Investigations on Deflagration to Detonation Transition in Porous Energetic Materials

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1 Introduction: Scope of Work and Report Format

This document is a final report that summarizes the research findings and research activities supported by the subcontract DOE-LANL-6730M0014-9, between the University of Illinois (D. Scott Stewart, Principal Investigator) and the University of California (Los Alamos National Laboratory, M-Division).

1.1 Scope of work

This contract with modifications was carried out for a five year period Jan. 1, 1994 to Dec. 30, 1998. The work is summarized succinctly by the task list as listed in the sub-contract:

1. Development of the BKS model
2. Development of the SPA model
3. DDT Theoretical and Computational Support
4. Numerical Implementation in LANL Hydrocodes
5. Characterization of Dispersive Acoustics
6. Multidimensional Studies of DDT
7. Convective Combustion
8. Theoretical and Computation Investigation of Microstructure in Energetic Materials
10. Properties of Structured Front Models
11. Interface code for DSD and documentation

1.2 Report format

The format of this report is as follows. First a narrative (overview) will be provided that will describe the efforts in the list given above with approximate dates. The purpose is to review the history and cite the various contributions to the LANL, DX-division, research mission afforded by this contract. Secondly a list of papers, reports and unpublished collaborative research note, written, wholly and partially supported by contract funds is included. For each listing an abstract is included and copies of two unpublished reports/research gs carried out in year 4 and 5 have been transmitted to John Bdzil. A list of relevant student Ph.D. theses is given. A list of personnel supported by the contract is given. Finally a representative list of trips and meetings, by the PI, wholly or partially supported by the contract is included.
1.3 Overview

The research carried out by this contract was part of a larger effort funded by LANL in the areas of deflagration to detonation in porous energetic materials (DDT) and detonation shock dynamics in high explosives (DSD). The managers for the UIUC effort were Drs. Blaine W. Asay and John B. Bdzil, both of DX-division. The DX-division manager was Dr. Phillip Howe.

The needs of the larger LANL effort dictated that frequent group meetings were held, and that the emphasis of individual research efforts be directed and changed according to the needs of the larger program. Frequent informal and formal reporting was done by phone, fax etc. Supporting calculations and notes were submitted to LANL and transferred directly to LANL, DX - staff members. Some information generated by this contract is in the form of unpublished notes. At the time of the writing of this final report some papers based on the research activities of this contract are being prepared for submission for publication.

The product of the contract has been to carry out directed research in collaboration and at the behest of LANL program managers, and to document the work in scholarly research articles, as far as possible. In addition, other materials in the form of computer research codes have been produced and transferred to LANL (including SEPTEMBER, AXS, TOYCODE, DKAPPA2D-3D, NONIDK.) DSD documentation has been drafted. A precursor of an extended work on DSD is the review article ref. [11] of Section 2. In the course of the contract, the program managers have been kept up to date on the current ongoing research and have suggested and approved changes in priorities amongst the tasks in the task list, with attendant effects on listed deliverables.

In the first three years of the contract the major focus was on DDT. However some researchers were carried out on DSD theory and numerical implementation. Blaine Asay carried out the program manager duties principally during this time period. In the last two years the principal focus of the contract was on DSD theory and numerical implementation. John Bdzil was the program manager during this time period. However, during the second period some work was also carried out on DDT.

1.4 DDT modeling

A previous contract, DOE-LANL-9-XG2-X5211-1, laid out model development for DDT research. The main line LANL experiment that was being modeled was a DDT tube experiment where powdered HMX crystals were packed in a DDT tube, and the HMX bed was then impacted by a piston driven by combustion gases. This experimental data base is referred to in the papers listed in this report. The SPA model referred to was a simple one-phase hydrodynamic model, where the purpose was to work out the basic thermodynamics and dynamics admitted by a model that admitted compaction and reaction as independent thermodynamic processes.

Later model DDT development, involved a group of LANL scientists and other academic researchers including but not limited to. John Bdzil, Ralph Menikoff, Steven Son (LANL) and Prof. Ash Kapila (Rensselaer Polytechnic Institute). One of the DDT models we jointly developed was given the internal LANL-recognized name. BKS for three of the originators. Bdzil, Kapila and Stewart. This class of models was based on a two-phase, hydrodynamics
modeling approach, where the HMX granular bed was considered to be a mixture of solids and gases, with conversion from solid to gas. The BKS model was based on a reduction of a two-phase DDT model originally posed by Baer and Nunziato, such that the solid phase and the gas phase had the same velocity. Indeed, both the SPA model and the BKS model had in common that they were one velocity model.

