One-Dimensional Detonation Initation

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The x-t observation of one-dimensional initiation behind a reflected shock wave in hydrogen-oxygen-argon mixtures is extended to include simultaneous density measurement using an optical interferometer. The wave pattern observed experimentally is reproduced theoretically using a nonsteady method-of-characteristics analysis with reasonable postulates for the reaction kinetics and shock heating. The close agreement between the theoretical and experimental results shows that in this case the initiation occurs as the result of an adiabatic explosion wave generated by reflected shock heating. The usefulness of this concept of initiation in understanding the mechanism of other initiation phenomena is discussed briefly.

Introduction

THE initiation of detonation has been studied quite extensively in recent years. Experimentally, three types of initiation have been observed: flame initiation in a long tube,[1-6] direct spark initiation,[7] and shock wave initiation when the shock is either a decaying shock generated by a neighboring detonation[8, 9] or an incident[10-12] or reflected[10, 13,14] step shock wave generated in a conventional shock tube. In all cases, shock waves are present in the gas prior to initiation.

Although the gross behavior of these various types of initiation has been documented quite well,[15] there have been only a few realistic attempts to relate the sudden appearance of a Chapman-Jouguet detonation to the exothermic reaction kinetics that must be responsible. Zeldovich, Kogarko, and Simonov[7] have constructed a theory for direct spark initiation which requires that the decaying spherical shock wave, generated by the spark, heats an element of the gas to a sufficient temperature for a long enough time to initiate rapid chemical reaction. The gas kernel that is responsible for flame initiation[16] does not contribute to their theoretical model. Strehlow and Cohen[13] have reported the observation of one-dimensional reflected shock initiation and indicate that in this case a detonation probably is formed from the adiabatic explosion wave generated by reflected shock heating.

In this paper, the study of Strehlow's and Cohen's ignition technique[13] is extended, and a more exact theoretical model is presented and compared to the experimental results.

Experimental Technique

The data were taken in the department's 4 × 15 × 600-in. rectangular shock tube using a 5-in. Mach Zehnder interferometer that is positioned across the 4-in. dimension of the tube. A 2-in.-thick plate of aluminum was placed in the shock tube at the window section to form the back wall for the experiments. Clearance to the inner shock tube wall was about 0.005 in. Strip film x-t photographs were taken through two 0.025-in. horizontal slits, one at the shock tube test section and the other in the second interferometer beam at the same optical distance as the first.[17] The interferometer used a 0.020-in.-diam pinhole and a #22GE flash bulb with no filter as a light source. Shock velocities were measured with a relatively insensitive schlieren system obtained by placing a 0.025-in. vertical slit at the image point of the light source (in the camera body). The horizontal slits were focused on the film plane. The system therefore allows one to obtain an x-t schlieren interferogram of the flow. Film velocity was determined by photographing a light flashing at 10,000 cps simultaneously with the shock reflection. Synchronization with the diaphragm burst was obtained by conventional electronic means, and the shock velocities were corrected for misalignment of the slit image in the usual manner.[18] Mixtures of 10% oxygen, 20% hydrogen, and 70% argon were prepared in a separate mixing bulb by partial pressure using high-quality commercial gases with no extra purification. The tube had a leak rate of less than 100 μ/min for all runs and was operated at an initial downstream pressure of 2 to 6 cm Hg. Premature detonation due to diaphragm burst was avoided by adding a quantity of pure argon in the neighborhood of the diaphragm just before measuring the final downstream pressure and subsequent firing. Since the downstream portion of the tube was 36 ft long, this did not dilute the explosive mixture observed in the test section. Bursting pressure was adjusted to produce shocks in the range $2.0 < M_s < 2.3$.

Discussion of Experimental Results

Figures 1 and 2 are representative x-t schlieren interferograms of initiation behind the reflected shock wave. In these photographs the incident shock, approaching from the right, generates a reflected shock when it contacts the back wall. Both of these shock waves produce a schlieren trace that can be used for velocity measurement and an interferometric fringe shift that can be used to determine density change. Some time after passage of the reflected shock, reaction occurs in the highly exothermic mixture. The photographs show an accelerating shock wave (which starts as a compression wave at the back wall; see Fig. 1) associated with and followed by a fringe shift that at first is in the direction of the earlier fringe shifts and then relaxes to a position between incident and reflected shock fringe positions.

