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The Time-Dependent Reaction Zones
of Ideal Gases, Nitromethane,
and Liquid TNT

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Charles L. Mader

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THE TIME-DEPENDENT REACTION ZONES OF IDEAL GASES,
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Charles L. Mader

ABSTRACT

The time-dependent reaction zones of ideal gases, nitromethane, and liquid TNT have been investigated using two-dimensional Lagrangian and Eulerian numerical hydrodynamics with Arrhenius chemical reaction and accurate equations of state.

The unperturbed two-dimensional Lagrangian and Eulerian calculations yield the same results regarding the stability of the steady-state solutions of the reaction zone as did previously reported calculations using one-dimensional Lagrangian and characteristic methods. The perturbed two-dimensional Lagrangian and Eulerian calculations reproduce the analytical predictions of Erpenbeck regarding the stability of the ideal gas reaction zone in two dimensions and indicate that the stabilities of the reaction zones of nitromethane and liquid TNT are identical in one and two dimensions.

I. INTRODUCTION

The study of the one-dimensional time-dependent reaction zone of ideal gases, nitromethane, and liquid TNT has been extended to two dimensions. The previous study¹ used the one-dimensional Lagrangian reactive hydrodynamic code called SIN.² The chemical reaction obeyed an Arrhenius rate law. The equation of state for nitromethane and liquid TNT was the HOM equation of state¹ which is described for an ideal gas in the Appendix. (A list of the nomenclature used is also provided in the Appendix.) This study uses the two-dimensional Lagrangian reactive hydrodynamic code called 2DL³ and the two-dimensional Eulerian reactive hydrodynamic code called 2DE.⁴ The two methods of numerically solving the hydrodynamics were used to establish, insofar as possible, that the stability results were independent of the method.

The stability of the ideal gas reaction zone has been studied analytically by Erpenbeck⁵ in one and two dimensions, by Fickett⁶ using a one-dimensional characteristic method, and by the author¹ using a one-dimensional Lagrangian method. This report describes an extension of the author's studies of the ideal gas detonation to two-dimensional slab and cylindrical geometry and shows that these studies agree with the predictions of Erpenbeck. Similar studies of the reaction zones of nitromethane and liquid TNT show that the two-dimensional perturbations of these two explosives do not grow, as opposed to those for ideal gases.

II. TWO-DIMENSIONALLY-UNPERTURBED CALCULATIONS

Previous results¹ of the SIN² one-dimensional Lagrangian reactive hydrodynamic code for nitromethane and TNT are summarized in Table I. The amount of overdrive necessary to stabilize the nitromethane

and liquid TNT detonation decreases with decreasing activation energy and is independent of the frequency factor.

As shown in Table I, nitromethane with an activation energy of 40 kcal/mole and a γ of 0.68 was found to be stable at C-J detonation velocity. With a stable detonation, we can study how the reaction zone is formed by constant-velocity pistons that initially shock the unreacted nitromethane to less than the C-J pressure.

The pressures at various positions of the flow as a function of time are shown in Fig. 1 for a 0.21-cm/ μ sec constant-velocity piston. The calculations with a resolved reaction zone show details of the process of shock initiation of nitromethane. The basic features are identical to those of the flow computed with an unresolved reaction zone.⁷ The shocked nitromethane first completely decomposes at the piston and results in a detonation with a "peak pressure" that builds up toward the C-J pressure of the high density shocked nitromethane. The detonation wave overtakes the shock wave, and the pressure at the end of the reaction zone decays toward the piston pressure.

Fig. 1. The shock initiation of nitromethane with a 0.21-cm/ μ sec constant-velocity piston. $E^* = 40$, $Z = 1.27 \times 10^6$, $\gamma = 0.68$, mesh = 4000 A, and reaction zone is resolved.

To study the stability of detonations, one uses a piston that is as close to the steady-state piston as possible so that the one-dimensional perturbations introduced by deviations from the steady state will be of tractable size. The magnitude of the one-dimensional perturbations depends upon the details of the piston profile, the numerical method, and the mesh size used. For a given mesh size, one-dimensional perturbations increase with the method in the order 2DL, SIN, 2DE. While attempts were made, by adjusting the mesh size, to obtain one-dimensional perturbations of approximately the same size in all three methods, this is unimportant to the question of whether the perturbations introduced grew or decayed. As described in Reference 4, to calculate the steady-state reaction zone piston in the 2DE method, it was necessary to use a lower mass fraction of undecomposed explosive, W , than the W of the cell above the piston for the piston's W . This was accomplished by multiplying by a constant called "piston factor."

