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Author(s):

K. A. Gonthier
R. Menikoff
S. F. Son
B. W. Asay

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MODELING ENERGY DISSIPATION INDUCED BY QUASI-STATIC COMPACTION OF GRANULAR HMX *

K. A. Gonthier[†], R. Menikoff, S. F. Son, and B. W. Asay

Los Alamos National Laboratory, Los Alamos, New Mexico 87545

A simple extension of a conventional two-phase (inert gas and reactive solid) continuum model of Deflagration-to-Detonation Transition (DDT) in energetic granular material is given to account for energy dissipation induced by quasi-static compaction. To this end, the conventional model equations, valid in the limit of negligible gas phase effects, are supplemented by a relaxation equation governing irreversible changes in solid volume fraction due to intergranular friction, plastic deformation of granules, and granule fracture. The proposed model constitutes a non-strictly hyperbolic system of equations, and is consistent with the Second Law of Thermodynamics for a two-phase mixture. The model predicts stress relaxation and substantial dissipation induced by quasi-static compaction; such phenomena are commonly observed in quasi-static compaction experiments for granular HMX. Predicted intergranular stress histories compare well with experimental data.

INTRODUCTION

There has been considerable research during the last thirty years addressing Deflagration-to-Detonation Transition (DDT) in granular energetic materials; much of this work has been motivated by concerns over the accidental detonation of damaged high explosives due to mechanical stimuli. Here, a damaged explosive refers to a cast explosive containing local granulated regions. It is widely accepted that various dissipative mechanisms induced by compaction of the granulated material give rise to local regions of thermal energy concentration termed hot-spots; such dissipative mechanisms include intergranular friction, plastic deformation of granules, and granule fracture. If the energy dissipation rate is sufficiently high, chemical reaction is initiated, and transition to detonation is possible. As such, models used to analyze DDT in these systems should accurately account for energy dissipation induced by material compaction.

To this end, we extend the two-phase DDT model of Baer and Nunziato (BN model) [1] to account for energy dissipation induced by quasi-static compaction of granular HMX. Though the BN model adequately accounts for compaction dynamics, Bdzil et al. [2] have recently shown that it improperly accounts for compaction energetics. In particular, the BN model predicts no energy dissipation due to quasi-static compaction, contrary to experimental data for granular HMX which indicate substantial dissipation [4]. A minimum requirement of any model is that it predict results commensurate with experiments in appropriate limits.

In this paper, we first give a brief description of the proposed model, valid in the limit of negligible gas phase effects (i.e., when the gas density is much smaller than the solid density). Though we only consider a granular solid, the model is equally applicable when gas phase effects are included. Next, we show that the model satisfies the Second Law of Thermodynamics for a two-phase mixture. Lastly, we demonstrate the model by applying it to the quasi-static compaction of granular HMX, and give comparisons of model predictions with experimental data.

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[†]Corresponding author. E-mail: gonthier@lanl.gov

MATHEMATICAL MODEL

The dimensional model equations, valid in the limit of negligible gas phase effects, are given by the following:

$$\frac{\partial}{\partial t} [\rho_s \phi_s] + \frac{\partial}{\partial x} [\rho_s \phi_s u_s] = 0, \quad (1)$$

$$\frac{\partial}{\partial t} [\rho_s \phi_s u_s] + \frac{\partial}{\partial x} [\rho_s \phi_s u_s^2 + P_s \phi_s] = 0, \quad (2)$$

$$\begin{aligned} \frac{\partial}{\partial t} \left[\rho_s \phi_s \left(e_s + \frac{u_s^2}{2} \right) \right] \\ + \frac{\partial}{\partial x} \left[\rho_s \phi_s u_s \left(e_s + \frac{u_s^2}{2} + \frac{P_s}{\rho_s} \right) \right] = 0, \end{aligned} \quad (3)$$

$$\frac{\partial \phi_s}{\partial t} + u_s \frac{\partial \phi_s}{\partial x} = \frac{\phi_s (1 - \phi_s)}{\mu_c} (P_s - \beta_s), \quad (4)$$

$$\frac{\partial \tilde{\phi}_s}{\partial t} + u_s \frac{\partial \tilde{\phi}_s}{\partial x} = \begin{cases} \frac{1}{\tilde{\mu}} (f - \tilde{\phi}_s) & \text{if } \tilde{\phi}_s \leq f \\ 0 & \text{if } \tilde{\phi}_s > f. \end{cases} \quad (5)$$