The photographs agreed in every respect with the information available from earlier results. Explosion delay times compared favorably with the data of Strehlow and Cohen[13] and Schott and Kinsey.[19] Velocities of the reaction wave were in the range observed by Strehlow and Cohen,[13] and the one-dimensionality of the initiation process was quite satisfactory.

The photographs also yielded information on the density field during the initiation process. Unfortunately, interpretation is complicated in this region because the gas composition is changing during reaction. Nevertheless, photographs taken with fringes near to and far from the back wall show that at the back wall the density does not increase before it decreases, whereas some distance from the wall shock compression

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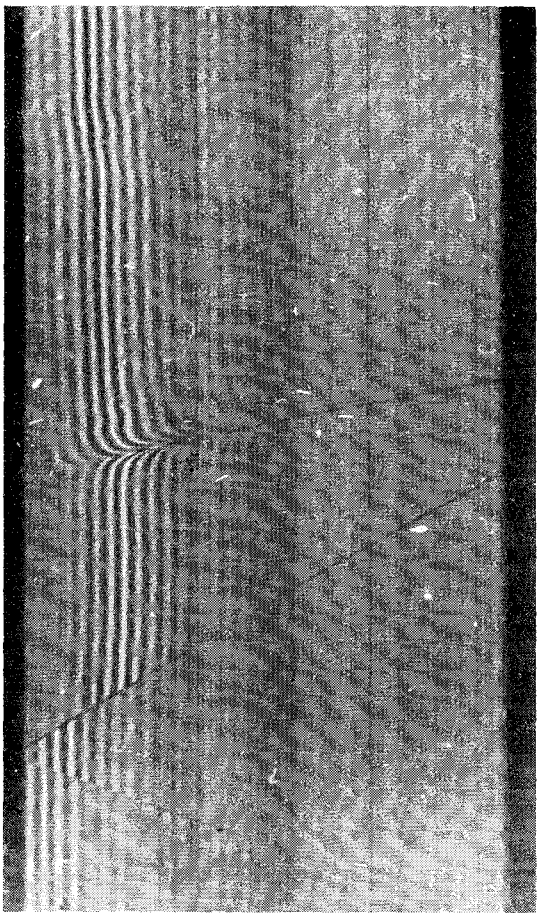

Fig. 1 x-t schlieren interferogram of detonation initiation behind the reflected shock in a shock tube showing interference fringes at the back wall; 10% oxygen, 20% hydrogen, 70% argon, P_0 = 20 mm Hg, T_0 = 298°K, M_s = 2.13, T_{rs} = 1017°K, delay to explosion = 251 μsec. Time increases upward, back wall on left. A fringe shift to the right represents a density increase in the unreacted gas

causes the density to increase markedly before it decreases. The photographs also show that some time after initiation the net fringe shift is relatively independent of the distance from the back wall.

The ability to follow the density throughout a one-dimensional nonsteady flow pattern is seen to be quite useful. The photographs distinctly show small density fluctuations due to weak waves which might otherwise go unobserved. In both Figs. 1 and 2 the rapid small-density decreases observed after the reaction zone can be related to the weak rarefaction fan generated by the reflected shock-reaction shock interaction. Figure 2 shows a small linear density increase ahead of and attached to the reflected shock which can be related to the slight reflected shock bifurcation to be expected in this gas mixture. Byron and Rott[20] observed a similar density shift in their experiments using a slightly different technique. The reflected shock region in Fig. 1 shows a slow increase in density with time which agrees with current concepts of the effect of incident shock attenuation on the gas properties in the reflected shock region. The forward traveling density increase observed in the incident shock region in Fig. 2 is caused by a spurious compression wave following the shock.

The Model

Schott and Kinsey,[19] Schott,[21] and Duff[22] have shown that at high temperatures the hydrogen-oxygen reaction consists of an almost thermally neutral chain branching explosion (with an induction period) followed by slow recombination reactions that produce most of the heat released by the reaction. They further observed that, for this reaction, the approach to equilibrium is approximately exponential in time. Strehlow and Cohen[13] have postulated that reflected shock initiation consists of the generation of a reaction wave which, if "undisturbed," would parallel the reflected shock on an x-t diagram. The following model therefore was used to construct a nonsteady one-dimensional flow pattern by a method-of-characteristics analysis.

