NUMERICAL STUDY OF BLAST INITIATION OF DETONATION USING A TWO-STEP CHEMICAL KINETICS MODEL

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Abstract   The effect of chemical reactions on the blast initiation of detonation in gaseous media has been investigated in this paper. Analytical method is based on the numerical solution of one-dimensional reactive Euler equations. So far, analyses on the blast initiation of detonation have modeled the combustion process as a one-step chemical reaction, which follows the Arrhenius rate law. Previous studies indicate that one-step model cannot predict single critical initiation energy. These results contradict with the experimental observation that a distinct value of critical initiation energy exists, below which no detonation occurs. Two-step chemical kinetics has been utilized in the present investigation. In two-step kinetics, first step is an induction step, while the chemical energy is released in second step via Arrhenius mechanism. With variation of activation energy in these two steps, the effect of induction length and energy release can be studied. This model predicts three regimes of blast initiation of detonation that have been observed in experimental observation. On the other hand, like one-step model, the present model cannot predict a limit for the initiation energy.

Key Words   Detonation, Initiation, Numerical Simulation, Two-Step Kinetics Model, Arrhenius

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چكيده   در اين مقاله تاثير سيتيک واكنشهاى شيميايى بر آغازش انفجارى دتونيشن در محيطهاى گازى بررسى شده است. روش تحليل بر مبناى حل عددى معادلات واكنشى اويلر يک بعدى است. محيط مورد بررسى به عنوان گاز كامل در نظر گرفته شده است. در تحقيقات قبلى احتراق به صورت مدل تک مرحله اى آرنيوس مدل شده بود. بر مبناى آن تحقيقات يک مقدار واحد به عنوان انرژى بحرانى براى آغازش انفجارى دتونيشن قابل پيش بينى نبود. اين نتيجه مخالف مشاهدات تجربى مى‌باشد. در كار حاضر پديده احتراق با يک مدل سيتيكى دو مرحله اى مدل شده است. مرحله اول زمان تاخير واكنش احتراق و مرحله دوم آزاد شدن انرژى واكنش را مدل مى كنند. تحقيق حاضر نشان مى‌دهد كه گرچه اين مدل قادر به پيشگويى سه رژيم مختلف آغازش انفجارى دتونيشن مى‌باشد، همچنان مانند مدل يک مرحله اى نمى‌تواند حداقل انرژى لازم را در توليد دنونيشن پيش بينى نمايد.

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1. INTRODUCTION

Direct initiation of detonation is a process at which a detonation wave is formed directly via rapid deposition of a large amount of energy in a small volume of combustible mixture. If the energy released by igniter is lower than a critical value, no detonation occurs [1].

Since the critical energy is a quantitative measure of detonability, there have been numerous experimental works in the past forty years devoted to measuring and cataloguing it for different mixtures [2]. The first analytical work to predict the critical initiation energy was the pioneer studying of Zeldovich et al. [3]. In the middle of 70's several models were proposed to calculate the critical initiation energy, which included the effect of the finite rate of chemical reactions through experimental length

and time scales. Among them the more popular are the kernel model of Lee et al. [4], the Sichel model, the hydrodynamic model of Edwards, and the chemical energy model of Korobeinikov. Benedick et al. [5] have comprehensively reviewed these models and compared them with experiments. None of the models discussed above can offer an appropriate criterion for critical energy.

Despite those attempts, a quantitative theory that can predict the critical energy required for direct initiation from first principles (i.e. basic thermochemical and kinetic rate data of the explosive gaseous mixture) is still lacking. Numerical solution of governing gas dynamics-chemical kinetics equations has an important role in the study of detonation. Mazaheri [6] used one-step chemical reaction and showed that this model cannot predict single critical initiation energy. Due to the important role of kinetics on detonation generation, as suggested by Lee [7], more complex chemical kinetics in the study of initiation process should be used. Short and Quirk [8] have studied the non-linear stability of a pulsating detonation using a three-step reaction model. They showed similar mechanism for regular and irregular modes of instability for both three-step and the standard one-step reaction model. However, they found that the use of the three-step model has an advantage over the standard one-step Arrhenius model, because a well-defined detonability limit can now be obtained. Recently Dionne [9] studied the propagation of non-ideal detonations arising from reaction steps involving a competition between exothermic and endothermic reactions. It was found that the propagation of detonation could be different by using more than one single-step rate law. Dionne reported that the pathological detonations, i.e., detonations can travel at a velocity greater than the Chapman-Jouguet (C-J) value, are possible when there are more than one-step kinetics present in the chemical model. More recently Mazaheri et al. [10] used a general two-step kinetic model to investigate the role of the induction length (or induction time) on stability of gaseous detonations. Their study showed that the ratio of reaction length to induction length characterizes general features of detonation stability. In view of the different qualitative features obtained from these studies, it appears of interest to investigate the direct initiation problem using a more realistic kinetics model. Therefore, in the present work the same two-step chemical reaction model as Mazaheri et al. [10] is utilized. Theoretical and numerical investigation of the blast initiation problem have already been carried out by many researchers, for instance He and Clavin [11], Mazaheri [6], and more recently Eckett et al. [12], all using a single-step Arrhenius rate law. Although He [13] also incorporated detailed kinetics of H_2-O_2 system in his numerical simulation, little attention is given on the influence of the chemical kinetics. Therefore the main objective of the present study is to elucidate further the direct initiation phenomenon and investigate the importance of the chemical kinetics scheme used. This study is performed via numerical simulation.

