A new concept for the modeling of detonation waves in multiphase mixtures


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This paper addresses two key issues of detonation wave modelling in hydrocodes. The first issues deals with the computation of the multiphase mixture variables. We develop a new model that deals accurately with non-equilibrium multiphase mixtures. The second issue lies in the computation of thermodynamic variables in so called mixture cells when using Eulerian hydrocodes. We show that the new multiphase model is able to deal with mixture cells resulting of numerical smearing of interfaces as well as physical mixtures, which result of explosive decomposition.

INTRODUCTION

A new numerical concept is presented to deal with two basic problems encountered in the computation of detonation waves in condensed energetic materials.

• The first problem is due to the multiphase feature of the condensed energetic material involving several compressible materials. The Euler equations (plus several species conservation equations) do not provide enough thermodynamic information to determine the mixture pressure and sound speed. The mixture EOS is most times very inaccurate.1,2,3.

• The second problem is related to the numerical treatment of material interfaces. Our numerical strategy adopts full eulerian methods without front tracking, Level Set or interface reconstruction. But it is well known that Eulerian methods create artificial mixtures due to numerical diffusion at the interfaces.1.

In a recent paper 4, some difficulties related to multiphase approaches have been cleared. The key idea is to discretize the multiphase mixture at the microscopic level and then to average the discrete equations. It provides a new discrete model as well as the numerical method. This is done in the opposite way of what was done before. Indeed, it was conventional to obtain a system of PDEs on the basis of averaging procedures, and then to discretize the corresponding PDE system. Nevertheless this method needs a lot of extensions to deal with the applications of confined multiphase detonations:

* The materials are reactive.
* The equations of state are complex.
* The mixture behaves essentially with a single velocity and pressure.2 It is then necessary to adopt a description of the mixture that relaxes intrinsically towards pressure and velocity equilibrium between all the phases.
* The mixture involves an arbitrary number of phases.
* The geometries are 2D axisymmetric.

In this paper, the reactive multiphase model is first written in a conventional continuous form. Then the basis of the new method is developed. It is then extended to account for the various relaxation effects. It is then validated upon a set of illustrative computations.

CONTINUOUS MODEL

In this section we recall a BN type 1,2,3,5 continuous multiphase model used for detonation waves computations.
Each pure phase $k$ is assumed to obey the reactive Euler equations. The mixture is composed of $N$ phases ($k \in [1, N]$). Each phase $k$ is composed of $N_k$ chemical species and the $m_k$ species is defined by its mass fraction $Y_{m,k}$.

A conventional averaging method provides the following system.

\[
\begin{align*}
\frac{\partial}{\partial t} \alpha_k + \vec{u_i} \cdot \nabla \alpha_k &= \dot{\alpha}_k + \dot{m}_k \\
\frac{\partial}{\partial t} (ap)_k + \text{div}(ap\vec{u})_k &= \dot{m}_k \\
\frac{\partial}{\partial t} (ap\vec{u})_k + \text{div}(ap\vec{u} \otimes \vec{u})_k + \nabla(ap)_k &= p_l \vec{u_i} \cdot \nabla \alpha_k + \dot{\vec{F}}_k + \dot{m}_k \vec{u_i} \\
\frac{\partial}{\partial t} (ap\rho)_k + \text{div}(ap\rho \vec{u})_k &= p_l \vec{u_i} \cdot \nabla \rho_k + \rho \vec{F}_k - \rho_l \dot{\vec{F}}_k + \dot{m}_k \vec{u_i} \\
\frac{\partial}{\partial t} (apY_m)_k + \text{div}(apY_m\vec{u})_k &= \dot{m}_k \\
\end{align*}
\]

Standard notations of the multiphase literature are used. The index $i$ refers to interfacial quantities which need to be modeled in order to close the system. The modeling of these interfacial quantities remains a difficult fundamental issue. The determination of the drag force ($\vec{F}_k$), pressure relaxation rate ($\dot{\alpha}_k$) is another difficulty. The numerical approximation of these equations is also difficult due to the presence of non-conservative terms and fluxes involving many elementary waves that render the system difficult to upwind. The new approach, developed in the next section solves these difficulties. A consequence is that numerical mixtures, resulting of the explosive decomposition and artificial smearing of interfaces will be computed accurately.