Consider an ideal reflected shock occurring in a constant heat capacity gas, each element of which is capable of reacting and thereby releasing heat. Label each element of the gas according to its dimensionless distance ξ_0 from the back wall at the time of reflected shock passage, where $\xi_0 = x_0/t_0 a_{rs}$, and x_0 is the distance of the reacting particle from the back wall at the time of shock passage, t_0 is a characteristic time for the system, and a_{rs} is the velocity of sound in the gas behind the reflected shock. Chemical reaction (or heat release) is assumed to occur after a delay time that is the same for each element irrespective of its subsequent motion. Now define a (ξ_0, τ) or undisturbed coordinate system where $\tau = t/t_0$ such that $\tau = 0$ at the instant reaction starts at the back wall. Therefore, in this coordinate system the leading edge of the reaction zone is the straight line $\tau = \xi_0/\eta$, where η = Mach number of the shock relative to the gas behind it. Further assume that after the induction period the rate of chemical reaction or heat release is the same for each element of gas and can be defined as an inverse exponential. This

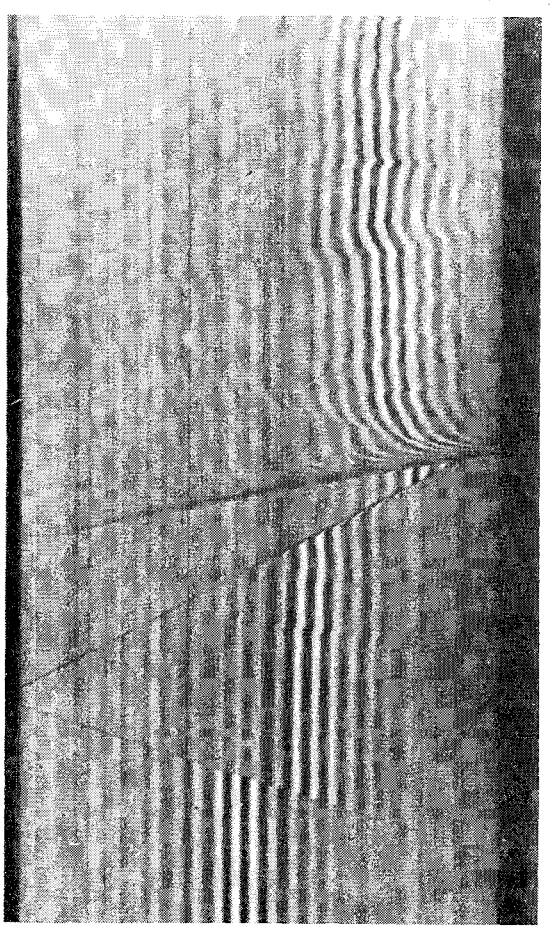

Fig. 2 x-t schlieren interferogram of detonation initiation behind the reflected shock in a shock tube showing interference fringes some distance from the back wall; 10% oxygen, 20% hydrogen, 70% argon , P_0 = 35 mm Hg, T_0 = 303°K, M_s = 2.25, T_{rs} = 1085°K, delay to explosion = 151 μsec. Time increased upward, back wall on left. A fringe shift to the right represents a density increase in the unreacted gas

yields the following heat release function in the (ξ_0, τ) or undisturbed coordinate system: $q = 0$ for $\tau < \xi_0/\eta$, $q = q_{max}\{1 - \exp[-(\tau - \xi_0/\eta)]\}$ for $\xi_0/\eta \leq \tau \leq 4 + \xi_0/\eta$, and $q = 0.98\, q_{max}$ for $\tau > 4 + \xi_0/\eta$, where q is the heat added to a gas element in calories per gram or other appropriate units. The reaction has been stopped arbitrarily at $\tau - \xi_0/\eta = 4$ or $q = 0.98\, q_{max}$ in order to simplify the analysis.

This information and the initial gas temperature now can be used to determine the flow pattern in the model gas. The flow was assumed to be one-dimensional and nonsteady, and Rudinger's[24] method-of-characteristics technique was used to determine the particle's actual position (ξ, τ) as a function of its initial position ξ_0 and the time τ. The method of determining the correct values for the parameters of the model and the details of the calculation are discussed below.