2. GOVERNING EQUATIONS

When a large quantity of energy E_O is suddenly deposited into a reactive gas mixture within a small size, the gas expansion generates a very strong blast wave and a highly overdriven detonation is formed instantaneously. As the blast wave expands, its intensity decreases. Experimental observations [13] show that the transition of this overdriven detonation to a self-sustained CJ detonation is possible only when the deposited energy is larger than a critical value E_C. In the present simulation of the direct initiation phenomenon, a plane blast model is used, where the early time is essentially the decaying of a strong blast governed by the similarity solution of Taylor [14]. The subsequent decay of the blast, where chemical reactions come into play, then will be described by numerical simulation.

Many investigators (e.g., Howe et al., [15]) have emphasized on the important role of the induction and reaction lengths on dynamics aspects of detonation waves. However, due to their one-step model the role of these kinetics length scales could not be observed independently. Therefore, in the present study the chemical reaction is modeled by two-step kinetics. The first step indicates a non-heat release induction delay where A is converted to an activated state A^*. In the second step, the energy of the reaction is released. These two steps
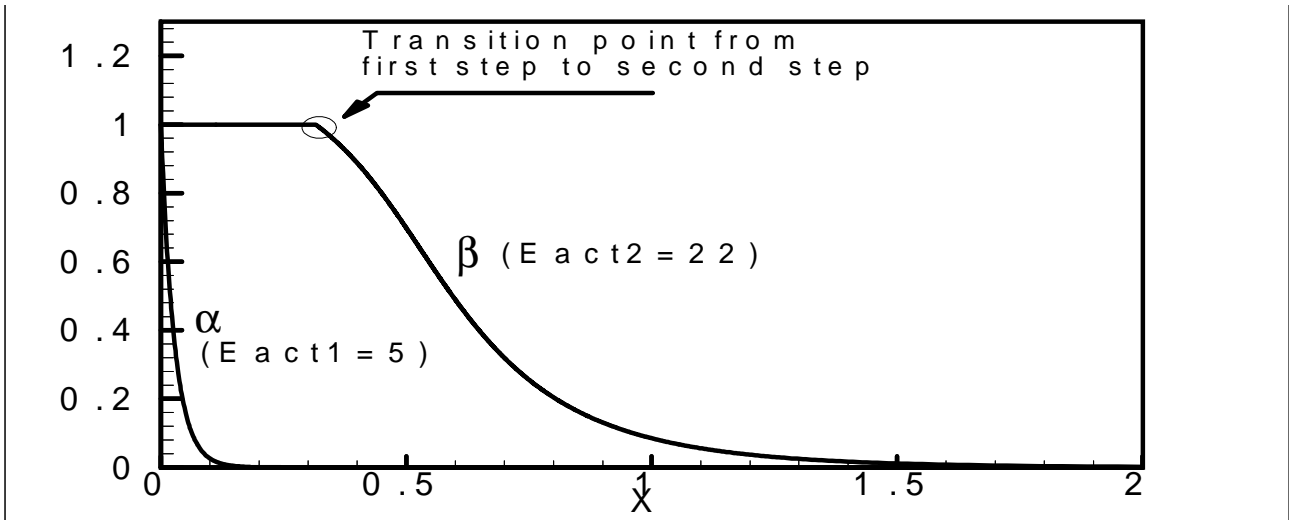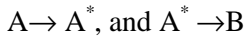
Figure 1. Variation of two steps reaction progress variables (α and β) in the reaction zone. Circle shows transition from the first step two the second step.

can be shown by two reactions:

$$A \rightarrow A^*, \text{ and } A^* \rightarrow B$$

The reaction rates are given by:

$$w_1 = \frac{d\alpha}{dt} = -K_1 \alpha \exp\left(\frac{-Eact_1}{RT}\right), \qquad \text{and}$$

$$w_2 = \frac{d\beta}{dt} = -K_2 \beta \exp\left(\frac{-Eact_2}{RT}\right)$$

where T denotes temperature and R the universal gas constant; K_1 and K_2 are the constants of reaction rates, and $Eact_1$ and $Eact_2$ are the activation energies. The progress parameters α and β for induction step are $0<\alpha<1$ and $\beta=1$, and for exothermic reaction are $\alpha=0$ and $0<\beta<1$. The variation of reaction progress parameters α and β, for a steady (ZND) detonation, is shown in Figure 1.