The different models had different features due to their formulations, and it was recognized that it was possible to design models that incorporated properties of the simple one phase SPA-like and the explicit constitutive structure of the more complex BKS model. It was possible to pose other models such as GISPA (Gas Interpolated SPA) model and the SVG (model).

Research was carried out that addressed tasks 1. through 8. and reported on in refs. [1], [2], [3], [4] and [5] of Section 2. Numerical implementations of DDT phenomena were carried out using the SEPTEMBER CODE that was prototyped at UIUC, and later the AXS code that was described in ref. [3]. Two dimensional DDT simulations were carried out by the AXS code and were described in ref. [3]. Elements of SEPTEMBER that specifically modeled the BKS model, were incorporated into the LANL code MESA through collaboration with Steve Son, John Bdzil, Ed Kober all of LANL.

The summary of research papers found in Section 2. give the findings in the attached abstracts, but one conclusion that stands out is that matching experimental DDT transients observed in LANL DDT-tube experiments, requires the treatment of compaction and reaction as independent thermodynamic processes.

1.5 DSD modeling

John Bdzil and the Principal Investigator are co-authors of a theory for propagation of detonation shocks in explosive materials. Of particular (but not exclusive interest) is the propagation of detonation shock waves in condensed high explosive. The theory leads to evolution equations that describe the motion of the detonation in terms of intrinsic geometric quantities such as the normal velocity of the shock and the curvature. The success of the theory in the description of physical experiment has led to an engineering implementation of the theory that is also referred to as DSD.

Advances in DSD theory were partially supported by the contract, and are described in refs. [6], [7], [8], [9] and [10]. Importantly we showed that higher order corrections to the theory included shock acceleration effects and higher order derivative in time of shock intrinsic quantities, see ref. [8]. We also worked out (albeit numerically) a DSD theory for real equation of state and critical curvature, see ref. [9].

The DSD implementation in high explosive design hydrocodes, is a major technology refinement of multi-dimensional explosive design technology that is being incorporated into LANL explosive code (as well as other lab codes used in the DOD/DOE community). Its implementation has two facets. The first is the construction of burn time tables for explosive pieces. One has a piece of explosive with known initial geometry. One has a detonation initiation scheme to start the detonation in the explosive piece. Then one calculate the time at which cells, that represent material elements are crossed by the detonation shock. The DSD implementation has included writing code/codes that propagate the detonation. One of the major innovations that we made in this area, with John Bdzil and Tariq Aslam
(formerly of UIUC, now a LANL staffer), is the use of Level Set Technology to propagate detonations in complex geometries, see refs. [12], [13], [14], [15].

The second facet of DSD implementation is the calculation of self-consistent states behind the detonation front. This work has been quite difficult and multi-disciplinary and it is fair to say our progress has been quite slow, since the problem itself is difficult to pose in an unambiguous mathematically consistent manner. In particular, one attempts to modify an existing hydrodynamic scheme in a way that attempts to be consistent with pre-existing program burn methods that are implemented in explosive design codes. This work has been recorded in unpublished research notes, refs. [16] and [17]. Ref. [17] is the first draft of a paper by John Bdzil, Tom Jackson (a senior research scientist supported by the contract in Year 5) and the PI that summarizes our joint efforts on discrete program burn.

What follows below is a complete listing of research papers, supported UIUC student theses, and unpublished research notes, with the published reference and the abstract or a brief description of the work.

2 Summary of research papers /unpublished research notes/ student theses

2.1 Modeling Deflagration to Detonation


(Note: The final revision of this paper was carried out under this contract, but the work was initiated in the previous contract.)