Here, subscript "s" denotes solid phase quantities. Independent variables are time, t , and position, x . Dependent variables are density, ρ_s ; volume fraction, ϕ_s ; particle velocity, u_s ; pressure, P_s ; specific internal energy, e_s ; no-load volume fraction, $\tilde{\phi}_s$; equilibrium no-load volume fraction, f ; and intergranular stress, β_s .

Equations (1-3) are conservation equations for the mass, momentum, and energy of the solid. Equation (4) is a relaxation equation for mechanical stresses, and Eq. (5) is a relaxation equation for the no-load volume fraction; the constant parameters μ_c and $\tilde{\mu}$ determine the relaxation rates. The inclusion of $\tilde{\phi}_s$ as an additional internal variable in the theory is not standard, but enables rate-independent dissipation induced by material compaction to be modeled. Equation (5) is not included in the BN model. Constitutive relations needed to mathematically close Eqs. (1-5) are constructed based on both rational thermodynamics and experimental data.

Using a rational thermodynamics development [3], we assume the granular system is 1) in local thermodynamic equilibrium, and 2) can be described in terms of a Helmholtz free energy

potential, ψ_s , having the following functional dependency:

$$\psi_s = \psi_s(\rho_s, T_s, \phi_s - \tilde{\phi}_s), \quad (6)$$

where T_s is the solid temperature. Here, ρ_s , T_s , ϕ_s , and $\tilde{\phi}_s$ are independent thermodynamic variables from which all dependent thermodynamic variables are determined. Given ψ_s , the dependent thermodynamic variables are defined by

$$P_s \equiv \rho_s^2 \frac{\partial \psi_s}{\partial \rho_s} \Big|_{T_s, \phi_s, \tilde{\phi}_s}, \quad (7)$$

$$\eta_s \equiv - \frac{\partial \psi_s}{\partial T_s} \Big|_{\rho_s, \phi_s, \tilde{\phi}_s}, \quad (8)$$

$$e_s \equiv \psi_s + T_s \eta_s, \quad (9)$$

$$\beta_s \equiv \rho_s \phi_s \frac{\partial \psi_s}{\partial \phi_s} \Big|_{\rho_s, T_s, \tilde{\phi}_s} = - \rho_s \phi_s \frac{\partial \psi_s}{\partial \tilde{\phi}_s} \Big|_{\rho_s, T_s, \phi_s}, \quad (10)$$

where η_s is the specific solid entropy. A functional form of ψ_s which is consistent with Eqs. (6-10), and can reasonably model the material response to both low and high pressure loading, is given by

$$\psi_s = \psi_{sp}(\rho_s, T_s) + \int \frac{\beta_s}{\rho_s \phi_s} d(\phi_s - \tilde{\phi}_s), \quad (11)$$

where

$$\frac{\beta_s}{\rho_s \phi_s} = h(\rho_s, \phi_s - \tilde{\phi}_s). \quad (12)$$

Here, ψ_{sp} is the Helmholtz free energy of the pure solid, assumed known, and h is an arbitrary function of ρ_s and the difference $\phi_s - \tilde{\phi}_s$.

Based on the quasi-static compaction data of Coyne et al. [4] for granular HMX, and the constraint of Eq. (12), we take

$$\beta_s = -\tau \phi_s (\phi_s - \tilde{\phi}_s) \frac{\ln [\kappa - (\phi_s - \tilde{\phi}_s)]}{\kappa - (\phi_s - \tilde{\phi}_s)}, \quad (13)$$

and

$$f = \frac{1}{1 - \phi_{so}} [(1 - \kappa - \phi_{so}) \phi_s + \kappa \phi_{so}], \quad (14)$$

where $\tau = 2.3 \text{ MPa}$, $\kappa = 0.03$, and ϕ_{so} is the initial solid volume fraction of the granular material. Equations (7-9), (11), (13), and (14) are sufficient to mathematically close Eqs. (1-5).