The shock-front pressure vs time for an ideal gas detonation (see Appendix for details) unstable to one-dimensional perturbations with an $f = D^2/D_{CJ}^2 = 1.6$ (where D is detonation velocity) is shown in Fig. 2 for the various numerical methods. In Fig. 3 the shock front pressure vs time is shown for an ideal gas detonation stable to one-dimensional perturbations with an $f = 2$ for various numerical methods.

TABLE I

SUMMARY OF PREVIOUSLY REPORTED RESULTS

Activation Energy, E^* (kcal/mole)	Frequency Factor, Z	Grüneisen γ	Thickness of Reaction Zone	Result
NITROMETHANE				
53.6	4×10^8	0.68 ^c	2400-A	{stable at 6750 m/sec, unstable at 6650
53.6	4×10^8	1.70	1600-A	{stable at 6750, unstable at 6650
40	1.27×10^6 ^a	0.68	77500-A	stable at 6460 (C-J)
40	1.27×10^6 ^a	1.70	66000-A	{stable at 6650, unstable at 6550
30	2.37×10^4 ^a	1.70	705000-A	stable at 6460 (C-J)
LIQUID TNT				
41.1	2×10^6	1.66 ^c	1.1×10^{-3} cm	{stable at 6600, unstable at 6500
30	3.14×10^4 ^b	1.66	7.3×10^{-3} cm	stable at 6406 (C-J)

^a Z adjusted to give 1.4 μ sec at 1180°K (85 kbar) to agree with shock initiation experimental data.⁷

^b Z adjusted to give 0.7 μ sec at 1227°K (125 kbar) to agree with shock initiation experimental data.⁷

^cFrom the relation

$$\gamma = -\frac{v}{c_p} \left(\frac{\partial p}{\partial v} \right)_s \left(\frac{\partial v}{\partial T} \right)_p$$

evaluated at $P = 1$ atm, $T = 300^\circ\text{K}$.

The SIN results are reproduced from Reference 1, and Fickett's characteristic method results are reproduced from Reference 6.

The shock front pressure vs time for nitromethane detonations with velocities of 0.6850, 0.6650, and 0.6465 cm/ μ sec is shown in Fig. 4. That for liquid TNT with a velocity of 0.6406 cm/ μ sec is shown in Fig. 5.

Since the magnitude of the one-dimensional perturbation introduced into the calculation varies, the shock front pressure vs time is not identical for the various methods. The stability results are related not to the magnitude but only to the growth or decay of the perturbations. The results in

ceeded to study two-dimensionally perturbed detonations.

III. TWO-DIMENSIONALLY-PERTURBED CALCULATIONS

To study the two-dimensionally perturbed flow, we used the two-dimensional Lagrangian code called 2DL³ and the two-dimensional Eulerian code called 2DE.⁴ The perturbation was introduced by increasing the initial density of a center cell near the piston interface. The density was increased by $1.1\rho_0$ and $1.3\rho_0$ without changing the stability results. Perturbations of all wavelengths are introduced by this procedure. In 2DL the viscosity constant was changed over half an order of magnitude without changing the stability results.

Fig. 2. The shock-front pressure vs time for an ideal gas detonation unstable to one-dimensional perturbations, with $f = 1.6$ and $Q = 50$, for various numerical methods.

Figs. 2 through 5 show that growth or decay of the perturbation is independent of the numerical method, and, incidentally, independent of the nature of the perturbation.

One can obtain identical stability results for two-dimensionally unperturbed flow using any of the four methods of numerically solving the reactive hydrodynamics. While this is not sufficient to guarantee that the stability results are independent of the numerical method, it is a necessary result. Encouraged by these results, we now pro-

The 2DL calculations were performed with 50 cells in the X or R direction and 250 cells in the Z direction. The 2DE calculations were performed with 100 cells in the R and Z directions. In Figs. 6 through 13, the bottom of the figure is the piston boundary, and the left side is the axis for cylindrical geometry or the half-slab boundary for slab geometry. The right and top sides are continuum boundaries. The calculations were continued until the perturbation interacted with the boundaries.

Fig. 3. The shock-front pressure vs time for an ideal gas detonation stable to one-dimensional perturbations, with $f = 2$ and $Q = 50$, for various numerical methods.

Fig. 4. The shock-front pressure vs time for nitromethane with various amounts of overdrive, for various numerical methods. $E^* = 53.6$, $Z = 4 \times 10^8$, and $\gamma = 0.68$. The mesh size is given for each method, and the detonation velocity is given in $\text{cm}/\mu\text{sec}$.