The governing one-dimensional reactive Euler equations are in the following form:

$$\frac{\partial U}{\partial t} + \frac{\partial F}{\partial x} = S$$

where,

$$U = \begin{bmatrix} \rho \\ \rho u \\ \rho e \\ \rho \alpha \\ \rho \beta \end{bmatrix}, F = \begin{bmatrix} \rho u \\ \rho u^2 + p \\ u(e+p) \\ \rho u \alpha \\ \rho u \beta \end{bmatrix}, \text{and } S = \begin{bmatrix} 0 \\ 0 \\ 0 \\ -\rho w_1 \\ -\rho w_2 \end{bmatrix}$$

Here, p, ρ, u and e are pressure, mass density, particle velocity and specific internal energy, respectively. A polytropic equation of state and an ideal thermal equation of state are assumed,

$$e = \frac{p}{\rho(\gamma-1)} + \frac{u^2}{2} + Q\beta, \text{ and } P = \rho RT$$

where, Q is the heat release per unit mass of reactant, and γ is the specific heat ratio.

The dependent variables are non-dimensionalized with respect to the unburned mixture properties, density with ρ_o, pressure with γp_o, and velocity with sound speed of the unburned mixture C_O. Heat release and activation energy are non-dimensionalized by RT_o. The characteristic length
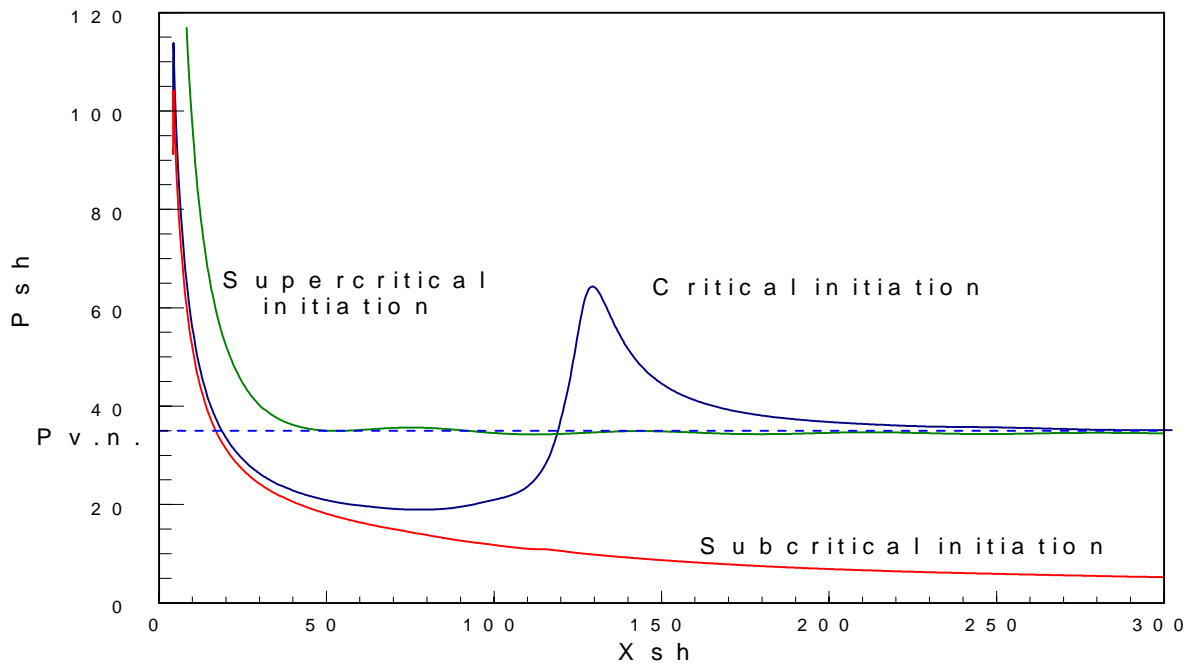
Figure 2. The three regimes of initiation using one-step reaction kinetics model. The corresponding initiation energies are 3415 for supercritical, 1765 for critical, and 1614 for the subcritical initiation ($Q/Rto = 50$, $\gamma = 1.2$ and $Eact/RT_0 = 24$).

scale, L_C, is the half-reaction length (hrl) of steady ZND detonation. The characteristic time scale is $t_c = L_C/C_O$. The dimensionless constants for mixture are $\gamma = 1.2$ and $Q/RT_O = 50$ in all calculations here.