Abstract A simplified model that can predict the transition from compaction to detonation and shock to detonation is given with the aim of describing experiments in beds of porous HMX. In the case of compaction to detonation, the energy of early impact generates a slowly moving, convective-reactive deflagration that expands near the piston face and evolves in a manner that is characteristic of confined deflagration to detonation transition. A single-phase state variable theory is adopted in contrast to a two-phase axiomatic mixture theory. The ability of the porous material to compact is treated as an endothermic process. Reaction is treated as an exothermic process. The algebraic (Rankine-Hugoniot) steady wave analysis is give for inert compaction waves and steady detonation waves in a piston supported configurations, typical of the experiments carried out for porous HMX. A structure analyst of the steady compaction wave is given. Numerical simulations of deflagration to detonation are carried out for parameters that describe an HMX-like material and compared with the experiments. The simple model predicts the high density plug that is observed in the experiments and suggests that the leading front of the plug is a secondary compaction wave. A shock to detonation transition is also numerically simulated.

**Abstract** A numerical study of the deflagration to detonation (DDT) in porous HMX materials is carried out. Three reactive-flow models varying from single phase to three phase formulations are chosen for the study. The GISPA model is a single-phase model and the BKS is a simplified two-phase, gas and solid model. The SVG model is a thre-phase model which is based on the evolution of solid, gas and void. The modeling assumptions made in construction of the SVG model are presented with a brief description of the other two models. In addition, a new reaction-kinetics model, or rate law, is presented to model energy release. The rate law accounts for autocatalytic decomposition of HMX and the pressure dependent shock-to-detonation transition kinetics. The model results are compared in detail against the DDT events observed in physical experiments. Numerical simulation of inert compaction waves and DDT is carried out for parameters suitable for powered HMX. The simulation shows that all three models can effectively predict: (a) the formation of secondary compaction wave and a high-density plug, (b) initiation of the transition to detonation in front of the plug, and (c) survival of the plug residual after the detonation. The SVG model compares the best against the measurable date of the physical experiment and is also computationally efficient and well-posed. Therefore it is a good candidate for multi-dimensional DDT calculation.


**Abstract** This paper explains the methodology used to develop a high-resolution, multi-dimensional Euler solver that is capable of handling non-ideal equation of state and stiff chemical source terms. We have developed a pointwise implementation that has computational advantages for our intended applications, as opposed to a finite volume implementation. Our solver allows for the placement of internal reflective boundaries and standard inflow and outflow and reflective boundaries at the edge of the domain. We discuss the spatial discretization and the temporal integration schemes, upwinding and flux splitting and the combined use of the Lax-Friedrichs and Rose schemes to solve for the required fluxes. A complete description of the pointwise internal boundary method is given. An overall summary of a representative code structure is given. We provide details on the verification of our integrated set of algorithms that resulting in an application code. We demonstrate the order of convergence for test problems. Two example applications from measurement of detonation shock dynamics and deflagration to detonation transition in porous energetic materials are presented.

The structure of the velocity relaxation zone in a hyperbolic, nonconservative, two-phase model is examined in the limit of large drag, and in the context of the problem of deflagration-to-detonation transition in a granular explosive. The primary motivation of the study is the desire to relate the end states across the relaxation zone, which can then be treated as a discontinuity in a reduced equivelocity mode that is computationally more efficient than its parent. In contrast to a conservative system, where the end states across thin zones of rapid variation are determined principally by algebraic statement of conservation, the nonconservative character of the present system requires an explicit consideration of the structure. Starting with the minimum admissible wave speed, the structure is mapped out as the wave speed increases. Several critical wave speed corresponding to changes in the structure are identified. The archetypal structure is partly dispersed, monotonic and involves conventional hydodynamics shocks in one or both phases. The picture is reminiscent of, but more complex than what is observed in such (simpler) two-phase media as a dusty gas.


**Abstract** The two-phase mixture model developed by Baer and Nunziato (BN) to study the deflagration-to-detonation transition (DDT) in granular explosives is critically reviewed. The continuum-mixture theory foundation of the model is examined, with particular attention paid to the manner in which its constitutive functions are formulated. Connections between the mechanical and energetic phenomena occurring at the scales of the grains, and their manifestations on the continuum averaged scale, are explored. The nature and extent of approximations inherent in formulating the constitutive terms, and their domain of applicability, are clarified. Deficiencies and inconsistencies in the derivation are cited, and improvements suggested. It is emphasized that the entropy inequality constrains but does not uniquely determine the phase interaction terms. The resulting flexibility is exploited to suggest improved forms for the phase interactions. These improved forms better treat the energy associated with the dynamic compaction of the bed and the single-phase limits of the model. Companion papers of this study [Kapila et al., Phys. Fluids 9, 3885 (1997); Kapila et al., in preparation; Son et al., in preparation] examine simpler, reduced models, in which the fine scales of velocity and pressure disequilibrium between the phases allow the corresponding relaxation zones to be treated as discontinuities that need not be resolved in a numerical computation.
2.2 DSD Theory


(Note: This work was relevant and was beneficial to this contract but was not directly supported. The work was supported with a grant from USAFRL Amment Directorate, Warheads Branch, Eglin AFB.)