Fig. 5. The shock-front pressure vs time for liquid TNT with a C-J velocity of 0.6406 cm/μsec. $E^* = 41.1$, $Z = 2 \times 10^6$, and mesh = 5000 A.

Visual presentation of two-dimensional numerical data is seldom completely satisfactory. Considerable study of the numerical results is usually necessary before one can interpret the visual representation. In the results described below the visual representations have been interpreted with the aid of the numerical results. Those calculations that were stable showed state variables that were constant with radius near the shock front after the wave had run some distance, while the unstable calculations showed state variables that were not constant with radius near the shock front or elsewhere in the flow.

A. Ideal Gas Detonations

Erpenbeck⁵ predicts that an ideal gas detonation with $f = 2$, $E^* = 50$, and $Q = 50$ will be stable to one-dimensional perturbations but unstable to two-dimensional perturbations, and that with $Q = 0.2$ it will be stable for all perturbations.

As an extra check, we ran a perturbed constant-pressure piston without chemical reaction and with a pressure equal to the ideal gas spike pressure for $f = 2$ of 84. The perturbation decayed as expected. The $f = 2$, $Q = 50$ ideal gas detonation that is sta-

ble to one-dimensional perturbations is unstable to two-dimensional perturbations as is shown using 2DL in Figs. 6 and 7, and 2DE in Fig. 8. The $f = 2$, $Q = 0.2$ ideal gas detonation is stable to two-dimensional perturbations as is shown using 2DL in Fig. 9.

Other calculations were performed for $f = 2$, $Q = 1$ (unstable); $f = 53$, $Q = 0.2$ (stable); $f = 35$, $Q = 10$ (stable); and $f = 1.6$, $Q = 50$ (unstable) ideal gas detonations using the two codes.

The results are in agreement with the predictions of Erpenbeck and give us further confidence in our numerical approach for determining stability of detonation waves. We shall now proceed to determine the stability of nitromethane and liquid TNT detonations to two-dimensional perturbations.

B. Nitromethane and Liquid TNT Detonations

Figs. 10, 11, and 12 show that two-dimensional perturbations in C-J nitromethane and liquid TNT detonations do not grow, and that the two-dimensionally perturbed flow decays to one-dimensionally perturbed flow.

Nitromethane with E^* 's of 40 and 53.6 at C-J velocity and with an E^* of 53.6 at 0.7130 cm/μsec was calculated using the two codes. In all cases the two-dimensional perturbation did not grow and the stability was identical with that observed for one-dimensional perturbations.

IV. CONCLUSIONS

The time-dependent reaction zones of ideal gases, nitromethane, and liquid TNT have been investigated using two-dimensional Lagrangian and Eulerian numerical hydrodynamics.

In those cases for which Erpenbeck's analysis showed the steady-state solution of ideal gas detonations to be unstable to infinitesimal longitudinal and transverse perturbations, flows started in a configuration approximating the steady-state solutions exhibited a bounded instability; in those cases for which Erpenbeck's analysis showed the steady-state solution to be stable, perturbations were found to decay. The two-dimensional ideal gas instability has not been numerically studied for a sufficient time to determine its final amplitude and period since large amounts of computer time would be required.

THE TIME IS 1.20000+000

Fig. 6. The 2DL-computed streamlines formed by the Lagrangian cell corners at 1.2, 4.0, and 8.4 for an ideal gas detonation, with $f = 2$, $Q = 50$, in cylindrical geometry. The perturbation grows, and therefore the detonation is unstable. The mesh is a square whose sides are 0.8.

THE TIME IS 4.00000+000

THE TIME IS 8.40000+000

THE TIME IS 4.00000+000

Fig. 7. The 2DL-computed streamlines formed by the Lagrangian cell corners of the ideal gas detonation shown in Fig. 6 (but in slab geometry) at 4.0 and 6.4.

THE TIME IS 6.40000+000

THE TIME IS 1.02000-001

Fig. 8. The 2DE-computed isobars (pressure difference between isobars is 1) at 0.102, 0.202, and 0.302, for an ideal gas detonation, with $f = 2.$, $Q = 50$, and piston factor = 0.9, in cylindrical geometry as in Fig. 6. The perturbation grows, and therefore the detonation is unstable. The mesh is a square whose sides are 0.04.

THE TIME IS 2.02000-001

THE TIME IS 3.02000-001

THE TIME IS 1.50000-001

Fig. 9. The 2DL-computed streamlines formed by the Lagrangian cell corners at 0.15, 0.9, 2.1, and 3.0 for an ideal gas detonation, with $f = 2$, $Q = 0.2$, in cylindrical geometry. The perturbation decays, and therefore the detonation is stable. The mesh is a square whose sides are 0.05.

