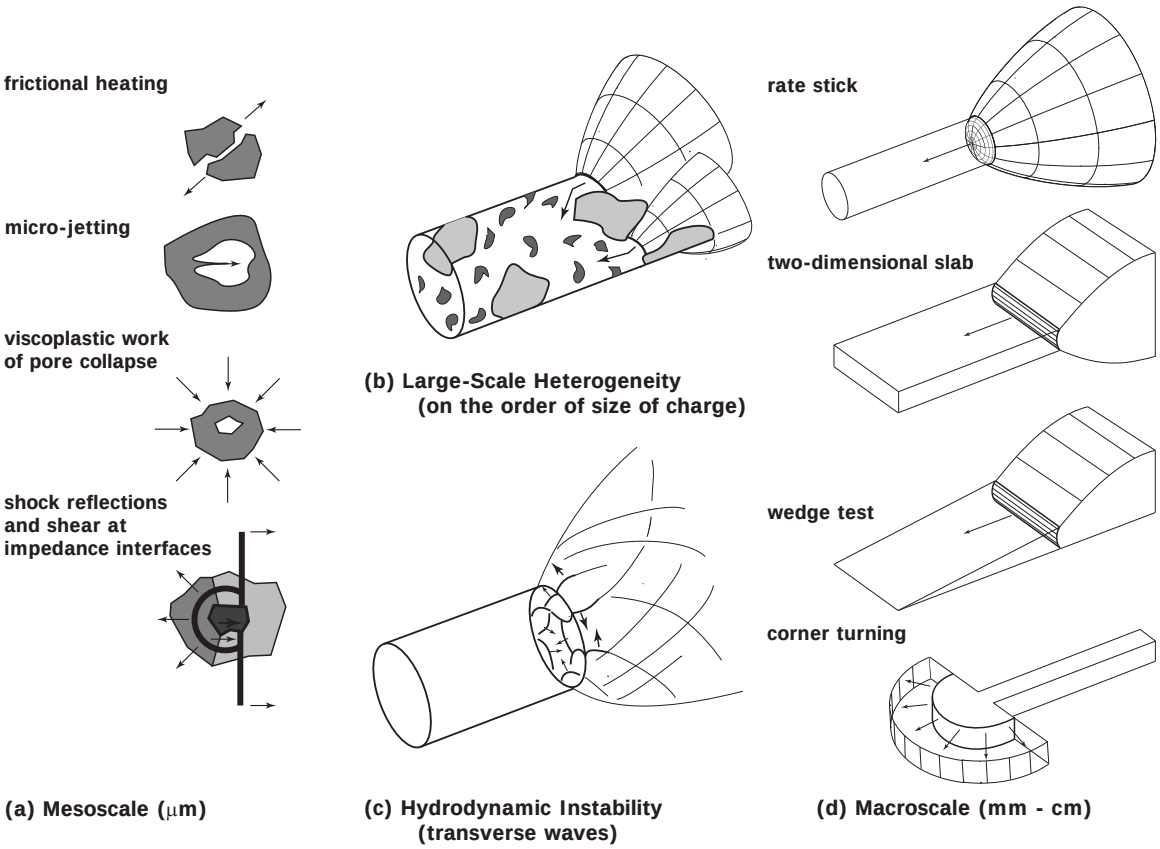

Figure 1. (a) Schematic representation of shock localization mechanisms occurring at the mesoscale, (b,c) phenomena that can bridge from mesoscale to macroscale, (d) continuum-level problems of engineering interest.

2. Explosives with Large-Scale Heterogeneities

One setting in which a clear delineation of scales between the mesoscale features (grains, voids, etc.) and the macroscopic dimension of the explosive charge is no longer possible is simply if the heterogeneities are on the order of the charge dimension itself, as shown schematically in figure 1(b). A sensitive primary explosive, for example, can continue to propagate as the dimension of the charge approaches that of the individual grains of explosive. A continuum-level model would not be expected to be valid in this case. Similarly, if very large scale heterogeneities are mixed with finer scale explosive grains, the description of the media as spatially uniform would no longer be valid, especially as the scale of the heterogeneity approached that of the charge. Examples of such explosives are encountered in commercial blasting emulsions and slurries, where often large ammonium nitrate prills approximately 2 mm in size are added to an emulsion explosive with emulsion droplets of micrometric size [11]. These large scale prills are not expected to burn in the reaction zone of the detonation, but rather in the expanding detonation products to provide additional push to rock. (Note that this description does not apply to ANFO-based explosives, where the ammonium nitrate prills themselves are the energetic component of the explosive.) A study of the detonation scaling between the cylindrical and slab geometry by Petel et al. [12] examined the commercial blasting explosive Apex Elite (Orica), an aluminized wax-based emulsion explosive containing glass microballoons ($\approx$ 50 μm) and

