NUREG/CR--4961

TI89 014713

NUREG/CR-4961
SAND87-7128
R3

A Summary of Hydrogen-Air Detonation Experiments

Manuscript Completed: June 1987
Date Published: May 1989

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Under Contract to:
Sandia National Laboratories
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Prepared for
Division of Systems Research
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555
NRC FIN A1246

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ABSTRACT

Dynamic detonation parameters are reviewed for hydrogen-air-diluent detonations and deflagration-to-detonation transitions (DDT). These parameters include the characteristic chemical length scale, such as the detonation cell width, associated with the three-dimensional cellular structure of detonation waves, critical transmission conditions of confined detonations into unconfined environments, critical initiation energy for unconfined detonations, detonability limits, and critical conditions for DDT. The detonation cell width, which depends on hydrogen and diluent concentrations, pressure, and temperature, is an important parameter in the prediction of critical geometry-dependent conditions for the transmission of confined detonations into unconfined environments and the critical energies for the direct initiation of unconfined detonations. Detonability limits depend on both initial and boundary conditions and the limit has been defined as the onset of single head spin. Four flame propagation regimes have been identified and the criterion for DDT in a smooth tube is discussed.

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1. INTRODUCTION

As part of a long-term continuing study of gaseous detonative combustion, the present report addresses the specific case of H_2 -air detonations. The objective is to seek out and compile in a comprehensive fashion a reliable pool of data for this specific fuel-air mixture over a broad range of parametric conditions. The report is intended to examine all aspects of detonative combustion that are commonly studied and are of practical interest. These include problems of initiation, transition, transmission, detonability limits, etc. An attempt has been made to include the results of the latest studies and to be up-to-date as far as developments in the field are at the time of writing. It is felt that this type of information has relevance to the nuclear industry in the context of the possibility of generation of hydrogen in a nuclear containment building.

As late as 1980, very few adequate experimental data on H_2 -air detonations were available. Most of the existing data were limited in scope and derived from relatively small-scale experiments similar to those used for detonation studies in highly reactive H_2 - O_2 mixtures. Modern developments in detonation theory indicate that, for the less sensitive H_2 -air mixtures, these data might not be accurate enough. Consequently, large-scale experiments might be required to determine on a more reliable quantitative basis, the appropriate dynamic detonation parameters (e.g., detonability limits, critical initiation energy, critical tube diameter) which constitute the practical information for detonation hazard assessments.

The equilibrium detonation parameters (i.e., detonation velocity, pressure, density and temperature ratios, and equilibrium composition of the reaction products) are adequately predicted by the classical Chapman-Jouguet (C-J) theory which models gaseous detonation waves as one-dimensional, steady combustion waves. However, the C-J theory, which is essentially an equilibrium calculation, cannot predict the nonequilibrium or dynamic detonation parameters which are related to the chemical reaction rates, i.e., to the structure of the detonation wave. A one-dimensional model for the detonation structure was formulated in the early 1940's by Zeldovich, [1] Doring, [2] and von Neumann, [3] the so-called ZDN model. The model assumes that a detonation wave consists of a planar shock wave followed by a reaction zone. The model allows the computation of the dynamic detonation parameters when a model for the physical processes involved is specified. However, experimental studies of the detonation structure only provide qualitative support for the ZDN model [4-8] because one-dimensional steady detonation waves are unstable as demonstrated by Schelkhin, [9] Zaidel and Zeldovich, [10] and Erpenbeck. [11]

Despite the observations of the three-dimensional structure of spinning detonations in tubes as early as 1929 by Campbell and Woodhead [12] and the subsequent studies of the spinning mode by Bone et al. [13] and Zeldovich, [14] the one-dimensional C-J model continued to enjoy a wide popularity for several decades. Spinning detonations were considered to be "marginal" combustion waves occurring under very restricted conditions near the detonability limits where the chemical reaction rates and the

heat release are just sufficient to maintain propagation in a tube of given diameter. Because the regular period of the spin structure suggested some form of resonant coupling with the tube dimensions, Manson [15] and Fay [16] formulated in the forties and fifties linearized acoustic wave theories for spinning waves which predict accurately the observed pitch-diameter ratios. Using some of the ideas developed by Manson and Fay, Chu [17] later performed a linear analysis of the vibration of a gas column subjected to a rotating heat source (the spinning detonation) which predicted many of the observed properties of spinning detonations. These theoretical studies were followed in the sixties by an extensive series of experimental investigations of spinning detonation by many researchers with the best contributions by Dove and Wagner, [18] Duff [19] and Schott [20] in the West, and Shchelkin and co-workers [21] and Voitzekhovskii and collaborators [22] in the Soviet Union.

In the 1960s, the classical concept of a detonation wave as a steady, one-dimensional combustion wave was superseded by the observations of the three-dimensional structure and nonsteady propagation of self-sustained detonation waves by White [23] (using interferometry) and Voitzekhovskii et al. [24] and Denisov and Troshin [25] (using the smoked foil technique). These new observations of detonation phenomena generated a large amount of experimental research in the subsequent years. [20, 26, 27] A new interpretation of the detonation structure emerged from these studies. As sketched in Figure 1, detonation waves are composed of curved triple wave front intersections. Transverse waves of finite amplitude travel across the leading front at the local sound speed and interact with the leading shock to produce Mach stems which propagate across the detonation front. The leading fronts behave like decaying blast waves with periodic reinitiation when two transverse waves collide. The leading shocks decay from an initial overdriven state to sub C-J conditions. As the temperature behind the leading shock decreases, the distance between the shock and the reaction fronts increases until collision of two transverse waves returns the decaying shock to the overdriven state and a new cycle repeats again. The trajectories of the triple points (i.e., the intersection of the leading shock, the transverse wave and the Mach stem) form a cellular network, characteristic of the three-dimensional structure of gaseous detonations. The cellular network can be recorded on smoked foils placed on the walls of the detonation tube for confined detonations [28] (Figure 2) and by open-shutter photography [29] in diverging (i.e., unconfined) detonations (Figure 3). The detonation structure is a consequence of the strong nonlinear coupling between gas dynamics and chemical kinetics.

In the sixties, the smoked foil technique was used extensively to measure detonation cell widths (i.e., the transverse wave spacing) in subatmospheric fuel-oxygen mixtures diluted with argon, helium and nitrogen. [30, 31] The fuels used in these experiments included hydrogen and the common hydrocarbons (CH_4 , C_2H_2 , C_2H_4 , and C_2H_6). It was observed that the regularity of the detonation structure increases as the diluent content is increased. [32] As shown in Figure 4, argon was found to be the best diluent to improve cellular regularity followed by helium and nitrogen. Among the various theories on detonation cellular structure,

Figure 1. Schematic Illustrating Local Decay of a Multiheaded Detonation Front Between Collisions of Transverse Waves

Figure 2. Smoked Foil Record of Detonation in Stoichiometric $\text{H}_2\text{-O}_2$ Diluted With 40 Percent Argon. Initial pressure (p_0) = 60 Torr (see Reference 28).

Figure 3. Open-Shutter Photographs of a Self-Sustaining Cylindrical Detonation Wave on the Left, Associated With Multiplication of Transverse Waves and a Decaying Wave on the Right, Devoid of Multiplication

Figure 4. Smoked Foil Records of Detonations in Stoichiometric Fuel- O_2 Mixtures (see Reference 36).

[33, 34, 35] the caustic theory developed by Barthel [35] is the only one to successfully predict the observed transverse wave spacings in subatmospheric H_2 - O_2 mixtures with or without argon dilution.

Detonation research in the sixties proved conclusively that all gaseous detonation waves possess a cellular three-dimensional structure. The characteristic scale of the structure is directly related to the rate of chemical reaction in a detonation wave via the induction time τ . In recent years, the detonation cell width λ has been adopted as the characteristic dimension of the cellular structure. Lee et al. [37] have shown the link between the detonation cell width and the dynamic detonation parameters and proposed the cell size as a measure of the sensitivity to detonation of gaseous mixtures.

In 1980 a joint program to investigate H_2 -air detonations was initiated between the Shock Wave Physics Research Group at McGill University and Sandia National Laboratories under the sponsorship of the U.S. Nuclear Regulatory Commission. The objective of the joint study is to determine the relevant dynamic detonation parameters such as the characteristic chemical length scale associated with the three-dimensional cellular detonation structure (i.e., the detonation cell width), detonability limits, critical initiation energy for unconfined detonations, critical transmission conditions of confined detonations into unconfined environments and requirements for the transition of deflagration into detonation in H_2 -air mixtures, pure or diluted with steam and CO_2 .

This report presents the results obtained to date. Section 2 deals with detonation cell sizes. The transmission of a detonation from a confined region into an unconfined environment (i.e., the critical tube diameter problem) is described in Section 3. Critical initiation energies of unconfined detonations are discussed in Section 4. Section 5 discusses detonability limits. The transition from deflagration to detonation is treated in Section 6. Section 7 presents some conclusions.

2. DETONATION CELL SIZE

In the joint program on H_2 -air detonations, small-scale experiments were carried out in the laboratory facilities at McGill University. Large-scale studies involving field test facilities were conducted at a test range at Sandia National Laboratories in Albuquerque (SNLA).

Direct measurements of detonation cell size λ from smoked foil records and pressure records have been performed in H_2 -air, H_2 -air- CO_2 and H_2 -air-steam mixtures over a wide range of mixture compositions and initial conditions. [38, 39, 40] The small-scale experiments were carried out in three detonation tubes (5, 15, and 30 cm in diameter and 5.5, 6, and 16 m long, respectively). The large-scale experiments were performed in the heated detonation tube (HDT) (43-cm internal diameter, 13.1 m long). As discussed in Section 5 on detonability limits, because the maximum cell size that can be observed in a circular tube of diameter D corresponds to the onset of single-head spin detonation, i.e., when $D = \lambda/\pi$, detonation tubes of increasing diameter were used to allow the study of the less reactive mixture compositions.

The variation of cell size as a function of equivalence ratio ϕ , plotted in Figure 5 for H₂-air mixtures at NTP, exhibits a characteristic U-shape with a minimum around the stoichiometric composition (29.6 percent H₂, $\phi = 1$). The cell size increases from $\lambda = 1.5$ cm for the stoichiometric composition to $\lambda = 1.03$ m for the leanest mixture tested (13.6 percent H₂, $\phi = 0.37$) and $\lambda = 1.35$ m for the richest mixture tested (70 percent H₂, $\phi = 5.55$). These composition limits correspond to the onset of single head spin in the larger (43-cm-diameter) detonation tube. They are much wider than previous estimates (18.3 to 59 percent H₂ measured in a 1.4-cm-diameter tube by Wendlant [41] and 15 to 63.5 percent H₂ measured in a 30.5-cm-diameter tube by Kogarko and Zeldovich [42]) and are determined by the tube diameter rather than any limiting chemical reaction of the H₂-air system. The cell size increases faster for lean than rich mixtures and reflects a larger detonation sensitivity of fuel-rich mixtures.