For a strong blast wave with the shock Mach number M_S at radius R_S, the initiation energy (E_0) is obtained from

$$\frac{E_0}{p_0} \cong R_s^{j+1} \gamma k_j M_s^2 I$$

where "I" is a function of only γ and should be calculated from the similarity solution of the strong blast wave, p_O is the initial pressure of the mixture, and (j, k_j) is (0, 1), (1, 2π), and (2, 4π) for the planar, cylindrical and spherical geometry respectively. In planar geometry, E_O/P_O, the non-

dimensional initiation energy has the same form as the so-called explosion length, which is non-dimensionalized with half reaction length [13].

3. THE METHOD OF STUDY

The piecewise parabolic method (PPM) of Colella and Woodward [16] is used as the main gas dynamics solver. In analyzing the propagation of a detonation wave, the tracking of shock front has an essential role. For this purpose, the conservative front tracking method of Chern and Colella [17] has been used. Since all reactions are completed in a narrow region close to the shock, it is more economical to use fine meshes only in this region and coarse grids elsewhere. To fulfill this requirement, a simple version of the adaptive mesh refinement of Berger and Colella [18] has been utilized. The developed code is validated via
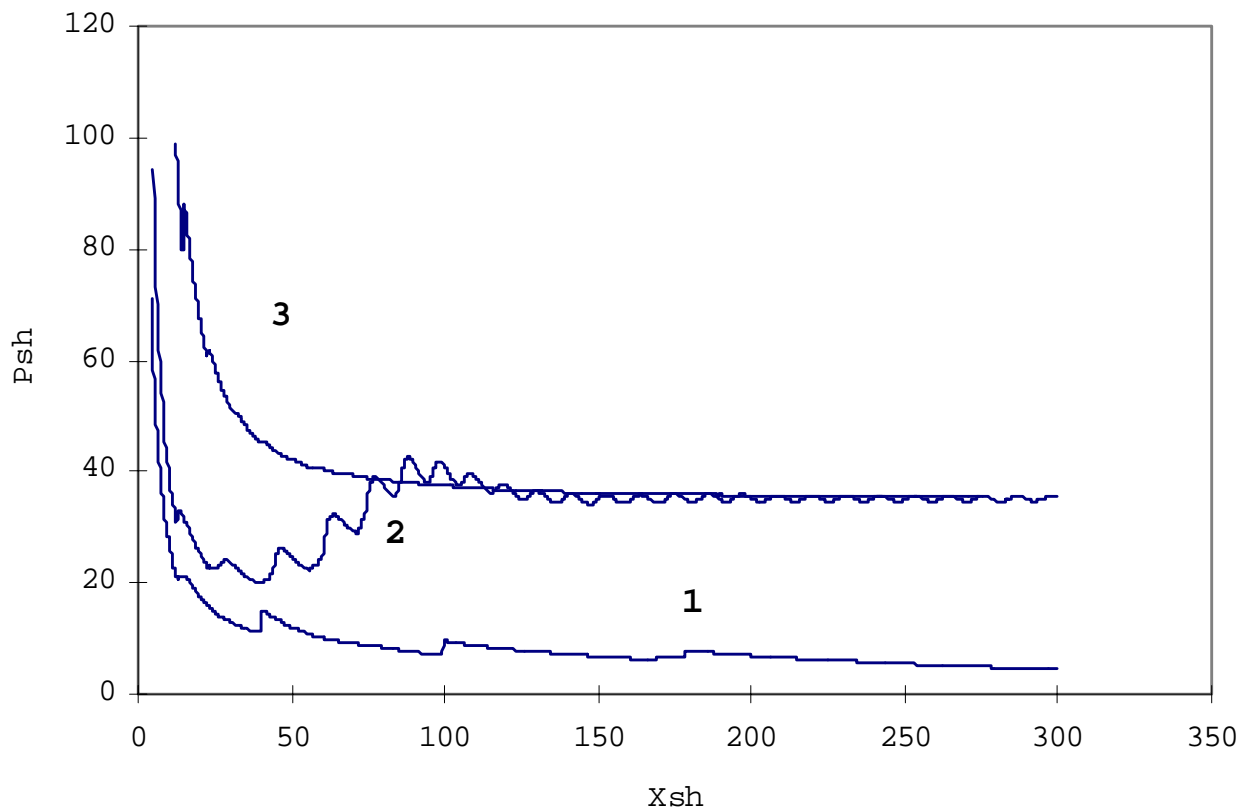
Figure 3. The three regimes of the initiation for two-step kinetics. The initiation energies of the three curves are $Eo_1 = 1081$, $Eo_2 = 1615$, $Eo_3 = 5340$. The kinetics parameters are: $Eact_1/Rto = 5$, $Eact_2/RTo = 20$, $Q/RTo = 50$, and $\gamma = 1.2$.

several test problems, including the mesh-independency test, Sod shock tube problem, Lax shock tube problem, and the numerical simulation of gaseous detonations [18].