ammonium nitrate (AN) prills ($\approx$ 2-3 mm). Charges were confined in acrylic tubes and wide, two-dimensional slabs that were initiated with a line wave generator. The critical diameter of Apex Elite was found to be approximately 13 mm, in good agreement with the critical diameter quoted by the manufacturer. In the two-dimensional slab, however, the explosive was able to continue sustaining detonation propagation in slabs as thin as 3 mm with a velocity still representative of a high-order detonation. The ratio of critical diameter to critical thickness was thus $d_c/t_c \approx 4$. The interpretation advanced by Petel et al. [12] is that in a two-dimensional slab of large aspect ratio, the detonation front is able to propagate around zones of local failure and exploit new avenues of propagation in comparison to a cylindrical stick.

Further investigation of the geometric scaling in emulsion explosives by Sil'vestrov et al. [13] found almost perfect 2:1 scaling in comparing the detonation velocity and critical dimension in cylindrical and slab charges, seemingly in contradiction to the results of Petel et al. [12]. However, the explosive used by Sil'vestrov et al. featured a very fine emulsion droplet size (1 μm) and a very large concentration of sensitizing glass microballoons (8% by mass) and lacked the large scale prills found in the explosive studied by Petel et al. [12]. Thus, the explosive studied by Sil'vestrov et al. [13] is an example of a system where the scale of heterogeneity is fine enough to yield ideal behavior.

Other explosives that consist of a mixing of very dissimilar scales of heterogeneities include explosives with large, metallic inclusions that are intended to react in the explosive products or in the surrounding air, providing an enhanced blast effect—the so-called thermobaric class of explosive [11]. The dynamics of these systems are expected to be heavily influenced by the presence of the large scale, nonreactive (on the timescale of the detonation wave) inclusions. As shown by Lee [11, 14, 15], in a packed bed of inert particles saturated with explosive, a regime of propagation exists wherein the detonation propagates through the tortuous path created by the explosive filling the interstitial spaces (i.e., large bead regime). Investigation of the geometric scaling in this system has shown a value of failure diameter to failure thickness in the range $3.3 < d/t < 3.6$ [16], far from the classically expected value of 2. In the other regime identified by Lee, wherein the detonation propagates via collective transmission through the admixture of liquid explosive and smaller particles (i.e., small bead regime), a scaling of 2.3 ± 0.1 was obtained [12, 16], more consistent with the classic scaling predicted by front curvature theory. Thus, the geometric scaling between the critical dimensions in cylindrical and slab geometries exhibited the ability to discriminate these two regimes of behavior.

3. Explosives with Transverse Instabilities

Another means by which features at the mesoscale can cascade upward to result in spatial inhomogeneity in the energy release is via instability in the detonation wave front itself, as sketched in figure 1(c). The behavior of gas-phase explosives is entirely determined by the dynamics of transverse waves; all known detonable gas mixtures exhibit a cellular structure in the shock front and reaction zone due to the ensemble of interacting transverse waves [17–19]. Most homogeneous liquid explosives (e.g., nitromethane) are believed to have a cellular structure as well, and the near-limit propagation and failure mechanisms of liquid explosives are exclusively controlled by the dynamics of failure waves (i.e., dark waves) that propagate transverse to the detonation front [20].

