

# An Application of Lagrangian Equations Coupled with Detailed Chemical Kinetics to the Simple Prediction of Transient Reaction Front Propagation

Je Ir Ryu

Department of Mechanical Engineering, University of California - Berkeley  
Berkeley, CA 94720, U.S.A.

## 1 Introduction

Detonation propagation properties, such as detonation speed, leading shock pressure, etc., are well predicted by the theory. However, detonation initiation and transition in an inhomogeneous mixture are not accurately estimated by a theory (e.g., Zel'dovich reactivity gradient) due to the transient effect and complex chemistry. The theory can only estimate the initiation condition roughly and may not be valid for a reactant with a long induction time [1].

On the other hand, numerical simulations of reaction front propagation with detailed chemical kinetics using sophisticated research codes (e.g., A-SURF [2]) have been carried out actively to investigate fundamentals of detonation initiation or deflagration to detonation transition (DDT) [1,3–5]. However, these simulations are computationally expensive and slow, because capturing shock front and stiff chemical reaction needs very small time step and grid size. Typically, the time step and grid size should be smaller than 1 ns and 10  $\mu\text{m}$  respectively, to achieve solutions with high fidelity. Finding a condition for the detonation initiation of an inhomogeneous mixture from these numerical simulations is time-consuming and empirical, so the less expensive method to find such a condition will be beneficial.

Utilizing Lagrangian equations may be a potential solution to accurately predict detonation initiation phenomena without expensive computational cost. Although the Lagrangian technique with simple Arrhenius chemistry (e.g., one-step, two-step) has been used for ages (e.g., [6]), the Lagrangian solver with detailed chemical kinetics has not been utilized actively. Using simple Arrhenius chemistry may have a limitation for the study of reaction front propagation modes, especially with a reactant that has complex chemistry behavior such as in the negative temperature coefficient region.

Therefore, a potential application of Lagrangian finite difference equations coupled with detailed chemical kinetics is discussed in the current study for the less expensive prediction method of transient reaction front propagation with complex chemistry behavior in advance of running expensive sophisticated simulations. In this work, governing equations, numerical schemes, and the solving procedure for Lagrangian finite difference equations are presented. Then, some examples and code validation are reported.

## 2 Methods

A simpler solver for trial simulations called the Solver for Lagrangian Equations of Deflagration/Detonation (SLED) was developed to investigate the potential application of Lagrangian equations coupled with detailed chemical kinetics for the simple prediction of transient reaction front propagation. SLED solves the Lagrangian finite difference equations for reactive compressible flow in one-dimensional spherical domain. The governing equations of a one-dimensional fluid motion using the time-independent Lagrangian invariant [7] are

$$u = \frac{\partial x}{\partial t}, \quad (1)$$

$$\frac{1}{\rho} = \frac{\partial x}{\partial M}, \quad (2)$$

$$\frac{\partial u}{\partial t} = -\frac{\partial P}{\partial M}, \quad (3)$$

$$\frac{\partial e}{\partial t} + P \frac{\partial(1/\rho)}{\partial t} = 0, \quad (4)$$

$$P = e\rho(\gamma - 1), \quad (5)$$

where  $u$  is the fluid velocity,  $x$  is the location in planar domain,  $t$  is the time,  $\rho$  is the density,  $M$  is the Lagrangian invariant mass,  $P$  is the pressure, and  $e$  is the internal energy per unit mass.

These equations were modified to solve spherical reactive wave propagation problem, and the finite difference forms are

$$\frac{r_l^{n+1} - r_l^n}{\Delta t^{n+\frac{1}{2}}} = u_l^{n+\frac{1}{2}}, \quad (6)$$

$$\Delta M_l = \rho_{l+\frac{1}{2}}^{n+1} \frac{4\pi}{3} \left( r_{l+1}^{n+1^3} - r_l^{n+1^3} \right), \quad (7)$$

$$\frac{u_l^{n+\frac{1}{2}} - u_l^{n-\frac{1}{2}}}{\Delta t^{n+\frac{1}{2}}} = -4\pi r_l^2 \left( \frac{P_{l+\frac{1}{2}}^n + A_{l+\frac{1}{2}}^{n-\frac{1}{2}} - P_{l-\frac{1}{2}}^n - A_{l-\frac{1}{2}}^{n-\frac{1}{2}}}{\Delta M_l} \right), \quad (8)$$

$$e_{l+\frac{1}{2}}^{n+1} - e_{l+\frac{1}{2}}^n + \left( \frac{P_{l+\frac{1}{2}}^{n+1} + P_{l+\frac{1}{2}}^n}{2} + A_{l+\frac{1}{2}}^{n+\frac{1}{2}} \right) \left( \frac{1}{\rho_{l+\frac{1}{2}}^{n+1}} - \frac{1}{\rho_{l+\frac{1}{2}}^n} \right) = q_{l+\frac{1}{2}}^{n+\frac{1}{2}}, \quad (9)$$