4. RESULTS AND DISCUSSION

4.1 Three Regimes of Initiation of Detonation
It is instructive to review the three regime of initiation using a single-step Arrhenius kinetics. The numerical simulation of the three different regimes of initiation is shown in Figure 2. In this figure the shock pressure is plotted as a function of the shock radius. This figure corresponds to a mixture with $Q/RT_o = 50$, $E/RT_o = 24$, and $\gamma = 1.2$.

The dotted line shows the non-dimensional pressure at the von-Neuman spike of the corresponding self-sustained CJ detonation. The three curves correspond to sub-critical, critical, and super-critical initiation regimes, where the non-dimensional initiation energies (i.e., E_o/P_o) are 1614, 1765, and 3415, respectively. In the early times of blast wave propagation, the shock pressure decays rapidly as in a strong non-reactive blast wave (i.e., $M_s \sim 1/R_s$). If the initiation energy is below a critical value, it is found that the blast wave will progressively decay to an acoustic wave. This regime is referred to as the sub-critical regime. On the other hand, if the initiation energy greatly exceeds the critical value, the blast decays asymptotically to the strength of the CJ value, forming a self-sustained detonation wave at x=50.

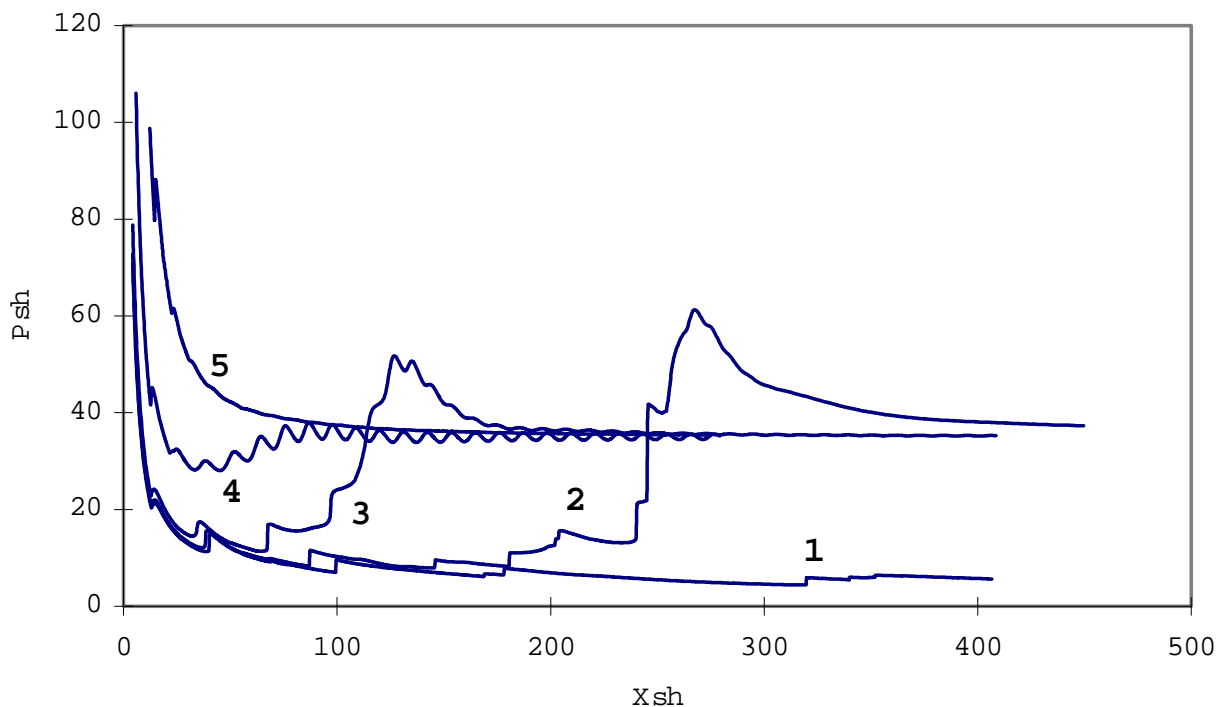**Figure 4**. The initiation for different initiation energies below the supercritical value. The initiation energies of the different curves are $Eo_1 = 1081$, $Eo_2 = 1110$, $Eo_3 = 1205$, $Eo_4 = 2256$ and $Eo_5 = 5340$. The kinetics parameters are:

$$Eact_1 / RTo = 5, \; Eact_2 / RTo = 20, \; Q/RTo = 50, \text{ and } \gamma = 1.2.$$

This regime is called super-critical initiation. Near the critical initiation energy, the phenomenon is more interesting. The decaying process slows down when shock reaches about x = 30. Then, for a short period, the shock front propagates at an almost constant velocity. This phase of the blast initiation is called the quasi-steady period. The quasi-steady period is terminated by an abrupt acceleration of the shock front to form an overdriven detonation wave. This overdriven detonation then decays asymptotically to a self-sustained CJ wave. This regime of initiation is called the critical regime of initiation. Bach et al. [19] first observed experimentally the three regimes of initiation.