The existence of a transient, multidimensional cellular structure in polycrystalline explosives is not as thoroughly established, however, a substantial amount of evidence pointing to the existence of transverse waves in solid explosives has been offered in the literature. Howe et al. [21] reported a transverse wave structure in TNT (both cast and pressed) nearly identical to that seen in the cellular structure of gaseous detonations. Howe et al. [21] used both the brass witness plate technique and an ingenious technique consisting of flushing gas around the charge, contained by thin sheets of vinylidene chloride, in order to create a thin layer of argon

on the charge surface surrounded by a thicker layer of propane. Upon shock breakout from the sides of the charge, the argon would flash with luminosity determined by the local shock strength that could be recorded via direct photography. The surrounding propane would not emit light, so the recorded luminosity could be directly attributed to the local structure of the front. The appearance of a transverse structure in cast TNT is perhaps not surprising, since TNT shares many characteristics with homogeneous explosives. The fact that a familiar structure was observed in pressed TNT, but with a scale about an order of magnitude smaller than that seen in cast TNT, is strong evidence that polycrystalline explosives may also exhibit a cellular instability similar to homogeneous explosives, but the scale of the instability can be reduced by the sensitizing nature of the polycrystalline media.

Transverse waves were also reported via the witness plate technique by Dunne in C-4 (91% RDX, grain size < 40 μm) [22]. In Dunne's experiment, the C-4 propagated into a semicircular wedge of explosive of decreasing thickness (from 3 mm to 0.5 mm) on a polished stainless steel disk. As the explosive layer thinned, the recorded cellular wave spacing observed on the witness plate increased from 0.05 mm to 1 mm. Thus, the instability in the detonation front in C-4 exists on a scale between the scale of the heterogeneity (grain size) and one order of magnitude larger. Held also reported an oscillating structure in RDX and HMX/TNT explosives in an early study using the transverse mirror method, in which light reflected off the bounding surface of an explosive charge is deflected by the application of detonation pressure [23]. Kozak et al. [24] documented the existence of a spinning wave structure in a number of explosives—trotyl (TNT) mixed with potassium picrate and trotyl mixed with hexogene (RDX)—when prepared in cylinders near the critical thickness necessary for propagation and recorded via a brass witness plate. Spinning detonation is another well recognized phenomenon in gaseous detonation that occurs when the dimension of the charge approaches the critical dimension necessary for detonation propagation. Kozak et al. [24] observed spinning detonation recorded as a spiral pattern on the witness plate with fixed angle; the wave pattern was less clear when rectangular prismatic charges were used.

A large body of evidence demonstrating the existence of transverse instabilities in solid explosives has been collected by Góis, Plaksin, and co-workers in Coimbra [25–27]. These studies had made extensive use of a technique wherein a strip of fiber optics (with up to 96 individual fibers) is in direct contact with the explosive or observes it via breakout through a thin Kapton layer, with the other end of the fiber fed into an electronic streak camera. These studies have examined a number of RDX and HMX-based plastic bonded explosives (PBX) with different binders or water as the interstitial material. An oscillating/transverse structure was observed in a number of these experiments, with the oscillation occurring on a scale 5 to 6 times that of the individual explosive grains that comprise the PBX. The Coimbra group has also observed cellular structure for detonation waves in nitromethane gelled with the addition of polymethyl methacrylate (PMMA) so that sensitizing glass microballoons (GMBs) can be held in suspension. In a corner turning experiment in which a detonation in a 12 $\times$ 12 mm channel of NM/PMMA/GMB emerged into a larger charge of explosive, a copper witness plate recorded a pattern of transverse waves [25] that is strikingly similar to the pattern of transverse waves observed when a gaseous detonation emerges from a channel [18, 19].

Use of the witness plate technique should always be viewed with considerable suspicion, since it is possible that the patterns observed may be a result of material instabilities in the high strain rate flow of the witness plate, rather than the imprint of an instability originating from the wave itself. Thus, it is necessary to verify the pattern seen in a particular witness plate can be reproduced in other plate materials using the same explosive. Recently, the author repeated the corner turning experiment of Góis et al. [25] using the same GMB concentration (NM/PMMA 96/4 with 3% GMB by mass) with similarly sized microballoons and using a variety of different plate materials (mild steel, stainless steel, and brass). Essentially, the same