Within the confines of this model, one is allowed to choose independently the speed of the reaction wave in the (ξ_0, τ) system and, for a particular gas temperature, the maximum quantity of heat to be added to the system. The rate of heat addition has been absorbed formally in the definition of q and is dependent on the characteristic time t_0. Also notice that the delay time for adiabatic explosion is unimportant to the model itself. An estimate of the explosion delay time simply will tell one when the accelerating wave intersects the reflected shock. The model is rather oversimplified in that heating due to the compression waves propagating ahead of the reaction wave is assumed not to affect either the delay time or rate of reaction in the (ξ_0, τ) coordinate system. However, the entropy increase due to reaction shock compression was included in the wave analysis.

Only four constants must be chosen to construct a flow diagram for comparison with x-t photographs obtained experimentally. The effective heat capacity ratio γ of the gas, the gas temperature just prior to reaction, the Mach number of the reaction wave in the undisturbed (ξ_0, τ) coordinate system, and the effective heat release q_{max} are the only variables of the model. The second and third of these were given the values $T_{rs} = 1010°K$ and $\eta = 0.6459$, corresponding to the gas properties for an incident shock of Mach number 2.15. This is the average Mach number of the experimental runs. The maximum heat release and the effective gamma are a little more difficult to determine. First, one must determine the state of the real gas at chemical equilibrium in the neighborhood of the back wall by calculating the theoretical fringe shift for an assumed thermodynamic path.

Since the gas was observed to expand only slightly during the explosion in the region near the back wall, it was assumed that the relation between the local pressure and local volume of any exploding element was linear; i.e., the thermodynamic reversible path on a PV plot was assumed to be a straight line. The locus of end points for this process (for straight line paths of different positive slopes) is simply the Hugoniot curve in the excluded quadrant (see Appendix). Therefore, Hugoniot calculations extended through this region were used to determine the pressure, volume, and composition of the equilibrium state for the real gas. Densities and compositions then were used to calculate a theoretical fringe shift (the data of White[23] were used for OH refractivity) and thereby determine the experimental pressure and density change across the reaction zone from the fringe shift measurements. These data were then interpolated to determine $T_r/T_{rs} = 2.582$, $\rho_r/\rho_{rs} = 0.855$, $P_r/P_{rs} = 2.208$ for $M_s = 2.15$. It must be emphasized that this calculation is not exact. In the first place, an assumption was made concerning the thermodynamic path, and secondly, some distance from the back wall the shock wave will change the entropy and therefore cause an additional error in the final end state.

Once the equilibrium state of the gas near the back wall was known, the equivalent gamma for the characteristic analysis was determined by averaging the gamma for the gas behind the reflected shock (1.529) and the gamma for the gas at complete equilibrium (1.422) to obtain $\gamma = 1.475$. To determine an equivalent heat release for a gas with this gamma, two of the three state variables may be matched, but the third cannot. The authors chose to match pressure and density (or specific volume). This, of course, causes a change in temperature and therefore an arbitrary difference in the velocity of sound of the real and model gases behind the reaction wave. The consequences of this assumption are discussed in the next section.

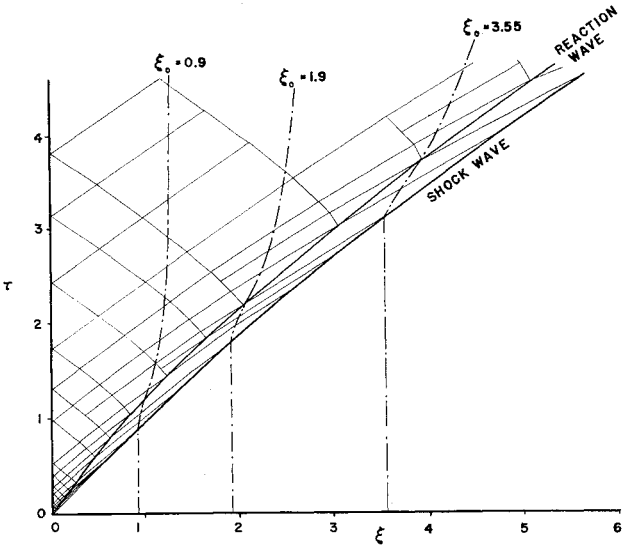

Fig. 3 Complete wave diagram showing the characteristics as light lines, particle paths as dashed lines, and shock wave and reaction wave as dark lines

In the actual calculation, a net size was chosen such that the change in the velocity of sound between neighboring points was always less than 10%. The resulting x-t diagram is shown in Fig. 3. Theoretical density-time profiles were calculated at four stations using the characteristic net and are shown in Fig. 4. After the analysis was completed, a direct calculation of the undisturbed reaction wave trajectory [in the (ξ_0, τ) coordinate system] using the nonsteady flow pattern showed that, for the calculation, the undisturbed (ξ_0, τ) trajectory accelerated (see Fig. 5). This effect was traced to a cumulative averaging error in the stepwise construction of the reaction wave position during the calculation. Its implications are discussed in the next section.