Two important parameters in the critical initiation regime are the shock "overshoot" and the "run up distance". The degree of overshoot in shock pressure at the end of the quasi-steady period or "overdriven", is measured with respect to the shock pressure of the corresponding ZND detonation. The distance between the center of initiation and the location of the maximum shock pressure shall be referred as the run up distance. The run up distance, corresponding to sub-critical initiation in Figure 2, is about 130.

The numerical simulation of the three regimes of initiation utilizing two-step kinetics is shown in Figure 3. This figure corresponds to a mixture with $Eact_1 = 5$ and $Eact_2 = 20$. The three curves 1, 2 and 3 correspond to sub-critical, critical, and super-critical initiation regimes, where the non-dimensional initiation energies (i.e., Eo/Po) are 1081, 1615, and 5340, respectively.

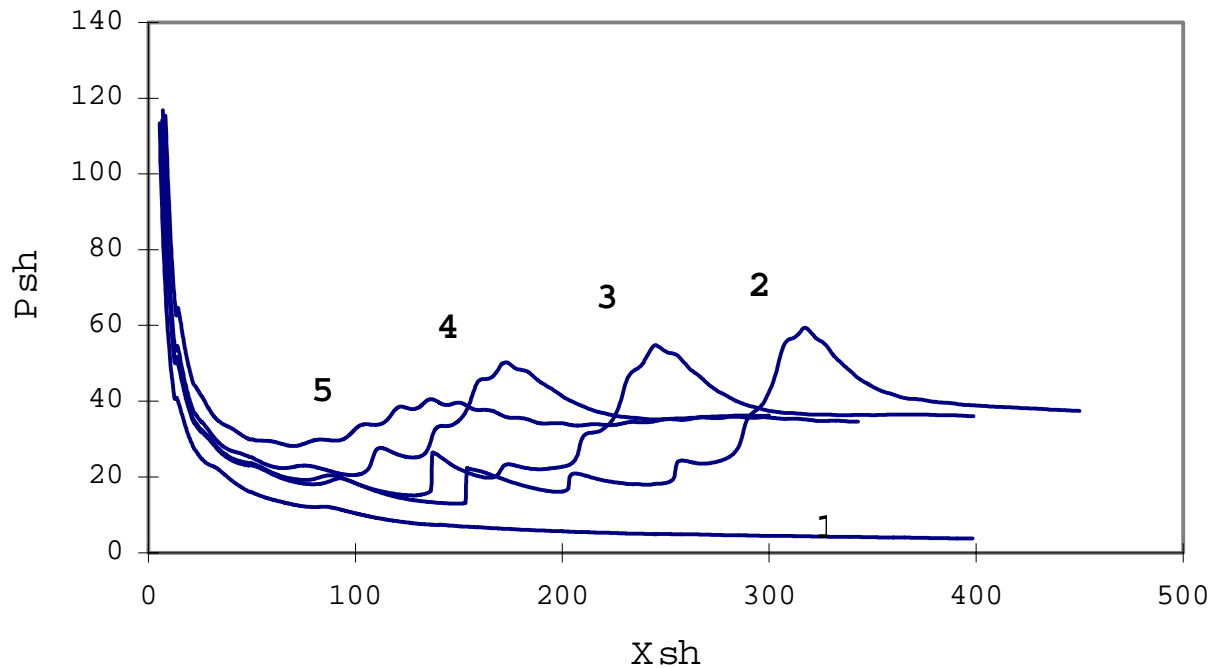**Critical Initiation Energy** It is a well recognized fact that the heat addition drives a one-dimensional

Figure 5. The initiation process when the activation energy of the second step is changed with respect to Figure 3. The initiation energies of the different curves are $Eo_1 = 2256$, $Eo_2 = 2865$, $Eo_3 = 2924$, $Eo_4 = 3125$, and $Eo_5 = 3858$.

The kinetics parameters are: $Eact_1/RTo = 5$, $Eact_2/RTo = 25$, $Q/RTo = 50$, and $\gamma = 1.2$.

subsonic flow behind a shock to the sonic condition, which is the equilibrium (i.e., the stable) condition for a shock initiated reaction complex (i.e., the CJ condition). The above conditions have some very important implications. The most important conclusion is that, in the framework of an exothermic Arrhenius model, regardless of the value of the initiation energy any blast initiation can initiate a detonation wave.

To further clarify this problem, the evolution process (i.e., the P_{sh}- X_{sh} curve) with different initiation energies are plotted in Figure 4. This figure is indeed an extension of Figure 3 to a wider range of initiation energies. Different curves correspond to different initiation energies. The non-dimensional initiation energies, Eo/Po, for curves 1, 2, 3, 4 and 5 are 1081, 1110, 1205, 2256 and 5340, respectively. However, it is observed that for these initiation energies a detonation has been formed. For curve 1 no formation of detonation was observed for the time of

calculation.