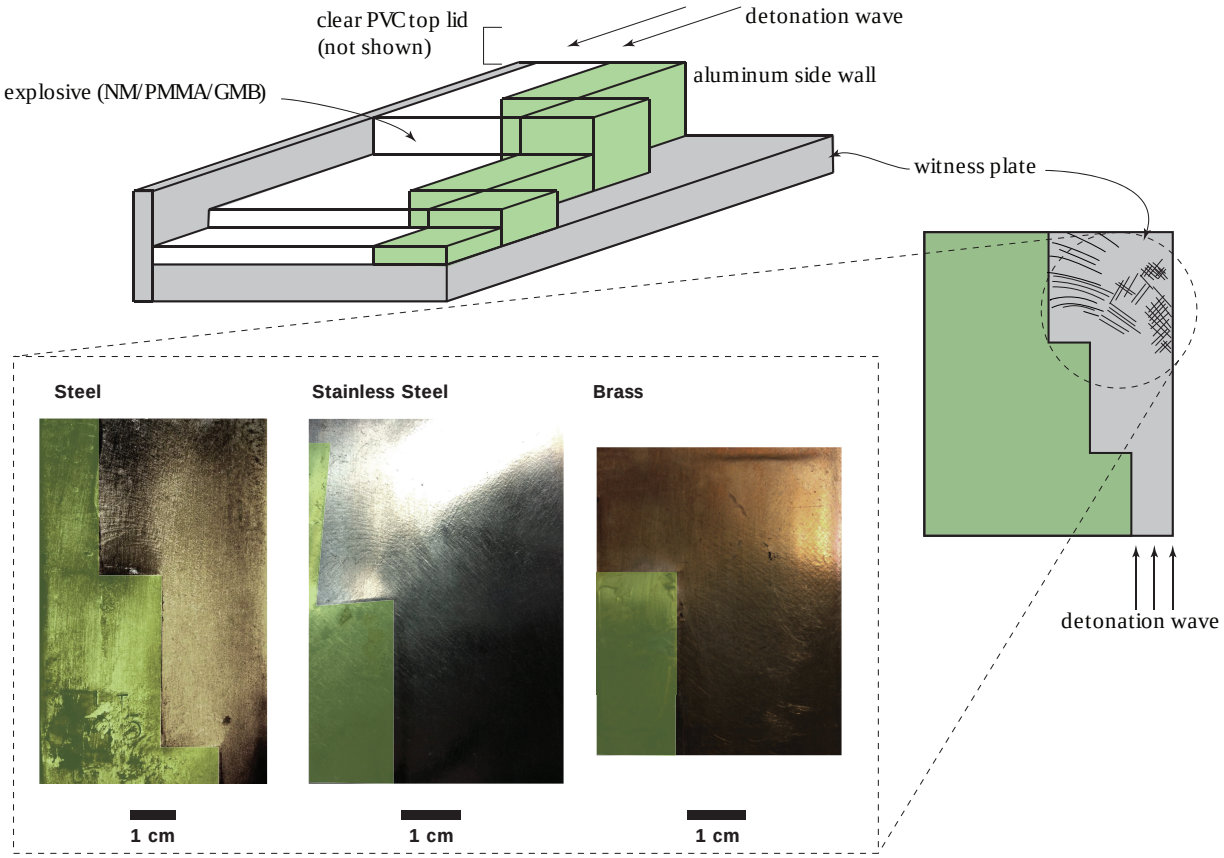

Figure 2. Experiment to record transverse waves on witness plates of different material. The charges were designed so that detonation in NM/PMMA/GMB explosive encountered steps of decreasing thickness and corner turns to accentuate the transverse wave dynamics. Inset shows locations of wave patterns with lines overdrawn to assist identifying the transverse waves in the photographs.

wave pattern reported by Góis et al. was observed, as shown in figure 2. Unfortunately, the patterns are not as clearly replicated and are very challenging to reproduce photographically. A line-drawing inset in figure 2 was added to assist in identifying the features of interest. The salient result is that these transverse waves have a spacing on the order of a millimeter, an order of magnitude greater than the average spacing between the individual microballoons.

Perhaps the most convincing demonstration of the existence of cellular structure in the gelled nitromethane system with particulate additives was the early study by Kato and Brochet [29], in which gelled nitromethane with aluminum particle (8-15 μm) additives was studied over mass loadings ranging from 5 to 15%. Side-on framing camera pictures clearly show a transverse wave structure existing in the detonation front, essentially identical to that reported by Howe et al. [21] in TNT and observed ubiquitously in gaseous explosives. The size of these transverse structures exist on a scale at least an order of magnitude larger than the average spacing between aluminum particles.