Cell size data for H₂-air mixtures diluted with CO₂ at ambient initial conditions are compared in Figure 6 with the data for undiluted mixtures. For undiluted mixtures, the fuel percentage is denoted by y whereas the fuel percentage in diluted mixtures is $(1 - x)y$ where x denotes the percentage of diluent. The addition of 5, 10, and 15 percent CO₂ to a stoichiometric H₂-air mixture (29.6 percent H₂) increases the cell size from $\lambda = 1.5$ cm (no dilution) to $\lambda = 2.16$ cm, 4.2 cm and 19.2 cm, respectively. In other words, 5, 10, and 15 percent CO₂ dilutions decrease the detonation sensitivity of stoichiometric H₂-air mixtures by factors of 1.5, 2.8 and 12.8, respectively. An order-of-magnitude reduction in detonation hazard is also observed in both fuel-lean (24 percent H₂) and fuel-rich (35 percent H₂)-air mixtures diluted with 15 percent CO₂, the limits of the present experiments. The present results demonstrate the efficiency of CO₂ dilution to desensitize H₂-air detonations.

For meaningful assessment of the influence of steam on the cell size of H₂-air detonations, cell sizes in preheated H₂-air mixtures were measured in the heated detonation tube (HDT) (i.e., the 43-cm-diameter detonation tube) at SNLA. Air at local conditions existing at the test site was first admitted into the tube. Hydrogen was then added to obtain the desired mixture composition. The mixture was then heated to the desired temperature prior to detonation initiation. Three tests were performed and the initial temperature and pressure of the mixture prior to detonation initiation were (1) 10°C and 744.5 torr, (2) 50°C and 843 torr, and (3) 97°C and 946 torr. The H₂ concentration was kept constant at 16.7 percent H₂ for all three tests.

As the initial temperature T_0 is increased, the detonation cell size is found to decrease from $\lambda = 18$ cm at $T_0 = 10^\circ\text{C}$ to $\lambda = 15$ cm for $T_0 = 50^\circ\text{C}$ and $\lambda = 13$ cm for $T_0 = 97^\circ\text{C}$. Therefore, increasing the initial temperature by 40°C (from 10 to 50°C) and 87°C (from 10 to 97°C) causes the cell size to decrease by about 23 and 30 percent, respectively. Hence, both preheating and pressurizing increase the detonation sensitivity of a reactive mixture.

Figure 5. Variation of Detonation Cell Size With Equivalence Ratio in H_2 -Air Mixtures at NTP

Figure 6. Variation of Detonation Cell Size With Hydrogen Percentage in H₂-Air-CO₂ Mixtures at NTP

A limited number of experiments were also performed at an initial temperature $T_0 \approx 100^\circ\text{C}$ and superatmospheric initial pressures in H_2 -air mixtures in the composition range 15.9 to 45.5 percent H_2 . The variations of cell size with respect to equivalence ratio ϕ are compared in Figure 7 for cold and preheated H_2 -air mixtures. With increasing fuel content, preheating to 100°C causes an increasing reduction of λ from about 30 percent for fuel-lean mixtures (16.7 percent H_2 , $\phi = 0.47$) to 67 percent for stoichiometric mixtures (29.6 percent H_2 , $\phi = 1$) and 80 percent for fuel-rich mixtures (45.5 percent H_2 , $\phi = 1.99$). Therefore, the detonation sensitivity of preheated mixtures (to 100°C) at superatmospheric initial pressures increases with increasing fuel content.

To assess the effects of initial pressure and initial temperature on detonation cell size, experiments were also conducted in the HDT at room temperature (298 K) and constant air density (i.e., the air density at NTP, viz., $\rho_{\text{air}} = 41.6 \text{ moles/cm}^3$). These data are compared in Figure 7 with those obtained at NTP (298 K, 1 atm) and in preheated mixtures (373 K, $\rho_{\text{air}} = 41.6 \text{ moles/cm}^3$). The two upper sets of data show a reduction in cell size due to an increase in initial pressure at constant temperature. On the other hand, the two lower sets of data indicate further reduction in cell size due to an increase in temperature at constant air density. Therefore, the increase in detonation sensitivity of preheated mixtures at superatmospheric pressures initially result from an increase in both initial pressure and initial temperature.

Detonation cell sizes of H_2 -air-steam mixtures have been measured in the HDT facility at SNLA. The loading procedure for air and H_2 was identical to that used for the tests in preheated H_2 -air mixtures. Air at local ambient conditions was first admitted into the tube, followed by H_2 to achieve the desired composition. The mixture was then heated to the desired final temperature (i.e., 100°C). Then, steam produced by vaporizing a measured volume of water into the recirculation line was admitted into the tube prior to detonation initiation. In the tests carried out thus far, the initial pressure of the mixture ranged from 0.89 to 2.89 atm and the initial temperature varied between 96° and 100°C . The fuel concentration prior to steam addition varied between $y = 14.7$ and 45 percent H_2 whereas the steam content in the mixture ranged from about $x = 4.7$ to 30 percent H_2O .

The detonation cell sizes for H_2 -air-steam mixtures are compared in Figure 3 with the cell size data for hot H_2 -air mixtures. The addition of 10, 20, and 30 percent steam to a stoichiometric H_2 -air mixture increases the detonation cell size by factors of 6, 30, and 60, respectively. Comparison of the efficiency of CO_2 and steam dilutions in desensitizing H_2 -air detonations cannot be made readily without some knowledge of the effect of CO_2 dilution on preheated H_2 -air mixtures. Therefore, detonation studies of preheated H_2 -air- CO_2 mixtures should be conducted to provide a valid assessment of both diluent efficiencies.

There exists no simple quantitative theory to predict cell sizes from first principles at present. In 1965, Shchelkin and Troshin [43] postulated that the cell size λ is proportional to the chemical induction

Figure 7. Variation of Detonation Cell Size With Equivalence Ratio in Cold and Hot H₂-Air Mixtures

Figure 8. Variation of Detonation Cell Size With Equivalence Ratio in Hot H₂-Air-Steam Mixtures at Superatmospheric Initial Pressures

length ℓ of a one-dimensional detonation wave:

$$\lambda = A \ell \quad , \quad (1)$$

where the constant A depends on the mixture composition. The chemical induction length ℓ in a one-dimensional detonation wave is simply related to the chemical induction time τ behind a normal shock wave propagating at the C-J detonation velocity V_{CJ} :

$$\ell = (V_{CJ} - u) \tau \quad , \quad (2)$$

where u is the particle velocity behind the shock wave. Recently, Westbrook, [44, 45] Roller and Shepherd, [46] and Shepherd [47] have developed detailed kinetic models to compute induction times. Westbrook assumes a volumetric explosion in a mixture initially compressed by a normal shock wave propagating at the C-J detonation velocity of the mixture. In other words, the initial state corresponds to the Von Neumann state. The kinetic calculation uses 17 elementary reactions to describe the hydrogen oxidation process. The induction time is defined as the time required to reach the maximum rate of temperature rise.

The kinetic model developed by Shepherd to predict detonation cell sizes calculates induction times from 19 elementary reactions for the oxidation of hydrogen. Shepherd essentially uses the ZDN model. Starting with the mixture in the Von Neumann state, Shepherd integrates the one-dimensional, unsteady gasdynamics equations and chemical reaction kinetics along the Rayleigh line. The induction length is defined as the distance behind the shock at which the Mach number reaches $M = 0.75$. This criterion gives an induction length slightly longer than that based on the point of maximum rate of temperature rise used by Westbrook. Using Schelkin's formula (Equation 1), the scaling constant is determined by matching the experimental cell size for the stoichiometric composition. Shepherd's model yields a value of $A = 22$ which is comparable to the value ($A = 20$) suggested by Westbrook and Urtiew. [45] Cell sizes predicted by both Shepherd and Westbrook for H_2 -air mixtures at ambient conditions are compared in Figure 5 with the experimental data. The cell sizes predicted by Westbrook overestimate the experimental data for lean mixtures by factors as large as 30 and underestimate rich mixture data by at most 25 percent. On the other hand, Shepherd's results never underestimate or overestimate the experimental data by more than a factor of 2.

For H_2 -air- CO_2 mixtures, similar agreement with the experimental data is achieved by Shepherd. Since with inert diluents, detonation properties are expected to be primarily dependent on the fuel-air ratio, Shepherd attempted to improve the reaction length cell size correlation for both CO_2 and H_2O dilutions by using the observed reaction length cell size ratios for H_2 -air mixtures at the same equivalence ratio. The close agreement with the experimental data shown in Figures 8 and 9 for H_2O and CO_2 dilutions, respectively, is encouraging and suggests that such a technique may be viable to predict the effects of inert diluents. However, none of the present kinetic models can provide any physical understanding of the three-dimensional structure of gaseous detonations

Figure 9. Variation of Detonation Cell Size With Equivalence Ratio in H₂-Air-CO₂ Mixtures at NTP

since none of them model the nonlinear coupling mechanism between gasdynamics and chemical kinetics. Although three-dimensional numerical codes could be developed to handle such complex modeling, the computer time and storage needed would exceed the capability of present computers. Therefore, direct experimental measurement remains the most convenient means of determining detonation cell sizes.

Although other methods have been tried to measure the cell size (e.g., measurements of the pressure fluctuations superimposed on the main pressure trace recorded by pressure transducers [48], measurement of the location of the sonic plane of a C-J detonation [49], or optical methods [50]), the simple smoked-foil technique still remains the only successful method. For near-limit fuel-air detonations, where the cells are very large and long foils (of the order of meters) must be used, the actual deposition of a uniform coating of soot from numerous burners poses a rather difficult engineering problem. A more serious problem in the measurement of cell size is the actual interpretation of the fish-scale pattern. For most fuel-air mixtures, this pattern is highly irregular, and thus the selection of the "correct" cell size requires a certain amount of experience. This introduces a subjective element into the measurement of λ which has to be resolved. The use of long foils which allows the recording of the detonation wave over a long travel length certainly facilitates the interpretation of the foil. Alternatively, if a large number of experiments are performed in order to accumulate an ensemble of records under identical conditions, then the accuracy of the measurement will be improved. In the HDT experiments, typical uncertainty bounds for detonation cell widths have been estimated by Tieszen et al. [40] at ± 25 percent. However, for some tests these authors' individual estimates could vary by as much as ± 100 percent. Therefore, other techniques should be developed to facilitate the measurement of this fundamental parameter which characterizes the structure of the detonation wave.

The detonation sensitivity of H_2 -air mixtures and some mixtures of common hydrocarbons (C_2H_2 , C_2H_4 , C_2H_6 , C_3H_8 , and C_4H_{10}) with air at NTP are compared in Figure 10 where the variations of cell size have been plotted with respect to the equivalence ratio ϕ . The cell size data for hydrocarbon-air mixtures were measured from smoked foil records obtained at McGill University. [48] The cell size data indicate that, for all mixture compositions, C_2H_2 is the most detonation-sensitive fuel followed by H_2 , C_2H_4 and the alkanes (C_2H_6 , C_3H_8 , and C_4H_{10}). For example, for the stoichiometric composition ($\phi = 1$), C_2H_2 has a cell size $\lambda \approx 0.6$ cm, $\lambda \approx 1.51$ cm for H_2 , $\lambda \approx 2.6$ cm for C_2H_4 and $\lambda \approx 5.35$ cm for the alkane group. The solid curves in Figure 10 represent cell size values estimated from Equation 1 using Westbrook's kinetic data. [44] For C_2H_2 , C_2H_4 and the alkanes, the scaling constant A (determined by matching the experimental cell size for the stoichiometric composition) is 23, 10, and 12, respectively.