**THE DISCRETE EQUATIONS METHOD (DEM)**

A numerical model for multiphase flows that relaxes intrinsically towards mechanical equilibrium (velocities and pressures equilibrium), is developed in this section.

The DEM considers the multiphase mixture at the microscopic/grain scale. At this scale, the interactions between phases are given by the solution of Riemann problems between two fluids governed by the reactive Euler equations. Thus, microscale interface problems are solved with the help of the Godunov scheme. The solution of these interface problems are averaged over the control volume and provide macroscopic discrete equations of the mixture.

For the sake of simplicity we first consider that mass transfer is absent. The multiphase control volume corresponds to the macroscopic computational cell $C_i$. It contains $N$ phases. The pure fluid equations are multiplied by an indicator function $X_k$ to select the appropriate fluid. In the computational cell $C_i$, between the time 0 and $\Delta t$, the evolution of the variables of the phase $F_k$ obey the following equation:

\[
\int_0^\infty \int_{C_i} X_k \left( \frac{\partial W}{\partial t} + \frac{\partial F}{\partial x} \right) dx dt = 0 \tag{1}
\]

where $W=(1, \rho, \rho u, \rho E, \rho Y_m)$ and $F(W)=(0, \rho u, \rho u^2 + p, \rho E u + p u, \rho Y_m)$. The first equation is a trivial identity. The indicator function obeys the topologic equation: $\frac{\partial X_k}{\partial t} + \sigma \frac{\partial X_k}{\partial x} = 0$

where $\sigma$ represents the local interface velocity $v_k$.

After some algebraic manipulations equation (1) becomes:

\[
\int_0^\infty \int_{C_i} \left( \frac{\partial (X_k W)}{\partial t} + \frac{\partial (X_k F)}{\partial x} \right) dx dt = \int_0^\infty \int_{C_i} (F - \sigma W) \frac{\partial X_k}{\partial x} dx dt
\]

We have to evaluate these three integrals.

\[
I_{\text{temp},i} + I_{\text{conv},i} = I_{\text{log},i}
\]

where
The domain \( D_{k_0} \) randomly contains the fluid \( \mathcal{F}_{k_0} \) for \( k_0 \in [1,N] \); the domain \( D_{k_i} \) randomly contains the fluid \( \mathcal{F}_{k_i} \) for \( k_i \in [1,N] \) and so forth. The fluids in the domains \( D_{k_j} \) and \( D_{k_{j+1}} \) lie on both sides of the \( j^{th} \) interface, \( j=0..M+1 \). In the domain \( D_{k_0} \), the fluid \( \mathcal{F}_{k_0} \) may come from the \( C_{j-1} \) computational cell. In the domain \( D_{k_{M+1}} \), the fluid \( \mathcal{F}_{k_{M+1}} \) may come from the \( C_{j+1} \) computational cell. This defines a random configuration \( \mathcal{P}(k_0,\ldots,k_{M+1}) \) of the considered topology in cell \( C_i \).

For computational purposes, we define a numerical projection of the microscopic variables \( x_i W \). This numerical projection is valid for any random configuration (event):

\[
((XW)_i^{n+1})(\text{event}) = \frac{1}{\Delta x} \int_{C_i} (XW)(\text{event})(x,t^{n+1})dx
\]

The macroscopic variables at the end of time step \( n+1 \) are given by:

\[
(xW)_i^{n+1} = \sum_{\text{event}} P(\text{event})*((XW)(\text{event}))
\]

where \( P(\text{event}) \) is the probability of event. For any configuration, a shock-capturing scheme is considered for the variables \( ((XW)_i^{n+1})(\mathcal{P}(k_0,\ldots,k_{M+1})) \) obtained by an average projection:

\[
((XW)_i^{n+1})(\mathcal{P}(k_0,\ldots,k_{M+1})) = \frac{1}{\Delta x} \int_{C_i} (XW)(\mathcal{P}(k_0,\ldots,k_{M+1}))(x,t^{n+1})dx
\]

The solution for the discrete value of \( (XW)_i \) requires the calculation of the three integrals defined previously: \( I_{\text{temp},i} \), \( I_{\text{conv},i} \), and \( I_{\text{lag},i} \).