One question that arises is which curve really corresponds to the critical initiation energy. This question will be addressed later in this section. It is useful to correlate the "run up distance" and the "overshoot" to the initiation energy. Curves 2, 3, and 4 are considered for this analysis. It is seen that by increasing the initiation energy, both run up distance and the amount of the overshoot are reduced. In the supercritical regime, almost no overshoot is observed. It has already been observed that when the initiating shock decays to a sub-CJ value a pressure pulse is formed between two fronts (Mazaheri, [6]). In general, when a pressure pulse-reaction front complex is formed behind the shock, its amplification depends on the property of particles crossing the pressure pulse. The particle, which crosses the pressure pulse, has already been processed by the shock front. Indeed, this particle re-processes by the pressure pulse; hence, its reaction rate will be very high. Whenever the
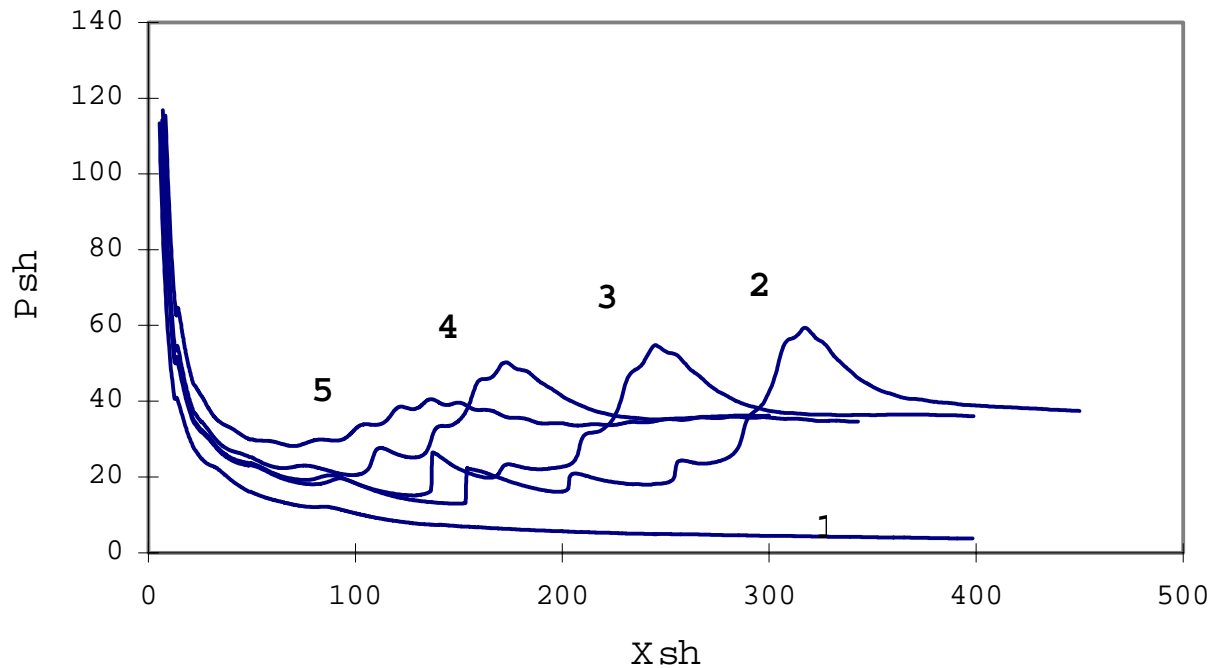
Figure 5. The initiation process when the activation energy of the second step is changed with respect to Figure 3. The initiation energies of the different curves are $Eo_1 = 2256$, $Eo_2 = 2865$, $Eo_3 = 2924$, $Eo_4 = 3125$ and $Eo_5 = 3858$.

The kinetics parameters are: $Eact_1/RTo = 5$, $Eact_2/RTo = 25$, $Q/RTo = 50$, and $\gamma = 1.2$.

"pressure pulse-reaction front" complex captures the shock its amplification will cease, since it processes a cold mixture. Thus, a longer quasi-steady period (for the same mixture) causes a higher final amplification. That is the reason for higher overshoot of curve 2 with respect to higher initiation energies.

There is no critical initiation energy in the framework of our analytical model. The present results contradict with the experimental observation that a distinct value for the critical initiation energy exists, below which no detonation can occur [13]. To understand the reason of this contradiction, two factors should be noted to; the present exothermic rate law and the effect of heat loss which are ignored in the analytical model. It has been shown by Clark *et al*. that the second factor can be ignored (Clark, [20]). However, the first factor plays a major role on the dynamic phenomenon of the initiation and must be modeled properly. A

proper model has to be involved an endothermic mechanism too. The competition between two mechanisms may produce both failure as well as successful initiation.