3. CRITICAL TUBE DIAMETER

The critical tube diameter d_c denotes the minimum diameter of a detonation tube from which a steady planar detonation wave can emerge into an

Figure 10. Variation of Detonation Cell Size With Equivalence Ratio in H_2 -Air and Hydrocarbon (C_2H_2 , C_2H_4 , C_2H_6 , C_3H_8 , and C_4H_{10})-Air Mixtures at NTP

unconfined volume containing the same mixture, transform into a spherical detonation wave and continue to propagate as a spherical detonation. In 1956, Zeldovich et al. [51] performed the first studies of this phenomenon in fuel-oxygen and fuel-oxygen-nitrogen mixtures at 800 torr, initially. A criterion for detonation transmission was formulated, namely, $d_c \approx 15\ell$ where ℓ is the induction distance behind a normal shock wave propagating at the C-J detonation velocity. The induction length was estimated from the measurements of the impulse and the pressure of the shock wave in spark initiation of spherical detonations. Friewald and Koch [52] also reported critical tube diameters in agreement with those of Zeldovich et al. In the mid-sixties, Mitrofanov and Soloukhin [53] found a relationship between the critical tube diameter and the detonation cell size in their studies on the diffraction of detonation waves in subatmospheric oxygen-acetylene mixtures. They observed that $d_c \approx 13\lambda$ for a circular tube and $d_c \approx 10\lambda$ for a square channel. The significance of these empirical correlations was not recognized until the mid-seventies when Edwards et al. [54] confirmed these earlier results and suggested the validity of the correlations for other chemical systems. In the early eighties, an extensive program was carried out at McGill University [55] to measure directly both the cell size and the critical tube diameter for most of the common hydrocarbon fuels (CH_4 , C_2H_2 , C_2H_4 , C_2H_6 , C_3H_6 , C_3H_8 , and C_4H_{10}) as well as hydrogen mixed with pure oxygen at low initial pressures (10 to 150 torr) and for fuel- O_2 - N_2 mixtures at atmospheric pressure, initially. These experiments extended the validity of the correlation $d_c \approx 13\lambda$ as suggested by Edwards et al.

Recently, in studying the transmission of detonations through circular orifice plates instead of from a straight tube, Liu et al. [56] found that the critical orifice diameter is identical to the critical tube diameter. For noncircular orifices (e.g., triangular, square, ellipse), the $d_c = 13\lambda$ correlation remains valid by defining an effective critical diameter as the arithmetic mean of the diameters of the inscribed and circumscribed circles of the orifice. For rectangular orifices of large aspect ratios ($L/W \geq 7$ where L and W are the length and width of the rectangle), the confined planar wave transforms into an unconfined cylindrical wave after transmission. Liu et al. [56] found that the critical width $W_c \approx 3\lambda$. As L/W decreases, W_c/λ increases and in the limit $L/W \rightarrow 1$ (i.e., a square orifice), $W_c \approx 10\lambda$ as observed by Mitrofanov and Soloukhin [53] as well as Edwards et al. [54]

To test the validity of the detonation transmission empirical laws in large-scale explosions, experiments were performed at the Sandia test site to determine the critical tube diameter [38] and the critical channel width [57] of H_2 -air mixtures. Cell size data were used to estimate the critical tube diameter from the empirical correlation $d_c \approx 13\lambda$ in order to provide some guidance for these large-scale field tests. Since the aim of these studies was to test the validity of the $d_c = 13\lambda$ correlation rather than to generate critical tube diameter data, only a limited number of tests were carried out in fuel-lean and fuel-rich mixtures. For the critical tube diameter tests, steady planar detonation waves were generated in steel tubes (ranging in diameter from 0.76 to 1.21 m) and then transmitted into thin-walled polyethylene bags. These bags were typically 60 cm larger than the tube diameter to provide some 30-cm radial

propagation of the detonation wave emerging from the steel tube before contacting the thin-wall plastic bag. To ensure the propagation of a planar detonation wave prior to transmission, rigid tubes must be used to prevent any wave curvature resulting from the soft confinement of the nonrigid tube. The detonation tube must also be long enough to ensure the propagation of a steady detonation wave prior to its transmission (especially for near-limit mixtures which require large initiation energies and may result in the transmission of an overdriven detonation if the tube is not long enough). In the present experiments, the detonation tubes were typically 3 to 4 m in length while the plastic bags were about 6 m long.

For the critical channel tests, the experiments were conducted in a rectangular channel made of two thick steel plates (2.5 cm wall thick x 2.36 m wide x 3.15 m long) held apart by spacers at the four corners to provide a range of channel widths W_c extending from 5 to 50 cm, i.e., aspect ratios L/W ranging from 63 to 6.3. Three sides of the channel were blocked with two plywood and one plexiglass strips (for photographic observation of the detonation propagation in the confining channel) to form the lateral confining walls. The open end of the channel was connected to a large thin-walled polyethylene bag similar to the critical tube diameter situation except that now the bag had a flat rectangular shape. In both transmission experiments, the entire deflated bag was first sucked into the inside of the confining duct by means of a vacuum pump to remove all the air from the system. A premixed explosive mixture from a large underground storage tank was then introduced to fill the tube or the channel and inflate the plastic bag. Thin strips of high explosive taped to the closed end of the confining duct were used to initiate detonation. The only diagnostic used was high-speed cinematography to record the propagation of the combustion wave.

The critical tube diameter for H_2 -air mixtures varies with fuel concentration as plotted in Figure 11, and exhibits the same U-shape dependence as the detonation cell size data from which it has been derived using the empirical correlation $d_c = 13\lambda$. As shown in Figure 11, good agreement is achieved between the direct measurements of d_c and the estimated values from the $d_c = 13\lambda$ relationship. For stoichiometric composition, a critical tube diameter of the order of 20 cm was directly measured in small-scale tests at McGill [55] in good agreement with the value $d_c = 19.6$ cm estimated from the $d_c = 13\lambda$ correlation using the value $\lambda = 1.51$ cm directly measured from smoked foil records. For off-stoichiometric mixtures, direct measurements of d_c in large-scale experiments were carried out at SNLA in tubes of diameters 76, 91, and 121 cm. The critical fuel concentrations are 21 and 51 percent H_2 for the 76-cm-diameter tube, 19.2 and 53.3 percent H_2 for the 91-cm-diameter tube, 18.1 and 55.5 percent H_2 for the 121-cm-diameter tube. As shown in Figure 11, these large-scale results do fall into the range predicted by cell size data using the $d_c = 13\lambda$ correlation.

Large-scale tests with mixtures of some common hydrocarbons (C_2H_2 and C_2H_4) and air at atmospheric pressure initially also confirm the validity of the $d_c = 13\lambda$ correlation. [58, 59] The results of the direct measurements are compared in Figure 12 with the estimated values from the

Figure 11. Variation of Critical Tube Diameter With Fuel Percentage in H_2 -Air Mixtures at NTP

Figure 12. Variation of Critical Tube Diameter With Equivalence Ratio in H_2 -Air and Hydrocarbon (C_2H_2 , C_2H_4 , C_2H_6 , C_3H_8 , and C_4H_{10})-Air Mixtures at NTP

$d_c = 13\lambda$ correlation for a range of equivalence ratios. From the practical point of view, the empirical law allows reliable estimates of critical tube diameters using cell size data obtained in 1/13-scale experiments. The correlation is particularly valuable for the less sensitive fuel-air mixtures which would require rather large-scale experiments.

The results for the successful transmission of a planar detonation confined in a rectangular channel and its transformation into a cylindrically diverging detonation wave in the same unconfined mixture are presented in Figure 13 where the variations of the critical channel width W_c (normalized with respect to the cell size λ of the mixture) have been plotted with respect to the channel aspect ratio L/W . For large aspect ratios ($L/W > 7$), the large-scale results for rectangular channels give a value $W_c/\lambda \approx 3$ in agreement with the small-scale data of Liu et al. [56] for rectangular orifices. The same critical width ratio $W_c/\lambda \approx 3$ was obtained with different fuel-air and fuel-oxygen-nitrogen mixtures (the fuels tested were H_2 , C_2H_2 , and C_2H_4) demonstrating the applicability of the scaling law to different chemical systems.

In the large-scale experiments, the symmetry of the transmission process about the center line of the channel was tested by sliding back the top plate of the channel configuration relative to the bottom plate to create a half channel configuration. In this situation, the bottom plate now plays a role equivalent to the center plane of symmetry for a full channel experiment and the critical width ratio should be $W_c/\lambda \approx 1.5$. Several experiments conducted in rectangular channels of aspect ratio $L/W \approx 18$ in lean H_2 -air mixtures (20.5 percent H_2) indicate a critical width $W_c \approx 1.55\lambda$ and confirm the symmetry of the transmission mechanism about the center plane.

As the aspect ratio is decreased below $L/W \approx 10$, the critical width ratio increases rapidly. A similar trend was observed by Liu et al. [56] in transmission experiments through rectangular orifices. For $L/W = 1$, Liu's results recover the value $W_c/\lambda \approx 10$ observed originally by Mitrofanov and Soloukhin [53] and Edwards et al. [54]. The similar trend for the results of the rectangular channel and rectangular orifice suggests that $W_c/\lambda \rightarrow 10$ as $L/W \rightarrow 1$ for both configurations.

Despite the lack of any quantitative theory to predict the critical tube diameter, a qualitative criterion for the transmission phenomenon has been developed by Lee. [60] The detonation emerging from the tube into the unconfined mixture is subjected to a radial gasdynamic expansion which increases the induction time of the unconfined shocked mixture and results in decoupling of the reaction zone from the leading shock front. The characteristic time t_q for the expansion waves to reach the tube axis and quench the detonation is of the order of the tube radius $d_c/2$ divided by the sound speed of the products c_b , i.e.,

$$t_q = d_c/2c_b$$

Figure 13. Variation of Critical Channel (or Rectangular Orifice) Width With Aspect Ratio

If the effective thickness, Δ , of the detonation is defined as the distance between the leading shock and the equilibrium C-J location, it is reasonable to assume that the detonation will not be quenched if, after emerging from the tube, the detonation propagates at least a distance 2Δ before the expansion waves reach the tube axis. This will guarantee that a small detonation core near the tube axis is not affected by the gasdynamic expansion and this core will serve as a kernel from which a spherical detonation will develop. The time t_p for the detonation wave to propagate a distance 2Δ is

$$t_p = 2\Delta/V_{CJ} \quad ,$$

where V_{CJ} is the C-J detonation velocity. Detonation transmission will be successful if $t_p \geq t_q$. Therefore, at criticality, $t_p \approx t_q$ and $d_c \approx 4c_b \Delta/V_{CJ}$. Since for most detonable mixtures, $V_{CJ} \approx 2c_b$, the transmission criterion reduces to

$$d_c \approx 2\Delta \quad .$$

An estimate for the effective thickness or the so-called hydrodynamic thickness of a detonation wave was reported by Edwards [61] from measurements of the decay of the transverse pressure vibrations (i.e., equilibration of the transverse shocks) in H_2-O_2 and $C_2H_2-O_2$ mixtures. Edwards found that $\Delta \approx (2.5-4)L \approx (5-8)\lambda$ (where L is the detonation cell length). These results are in good agreement with earlier independent measurements by Vasiliev et al. [62]. Taking an average value for $\Delta \approx 6.5\lambda$, Lee's criterion ($t_q \approx t_p$) yields

$$d_c \approx 2\Delta \approx 13\lambda \quad ,$$

which is in agreement with experiments.