Second Law of Thermodynamics

The Second Law of Thermodynamics for a thermally isolated two-phase (gas and solid) mixture reduces to the following mathematical expression in the limit of negligible gas phase effects [2]:

$$\dot{\Sigma}_\eta \equiv \frac{\partial}{\partial t} [\rho_s \phi_s \eta_s] + \frac{\partial}{\partial x} [\rho_s \phi_s u_s \eta_s] \geq 0, \quad (15)$$

where $\dot{\Sigma}_\eta$ is the volumetric entropy production rate. Using Eqs. (1), (3), and (11), we obtain the following (here, we omit details of the derivation for brevity):

$$\dot{\Sigma}_s = \frac{1}{T_s} \left[(P_s - \beta_s) \frac{d\phi_s}{dt_s} + P_s \frac{d\tilde{\phi}_s}{dt_s} \right], \quad (16)$$

where the Lagrangian differential operator is defined as

$$\frac{d}{dt_s} \equiv \frac{\partial}{\partial t} + u_s \frac{\partial}{\partial x}.$$

It is seen from Eqs. (4) and (5) that $\dot{\Sigma}_\eta$ is non-negative provided that $P_s \geq 0$; this condition holds for granular materials as they cannot support tensile stresses. As discussed in the following section, $P_s = \beta_s$ in the quasi-static compaction limit, and Eq. (16) reduces to

$$\dot{\Sigma}_\eta = \frac{\beta_s}{T_s} \frac{d\tilde{\phi}_s}{dt_s}; \quad (17)$$

thus, the model is dissipative in this limit.

QUASI-STATIC COMPACTION

To demonstrate the model, we simulate a quasi-static compaction experiment performed by Coyne et al. [4] on a 15 g sample of *Class D* granular HMX. The sample, strongly confined by a movable piston-cylinder apparatus (ID = 2.54 cm), has an initial solid volume fraction of $\phi_{so} = 0.655$ (based on a crystal density of $\rho_s = 1.903 \text{ g/cm}^3$), and an initial

length of $L_o = 2.36 \text{ cm}$. The sample undergoes a (I)loading-(II)unloading-(III)reloading cycle at a constant extension rate of $u_p = 18.8 \text{ cm/min}$ during loading, where u_p is the piston velocity.

To this end, we assume 1) the solid is incompressible; 2) $\mu_c u_p / L_o \ll 1$; and 3) all variables depend only on time. The second assumption implies that the time scale associated with equilibration of mechanical stresses is much smaller than that associated with volume changes due to piston motion; consequently, it can be shown from Eq. (4) that $P_s = \beta_s$ in this limit. With these assumptions, Eq. (1) reduces to a homogeneous ordinary differential equation which can be directly integrated, and the resulting algebraic equation solved for ϕ_s , to get

$$\phi_s(t) = \left(\frac{L_o}{L(t)} \right) \phi_{so}, \quad (18)$$

where

$$L(t) = L(t_o) - u_p(t - t_o). \quad (19)$$

Here, t_o is the time at which the loading or unloading process is initiated, and $L(0) = L_o$. Knowing $\phi_s(t)$, then $f(t)$ is given by Eq. (14). Equation (5) can then be solved directly:

$$\tilde{\phi}_s(t) = \tilde{\phi}_s(t_o) e^{-\frac{t-t_o}{\tilde{\mu}}} + \frac{1}{\tilde{\mu}} \int_{t_o}^t f(t') e^{-\frac{t-t'}{\tilde{\mu}}} dt', \quad (20)$$

where $\tilde{\phi}_s(0) = \phi_{so}$, and $\tilde{\mu} = 0.5 \text{ s}$. With both $\phi_s(t)$ and $\tilde{\phi}_s(t)$ known, Eq. (13) gives $\beta_s(t)$.