**Approximation of the temporal terms**

\[
I_{\text{temp},i} = \Delta x \left( X_i W_i \right)_i^{n+1} + \sum_{j=0}^{M-1} \left( (X_{i,j})_i W_{i,j}^u (x_{i,j} - x_i) \right)
\]

where \( (X_{i,j})_i = \delta_{i,j} \), with \( \delta_{i,j} \) the Kronecker symbol.

**Approximation of the convective flux**

\[
I_{\text{conv},i} = \int \left( \sigma \frac{\partial (X_i W_i)}{\partial t} \right) dx dt + \sum_{j=0}^{M-1} \int \left( \sigma \frac{\partial (X_i W_i)}{\partial x} \right) dx dt
\]

\[
I_{\text{lag},i} = \int \left( \sigma \frac{\partial \sigma}{\partial t} \right) dx dt + \sum_{j=0}^{M-1} \int \left( \sigma \frac{\partial \sigma}{\partial x} \right) dx dt
\]
where

\[
Lagrangian \text{ flux is given by }
\]

\[
\text{The convective flux is of the form: }
\]

\[
\text{and the interface is constant across the interface and } \sigma \text{ is function of velocity and mass transfer of the phase across the interface and } \text{the sign of the velocity of the interface } B .
\]

\[
(F - \sigma W)^{i+1/2}_{j+1/2} \left[ X_k \right]^{i+1/2}_{j+1/2} =
\]

\[
(F - \sigma W)^{i+1/2}_{j+1/2} (F_{p}, F_{q}) [(X_{j})_{k} - (X_{j+1})_{k}] =
\]

\[
\text{The numerical scheme reads}
\]

\[
\Delta t \left( X_{j+1} W_{j}^{m} \right) =
\]

\[
\text{The other Lagrangian fluxes are computed in the same way. Thus, for a specific event } \mathcal{P}(k_{i0}, \ldots, k_{M+1}), \text{ the}
\]

\[
\text{numerical scheme reads}
\]

\[
\sum_{k_{i0}}^{M} \left( X_{j+1} W_{j}^{m} \right) =
\]

\[
\text{The numerical scheme for the microscopic variables of a particular event has been developed. It must be averaged in a proper manner in order to get the numerical scheme for the macroscopic variables. Each numerical scheme is multiplied by the discrete probability } \mathcal{P}(k_{i0}, \ldots, k_{M+1}) \text{ of the particular event:}
\]

\[
\mathcal{P}(k_{i0}, \ldots, k_{M+1}) =
\]

\[
\text{In this definition, the notation of the event and its probability are the same for the sake of simplicity in the notations.}
\]

\[
\text{The numerical multiphase model is finally developed. It must be averaged in a proper manner in order to get the numerical}
\]

\[
\text{scheme for the macroscopic variables. Each numerical scheme is multiplied by the discrete probability } \mathcal{P}(k_{i0}, \ldots, k_{M+1}) \text{ of the}
\]

\[
\text{particular event:}
\]

\[
\mathcal{P}(k_{i0}, \ldots, k_{M+1}) =
\]

\[
\text{In the following, we note:}
\]

\[
\sum_{k_{i0}}^{M} \mathcal{P}(k_{i0}, \ldots, k_{M+1}) =
\]

\[
\text{and}
\]

\[
P_{k_{i0}k_{j0}} = P(k_{i0}, k_{j0}) = \alpha_{k_{i0}} \alpha_{k_{j0}} .
\]

\[
\text{Note that}
\]

\[
P_{k_{i0}k_{j0}} = P_{k_{i0}k_{j0}} = \sum_{k_{i0}}^{M} P_{i0,k_{j0}}
\]

\[
\text{for all } k_{i} \leq k_{j} \leq k_{j} . \text{ Note also that}
\]

\[
\sum_{k_{i0}}^{M} P_{i0,k_{j0}} = 1 .
\]

\[
\text{After some algebraic manipulations of the temporal term we get:}
\]
The numerical scheme thus reads:

\[
(aW)_i^0 = (aW)_i^1 - \frac{\Delta t}{\Delta x} \left( I_{\text{conv},i+1/2}^D - I_{\text{conv},i-1/2}^D \right)
\]