The work of Liu et al. [56] on detonation transmission through noncircular orifice plates indicates that the $d_c = 13\lambda$ correlation still holds if d_c denotes an effective diameter d_{eff} (defined as the mean value of the largest and smallest dimensions that characterize the orifice opening), i.e., $d_{eff} \approx 13\lambda$. For a square orifice of side W , the smallest characteristic dimension is W and the largest is the diagonal $\sqrt{2} W$. Thus $d_{eff} = 1/2(1 + \sqrt{2})W = 1.2 W$. From $d_{eff} \approx 13$, $W_c \approx (13/1.2)\lambda \approx 10.8\lambda$ in close agreement with the earlier observation of $W_c = 10\lambda$ by Mitrofanov and Soloukhin [53] and Edwards et al. [54]. Although the time for the rarefaction to attenuate the detonation is controlled by the smallest dimension, when the two characteristic dimensions are not too different, the expansion is three-dimensional. Therefore, the mean value is appropriate to characterize the gasdynamic process.

To generalize the transmission results, Lee [60] has proposed the concept of wave curvature. If the curvature of the diffracted wave created by the rarefaction wave exceeds a certain critical value, the wave will fail. For the three-dimensional expansion associated with arbitrary openings, the maximum curvature (or minimum radius of curvature R for the transformation of a planar detonation into a spherical one) would be of the order of the hydrodynamic thickness Δ (i.e., $R \approx 6.5\lambda$). For rectangular orifices (or slots) of large aspect ratios ($L/W > 7$), the transformation of a planar detonation into a cylindrical one is now controlled by the smallest dimension W . Based on the wave curvature concept, failure of a two-dimensional cylindrical wave will occur for the same critical curvature as the spherical wave, i.e., when the radius of curvature of the cylindrical wave is one half that of the spherical wave. Thus, if $R = \Delta \approx 6.5\lambda$ for the three-dimensional case, $R = \Delta/2 \approx 3.25\lambda$ for the two-dimensional case. This is in agreement with the experimental result of Liu et al. [56] and Benedick et al. [57] that $W_c \approx 3\lambda$.

The critical tube diameter problem provides much insight into the coupling mechanism between gasdynamics and chemical kinetics. The sudden ejection of a detonation from a confined medium into an unconfined environment essentially subjects the wave to a finite perturbation to which it will or will not adjust depending on the severity of the gasdynamic expansion. In a recent study on detonation transmission from rigid tubes to plastic tubes of different thickness to control the degree of gasdynamic expansion, Murray and Lee [63] demonstrated that detonation failure can be related to the wave curvature. Even in rigid tubes, the negative displacement thickness of the boundary layer causes flow divergence similar to the yielding plastic wall experiments of Murray and Lee. Rarefaction waves generated at the walls will propagate towards the tube axis and cause the wave to become curved. For waves with radii of curvature much larger than the critical value, a velocity deficit results. Thus, detonation failure in rigid and plastic tubes, as well as sudden expansion into unconfined space, is one process which differs only by the degree of transverse expansion. Since the lateral expansion causes the wave front to become curved, the criteria for detonation propagation under different boundary conditions can all be formulated using the wave curvature concept.

4. CRITICAL ENERGY FOR DIRECT INITIATION OF DETONATION

Among the possible modes of initiation of detonation, there exist two modes which have been investigated quite extensively: a slow mode when the detonation results from the acceleration of a flame and a fast mode when the detonation is formed almost instantaneously using a sufficiently powerful igniter. The slow mode is usually referred to as the transition from deflagration to detonation (or DDT). Turbulence and interactions between pressure waves and flame are the main flame-acceleration mechanisms that generate the critical states for the onset of detonation. In general, the ignition source plays no role in the transition process. On the other hand, the ignition source plays the dominant role in the fast mode of initiation. The critical states for the onset of detonation in the fast mode are created by the blast wave generated by the igniter. The fast mode is called direct initiation since the detonation is formed

directly without a deflagration- detonation regime. It is also referred to as blast initiation to emphasize the role the blast wave plays in the initiation process. Lee [64] has suggested to call self-initiation the slow mode of transition from deflagration to detonation because the detonation results from the energy released by the combustible mixture itself in the predetonation regime. The parameters that characterize these two modes of initiation are the final steady-state flame velocity and the ratio of the tube diameter to the detonation cell size for self-initiation, and the igniter energy for direct initiation. The basic initiation mechanisms associated with these two modes are quite well understood on a qualitative basis. Quantitative models have also been formulated to predict the initiation parameters. Self-initiation will be discussed in Section 6. The remainder of this section deals with direct initiation.

In direct or blast initiation, the detonation is formed in the immediate vicinity of the igniter. The igniter must be powerful enough to generate a strong shock and maintain the shock above a certain minimum strength for a certain duration. Depending on the strength of the igniter, three initiation regimes have been identified: the subcritical regime, the critical regime and the supercritical regime. In the subcritical regime, the energy of the igniter is below a certain threshold value. The blast wave generated by the igniter progressively decouples from the reaction front and degenerates to a sound wave. The subsequent propagation of the reaction front is identical to an ordinary flame. In the supercritical regime, the igniter energy exceeds the critical threshold value. The blast and the reaction fronts are always coupled in the form of a multi-headed detonation that starts at the source and expands at about the C-J velocity. The most interesting phenomenon is observed in the critical regime when the ignition energy is at the critical threshold value. For very early times, the blast and the reaction fronts are coupled. As the blast expands, decoupling occurs and the reaction front recedes from the shock front until the heat released by combustion begins to contribute significantly to the blast motion. After a quasi-steady regime in which the shock wave and the reaction front propagate as a coupled complex at a constant velocity, a localized explosion occurs in the shocked layer between the shock and the reaction fronts. The blast from the localized explosion immediately forms a multiheaded detonation bubble that grows and engulfs the entire layer of shock-heated mixture resulting in the formation of an asymmetrical detonation. This last stage is referred to as "detonation reestablishment" because after decoupling of the shock and reaction fronts, the multiheaded detonation is regenerated by the localized explosion.

The classification of the direct initiation phenomena into three initiation regimes is based on the total energy of the igniter. The igniter energy can, in general, be readily measured experimentally. For each mixture at given initial conditions, there corresponds a critical energy for direct initiation. The earlier studies [65] on direct initiation were mainly performed in very sensitive fuel-oxygen mixtures which do not require extremely powerful igniters. These studies indicate that the variations of critical initiation energy with mixture composition exhibit a typical U-shape with a minimum around the stoichiometric

composition. The critical initiation energy increases rapidly as the mixture composition departs from the stoichiometric composition. Furthermore, it has been found that the critical energy strongly depends on the igniter characteristics such as the nature of the igniter (e.g., electrical spark, laser spark, exploding wire), the igniter geometry and the energy-time profile of the ignition source. This is a consequence of the nonideal blast wave generated by a real initiation source of finite power density. However, the blast wave generated by an igniter whose time of energy deposition is short compared with the characteristic time of the blast wave R_0/c_0 (where $R_0 = (E_c/p_0)^{1/3}$ is the explosion length, E_c the initiation source energy and c_0 the initial sound speed of the mixture) can be considered as an ideal point blast characterized by the total energy E_c alone. Concentrated solid explosive charges satisfy this condition and have been used extensively to directly initiate detonations in less sensitive mixtures such as fuel-air mixtures. In the early seventies, large-scale experiments in unconfined fuel-air mixtures conducted by Collins et al. [66] and Benedick et al. [67] provided the first measurements of the critical initiation energy of hydrocarbon-air mixtures at NTP. The fuels tested were propane, propylene, butane and MAPP gas. In the following years, increasing interest in unconfined vapor cloud explosions led to the measurement of the critical initiation energy of spherical detonations in stoichiometric hydrocarbon-air mixtures at NTP by Bull et al. [68]. The fuels tested were ethylene, ethane, propane, n-butane and isobutane.

Recently, some data on the critical charge weight for the direct initiation of H_2 -air detonations were reported by Atkinson et al. [69]. Due to the small size of their detonation vessel ($\approx 1.5 \text{ m}^3$), they were limited to a maximum charge weight of only 2.4 g of tetryl and were able to initiate detonation in a stoichiometric H_2 -air mixture (29.6 percent H_2) with a charge of 1.1 g of tetryl. By kinetic modeling, they estimated that a charge of 10 g of tetryl (heat of detonation $\approx 4.27 \text{ kJ/g}$) would initiate mixtures in the concentration range from 22.7 to 49.1 percent H_2 , whereas for a 100 g charge, the corresponding range would be from 17.3 to 56.5 percent H_2 .

In a private communication to Lee in 1982, Elsworth indicated successful initiation by tetryl charges of H_2 -air mixtures over the composition range from 17.3 to 56.5 percent H_2 . The smallest tetryl charge of 1.09 g initiated a slightly fuel-rich mixture (31.5 percent H_2) in slight disagreement with the predictions of the Atkinson model which indicate that the most detonable composition occurs at approximately 33.47 percent H_2 and requires about 1 g of tetryl. Direct initiation of the leanest mixture tested by Elsworth (17.33 percent H_2) requires 36.4 g of tetryl in contrast with the value of about 100 g predicted by the Atkinson model. On the other hand, the richest mixture tested by Elsworth (56.5 percent H_2) was initiated by a tetryl charge of 116 g in good agreement with the estimate of Atkinson et al. However, the experimental accuracy of Elsworth's results is limited by the size of the explosion vessel ($\approx 1.5 \text{ m}^3$) vis-a-vis the initiating charge used. There are indications that at the larger charge weights, the results may represent overdriven conditions and thereby the initiation energy would be underestimated. In view of the specific interest in H_2 detonations in connection with safety

considerations in light water reactors, a series of large-scale experiments were carried out at the field test facility of SNLA. [70] These tests were designed to check and extend Elsworth's results for the direct initiation of unconfined detonations in hydrogen-air mixtures to larger scale and to broader composition limits.

The experiments were carried out in large, vertically mounted, cylindrical polyethylene bags 2.4 m in diameter, 4.2 m long) slightly tapered at the top (to a diameter of 2.7 m). Spherical charges of Comp. C-4 (heat of detonation ≈ 4.87 kJ/g) backed by a booster set off by a bridge wire detonator was attached to the end of a 1-m long thin wooden rod installed vertically inside the deflated bag along the axis of the cylindrical bag. Mounting the charge 1-m above ground level ensured that the ground reflection of the blast wave generated by the charge would not serve as an initiating source so that detonation initiation in the upper portion of the bag would correspond to unconfined initiation. The air in the mounted bag was first removed with a vacuum sweeper to collapse the bag around the charge. The reactive mixture, prepared in an auxiliary tank, was then loaded into the bag up to the local ambient pressure ($p_0 \approx 630$ torr). The charge was then detonated. High-speed cameras provided a go/no-go assessment of detonation initiation. No other diagnostic was used.

The currently available results for the critical energy for direct initiation of spherical detonations in H_2 -air mixtures are presented in Figure 14 and tabulated in Tables 1 and 2. In lean H_2 -air mixtures, Elsworth's results are substantially lower than the Sandia measurements and demonstrate, as suspected, that in the limited size of Elsworth's apparatus (≈ 1.5 m³) and at the high charge levels (> 10 g of tetryl), the wave was still overdriven when it reached the vessel wall. For example, to initiate a detonation in a lean H_2 -air mixture (20 percent H_2), the Sandia tests required a charge of about 30.5 g of Comp. C-4 as compared to about 10 g of tetryl in Elsworth's experiments. For a leaner mixture (17.4 percent H_2), the Sandia measurement increases more than twelvefold with a charge of about 462 g of Comp. C-4. On the rich compositional side, although both sets of results do not overlap directly, the agreement between both sets of data is better. For example, Elsworth mentioned that a tetryl charge of about 117 g would initiate a rich H_2 -air mixture (56.6 percent H_2) which compares well with a charge weight of about 153 g of Comp. C-4 measured in the Sandia tests for a similar H_2 -air mixtures (57 percent H_2).