$$P_{l+\frac{1}{2}}^{n+1} = e_{l+\frac{1}{2}}^{n+1} \rho_{l+\frac{1}{2}}^{n+1} \left( \gamma_{l+\frac{1}{2}}^{n+1} - 1 \right), \quad (10)$$

where  $r$  is the location in the spherical domain,  $\Delta t$  is the size of time step,  $A$  is the artificial viscosity term,  $\Delta M$  is the Lagrangian invariant mass of a fluid element,  $q$  is the heat release rate per unit mass, the subscript  $l$  denotes the mesh point, and the superscript  $n$  denotes time step. Note that staggered mesh points are represented using  $\frac{1}{2}$  in subscript and superscript.

The initial values of the Lagrangian invariant mass of each fluid element are time-independent, so the size of each mesh changes every time step to keep the value of the Lagrangian invariant mass constant. The high pressure zone has finer meshes, while meshes in the low pressure zone are coarse. This approach is beneficial in the compressible wave propagation problem, since the high pressure zone, i.e., shock area, is of interest. By the Lagrangian finite difference approach, the problem can be solved with a smaller number of meshes.

Equations 8 and 9 include the artificial viscosity term [7]. The term is added to the pressure terms of the equations for the artificial dissipation to resolve the stiff shock front. It smooths out the pressure cliff to solve the equations numerically. Typically,  $A$  is a function of  $u$ ,  $\rho$ , and  $a$ , including quadratic and linear terms as:

$$A_{l+\frac{1}{2}}^{n+\frac{1}{2}} = \frac{c_0^2 \left( u_{l+1}^{n+\frac{1}{2}} - u_l^{n+\frac{1}{2}} \right)^2}{\frac{1}{2} \left( \frac{1}{\rho_{l+\frac{1}{2}}^{n+1}} + \frac{1}{\rho_{l+\frac{1}{2}}^n} \right)} - \frac{c_1 \left( u_{l+1}^{n+\frac{1}{2}} - u_l^{n+\frac{1}{2}} \right)}{\frac{1}{2} \left( \frac{1}{\rho_{l+\frac{1}{2}}^{n+1}} + \frac{1}{\rho_{l+\frac{1}{2}}^n} \right)} a_{l+\frac{1}{2}}^{n+1}, \quad (11)$$

where  $c_0$  and  $c_1$  are constants [7, 8]. The artificial viscosity term is only applied for the meshes that  $\frac{\partial u}{\partial r} < 0$ . In Equation 11, the sound speed,  $a$ , can be calculated using the value of  $A$  in the previous time step as

$$a_{l+\frac{1}{2}}^{n+1} = \gamma_{l+\frac{1}{2}}^{n+1} \sqrt{\frac{P_{l+\frac{1}{2}}^{n+1} + A_{l+\frac{1}{2}}^{n-\frac{1}{2}}}{\rho_{l+\frac{1}{2}}^{n+1}}}. \quad (12)$$

For examples and code validation,  $c_0^2 = 1$  and  $c_1 = 0.5$  were used.

SLED utilizes the CHEMKIN [9] subroutine libraries to load thermodynamic data and calculate chemical properties from the chemical kinetic models. To resolve the stiff chemistry term, the adaptive time refinement method was employed based on the mesh locations and mass fractions of species obtained.

It was performed successfully and produced reasonable results with a much shorter calculation time than the sophisticated research code.

### 3 Results and Discussions

Several examples were calculated using SLED. A reaction propagation wave was initiated from the temperature gradient of the center hot spot, i.e., a zone at a higher temperature than the surrounding. The mixtures at different temperatures have different auto-ignition delay times, and the auto-ignition delay time is exponentially decreased as temperature increases. Thus, the initial temperature gradient can produce reaction front propagation from the center to the outside of the hot spot. The hot spot works as an ignition source.

In these examples, the radius for the one-dimensional spherical domain is 10 cm. An isotropic flow is assumed and the reflective boundary condition at the center was applied. The code can solve for both open and closed boundary conditions at the outer boundary, and the open boundary condition was used here. Outside of the hot spot, the temperature is uniformly 1180 K, and the pressure is 50 atm everywhere. Initially, the size of every cell is 0.2 mm. The initial mixture is a stoichiometric 50% $H_2$ -50% $CO$ /air mixture, so the initial mole fractions are  $X_{CO} = 0.148$ ,  $X_{H_2} = 0.148$ ,  $X_{O_2} = 0.148$ ,  $X_{N_2} = 0.556$ . The size of the time step is initially 2.6 ns, and it adaptively reduces to the order of 0.01 ns for stability when the ignition happens. Several cases with different maximum hot spot temperatures were solved to capture different modes of reaction front propagation. The temperature is maximum at the center of the hot spot and linearly decreases to the outer temperature. The size of the hot spot is 5 mm for all cases. For chemistry, 13 species skeletal mechanism for 50% $H_2$ -50% $CO$ /air mixture from GRI-Mech 3.0 [10] was used.