THE TIME IS 9,00000-001

THE TIME IS 2.10000+000

THE TIME IS 3.00000+000

THE TIME IS 1.00000-003 MICROSECONDS

Fig. 10. The 2DL-computed streamlines formed by the Lagrangian cell corners at 1×10^{-5} and 1.8×10^{-4} μsec for a nitromethane C-J detonation in cylindrical geometry. The mesh is a square whose sides are 100 A, $E^* = 53.6$, and $Z = 4 \times 10^8$. The perturbation decays, and the shock front pressure closely approximates that shown in Fig. 4 for a one-dimensional perturbation.

THE TIME IS 1.00000-004 MICROSECONDS

THE TIME IS 1.02000-005 MICROSECONDS

Fig. 11. The 2DE-computed isobars (pressure difference between isobars is 10 kbars) at 1.02×10^{-5} , 2.02×10^{-5} , 5.02×10^{-5} μsec for a nitromethane C-J detonation in cylindrical geometry. The mesh is a square whose sides are 40 A, piston factor = 0.8, $E^* = 53.6$, and $Z = 4 \times 10^8$. The perturbation decays, and the shock front pressure closely approximates that shown in Fig. 4 for a one-dimensional perturbation.

THE TIME IS 2.02000-005 MICROSECONDS

THE TIME IS 9.02000-005 MICROSECONDS

THE TIME IS 1.00000-003 MICROSECONDS

Fig. 12. The 2DL-computed streamlines formed by the Lagrangian cell corners at 1×10^{-3} and 6.5×10^{-3} μsec for a liquid TNT C-J detonation in cylindrical geometry. The mesh is a square whose sides are 5000 A, $E^* = 41.1$, and $Z = 2 \times 10^6$. The perturbation decays, and the shock front pressure closely approximates that shown in Fig. 5 for a one-dimensional perturbation.

THE TIME IS 0.50000-003 MICROSECONDS

The nitromethane and liquid TNT detonations exhibited the same stability to transverse and longitudinal perturbations as to longitudinal perturbations alone. It is obviously possible that this surprising result is an artifact of the numerical methods used to solve the reactive hydrodynamics, or of faulty interpretation of the numerical results.

The general behavior of time-dependent solutions to the reactive hydrodynamic equations for ideal gases is not necessarily similar to that for condensed explosives, and, therefore, the particular form of the equation of state is one of the determining features of the stability of the flow.

Since the details of the chemical kinetics are crucial to the stability behavior, one probably can not expect the real detonations to closely reproduce the time-dependent behavior of the theoretical model with simple Arrhenius kinetics.

Perhaps the most important conclusion to be obtained from this study is that there is every reason to believe that real detonations of condensed explosives can exhibit unstable periodic behavior. The time-dependent solution to the reactive hydrodynamic equations does not necessarily result in a steady-state solution for real explosives.

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APPENDIX

THE IDEAL GAS MODEL

NOMENCLATURE

C_v	heat capacity
D	detonation velocity
E^*	activation energy
f	degree of overdrive = $(D/D_{CJ})^2$
I	internal energy
P	pressure
Q	heat of explosion
R	gas constant
T	temperature
t	time
U	particle velocity
V	volume
W	mass fraction of undecomposed explosive
Z	frequency factor
γ'	C_p/C_v

Subscripts

o	initial state
p	constant pressure
v	constant volume

The equation of state for an ideal gas of constant heat capacity undergoing an exothermic, irreversible, unimolecular reaction is as follows. Given I , W , and V , one calculates P and T from

$$P = \frac{[I + (1 - W)Q](\gamma' - 1)}{V}$$

and

$$T = \frac{[I + (1 - W)Q]}{C_v}$$

$$\begin{array}{ll} \gamma' = 1.2 & T_o, V_o, P_o = 1 \\ C_v = 5 & I_o = 5 \\ R = 1 & U_o = 0 \end{array}$$

The Arrhenius rate law is $\frac{dW}{dT} = ZW e^{-E^*/RT}$.

The results of some of the calculations are summarized below.

Q	50	50	0.2
Z ⁽¹⁾	206	82.5	3×10^{17}
E*	50	50	50
f	1.6	2.0	2.0
D	8.613	9.63	1.873
D _{CJ}	6.809	6.809	1.325
Results	Unstable 1-D Unstable 2-D	Stable 1-D Unstable 2-D	Stable 1-D Stable 2-D
P _{W=1}	67.5	84.0	3.20
P _{W=0}	55.0	72.0	3.14

(1) Z is adjusted so that W = 0.5 when the time is 1.0.