Presently, there is no direct a priori means to predict quantitatively the magnitude of the critical initiation energy for a given explosive mixture based solely on its thermochemical properties. There do exist, however, a considerable number of phenomenological models which were developed using the physical picture of initiation deduced from experimental observations. The differences between these models lie in the definition of the details of the criticality condition. They are essentially variations of the original postulate of Zeldovich et al. [51] formulated more than thirty years ago. According to Zeldovich, in direct initiation of a detonation, the ignition source must generate a spherically divergent shock wave (i.e., a blast wave) of sufficient strength and adequate duration for the chemical reactions, occurring in the expanding flow field behind the blast

Figure 14. Variation of Critical Energy For Direct Initiation of Spherical Detonation With Fuel Percentage in H₂-Air-CO₂ Mixtures at NTP

Table 1
 Elsworth's Results ($p_0 = 760$ torr)

% H ₂ in air	ϕ	Charge Weight g (tetryl) ^a	Detonation
17.36	0.5	26.50	NO
"	"	36.40	YES
20.13	0.6	6.95	NO
"	"	9.55	YES
22.73	0.7	2.72	NO
"	"	3.73	YES
23.96	0.75	1.92	NO
"	"	2.64	YES
25.16	0.8	1.41	NO
"	"	1.94	YES
27.44	0.9	0.97	NO
"	"	1.33	YES
28.53	0.95	0.86	NO
"	"	1.18	YES
29.57	1.0	0.80	NO
"	"	1.10	YES
30.61	1.05	0.79	NO
"	"	1.09	YES
31.61	1.10	0.80	NO
"	"	1.10	YES
32.58	1.15	0.83	NO
"	"	1.14	YES
33.52	1.20	0.88	NO
"	"	1.22	YES
35.33	1.30	1.03	NO
"	"	1.41	YES
36.19	1.35	1.13	NO
"	"	1.56	YES

^aHeat of detonation = 4.27 kJ/g

Table 1 (Continued)

Elsworth's Results ($p_0 = 760$ torr)

% H ₂ in air	ϕ	Charge Weight g (tetryl) ^a	Detonation
37.04	1.40	1.27	NO
"	"	1.75	YES
37.86	1.45	1.42	NO
"	"	1.96	YES
39.44	1.55	1.79	NO
"	"	2.46	YES
40.20	1.60	1.97	NO
"	"	2.71	YES
41.67	1.70	2.59	NO
"	"	3.56	YES
43.06	1.80	3.34	NO
"	"	4.59	YES
44.39	1.90	4.20	NO
"	"	5.77	YES
45.66	2.00	5.47	NO
"	"	7.51	YES
48.60	2.25	10.20	NO
"	"	14.10	YES
51.23	2.50	19.60	NO
"	"	26.90	YES
56.57	3.10	84.80	NO
"	"	116.60	YES

^aHeat of detonation = 4.27 kJ/g

Table 2
Sandia Results ($p_o \sim 630$ torr)

Shot #	% H ₂ in Air	ϕ	Charge Weight g (Comp. C-4) ^a	Detonation
C-01	20.0	0.59	30.5	YES
C-02	20.0	0.59	14.5	NO
C-03	18.5	0.54	60.5	NO
C-04	18.5	0.54	88.5	NO
C-05	18.5	0.54	151.0	YES
C-06	17.4	0.50	461.7	YES
C-07	16.7	0.48	461.0	NO
C-08	58.1	3.30	461.0	YES
C-09	60.0	3.57	461.0	YES
C-10	57.0	3.15	152.7	YES
C-11	60.5	3.65	152.7	NO

^aHeat of detonation = 4.87 kJ/g

wave, to couple with the shock wave and develop into a spherical detonation. In essence, the criticality condition amounts to a balance between a characteristic chemical time scale and the corresponding hydrodynamic time scale. The initiation criterion that results from such a balance is then invariably expressed in terms of the energetics of the source. Choosing the induction time τ as the characteristic chemical time scale, Zeldovich postulated that for direct initiation, the time for the blast wave to decay to the C-J Mach number M_{CJ} must be at least equal to the induction time of the mixture. From the above criterion, Zeldovich demonstrated that the critical energy for direct initiation of spherical detonations must be proportional to the cube of the induction time, i.e.,

$$E_c \sim \tau^3$$

Since a comprehensive review of existing phenomenological models can be found in Reference 70, only the model which provides the best estimates of critical initiation energy data will be discussed in the present report.

The surface energy model developed by Lee et al. [37] relates the point blast initiation mode with the planar detonation wave initiation mode. The model postulates that in point blast initiation of a spherical detonation, at criticality (i.e., when the chemical energy release becomes equal to the igniter energy) the blast wave has decayed to the C-J state from its overdriven state. Most of the reacted mass is concentrated in a thin layer behind the strong blast wave which has expanded to a radius R_s^* . To estimate the critical blast radius R_s^* , Lee et al. further postulated the equivalence of the surface energy in point blast initiation and planar detonation wave initiation. In the latter mode, the initiation of an unconfined spherical detonation by transmission of a confined planar detonation requires a critical tube diameter $d_c \approx 13\lambda$. At criticality, equating the surface energies for both modes of initiation yields an estimate for the critical blast radius R_s^* , viz.

$$4\pi R_s^{*2} = \pi d_c^2/4$$

or

$$R_s^* = d_c/4$$

Combining Taylor's solution for the trajectory of a strong blast wave,

$$R_s = A t^{2/5} ,$$

with the shock strength,

$$R_s = c_o M_s = 2A/5t^{3/5} ,$$

yields the critical initiation energy,

$$E_c = 4\pi\gamma_o p_o M^2 I R_s^3 ,$$

where

$$A = (25 E_c / 16\pi\rho_o I)^{1/5}$$

I is the blast energy integral, γ , ρ and p are the specific heat ratio, density, and pressure, respectively, and subscript o characterizes the unshocked mixture. At criticality, where $R_s = R_s^*$ and $M_s = M_{CJ}$, the critical initiation energy can be written as

$$E_c = \frac{\pi}{16} \gamma_o p_o I M_{CJ}^2 d_c^3 ,$$

or using the empirical law $d_c \approx 13 \lambda$,

$$E_c = \frac{2197}{16} \pi \gamma_o p_o I M_{CJ}^2 \lambda^3 .$$

The currently available data for the critical energy for the direct initiation of spherical detonations in H₂-air mixtures presented in Figure 14 are well predicted by the surface energy model, especially in fuel-lean mixtures. The surface energy model has also been used to estimate the critical initiation energies of H₂-air-CO₂ mixtures. The results are compared in Figure 14 with the predictions for undiluted H₂-air mixtures. For stoichiometric composition (i.e., 29.6 percent H₂), the addition of 5, 10, and 15 percent CO₂ increases the critical initiation energy of the undiluted mixture by factors of 3, 22 and 2100, respectively due to the cubic dependence of E_c on cell size λ . For comparison, critical initiation energy results for a range of hydrocarbon-air mixtures are shown in Figures 15 and 16. Figure 15 presents critical initiation energy data for the ethylene-air system derived from the experiments of Elsworth* and Murray et al. [71] The agreement between both sets of data is good. Figure 16 presents the initiation energy measurements of Elsworth* for the heavier hydrocarbon-air mixtures (i.e., C₂H₆-air, C₃H₈-air, and C₄H₁₀-air). The detonation sensitivities for the alkane-air mixtures are quite comparable

*Private Communication, J. E. Elsworth, 1982.

Figure 15. Variation of Critical Energy for Direct Initiation of Spherical Detonation With Fuel Percentage in Ethylene-Air Mixtures at NTP

Figure 16. Variation of Critical Energy For Direct Initiation of Spherical Detonation With Fuel Percentage in Alkane (C_2H_6 , C_3H_8 , and C_4H_{10})-Air Mixtures at NTP

and they are all presented together. Once again the experimental results are well correlated by the surface energy model except for the rich mixtures of the ethylene-air system where substantial deviations are observed.

The surface energy model has also been used to estimate the relative detonation sensitivity of various fuels. As proposed by Matsui and Lee, [72] the relative detonation sensitivity can be characterized by a dimensionless parameter D_H defined as the ratio of the critical initiation energy of a fuel-oxidizer mixture to that of the most detonable common gaseous mixture, namely, the stoichiometric acetylene-oxygen mixture. The results have been tabulated in Table 3 for the stoichiometric compositions. The values of D_H for fuel-air mixtures increase from about $D_H = 10^5$ for C_2H_2 to $D_H \approx 8 \times 10^7$ for the alkanes (C_2H_6 , C_3H_8 , and C_4H_{10}). With a value $D_H \approx 10^6$, H_2 is approximately 10 times less sensitive than C_2H_2 but 10 times more sensitive than C_2H_4 . However, a stoichiometric H_2 -air mixture diluted with 5 percent CO_2 is only two times more sensitive than a stoichiometric C_2H_4 -air mixture. With 10 percent CO_2 dilution, H_2 -air- CO_2 mixtures become slightly less sensitive than C_2H_4 -air mixtures. The addition of 15 percent CO_2 makes H_2 -air- CO_2 mixtures 40 times less sensitive than the alkane-air mixtures. However, because of the cubic dependence of the critical initiation energy, E_c , on the detonation cell size, λ , and the increasing difficulty in measuring cell sizes accurately in the less reactive mixtures, direct measurements of E_c should be performed to provide reliable assessments of the detonation sensitivity of reactive mixtures.

5. DETONABILITY LIMITS

By definition, the detonability limits of a reactive mixture are the critical conditions for the propagation of self-sustained detonation. The critical conditions denote both the initial and boundary conditions of the explosive mixture. The initial conditions are the nature of the fuel, its concentration, the initial thermodynamic state, the initial fluid mechanical state (i.e., mean flow and turbulence characteristics), the ignition source properties and all other relevant parameters which characterize the explosive mixture prior to ignition. The boundary conditions are the size and geometry of the volume of explosive, the degree of confinement, the surface topology of the confining walls, as well as all other relevant parameters characteristic of the boundary of the explosive. At present, no theory can predict detonability limits from first principles. Moreover, the experimental determination of the limits suffers from the lack of a unique definition of an experimental criterion. Whereas the flammability tube of Coward and Jones [73] is usually regarded as the standard apparatus to determine flammability limits, no standard detonation tube and procedure exist for measuring detonability limits. Detonability limit experiments are usually performed in long tubes using powerful igniters. The limits are usually defined as the critical fuel concentrations beyond which a detonation can no longer propagate in a given tube.