Figure 1 shows the calculation result from the hot spot with the center temperature of 1183 K. The temperature difference between the center and the outside of the hot spot,  $\Delta T$ , was 3 K. Due to the relatively low temperature gradient, the autoignition delay times at neighboring meshes in the hot spot were similar. Thus, the reaction front propagation speed was very fast, and the reaction wave was ahead

of the pressure wave. The center of the hot spot started to ignite at  $\sim 65 \mu\text{s}$ , and the reaction front propagated very fast until  $\sim 67 \mu\text{s}$ . At about  $67 \mu\text{s}$ , autoignition occurred simultaneously in the unburnt mixture, and the reaction front propagation became a thermal explosion without detonation initiation.

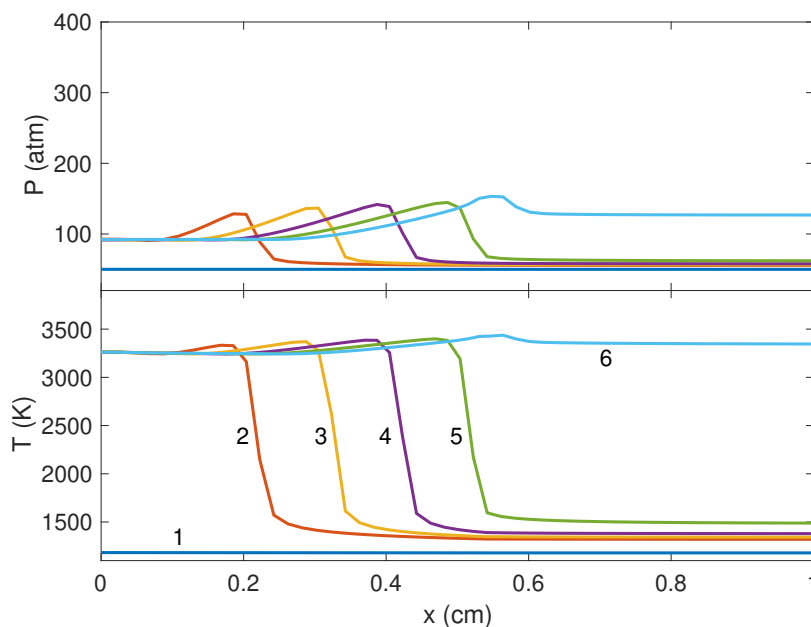


Figure 1: Deflagration and thermal explosion from SLED calculation with  $\Delta T = 3 \text{ K}$ . Time sequences 1:  $0.00 \mu\text{s}$ , 2:  $65.52 \mu\text{s}$ , 3:  $65.99 \mu\text{s}$ , 4:  $66.43 \mu\text{s}$ , 5:  $66.84 \mu\text{s}$ , 6:  $67.23 \mu\text{s}$ .

$\Delta T$  was increased to  $20 \text{ K}$  (maximum temperature of  $1200 \text{ K}$ ) for the second example as shown in Figure 2. A detonation propagation was obtained. The reaction front propagation speed and sound speed were on the same order of magnitude, and the resonance of two waves produced the detonation wave [11, 12]. High pressure peaks and temperature cliffs were observed compared to the previous example. As the hot spot temperature was higher than that of the deflagration and thermal explosion case, the hot spot ignited earlier. Thus, the reaction front propagated longer before the simultaneous autoignition. These two results are consistent with the previous simulation work [1].

The code was further validated using the example result shown in Figure 2. A simulation with the same conditions was performed using a more sophisticated research code [2], which are widely used for solving one-dimensional compressible reacting flows. The sophisticated code is an in-house code using a finite-volume method with chemical kinetics and transport models, which requires expensive computational cost (typically on the order of  $1000 \text{ CPU-hour}$ ). The code was utilized successfully in the recent detonation study [3–5], and the details about the code can be found in [2].

The result from the research code is presented in Figure 3. The magnitude of pressure peaks and detonation wave speed were very close to the result from SLED. Even though the result in Figure 3 oscillates less and describes the leading shock front more accurately, the solution from SLED is still reasonable and captures a similar trend in a very short computation time (on the order of  $1 \text{ CPU-hour}$ ).