The most important factor, which governs the present kinetics model, is the activation energy. Many researchers (e.g. J. H. Lee [21] and Lee and Stewart [22] have investigated the effect of this parameter on dynamic parameters of detonation [21] such as initiation energy and instability boundaries. The effects of activation energy on the initiation process are investigated in two stages in this work. In each stage one of the activation energies (either $Eact_1$ or $Eact_2$) is kept constant and the other one is changed. Then, the effect of this variation on the initiation process is studied. The purpose of this study is to see if the uniqueness of the critical initiation energy depends on the choice
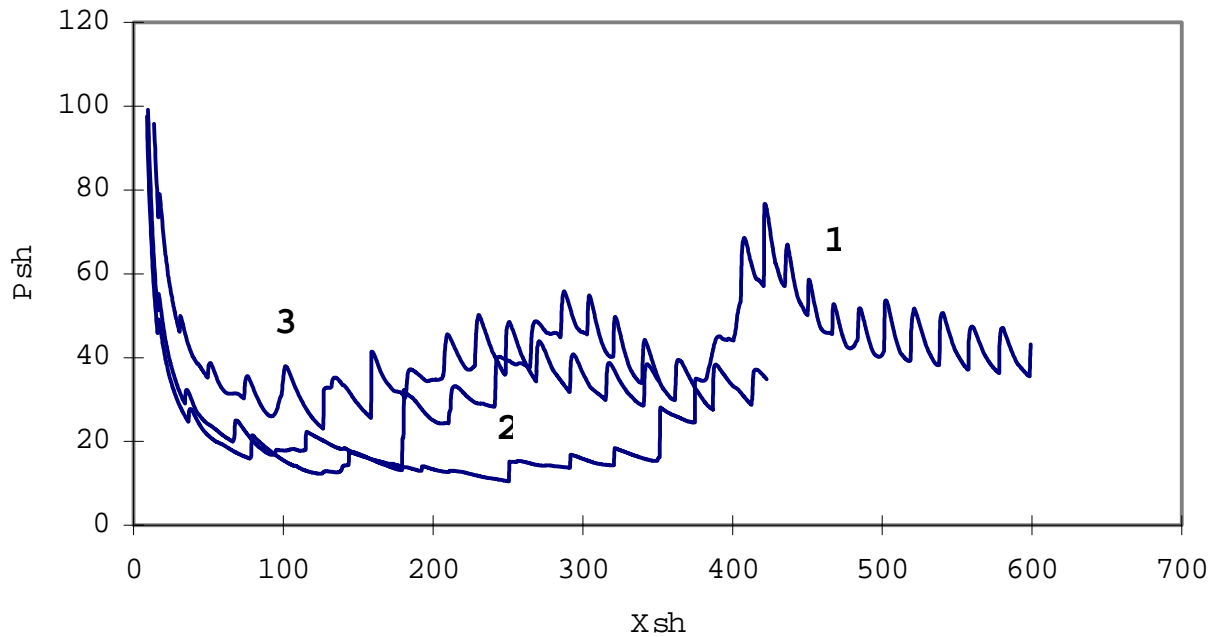
Figure 6. The initiation process when the activation energy of the first step is changed with respect to Figure 3. The initiation energies of the different curves are $Eo_1 = 3418$, $Eo_2 = 3858$, and $Eo_3 = 5887$. The kinetics parameters are: $Eact_1/RTo = 10$, $Eact_2/RTo = 20$, $Q/RTo = 50$, and $\gamma = 1.2$.

of activation energies or not. The results are demonstrated in Figures 5 and 6.

In Figure 5 the activation energy of the first step is the same as Figure 4 (i.e., 5) while the activation energy of second step is changed from 20 to 25. The same behavior as Figure 3 is observed. This means that no unique value for the initiation energy is found.

In Figure 6, the second step activation energy is the same as Figure 4 and the activation energy of the first step is varied from 5 to 10. No significant difference is observed with respect to the previous cases. Therefore, regardless of the value of the activation energies of two steps, with the present kinetics model, no single value can be found as the critical initiation energy.

numerical simulation of the reactive Euler equation with a two-step chemical reaction model can predict no unique valve for the critical initiation energy. This fact is independent of the value of the activation energies of the two steps. The presented two steps model has the main drawback of a single step model that is both models represent only exothermic processes. In order to predict a real failure, the kinetic model must be able to capture the competition between chain branching and recombination reactions. Below about 1100 K, chain-branching reactions become ineffective and hence give a sudden decrease in the global reaction rate, which is an essential for true simulation of the critical initiation. This point, in fact, shows the route for the future research.

5. CONCLUSION

The results of the present study indicate that the

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