Detonability limit data are usually reported without reference to the experimental conditions although it is generally acknowledged that

Table 3
Relative Detonation Sensitivity

Mixture	Fuel (%)	E_c (Joules)	D_H
C_2H_2/O_2	28.57	427	1.
C_2H_2/Air	7.75	427	1.67×10^5
H_2/Air	29.6	4270	1.67×10^6
C_2H_4/Air	6.54	4.27×10^4	1.67×10^7
C_2H_6/Air	5.66	2.135×10^5	8.33×10^7
C_3H_8/Air	4.03	2.135×10^5	8.33×10^7
C_4H_{10}/Air	3.13	2.135×10^5	8.33×10^7

$H_2-Air-CO_2$			
Fuel (%)	CO_2 (%)	E_c (Joules)	D_H
29.6	5	1.37×10^4	5.35×10^6
29.6	10	9.55×10^4	3.73×10^7
29.6	15	8.55×10^6	3.34×10^9

detonability limits are dependent on the apparatus (e.g., tube diameter) and ignition source strength.

For example, Kogarko and Zeldovich [42] were able to observe an increase in the range of detonability limits of the H_2 -air system from 18.3 to 59 percent H_2 [41] to 15 to 63.5 percent H_2 by initiating detonations in larger diameter tubes (30.5 cm instead of 1.4 cm). Under the present research program, the detonability limit range of the H_2 -air system has been further extended from 13.6 to 70 percent H_2 using a 43-cm-diameter tube at SNLA [40] as discussed in Section 2. Hence, detonability limits of confined mixtures are strongly dependent on the boundary conditions of the medium in which the detonation propagates. In other words, detonability limits of confined mixtures are not unique.

On the other hand, the detonability limits of unconfined mixtures should be regarded as the "true" detonability limits of the mixture since spherical and cylindrical detonations should be, in principle, free from boundary effects. In practice, the experimental determination of the detonability limits of unconfined mixtures cannot be rid of the influence of the characteristics of the initiation source. Therefore, it is only meaningful to speak of detonability limits associated with a given apparatus (e.g., in a smooth cylindrical tube of a given diameter) using a certain reasonable criterion. We shall first discuss detonability limit criteria for confined mixtures.

Meaningful criteria for the detonability limits in a specific apparatus have been formulated based on certain identifiable characteristics of the detonation as the limits are approached. Whereas in small tubes, heat and momentum losses through the boundary layer are responsible for detonability limits, this mechanism has negligible effect in large tubes. It has been observed that, in a given smooth circular tube, as the mixture composition becomes leaner or richer, a multi-headed, self-sustained detonation is finally replaced by a single-head spinning detonation propagating at about the C-J velocity. However, using powerful igniters, single-head spinning detonations can be propagated over a wide range of mixture compositions beyond the composition for the onset of single head spin. The experiments of Wolanski et al. [74] on methane-air detonations are a typical example. These authors observed the propagation of single-head spins in a 6-m long, 6.35-cm square tube over the composition range from 8 to 14.5 percent CH_4 without any noticeable attenuation. Hence, there exists in a given tube a range of compositions between the onset of the fundamental spinning mode and the composition beyond which a detonation can no longer be initiated. Over this range, a single-head spinning detonation can be propagated without attenuation. This range of compositions will be referred to as the "detonation-like" regime as opposed to the "self-sustained" detonation regime which corresponds to multi-headed detonations. The onset of single-head spinning detonation defines the boundary of the two regimes. Being the lowest possible stable mode in a given tube, the single-head spinning detonation compositions were proposed long ago by Dove and Wagner [18] as the detonability limits of confined mixtures.

The recent studies of Moen et al. [75] on the stability of near-limit detonations indicate that the detonation structure in the "detonation-like" regime is unstable to finite perturbations. Using a short length of Shchelkin spiral to perturb the detonation, Moen et al. observed that the single-head spinning detonation recovers its structure immediately after the obstacles for mixture compositions near the onset of single-head spin only. For mixtures beyond the critical composition, the detonation fails, then recovers its spinning structure far downstream of the Shchelkin spiral. This behavior is similar to the galloping detonations observed by Mooradian and Gordon, [76] Saint-Cloud et al., [77] and Edwards and Morgan. [78] The "galloping" mode is a longitudinal mode with periodic destruction and formation of the detonation. As reported by Urtiew and Oppenheim, [79] the reformation process is identical to the transition from deflagration to detonation. Therefore, it is quite reasonable to define the detonability limits of confined mixtures as the compositions for the onset of single-head spinning detonations.

As early as 1948, Kogarko and Zeldovich [42] proposed without any derivation that, at the onset of single-head spin in a smooth tube, the detonation cell width λ must be equal to the tube circumference, i.e., $\lambda = \pi D$. Recently, Lee [80] provided a derivation of the $\lambda = \pi D$ criterion. Lee argued that since the tube circumference πD represents the largest characteristic dimension for the tube, the longest characteristic acoustic time is $\pi D/c$, where c is the sound speed of the detonation products. Assuming a resonant coupling between the acoustic vibration and the periodic chemical processes in the detonation cell, the characteristic acoustic time, $\pi D/c$ must be equal to the characteristic chemical time, λ/c , i.e., $\lambda = \pi D$. The $\lambda = \pi D$ criterion is in the same spirit as the criterion $\lambda = D$ proposed by Shchelkin [81] for the existence of a single-head spin in a circular tube. Another limit criterion $\lambda = 1.7D$ was derived by Moen et al. [75] based on Manson [15] and Fay [16] acoustic theories. These theories predict the spin pitch to diameter ratio, viz., $P/D = \pi U/K_1 c$ where U is the C-J detonation velocity and $K_1 = 1.841$ is the first root of the derivative of the Bessel function of order one. Since for most fuel-air mixtures, $U/c \approx 1.6$, Moen et al. arrived at the limit criterion $\lambda \approx 1.7D$ by assuming that the spin pitch corresponds to the cell length ($P \approx 1.6\lambda$). It should be pointed out that there is no basis for this assumption. For a given tube diameter, the detonability limits are wider using the $\lambda = \pi D$ criterion than the $\lambda = 1.7D$ criterion. In other words, the onset of single-head spin occurs in less sensitive mixtures according to the $\lambda = \pi D$ criterion than those predicted by the $\lambda = 1.7D$ criterion. Irrespective of the different criteria stated above, all criteria indicate that the detonability limits in smooth round tubes occur when the tube diameter is of the order of the cell size λ .

Recent experiments by Dupre et al. [82] suggest that the $\lambda = \pi D$ criterion could be the most reasonable one. The experiments were conducted in lean hydrogen-air mixtures (15 to 25 percent H_2) at NTP initially in five cylindrical smooth tubes of decreasing diameters (15.2 to 3.8 cm). The aspect ratios L/D (length to diameter) were greater than 37 to ensure that the propagation of the detonation was stable. Ion gauges and pressure

transducers were used to measure detonation velocity and pressure. The detonation structure was monitored in each tube by using long smoked foils ($\ell/D > 14$, where ℓ is the smoked foil length). A schematic of the apparatus is shown in Figure 17.

For mixtures less sensitive than that specified by the criterion $\lambda = \pi D$ (i.e., for $\lambda > \pi D$), Dupre et al. found that the detonation is highly unstable to finite perturbations with velocity fluctuations in excess of 10 percent from the C-J value. These results are shown in Figure 18 where the velocity fluctuations, $\Delta V/V_{CJ} = 1 - V/V_{CJ}$ (V being the measured velocity), have been plotted with respect to the cell size, λ , normalized with the tube diameter, D . By reducing the degree of confinement using thin plastic wall tubes, Murray [83] observed detonation failure when the velocity deficit $\Delta V/V_{CJ}$ exceeds 10 percent. It may be concluded that for every reactive mixture, one can infer the detonability limits for stable propagation of a detonation initiated by a strong source in a cylindrical rigid smooth-walled tube in terms of the limiting tube diameter, D^* , which can be estimated directly from detonation cell size data using the relationship

$$D^* = \lambda/\pi$$

Estimates of D^* for H_2 -air, H_2 -air- CO_2 (at $25^\circ C$) and H_2 -air-steam mixtures (at $100^\circ C$) are plotted in Figures 19 and 20 as a function of the fuel concentration. The values of D^* exhibit the same U-shape as the detonation cell size data shown in Figures 6 and 8 since they represent a λ/π scaling of the cell data. For the stoichiometric composition at NTP (29.6 percent H_2), the minimum tube diameter is $D^* \approx 0.5$ cm. With 15 percent CO_2 dilution, this value increases by one order of magnitude to $D^* \approx 6.1$ cm. For hot H_2 -air mixtures (at $100^\circ C$ initially) the minimum tube diameter for the stoichiometric composition is $D^* \approx 0.16$ cm. With 30 percent steam dilution, this value increases by a factor of 60 to $D^* \approx 9.6$ cm. For the hydrocarbon-air mixtures at the stoichiometric composition, the minimum tube diameters are $D^* \approx 0.19$ cm for C_2H_2 , $D^* \approx 0.83$ cm for C_2H_4 and $D^* \approx 1.7$ cm for the alkanes. With a cell size $\lambda \approx 33$ cm for stoichiometric CH_4 -air mixtures, [84] the minimum tube diameter for stable propagation of a detonation should be $D^* \approx 10.5$ cm. Therefore, the spinning detonations observed by Wolanski et al. [74] in a 6.35 cm tube should correspond to overdriven transient waves. This is supported by the fact that they observed single-head spinning waves over the whole range of fuel concentration studies. Based on stability considerations, the composition limits of confined mixtures initiated by a strong source in circular tubes can now at least be defined experimentally.

It should be noted that the $\lambda = \pi D$ criterion applies to a detonation initiated by a strong source in a cylindrical rigid, smooth-walled tube. For weak initiation sources, like in deflagration to detonation transition (DDT), it is reasonable to expect that the limits should correspond to more sensitive mixture compositions. The recent experiments on DDT of Knystautas et al. [85] discussed in Section 6 indicate that the detonability limit criterion for transition in a cylindrical smooth-walled tube is $\lambda \approx D$ in agreement with Shchelkin criterion. [81]

Figure 17. Schematic of Experimental Set Up For Detonability Limit Studies

Figure 18. Variation of Velocity Fluctuations With Detonation Cell Size

Figure 19. Variation of Limiting Tube Diameter With Fuel Percentage in H_2 -Air and H_2 -Air- CO_2 Mixtures at NTP

Figure 20. Variation of Limiting Tube Diameter With Fuel Percentage in Hot H_2 -Air-Steam Mixtures at Superatmospheric Initial Pressures

For two-dimensional planar channels, Vasiliev [86] recently found that the critical condition for a stable wave corresponds to a channel width, W , equal to the detonation cell size, λ . Therefore, $W = \lambda$ represents the criterion for the detonability limits of mixtures confined in two-dimensional channels of large aspect ratios. Limit criteria for other tube geometries have yet to be established.

The criterion for the detonability limits for unconfined cylindrical and spherical detonations is more difficult to formulate. With the absence of boundaries, composition limits should mainly depend on the detailed processes of the cellular structure. Since the average cell size for a given mixture must be constant for the stable propagation of a detonation, the number of detonation cells in diverging detonations must increase at the same rate as the surface area increases. Therefore, failure of the cells to multiply at the critical rate might be used to define the composition limits. Because the mechanism for cell multiplication results from the coupling of the complex shock pattern (double Mach reflections) with the flame front, no quantitative theory has been formulated so far to predict the detonability limits for unconfined detonations. Furthermore, no experimental method has been devised to measure limits for diverging waves. The composition limits of spherical detonations are usually determined from the U-shaped curve of initiation energy versus fuel concentration by choosing some arbitrary value for the maximum initiating energy. Because of the extremely rapid increase in the initiation energy as the limits are approached, the actual values for the limiting composition are rather insensitive to the maximum value of critical charge weight chosen. This is an indirect way to link the composition limits to the detonation cell size. However, there is no doubt that the limit conditions should be directly correlated to the fundamental chemical length scale of the mixture, i.e., the detonation cell size.