Because \( \mathcal{F}_{ij} \) belongs to \( C_i \) and \( \mathcal{F}_{M+1,j} \) belongs to \( C_{i+1} \), the convective flux \( I_{\text{conv},i+1/2}^D \) can be written in a more convenient way:
Let us split the contribution of these Lagrangian fluxes from the convective and Lagrangian fluxes of the boundaries. Their contribution to the evolution of conservative variables is given by the algebraic equation:

\[ \Delta x \left\{ (aw)_{i+1/2} - (aw)_{i-1/2} \right\} = \Delta t I_{xxj}^m \]

For a two-phase flow, the corresponding semi-discrete equations associated to fluid 1 are:

\[ \frac{\partial}{\partial t} \alpha_i = \mu(p_2 - p_1) \]

\[ \frac{\partial}{\partial t} (\alpha p_i) = 0 \]

\[ \frac{\partial}{\partial t} (\alpha p E_i) = \lambda V_i (u_2 - u_1) - \mu P_i (p_2 - p_1) \]

When the number of internal interfaces tends to infinity ( \( M \to \infty \)), it can be easily shown that the various differences of Lagrangian fluxes of the Riemann problem for the Euler equations provide the preceding system where \( \lambda \) and \( \mu \) tend to infinity. It means that the pressures and the velocities relax instantaneously. This result can be generalized for \( N \) phases.

**NUMERICAL STRATEGY**

The numerical model stiffens as the number of internal interfaces increases. Indeed, the direct resolution of the numerical model would require infinitely small time step. We propose a numerical strategy to get free from this constraint.

The scheme is be written in the following form

\[
\frac{\partial(aw)}{\partial t} + \text{Transport}(aw) = 0
\]

It is followed by the Internal step:

\[
\frac{\partial(aw)}{\partial t} + \text{Internal}(aw) = 0
\]

The scheme is extended to second order by using a classical MUSCL strategy. A special care is brought for the extension to second order of the various non conservative terms.

The extension to 2D configurations is obtained by using a dimensional splitting. The source terms involving chemical reactions, mass transfer or other phenomena that are not included in the Lagrangian fluxes, are integrated after a complete time step by using a well adapted ODE solver for stiff equations.

**RESULTS**

**Detonation reaction zone**

In this test, the solution of the multiphase model is compared to the exact solution for a steady detonation wave. A shock wave is transmitted to an explosive by a piston with a given velocity.

We compute the shock to detonation transition and the stabilisation of the detonation wave. The solid and the gaseous phase (detonation products) obey the same equation of state : \( p = (\gamma - 1) p \rho e \), with a polytropic coefficient \( \gamma \) equal to 3.

This calculation corresponds to the ZND model solution and the conditions used here are those given in Fickett and Davis.

In this specific case, the source terms representing the mass transfer terms for the gaseous products are:

\[
\text{Mass}_{\text{GAS,PROD}} = -\left( m_{\text{exp,.flat}}, m_{\text{gus,flat}}, m_{\text{exph},\text{exp}}, m_{\text{exp},\text{exp}} (E_{\text{exp}} + q_{\text{react}}) \right)
\]

The term \( q_{\text{react}} \) corresponds to the heat of combustion ( \( q_{\text{react}} = 4.5156 \text{MJ/kg} \)). The mass transfer is given by

\[
m_{\text{exp}} = -k \left[ (\rho_m \alpha_{\text{exp}} p_{\text{exp}}) \right] \text{ with } k = 2.10^6 \text{ s}^{-1}
\]

and \( \rho_m = \sum \alpha_k \rho_k \) is the mixture density.
Initially, the pressure is set at 1 bar and the densities of the both phases are equal to 1600 kg/m³. The initial volume fraction of the gaseous product is \( \alpha = 10^{-6} \). The piston velocity is 1000 m/s. The total length of the domain is 0.3 m. 1000 cells are used for the mesh, with a constant length (3.3 cells/mm).