## 4 Conclusions

A solver for deflagration/detonation simulations, SLED, was developed to investigate a potential application of Lagrangian finite difference equations coupled with detailed chemical kinetics to the simple

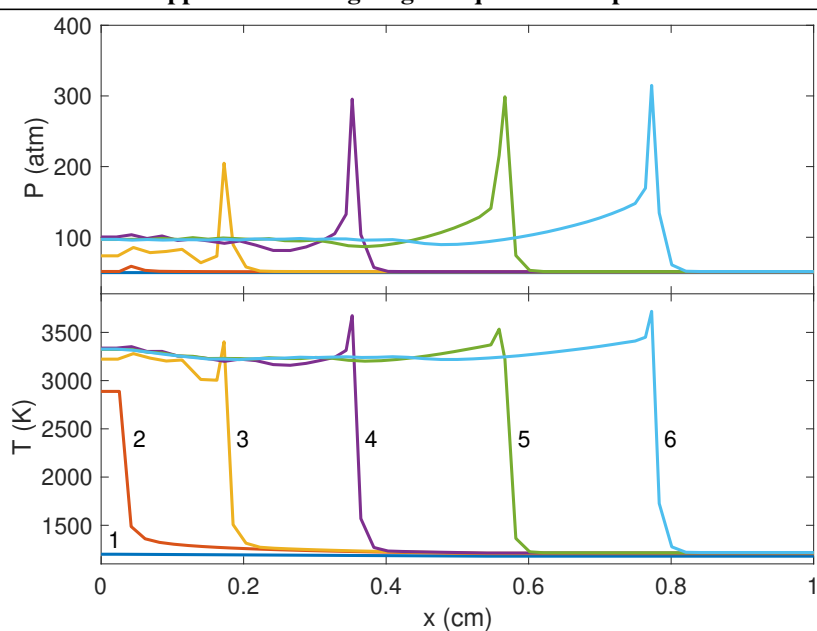


Figure 2: Detonation initiation from SLED calculation with  $\Delta T = 20$  K. Time sequences 1:  $0.00 \mu\text{s}$ , 2:  $50.51 \mu\text{s}$ , 3:  $52.21 \mu\text{s}$ , 4:  $53.37 \mu\text{s}$ , 5:  $54.56 \mu\text{s}$ , 6:  $55.71 \mu\text{s}$ .

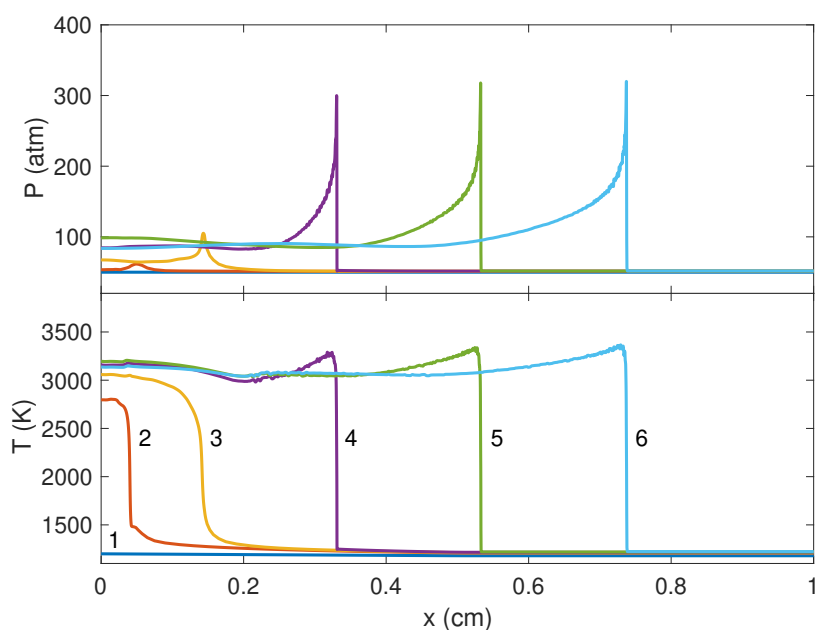


Figure 3: Detonation initiation from sophisticated research code calculation with  $\Delta T = 20$  K. Time sequences 1:  $0.00 \mu\text{s}$ , 2:  $52.34 \mu\text{s}$ , 3:  $53.90 \mu\text{s}$ , 4:  $55.07 \mu\text{s}$ , 5:  $56.25 \mu\text{s}$ , 6:  $57.42 \mu\text{s}$ .

prediction of transient reaction front propagation as a preliminary stage. The time-independent Lagrangian invariant, the artificial viscosity, and the adaptive time refinement method were employed. The solutions from SLED were compared with those from sophisticated research codes, showing that SLED can capture similar detonation wave propagation with complex chemistry behavior in a relatively short computing time. Therefore, it can be utilized as a good trial solver to estimate a primary trend or result before running the sophisticated research code.

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