6. TRANSITION FROM DEFLAGRATION TO DETONATION

Because very powerful ignition sources such as solid explosives are usually required to initiate unconfined fuel-air detonations, most accident scenarios assign a very low probability to the detonation mode in accidental explosions. However, detonations cannot be completely ruled out, since a flame can accelerate to a sufficiently high speed and transit to detonation. An explosive mixture at given initial and boundary conditions is expected to undergo transition from deflagration to detonation (DDT) if the following conditions are satisfied: (1) The mixture composition must be within the detonability limits, and (2) powerful acceleration mechanisms such as turbulence generated by obstacles and adequate boundary conditions (confinement) must be present. Therefore, the transition phenomenon involves detonability limits, flame acceleration mechanisms and the mechanisms responsible for the onset of detonation itself. Detonability limits have been discussed in Section 5.

The different flame acceleration mechanisms are fairly well understood on an individual basis, [87, 88] but their effects when they exist simultaneously are still not clear. A future report on hydrogen-air deflagrations will provide a detailed review on flame acceleration processes. The most powerful flame acceleration mechanism is the

continuous generation of large-scale flame folds which can be provided by either multiple shock wave-flame interactions (Taylor's interface instability [89]) or by placing periodically spaced large obstacles in the path of the flame. As the flame accelerates, it finally reaches a maximum velocity at which abrupt transition to detonation occurs.

All methods of initiation of a detonation (i.e., DDT, blast initiation, photochemical initiation [90] and initiation by a hot jet of combustion products [91]) indicate that the onset of detonation consists of two phases. The first phase is the creation of the critical conditions for the onset of detonation, whereas the second phase is the actual formation of the detonation wave itself. In transition from deflagration to detonation, the critical states are achieved by the positive feedback mechanism which couples the flow field and the accelerating flame. The critical states are characterized by the formation of auto-explosion centers and the amplification of the blast waves from the auto-explosions to form a detonation wavelet. The formation of auto-explosion centers in DDT is caused by localized fluctuations due to turbulence in a region which has been brought close to its auto-ignition limit by turbulent mixing with hot combustion products. The mechanism responsible for the amplification of the blast waves created by the auto-explosions is given by the Rayleigh's criterion. [92] More explicitly, the mechanism is the Shock Wave Amplification by Coherent Energy Release [90] (the so-called SWACER mechanism). In other words, the region surrounding the initial auto-explosion center releases its chemical energy in phase with the expanding shock wave. The SWACER mechanism requires the existence of spatial gradients in induction time over a finite distance (or coherence length) in order for the shock wave and the reaction zone to become self-coherent and amplify to a detonation wavelet.

Under a joint program between McGill University and Sandia National Laboratories, an extensive experimental study has been undertaken to investigate the flame acceleration processes in confined H_2 -air mixtures over a wide range of mixture compositions in order to formulate some criteria for the onset of DDT. In this section, the results of small-scale experiments on flame acceleration by repeated obstacles carried out at McGill [85, 93] will be discussed. The corresponding large-scale experiments conducted at SNLA will be discussed in a future report on H_2 -air deflagrations. Some accidental DDT resulting from turbulent jet mixing in large-scale experiments will also be reviewed. To put the present results in proper perspective, a brief review of earlier studies on turbulent deflagrations propagating in smooth and rough tubes will be given first.

While studying flame propagation in long smooth tubes, Mallard and Le Chatelier [94] and Berthelot and Vieille [95] observed flame acceleration and abrupt transition to a steady-state supersonic combustion wave and thus discovered the phenomenon of detonation. A typical self-luminous streak photograph of an accelerating flame and the abrupt transition to detonation in a smooth tube is shown in Figure 21. In these studies, little interest was shown for the steady-state high-speed deflagrations reached in less sensitive mixtures in which DDT was not achieved. Chapman and Wheeler [96] were the first to study flame propagation in rough tubes.

Figure 21. Self-Luminous Streak Photograph of Accelerating Flame and Abrupt Transition to Detonation in a Smooth Tube

By using a sequence of orifice plates spaced about one diameter apart along the tube to roughen the walls, they observed turbulent deflagration speeds of about 420 m/s in methane-air mixtures in a 5-cm-diameter tube. Similar studies by Shchelkhin [97] in tubes roughened by insertion of a wire spiral indicated a significant reduction in the transition distance from flame to detonation and steady-state detonation velocities in the rough tube as low as half the normal C-J detonation value. These low velocity detonations (or quasi-detonations) were studied later by Guenoche and Manson [98] and Brochet. [99] Recent interests in high-speed turbulent deflagrations in relation with vapour cloud explosions have prompted further studies of flame propagation in rough tubes. [85, 93, 100-106] A typical streak photograph of the flame acceleration process and the final steady turbulent deflagration in a CH₄-air mixture in a 4-cm-diameter tube partially blocked by a sequence of orifice plates (3 cm in diameter) spaced about 19 cm apart, is shown in Figure 22. In contrast to Figure 21, the approach to the final steady-state flame speed is continuous with no distinct transition from one regime (deflagration) to another (detonation). The final steady-state deflagration speed in Figure 22 is about 770 m/s whereas in a smooth tube, the turbulent deflagration speed for CH₄-air mixtures is typically a few tens of meters per second. It is interesting to note in Figure 22 that the slope of the trajectory of the deflagration front is more or less parallel to the trajectories of the sound waves in the burnt gases. This indicates that the flow of burnt gases is moving at the local sonic velocity with respect to the deflagration front. In other words, the flow of combustion products is choked. This regime of flame propagation will be discussed in detail later in this section.

Under the joint research program between McGill University and SNLA, small-scale experiments [85, 93] were carried out in long steel tubes (11 to 19 m in length) of diameters ranging from 5 to 30 cm. Wire spirals and orifice plates were used to roughen the walls. The orifice plates were spaced about one tube diameter apart and the wall roughness was controlled by the blockage ratio BR of the orifice plate ($BR = 1 - d^2/D^2$, where d and D are the orifice and tube diameters, respectively). Similarly for the spirals, the pitch was about one tube diameter and the wall roughness was controlled by the wire diameter of the spiral. The blockage ratios were 0.43 for the orifice plates and 0.44 for the spiral. The spiral coil in the 5 cm diameter tube was 3 m long. The orifice plates extended for distances of 6 and 9 m in the 5- and 15-cm-diameter tubes, respectively. In the 30-cm-diameter tube, the obstacle field extended over the whole length of the tube, i.e., 17 m. A range of fuels (CH₄, C₂H₂, C₂H₄, C₃H₈, and H₂) of different compositions in air at NTP initially were used. Flame speeds were measured by ionization probes spaced about 0.5 m apart along the tube. Pressure transducers were used to record the pressure-time profiles at various locations along the tube. A schematic of the experimental apparatus is given in Figure 23.

The behaviour of the flame propagation in the rough tubes, (i.e., within the obstacle field) will be discussed first. Two different behaviours of flame propagation were observed. Upon ignition, the flame accelerates but after propagation through a certain number of obstacles, the flame either extinguishes itself or approaches a steady-state velocity. The regime of

Figure 22. Streak Photograph of Flame Acceleration and Final Steady Turbulent Deflagration in Methane-Air Mixture

Figure 23. Schematic of Experimental Apparatus for Studies on Transition From Deflagration to Detonation

flame extinction has been called the "self-quenching" regime. This regime was observed in near-limit compositions of the less sensitive mixtures (i.e., CH_4 , C_3H_8 , and C_2H_4 -air). It has been established [107] that quenching is due to the rapid turbulent mixing between the hot product gases and the cold unburned mixture as the flame exits the orifice.

With increasing fuel concentration, the flame achieves a final steady-state velocity in the obstacle field. It should be emphasized that this steady-state velocity has been averaged over a distance of about half to one meter (depending on the tube diameter). Thus the flame velocity is averaged over at least three or four obstacles even for the largest tube diameter (i.e., the 30-cm-diameter tube). The local combustion phenomenon is extremely complex and it is meaningless to speak of a local flame velocity. Figure 24 shows typical plots of flame speed versus fuel concentration for H_2 -air mixtures in the three tubes (5, 15, and 30 cm in diameter) with orifice plates spaced one diameter apart and blockage ratio $\text{BR} = 0.43$. Depending on the magnitude of the final steady-state velocity of the flame, several distinct regimes of flame propagation can be identified in Figure 24. The transition from one regime to another appears quite distinct and corresponds to critical values of the fuel concentration for a given tube diameter and obstacle configuration. Furthermore, Figure 24 indicates that not all regimes can be observed for a given tube and obstacle configuration. For the same blockage ratio ($\text{BR} = 0.43$), two regimes were observed in both 5- and 30-cm-diameter tubes whereas a third regime was also observed in the 15-cm-diameter tube. In very lean mixtures (≤ 12.5 percent H_2) a weak turbulent deflagration regime characterized by steady-state flame speeds of the order of a few tens of meters per second was identified. As the fuel concentration was increased, a second regime with steady flame velocities of the order of several hundred meters per second ($V \approx 800$ to 1000 m/s) occurs. In this regime, the flame trajectory is approximately parallel to the trajectories of the sound waves in the combustion products as shown in Figure 22. This indicates that the velocity of the combustion products with respect to the flame is sonic. In other words, the flow is choked. Therefore this regime has been called the "sonic" or "choking" regime. This regime was observed in the three tubes. In the 5-cm-diameter tube, this regime occurs in the fuel range from 10 to 20 percent H_2 on the fuel-lean side and from 47.5 to 55 percent H_2 (the limit of the present experiment) on the fuel-rich side. In the 15-cm-diameter tube, the fuel range in the choking regime extends from 12.5 to 18 percent H_2 on the fuel-lean side and from 58 to 70 percent H_2 (the limit of the present experiment) on the fuel-rich side. In the 30-cm-diameter tube, the choking regime occurs on the fuel-lean side from 12.5 to 16 percent H_2 and on the fuel-rich side from 60 to 70 percent H_2 (the limit of the present experiment). It is interesting to note that the steady flame speeds in the choking regime are nearly equal to the isobaric sound speeds of the combustion products which have also been plotted in Figure 24. This observation provides a good approximation for estimating flame speeds in the choking regime.

With further increase in mixture sensitivity, a third regime characterized by steady velocities $V > 1100$ m/s but substantially below the normal C-J detonation velocity of the mixture ($V_{\text{CJ}} - V \approx 200$ to 500 m/s) was observed. This is the regime of quasi-detonations already studied by

Figure 24. Variation of Thermal Flame Velocity With Fuel Percentage in H₂-Air Mixtures at NTP

Guenoche and Manson [98] and Brochet. [99] The large velocity deficits in the quasi-detonation regime can be attributed to severe momentum and heat losses to the walls caused by the obstacles. This regime was observed in the three tubes in the fuel concentration ranges from 25 to 47.5 percent H_2 in the 5-cm-diameter tube, from 18.5 to 55 percent H_2 in the 15-cm-diameter tube and from 16 to 60 percent H_2 in the 30-cm-diameter tube. Similar results were obtained in the 5-cm-diameter tube roughened by a wire spiral ($BR = 0.44$). [98] For clarity, these data have not been plotted in Figure 24.