Discussion

The theoretical asymptotic density ratio after reaction is 0.75, whereas the experimental asymptotic density ratios averaged 0.85. Exact quantitative agreement therefore was

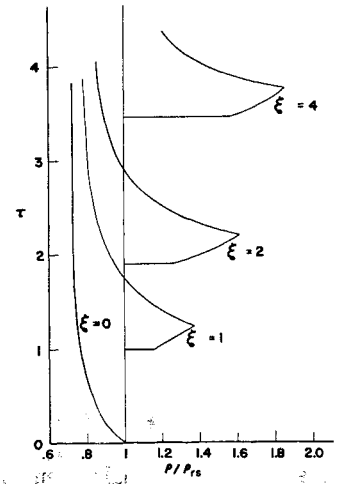

Fig. 4 Timewise density fluctuation at four stations as calculated from the wave diagram; ρ_{rs} is the density in the quiescent gas behind the reflected shock

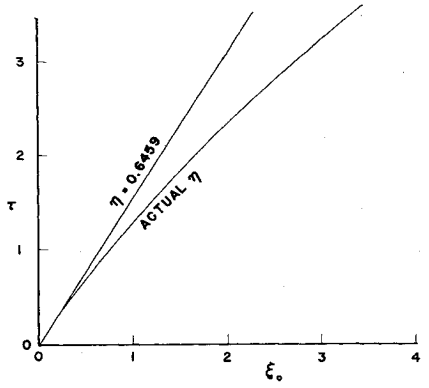

Fig. 5 Undisturbed reaction wave trajectory; $\eta = 0.6459$ assumed for the calculation of Fig. 3; the actual velocity plotted was obtained from Fig. 3. Both lines are in the undisturbed ($\xi_0\tau$) plane. The discrepancy was caused by a cumulative averaging error discovered after completion of Fig. 3

not obtained using this model. There are two reasons why one might expect this deviation from observed behavior. In the first place, the effects of reaction shock heating on the explosion delay time and on the local exothermic reaction rate intentionally were not included in the analysis. However, the acceleration of the heat release wave in the (ξ_0, τ) system produced by the cumulative averaging error during the analysis had the same effect as an arbitrary acceleration generated by reaction-shock heating of the gas. It must be stated, though that there is no way of justifying the acceleration in the (ξ_0, τ) system as shown in Fig. 5. This therefore represents a fault of the model which could account for the lack of exact quantitative agreement. Second, the procedure of balancing density and pressure in constructing the model leads to an incorrect velocity of sound in the model gas. Since nonsteady wave development is related intimately to the local velocity of sound in the gas, one might expect an error from this source. It is probable that pressure (as the driving force) and temperature (as an indication of the velocity of sound) would be better variables to match in future representations of this type.

The analysis does show, however, that the behavior of the experimentally observed wave pattern can be explained in qualitative detail as being the result of an adiabatic explosion wave generated by the reflected shock. This wave subsequently is observed to interact with the reflected shock wave, producing a Chapman-Jouguet detonation in the incident shock region. Strehlow and Cohen[13] observed three types of initiation phenomena in the reflected shock region. The model presented here agrees with the first two classes of initiation which they described. Their third observation, that highly exothermic mixtures quickly produce *CJ* detonation waves, could be investigated by assuming a larger heat release function than that used in this analysis.