Shown in Fig. 1 are comparisons of the predicted and experimental histories for ϕ_s and β_s ; good agreement exist. It is noted that the model predicts experimentally observed stress relaxation, indicated by the small decrease in β_s with time immediately following cessation of loading.

Figure 2 gives a plot of the predicted result in the $\beta_s - \ln \phi_s$ phase plane. The entire area under the loading curve is directly proportional to the total work per unit mass, w_T , required to compact the sample from the initial unloaded state to the final loaded state. Using Eqs. (1) and (3), and assuming an incompressible solid, it can be shown that w_T is given by:

$$w_T = \frac{1}{\rho_s} \int_{\ln \phi_{so}}^{\ln \phi_s} \beta_s d(\ln \phi'_s)$$

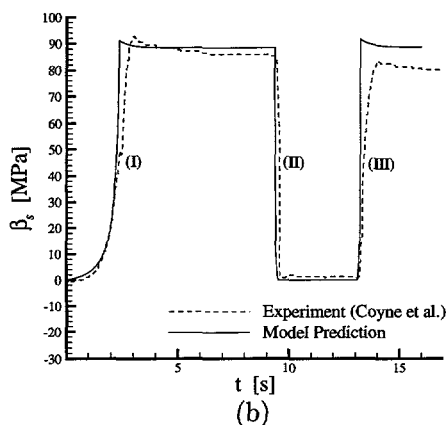
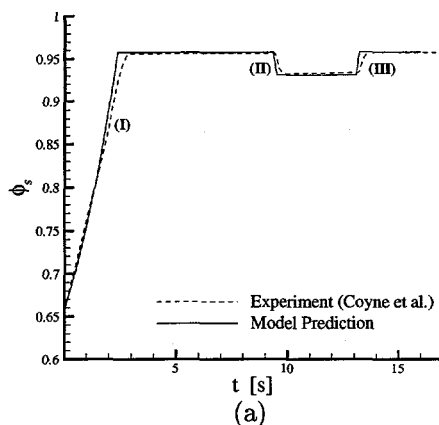


FIGURE 1. Predicted and experimental histories for (a) solid volume fraction and (b) intergranular stress.

$$= \frac{1}{\rho_s} \int_0^{\phi_s - \bar{\phi}_s} \frac{\beta_s}{\phi_s} d(\phi'_s - \bar{\phi}'_s) + \frac{1}{\rho_s} \int_{\phi_{s0}}^{\bar{\phi}_s} \frac{\beta_s}{\phi_s} d\bar{\phi}'_s. \quad (21)$$

Here, the total work consists of elastic and dissipative energy components, given by the first and second terms in the latter expression of Eq. (21), respectively. Only the elastic energy is recovered upon unloading of the sample; consequently, the sample does not uncompact to its initial solid volume fraction due to dissipated mechanical energy. In this figure, the shaded area is proportional to the dissipated energy associated with the initial loading cycle. Most conventional two-phase DDT models do not properly account for this dissipated energy, and thus do not accu-

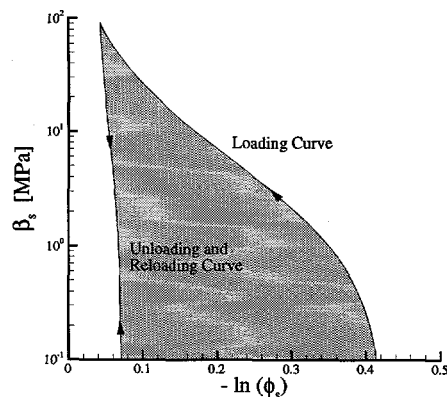


FIGURE 2. Predicted compaction work.

rately model compaction energetics.

FUTURE WORK

This paper has outlined the modification of a two-phase DDT model to account for energy dissipation induced by quasi-static compaction. Future work will address 1) energy dissipation induced by dynamic compaction, and 2) localization of the dissipated energy to form hot-spots. The long term goal of this work is to increase the predictability of compaction-initiated combustion in damaged energetic materials.

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