In Table 1, relevant analytical values are reported. They are compared with the numerical results.

<table>
<thead>
<tr>
<th></th>
<th>Pressure (10^8 Pa)</th>
<th>Density (kg/m³)</th>
<th>Velocity (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CJ Point</td>
<td>289</td>
<td>2133</td>
<td>2125</td>
</tr>
<tr>
<td>Neumann spike</td>
<td>578</td>
<td>3200</td>
<td>4250</td>
</tr>
</tbody>
</table>

Table 1: Characteristic values of the ZND model for detonation wave

The numerical velocity of the detonation zone and the reaction zone thickness are in very good agreement with the analytical solution (Table 2).

<table>
<thead>
<tr>
<th></th>
<th>Analytical</th>
<th>Numerical</th>
</tr>
</thead>
<tbody>
<tr>
<td>Velocity of the detonation front</td>
<td>8500 m/s</td>
<td>8486 m/s</td>
</tr>
<tr>
<td>Length of the reaction zone</td>
<td>5.31 \times 10^{-3} m</td>
<td>5.2 \times 10^{-3} m</td>
</tr>
</tbody>
</table>

Table 2: Comparison between ZND characteristic values and numerical characteristics values

The numerical velocity of the detonation zone and the reaction zone thickness are in very good agreement with the analytical solution (Table 2).

In Figure 2, the numerical pressure is plotted with solid lines while the analytical solution in the reaction zone is plotted with symbols. The Neumann spike and the Chapman-Jouguet point are well reproduced by the numerical model. The numerical solution converges to the exact one in the reaction zone. The density and of course the temperature of the gaseous product is not equal to the ones of the explosive. This obvious remark is however a proof that models based on the Euler equations and mixture equations of state based on thermal equilibrium are wrong for this type of application.

Detonation of a condensed CHNO-PA-Al non ideal energetic material

The multiphase model is used to calculate the detonation properties of a condensed energetic material. For a highly sub-oxygenated material, the chemical decomposition is given by:

1. \( \text{CaH}_b\text{N}_c\text{O}_d \rightarrow d\text{H}_2\text{O} + (b/2-d)\text{H}_2 + c/2\text{N}_2 + a\text{C} \)
2. \( \text{Al} + \text{G} \rightarrow \text{Al}_2\text{O}_3 + \text{C} + \text{G}' \)
3. \( \text{NH}_4\text{ClO}_4 \rightarrow 1.5 \text{H}_2\text{O} + 0.5 \text{N}_2 + 1.25 \text{O}_2 + \text{HCl} \)
4. \( \text{C} + \text{O}_2 \rightarrow \text{CO}_2 \)
5. \( \text{H}_2 + 0.5 \text{O}_2 \rightarrow \text{H}_2\text{O} \)

A piston impacts the reactive material with the velocity of 400 m/s. The overall computational domain (one meter long) is at the initial pressure of 1 bar. Both phases are at rest. We have chosen the initial gaseous composition: \( Y_{H_2O} = 1, Y_i = 0 \) for the other gases with an initial volume fraction of 10⁻⁵.

We compute the shock to detonation transition and the stabilisation of the detonation wave. The spatial resolution is one cell per millimetre. The reactive material is governed by a multi-component Mie-Gruneisen equation of state whereas the detonation gaseous products are
governed by a multi-component H9 equation of state. All the chemical decomposition are based on a Vieille’s law. The different rates of change of the species concentrations \([X_i]\) are modelled by:

\[
[X_i] = \sum_{j=1}^{N_m} \frac{V_a - V_d}{\tau_i} \left( \prod_{i=1}^{N_r} [X_i]^{\eta_i} \right)
\]

\(\tau_i\) is a function of the relaxed pressure:

\[
\frac{1}{\tau_i} = A_i \left( \frac{p}{p_o} \right)^n, \quad n=2.
\]