It is interesting to note that shock heating always precedes the formation of detonation waves in exothermic gas mixtures. In addition, in a large fraction of observed flame initiations, the detonation suddenly appears as a very high (above *CJ*) velocity forward traveling wave accompanied by a rearward traveling detonation commonly called a retonation. If one assumes that the primary shock is accelerating continuously during flame-to-detonation transition, the formation of this high velocity initial detonation wave can be explained qualitatively in terms of the "reaction wave" model of this paper. Consider successive elements of gas being heated by an accelerating shock. The region between the shock and the driving piston (i.e., accelerating flame) is truly nonsteady and contains a pressure, velocity, and thermal gradient. Since for an accelerating shock the temperature at the shock front is higher than the temperature in the gas behind the shock front, the locus of the initial adiabatic explosion points for each gas element generates a "reaction wave," which, if not disturbed by the occurrence of reaction [(ξ_0, τ) system], will be traveling at a velocity greater than the shock velocity. In fact, one can conceive of conditions where the velocity of this wave in room coordinates could be infinite or even negative relative to the shock travel. Therefore, an ignition delay concept applied to each gas particle in the nonsteady flow regime ahead of an accelerating flame can explain qualitatively the observed behavior. The accelerating shock initiation recently reported by Bazhenova and Soloukhin[25] shows a behavior typical of what one would expect for accelerating shock initiation using the nonsteady model of this paper.

With this "reaction wave" model in mind, it should be possible to reverse the usual procedure used in detonation initiation studies. By measuring wave developments experimentally and using a nonsteady flow characteristic analysis, the ignition delay time of each gas element could be measured and, in addition, related to its temperature and pressure history. Therefore, chemical reaction rate constants for comparison to kinetic data from other sources should be obtainable from initiation experiments. Most important, though, is the fact that this model clearly shows the nonsteady nature of the last stage of the initiation process and predicts shock initiation without requiring either a Chapman-Jouguet deflagration or a shock velocity above the Chapman-Jouguet detonation velocity. In fact, it is difficult to see how the strictly steady-state concept of a Chapman-Jouguet deflagration or detonation applies to nonsteady initiation phenomena. This implies that there should be a re-evaluation of the pseudo-steady flow assumptions used by Adams and Pack,[26] Oppenheim,[3] and Brinkley and Lewis[15] to discuss the mechanism of detonation initiation.

Conclusions

A nonsteady analysis based on reasonable postulates of one-dimensional flow, exothermic reaction, and wave initiation by a reflected shock has produced a theoretical flow pattern that agrees in qualitative detail with the experimentally observed phenomena. An analysis of this type should be applicable to the later stages of other initiation processes and should allow the prediction of when and where detonation first will appear under any set of experimental conditions. The importance of treating initiation as a truly nonsteady phenomena is emphasized.

A new photographic technique, strip film schlieren interferometry, was used to determine simultaneously the wave processes and density field in a one-dimensional nonsteady flow.

Appendix

The Hugoniot equation for steady inviscid flow in a constant area duct is

$$E_2 - E_1 - q = (P_2 + P_1)(V_1 - V_2)/2$$

where E is internal energy, q is heat added (both in units of ergs per gram), P is the pressure in dynes per square centimeter, and V is the specific volume in cubic centimeters per gram. This equation places no restriction on the nature of the process occurring between stations 1 and 2 or the equation of state of the substance.

For a static process, such as homogeneous reaction or homogeneous heat addition, the equation for the energy change is

$$E_2 - E_1 - q = -\int_1^2 PdV$$

where the path of integration must be known to evaluate the

right-hand quantity. This equation is valid for any reversible path in the PV plane independent of the nature of the process or the equation of state of the substance. Evaluate the integral along the straight line path $Y = A\epsilon + 1 - A$, where $Y = P/P_1$, $\epsilon = V/V_1$, and $A = (Y_2 - 1)/(\epsilon_2 - 1)$ is the slope of the path. The integral then reduces to

$$-P_1V_1\int_1^2 (A\epsilon + 1 - A)d\epsilon$$

which yields on integration

$$-P_1V_1(\epsilon_2 - 1)(A\epsilon_2 + 2 - A)/2$$

Substituting for A, one gets

$$-P_1V_1[(Y_2 - 1)\epsilon_2 + 2(\epsilon_2 - 1) - (Y_2 - 1)]/2$$

or

$$E_2 - E_1 - q = (P_2 + P_1)(V_1 - V_2)/2$$

which is the same as the Hugoniot relation for steady flow. In other words, the Hugoniot also represents the locus of end states for homogeneous processes occurring along a straight line path in the PV plane irrespective of the other details of the process or the equation of state of the substance. In addition, one sees that the portion of the usual Hugoniot excluded because of the existence of imaginary velocities simply represents possible homogeneous processes where the pressure and volume increase simultaneously. This allows one to use real gas Hugoniot calculations in the excluded quadrant to calculate the end state for the adiabatic explosions observed near the back wall.

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