We derive the normal detonation shock velocity-curvature relation for a near-Chapman-Jouguet detonation for an explosive material with Arrhenious kinetics and a large activation energy. Large activation energy asymptotics are used to develop an explicit exponential formula relation the shock curvature, \( \kappa \), to the normal detonation shock velocity \( D_n \). In this case, the \( D_n - \kappa \) relation is multi-valued and has a turning point with a critical curvature \( \kappa_{cr} \) such that for \( \kappa > \kappa_{cr} \), the possibility of a detonation extinction arises. The asymptotic formula is in excellent agreement with the exact solution found by a numerical shooting procedure.


This paper is a precursor of ref. [8].


(Note: This work was relevant to this contract and was beneficial, but was not directly supported. The work was supported with a grant from USAFRL Amment Directorate, Warheads Branch, Eglin AFB. Importantly this work stimulated the investigations now carried out at Los Alamos on the "Extended Theory of Detonation Shock Dynamics", where shock acceleration effects are considered.)

**Abstract** We present an asymptotic theory for the dynamics of detonation when the radius of curvature of the detonation shock is large compared to the one-dimensional, steady, Chapman-Jouguet (CJ), detonation reaction-zone thickness. The analysis considers time-dependence in the slowly-varying reaction-zone, not considered in previous works. The detonation is assumed to have a sonic point in the reaction-zone structure behind the shock, and is referred to as an *eigenvalue* detonation. A new iterative method is used to calculate the eigenvalue relation, that ultimately is expressed as intrinsic partial differential equation (PDE) for the motion of the shock surface. Two cases are considered for an ideal equation of state. The first corresponds to a model of a condensed phase explosive, with modest reaction rate sensitivity, and the intrinsic shock surface PDE is a relation between the normal
detonation shock velocity, \( D_n \), the first normal time derivative of the normal shock velocity, \( \dot{D}_n \), and the shock curvature, \( \kappa \). The second case corresponds to a gaseous explosive mixture, with the large reaction rate sensitivity of Arrhenius kinetics, and the intrinsic shock surface PDE is a relation between the normal detonation shock velocity, \( D_n \), its first and second normal time derivatives of the normal shock velocity, \( \dot{D}_n, \ddot{D}_n \), and the shock curvature, \( \kappa \) and its first normal time derivative of the curvature, \( \dot{\kappa} \). For the second case, one obtains a one-dimensional theory of pulsations of plane, CJ detonation and a theory that predicts the evolution of self-sustained cellular detonation. Versions of the theory includes the limits of near-CJ detonation, and when the normal detonation velocity is significantly below its CJ-value. The curvature of the detonation can also be of either sign corresponding to both diverging and converging geometries.


**Abstract** We present a model and a simple-to-implement numerical procedure that obtains the normal detonation shock velocity (\( D_n \))-curvature (\( \kappa \)) relationships for an explosive material with nonideal equation of state and an arbitrary reaction rate law. In addition we illustrate numerically (for a nonideal equation of state) and analytically (for an ideal equation of state with a large activation energy rate law) that for sufficient rate-state-sensitive explosive, the correspond \( D_n, \kappa \) response curve can have two turning points, at \( D_n, \kappa \)-pairs \( [(D_n)_1, \kappa_1], [(D_n)_2, \kappa_2] \), such that \( \kappa_1 > \kappa_2 > 0 \) for \( D_{CJ} \geq (D_n)_1 \geq (D_n)_2 \) and such that the curve has a Z-shape. The top branch of the Z-response curve has been previously associate with detonation extinction at a critical curvature. The bottom branch can be possibly associated with low-velocity detonation and rapid change from low-order detonation to high-order.