\(p_o\) is a reference pressure. The various rates of change of the mass fractions \(Y_i\) are modelled by:

\[
m_i = \sum_{j=1}^{N_m} \frac{V_a - V_d}{\tau_i} \left( \prod_{i=1}^{N_r} (\alpha p) Y_{i_j}^{\eta_i} \prod_{i=1}^{N_r} (\alpha p) Y_{i_k}^{\eta_i} \right)
\]

After integrating the whole system of equations in a 1D code, the 1D numerical simulation represents a detonation wave in a section of infinite area. The numerical results are shown hereafter. A magnified view of the variables inside the reaction zone is shown on Figure 3: Two characteristic time scales are visible for the chemical decomposition condensed material (3a). The decomposition of the organic molecule CHNO is the fastest. Its decomposition releases part of the energy contained into the solid. Without this initial released energy, the other highly energetic components of the solid (Ammonium Perchlorate PA and Aluminium Al) cannot decompose. The PA decomposes totally but there is not enough oxygen in the reactive mixture to allow the Al to burn totally. There is still energy that can be released in the system.

\[\text{Figure 3: Solid components mass fractions:}\]
\[\text{a): CHNO (circles), PA (triangles), Al (squares)}\]
\[\text{b): Al}_2\text{O}_3 \text{ (circles), C (squares)}\]

Gas components mass fractions:
\[\text{c): CO}_2 \text{ (circles), H}_2\text{O (triangles), O}_2 \text{ (squares)}\]
\[\text{d): N}_2 \text{ (circles), HCl (triangles), H}_2 \text{ (squares)}\]

The combustion of both PA and Al has thickened the reaction zone because the rates of their decomposition are lower. The condensed material volume is about 30 % (cf. solid and gas volume fractions in Figure 4) even far from the reaction zone.
Two-dimensional detonation simulation

In this section a two-dimensional simulation is presented. We simulate a projectile high-velocity impact (1930 m.s⁻¹) onto a tank filled with a reactive liquid (Nitromethane). The transmitted shock wave elevates the liquid temperature and pressure. This pressure and temperature elevation initiates the chemical decomposition of the liquid explosive. The shock transits to a detonation. The increase in pressure will put into motion the different walls of the tank. A schematic view of the system is presented in Figure 5.

The chemical decomposition is based on the theory of the state transition. The reactive material and the detonation products obey a Mie-Gruneisen EOS (Cochran-Chan and JWL). The copper obeys a modified stiffened gas EOS. The ideal gas law is used for the surrounding air.

Figure 5: Presentation of the system simulated by a two-dimensional calculation.

The total amount of equations is large: twenty equations –five equations per phase- are solved at each grid point. But the method is able to solve all flow features (physical mixtures inside the reaction zone) as well as material interfaces under a unique mathematical formulation and numerical scheme.

The mixture density is depicted on Figure 6. In Figure 6a the initial mixture density is plotted and one can observe the envelope as well as the projectile. In the next view, the projectile has penetrated the envelope, which is deforming. The initial shock wave is now transmitted to the explosive (6b) in Figure 6c, a curved detonation wave is observed which propagates into the liquid. One can also see in this figure, that a very low-pressure zone is setting in the projectile. This is due to the interaction of two rarefactions wave. The first one comes from the depressurisation behind the projectile because of its motion. The second one is a reflected rarefaction wave induced by the interaction of the shock and the liquid.

In Figure 6d, the detonation wave has reached the upper wall. It is noticeable that the left wall above the projectile has a motion toward the left: it is due to the pressure difference with the atmosphere.

In the last view (6e), the entire explosive is burnt. The upper wall has now a motion that will turn to a complete destruction of the tank.
The new approach proposed in the present paper is able to solve accurately very different problems like physical multiphase mixtures as well as material interfaces and artificial mixtures, produced by numerical diffusion zones of contact discontinuities. It has been extended to the resolution of turbulence problems in plasma physics and phase transition fronts propagation in superheated liquids. We believe that this new approach may have important applications in several problems of physical, mathematical and industrial importance.

REFERENCES
8 Fickett W. and Davis W.C., 1979, Detonation, Univ. of California Press.