The NM/PMMA/GMB system is in many ways an ideal system for studying heterogeneous detonation, since the base explosive is a well characterized, insensitive homogeneous explosive. By adding GMBs, the concentration of heterogeneities can be precisely controlled. The observation of detonation occurring in charges much smaller than the critical diameter of neat nitromethane ensures that heterogeneous behavior, dictated by the presence of the GMBs, is

observed. (To the author's knowledge, detonation of gelled nitromethane without the addition of GMBs has never been reported.) The NM/PMMA/GMB system has now been thoroughly characterized by its detonation velocity/charge diameter dependences over a wide range of GMB concentrations in rate stick experiments [30–35]. The study by Góis et al. [9] verified that the expected 2:1 scaling between cylindrical charges and two-dimensional charges, until the concentration of microballoons became less than 0.5% by mass. In that case, the scaling between the critical diameter and thickness increased to greater than 3:1, similar to the anomalous scaling reported earlier in systems with large-scale heterogeneities. This result was later reproduced by the author [36]. It may be that as the concentration of microballoons becomes low, the inherent instability of the base nitromethane explosive becomes manifest or that at a low concentration of microballoons, the density of hot spots is insufficient to homogenize the explosive and nonideal behavior is observed. Again, examining the d_c/t_c parameter may be a means to diagnose the regime of behavior as ideal or nonideal.

4. Modeling

Computational simulations of the shock-localization processes occurring at the mesoscale has enormous potential to resolve the details of mechanisms that are currently only the subject of conjecture [37]. At present, however, these are *tour de force* computational exercises and cannot be considered models, but rather computational experiments. A model should strive to reproduce the key phenomenon using a simpler parameter set; otherwise, the model will be as unwieldy as the experiment itself.

A possible approach is the use of percolation theory, a branch of statistical mechanics, that was originally formulated to understand how the random formation of clusters can give rise to connectivity that spans the entire domain in problems such as polymerization, flow of liquid through porous media, and contagious spread of disease through a population. Dlott and co-workers [38, 39] recently proposed a percolation model for how a critical concentration of hot spots allows a reaction front to spread from ignition centers. The present author has explored a simple "Swiss cheese" model wherein detonation can propagate through the connected pores in a medium randomly filled with spherical holes [40, 41]. When finite sized cylindrical sticks and two-dimensional slabs of Swiss cheese where compared in finding the critical dimensions necessary for propagation through the domain, the $d_c/t_c = 4$ scaling was recovered. This simple model may have some utility in explaining the anomalous $d_c/t_c \approx 4$ scaling observed in many of the experiments reported in sections 3 and 4 of this paper.

5. Conclusions

The evidence reviewed in this paper can leave little doubt that a number of explosive systems do not exhibit "classical" behavior wherein the front dynamics can be described by the effective global curvature of the detonation front, and instead are profoundly influenced by local structures governed by the heterogeneity of the explosive media or instability of the wave itself. The number and type of explosives that fall into this category is unknown. The majority of studies have examined explosives of military interest which are typically comprised of fine grains much smaller than the scale of the explosive. Even then, front curvature studies using streak cameras sometimes observe structures in the wave front breakout that are reported as "ragged and turbulent" [42] but that are still fit with a monotonic function to describe the front curvature. The shock front in polycrystalline explosive, examined on the scale of individual grains, is expected to exhibit a roughened structure. The evidence reviewed in this paper, however, has suggested that an instability can exist on a scale at least an order of magnitude larger than the individual grains. The possibility that all detonation waves and shock waves of sufficient strength exhibit turbulent-like behavior has been advanced by Lee [43].

The use of "sub-grid" models to treat the shock energy localization and subsequent reaction that have been successfully applied to explosives with fine scale heterogeneities cannot be expected to work for all classes of explosives. Mesoscale calculations remain computationally challenging for the foreseeable future and yield such a vast amount of information that they are as complex as the experimental phenomena they seek to model. The need for simplifying models that can capture this multiscale behavior represents a considerable challenge to the development of predictive tools for modeling detonation dynamics.

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