**Abstract** A detonation shock-evolution equation that predicts both pulsating and cellular detonation has been derived in the limit of near-Chapman-Jouguet detonation, weak curvature, slow temporal variation, and large activation energy with the newly applied technique of the method of successive approximation. The evolution equation describes a wave hierarchy that is consistent with the linear stability theory of the evolution equation. We define the parameter regime for which the equations applies. The transverse wave instability as indicated from analysis, leads to cellular detonation. Triple-point tracks correspond to shock-shock interactions of the dynamic solutions of smooth portions of the front. The dynamics of the cellular reaction
zone and the triple points are generated as the interaction of the independently propagating fronts and the consequent shock-shock interactions, not as the centers of blast waves. Explicit criteria for the prediction of cell widths and cell aspect rations are given.


Abstract Detonations are comprised of broad detonation shocks supported by thin reaction zones. Approximations based on weak shock curvature measured on the inverse reaction zone scale, and quasi-steady flow, measured on the particle passage time through the reaction zone can be used to simplify the mathematical description of detonations that are governed the gasdynamic equations for a reacting flow. When the detonation reaction zone contains a sonic locus it is possible to derive intrinsic (coordinate independent) partial differential equations for the lead detonation shock's motion in terms of the normal detonation shock velocity, the shock curvature and higher normal time derivatives. We refer to this collection of theory and supporting experimental results as Detonation Shock Dynamics after Whitham's Geometrical Shock Dynamics. The reduced detonation dynamics is based on the concept of a eigenvalue (sonic) detonation, an idea that goes back to the original investigations in the 1940's. We present a review of the theoretical and experimental developments and attempt to update Fickett and Davis' discussion of work prior to 1980. We give examples of the theory and applications which include: i) weakly-curved, quasi-steady, near-CJ detonation ii) critical detonation curvature iii) quasi-steady extinction and ignition (and low velocity detonation) iv) shock acceleration effects and v) cellular and pulsating detonation in gases. We also review the engineering method of Detonation Shock Dynamics as it is applied to explosive systems.

2.3 DSD Wave front tracking


*Note:* This paper was a precursor proceeding paper that introduced the idea of using level-set techniques for multi-dimensional detonation propagation. A complete analysis of the level-set techniques are given in ref. [13].


Abstract We give an extension of the level set formulation of Osher and Sethian, which describes the dynamics of surfaces that propagate under the
influence of their own curvature. We consider an extension of the original algorithms for finite domains that includes boundary conditions. We discuss this extension in the context of a specific application that comes from the theory of detonation shock dynamics (DSD). We given an outline of the theory of DSD which includes the formulation of the boundary conditions that comprise the engineering model. We give the formulation of the level-set methods, as applied to our application with finite boundary conditions. We develop a numerical method to implement arbitrarily complex 2D boundary conditions and give a few representative calculations. We also discuss the dynamics of level curve motion and point out restrictions that arise when applying boundary conditions.


**Abstract** Detonation Shock Dynamics (DSD) can be used to model the effects that shock curvature $\kappa$ has on detonation speed $D_n(\kappa)$. At the edges of the explosive, $D_n(\kappa)$ is supplemented with boundary conditions. By direct numerical simulation (DNS), we study how the reaction zone interacts with the edge. DSD theory has been integrated with the level-set method of Osher & Sethian and the Los Alamos DNS code Mesa to create a powerful tool for simulating complex explosive-containing systems.


**Abstract** Comparisons between direct numerical simulation (DNS) of detonation and detonation shock dynamics (DSD) is made. The theory of DSD defines the motion of the detonation shock in terms of intrinsic geometry of the shock surface, in particular for condensed phase explosives the shock normal velocity $D_n$, the normal acceleration $\dot{D}_n$ and the total curvature, $\kappa$. In particular, the properties of three intrinsic front evolution laws are studied and compared. These are 1) Constant speed detonation (Huygens’ construction), 2) Curvature dependence speed propagation ($D_n - \kappa$ relation), and 3) Curvature and speed dependent acceleration ($\dot{D}_n - D_n - \kappa$ relation). We show that it is possible to measure shock dynamics directly from simulation of the reactive Euler equations and that subsequent numerical solution of the intrinsic partial differential equation for the shock motion (e.g. a $\dot{D}_n - D_n - \kappa$ relation) reproduces the exact shock motion with high precision. Finally, a few numerical examples will be give to demonstrate the properties of these DSD intrinsic evolution laws.
2.4 DSD Program Burn Implementation

16. Stewart, D. S., Notes on: The dynamics of a model of curved detonation, discrete and continuous

(Note: This work has been transmitted to John Bdzil of LANL. The work is still unpublished and hence should be considered not for general distribution.)

These notes record a study of a two equation model in space and time that mimics multidimensional detonation shock dynamics. The analog is posed and its characteristics studied in PART II: "Results for a continuous analog". A discrete version of the analog is studied in PART I: "Results for the discrete analog". Importantly in PART II, the logic and properties of a Program Burn algorithm are stated and studied.

17. Bdzil, J. B., Jackson, T. L., and Stewart, D. S., Discrete approximations of detonation flows with structured detonation reaction zones by discontinuous front models: A program burn algorithm based on Detonation Shock Dynamics

(Note: This work has been transmitted to John Bdzil of LANL. The work is still unpublished and hence should be considered not for general distribution.)

Abstract In the design of explosive systems the generic problem that one must consider is the propagation of a well-developed detonation wave sweeping through an explosive charge with a complex shape. At a given instant of time the lead detonation shock is a surface that occupies a region of the explosive and has a dimension that is characteristic of the explosive device, typically on the scale of meters. The detonation shock is powered by a detonation reaction zone, sitting immediately behind the shock, which is on the scale of 1 millimeter or less. Thus, the ratio of the reaction zone thickness to the device dimension is of the order of 1/1000 or less. This scale disparity can lead to great difficulties in computing three-dimensional detonation dynamics. An attack on the dilemma for the computation of detonation systems has lead to the invention of sub-scale models for a propagating detonation front that we refer to herein as program burn models. The program burn model seeks not to resolve the fine scale of the reaction zone in the sense of a DNS simulation. The goal of a program burn simulation is to resolve the hydrodynamics in the inert product gases on a grid much coarser than that required to resolve a physical reaction zone. We first show that traditional program burn algorithms for detonation hydrocodes used for explosive design are inconsistent and yield incorrect shock dynamic behavior. To overcome these inconsistencies, we are developing a new class of program burn models based on detonation shock dynamic (DSD) theory. It is hoped that this new class will yield a consistent and robust algorithm which reflects the correct shock dynamic behavior.
2.5 Persons supported by the contract

All persons listed were either U.S. citizens or U.S. permanent residents. The salary or material support was provided to the following individuals during the contract.

- D. S. Stewart, Principal Investigator
- Shaojie Xu, former TAM Research Assistant
- Jin Yao, former TAM Research Assistant
- Tariq Aslam, former TAM Research Assistant
- Brett Okhuysen, former TAM Research Assistant
- Dr. Thomas L. Jackson, Senior Research Scientist, Center for Simulation of Advanced Rockets, UIUC and Adjunct Professor of Theoretical and Applied Mechanics

2.6 Student theses


3 Partial listing travel

Here we list meetings were the research results of this contract was discussed. Travel to LANL for program review is not listed here. Travel support for the PI enables dissemination of scientific results generated and of interest to LANL and to a broader scientific community. The PI does his best to represent LANL’s interest in an appropriate way and report new information back to LANL program managers and staff LANL scientists. Broader communication of the scientific base of this research is an effective way to generate interest in creating a larger knowledge base of interest to LANL, DX programs. The travel support was sometimes shared between this contract and other contracts held by the PI. The foreign travel listed was approved by the DOE. This listing of travel is representative and is not complete.

- PI travel to Snowmass CO to attend the Detonation Symposium 9/98.
- PI travel to Boulder CO to attend 27th International Symposium on Combustion 8/19/98 - 7/21/98.
- PI travel to American Physical Society Syracuse. NY 11/96
• PI travel to 26th Combustion Symposium, Naples Italy, 7/96
• SIAM National meeting, July 1996
• PI travel to Gordon Conference on Engertic Materials, June 1996
• PI travel to APS Division of Fluid Dynamics Meeting, Irvine CA 11/95
• PI travel to American Physical Society topical meeting on Shock Compression of Condensed Matter, Aug 95, Seattle, WA.