The steady flame speeds for H_2 -air mixtures in the obstacle field of the 5- and 15-cm-diameter tubes are compared with the corresponding results for mixtures of the common hydrocarbons (CH_4 , C_2H_2 , C_2H_4 , and C_3H_8) and air in Figures 25 and 26 where the steady flame speeds have been plotted with respect to the equivalence ratio. For a given fuel, in a given tube with a given obstacle configuration, it can be seen that not all flame propagation regimes can be achieved. In both tubes, the low-deflagration regime and the quasi-detonation regime were not observed in the least sensitive mixtures, i.e., the methane-air mixtures. On the other hand, the self-quenching regime was never observed in the most sensitive mixtures, i.e., acetylene-air and hydrogen-air mixtures. A discussion of the mechanisms of flame propagation in the different regimes has been given by Lee [108] and will be reviewed in a future report on hydrogen-air deflagrations.

The studies on turbulent deflagration propagation reported above have allowed the formulation of a criterion for DDT in rough tubes. [85] The conditions under which DDT can occur in a rough tube have been assessed by estimating the ratio of the characteristic length scales for stable propagation of a detonation wave through obstacles. The relevant physical length scale is the transverse dimension d of the orifice obstacle. The characteristic chemical length scale is the detonation cell size, λ , of the explosive mixture. From the experiments carried out so far, it appears that transition to detonation within the obstacles occurs when $\lambda/d \leq 1$. In other words, when $\lambda/d > 1$, the flame propagation is in the choking regime where the flame speed is of the order of the isobaric sound speed of the combustion products. However, when the mixture becomes more sensitive (i.e., when λ decreases), then an abrupt transition to the quasi-detonation regime occurs when $\lambda/d \leq 1$. Table 4 gives the experimental values for λ/d for DDT. Except for C_2H_2 (where a discrepancy in cell size not yet resolved yields a value $\lambda/d \approx 0.5$ for DDT), the $\lambda/d \leq 1$ criterion is confirmed for the other fuels within experimental errors in cell size measurements. For H_2 -air mixtures, DDT occurs in the 5-, 15-, and 30-cm-diameter tubes at 22, 18, and 16 percent H_2 on the fuel-lean side and 47.5, 57, and 60 percent H_2 on the fuel-rich side, respectively. It should be noted that the value of the quasi-detonation velocity may not differ much from that of the choking regime for a given tube and orifice diameter. The transition between the two regimes occurs at a critical value of the fuel concentration indicating a changeover in the dominant propagation mechanism. Lee [108] has identified this as a detonation regime on the basis of the transition criterion $\lambda/d \leq 1$. The criterion for detonability limits in tubes postulated by Schelkin [81] states that $\lambda/d \approx 1$ since the tube diameter must at least accomodate one detonation

Figure 25. Variation of Terminal Flame Velocity With Equivalence Ratio in H_2 -Air and Hydrocarbon (CH_4 , C_2H_2 , C_2H_4 , and C_3H_8)-Air Mixtures at NTP in 5-cm-Diameter Tube

Figure 26. Variation of Terminal Flame Velocity With Equivalence Ratio In H₂-Air and Hydrocarbon (CH₄, C₂H₂, C₂H₄, and C₃H₈)-Air Mixtures at NTP in 5-cm-Diameter Tube

Table 4
Transition in Rough-Walled Tube

	Mixture	Concentration (%)	λ (mm)	λ/d
D = 5 cm d = 3.74 cm (BR = 0.43)	C ₂ H ₂ -Air	4.75	19.8	0.51
	H ₂ -Air	22	30.7	0.82
		47.5	41.2	1.10
	C ₂ H ₄ -Air	6	37.8	1.01
		9	30.1	0.81
	C ₂ H ₂ -Air	4	58.3	0.51
D = 15 cm d = 11.4 cm (BR = 0.43)	H ₂ -Air	18	111	0.97
		57	120	1.05
	C ₂ H ₄ -Air	4.5	100	0.88
		13.5	115	1.01
	C ₃ H ₈ -Air	3.25	112	0.98
		5.5	116	1.02
D = 30 cm d = 22.6 cm (BR = 0.43)	H ₂ -Air	16.0	245	1.08
		60.0	189.2	0.84

cell. Schelkin's criterion is supported by Vasiliev's observation [86] that self-sustained detonations can only propagate in a channel of width $W \geq \lambda$. Thus the transition to the quasi-detonation regime for $\lambda/d \lesssim 1$ suggests that the wave is a detonation. However, the severe momentum losses reduces its velocity to a value which can be significantly below the C-J detonation velocity. The fact that the orifice diameter is of the order of the detonation cell size implies that the detonation mechanism dominates. A theory has yet to be developed to predict the quasi-detonation velocity for a given mixture with prescribed momentum losses. The necessary conditions for transition to detonation in a rough tube are now clear. First, the flame must achieve a high supersonic velocity of the order of the isobaric sound speed of the burnt products. Furthermore, the minimum transverse dimension of the tube must be able to accommodate at least one detonation cell of width λ , i.e., $\lambda/d \lesssim 1$.

The phenomenon of DDT was also studied in the smooth-walled section of the tube after the combustion wave exits from the rough-walled tube. The propagation of the combustion wave in the smooth-walled tube depends critically on its regime of propagation in the rough-walled tube upstream and on the mixture sensitivity. If the flame was in the quenching regime in the rough-walled tube, clearly the flame will not emerge in the smooth-walled portion. When a flame in the choking regime in the obstacle field is transmitted into the smooth tube, the initially supersonic turbulent flame either decays to a low velocity flame or, after a short decay (over a distance of the order of a 1 m in the 5-cm-diameter tube), reaccelerates and undergoes transition to detonation with the typical overdriven overshoot and asymptotic approach to the C-J level. If the quasi-detonation regime was achieved in the obstacle field, then the combustion wave on emerging from the rough-walled tube accelerates immediately to the C-J detonation velocity of the mixture. This is illustrated in Figure 27 for H_2 -air mixtures where the combustion wave velocity in both obstacle field and smooth-walled portion of the 5-cm-diameter tube have been plotted as a function of the distance along the tube. Transition to detonation in the smooth-walled tube occurs at 20 and 51 percent H_2 .

Under certain conditions for a narrow range of concentrations, the transmission of a flame from the obstacle field resulted in a galloping detonation in the smooth-walled tube. As illustrated in Figure 28 for a lean H_2 -air mixture (18 percent H_2), the reaccelerating wave after overshooting the C-J level does not decay asymptotically to the C-J velocity V_{CJ} but decreases significantly below V_{CJ} . In other words, a stable C-J wave is never established in this case within the length of tube available in the present study. Such a behavior was not recognized in the earlier experiments using a wire spiral in the 5-cm-diameter tube. [93] This led to the erroneous conclusions that DDT in H_2 -air mixtures in smooth-tube occurs at 17 percent H_2 and the criterion for DDT is $\lambda = \pi D$ and is identical to the criterion for detonability limits in rigid circular tubes using strong initiation sources.

The present results for DDT in a smooth-walled tube [85] indicate that the criterion for DDT is $\lambda/D \lesssim 1$ where D is the tube diameter. Table 5 summarizes all the available data for transition in the smooth-walled tube

Figure 27. Variation of Combustion Wave Velocity With Distance (in a 5-cm-Diameter Tube) in H_2 -Air Mixtures at NTP

Figure 28. Variation of Combustion Wave Velocity With Distance (in a 5-cm-Diameter Tube) for 18 percent H_2 -Air Mixtures at NTP

Table 5
Transition in Smooth-Walled Tube

D = 5 cm			
Mixture	Concentration (%)	λ (mm)	λ/D
C ₂ H ₂ -Air	4	58.3	1.18
	5	65.1	1.32
C ₂ H ₄ -Air	10	39.7	0.80
	4	52.2	1.06
C ₃ H ₈ -Air	5	59.0	1.19
	20	55.4	1.12
H ₂ -Air	51	52.5	1.06

for H₂-air mixtures and mixtures of air and hydrocarbons (C₂H₂, C₂H₄, and C₃H₈). It is to be noted that no data is available for the 15- and 30-cm-diameter tubes. To achieve DDT in the obstacle field of the 30-cm-diameter tube, the rough-walled section had to be extended over the whole length of the tube (17 m). In the 15-cm-diameter tube, the smooth-walled tube length (≈ 10 m) was insufficient to achieve DDT. Furthermore, because the fractional difference between the orifice diameter, d , and the tube diameter, D , was too small for the particular blockage ratio used, it was very hard to achieve the bifurcation in the first place because the appropriate changes in mixture composition proved to be too small and within the inherent experimental errors in measurement. Experiments in the larger tubes (i.e., 15- and 30-cm-diameter tubes) should be carried out with larger obstacle blockage ratios and/or longer tubes.

The different criteria on detonability limits ($\lambda/D \approx \pi$) and DDT ($\lambda/D < 1$) derived from the present studies demonstrate the influence of the initiation source strength on detonation propagation in smooth rigid circular tubes. These criteria indicate that for the propagation of a C-J detonation, the stronger the initiation source, the less sensitive the mixture that may propagate a detonation in a given tube.

Whereas DDT can be achieved via flame acceleration in mixtures confined in long channels filled with obstacles, other mechanisms must be responsible for DDT in partially or totally unconfined environments or in confined channels too short for flame acceleration to achieve the required flame

speed for transition. The experiments of Knystautas et al. [91] on the initiation of a spherical detonation by a turbulent jet of hot products (of the same mixture) have identified jet initiation as a mechanism for DDT to occur in an unconfined cloud. In jet initiation, the onset of detonation results from the rapid turbulent mixing between the hot products and the cold mixture. Jet initiation requires a critical jet diameter, characteristic of the scale of the turbulent mixing region which depends on the sensitivity of the mixture. Three recent reports of large-scale experiments where DDT occurred support the jet turbulent mixing mechanism.

Pförtner observed DDT in a flame propagation experiment in an H₂-air mixture (≈ 40 percent H₂) partially confined in a rectangular channel.* The local turbulent mixing region resulted from a ventilation fan in operation during the flame propagation.

DDT was also observed by Geiger in a partially confined stoichiometric H₂-air mixture.** The experiment was carried out in a rectangular box (0.5 x 0.5 x 1 m) closed at the ignition end and connected through a small orifice at the other end to a large volume of the same explosive mixture contained in a plastic bag (1 x 1 x 4 m). Although the turbulent jet discharging through the orifice provided a powerful ignition source, DDT was not observed in the jet mixing region but rather after a fast deflagration propagated some distance down the bag.

While studying direct initiation of detonation, Moen observed a DDT in a partially confined, lean C₂H₂-air mixture (≈ 5 percent C₂H₂).⁺ A misfire of the initiating charge resulted in flame propagation in a lean C₂H₂-air mixture (≈ 5 percent C₂H₂) confined in a tube (0.63 m in diameter, 3 m long) with subsequent discharge into a large plastic bag (2 m in diameter, 3.5 m long) containing the same mixture. As in Geiger's experiment, DDT was not achieved in the immediate vicinity of the tube exit. The jet was observed to impinge on a vertical wall at the end of the plastic bag, creating a recirculation zone (vortex) at the base (i.e., between the wall and the ground). The onset of detonation occurred in this recirculation zone and once formed, the detonation propagated throughout the entire volume of mixture at about 1700 m/s, close to the C-J value.

In the large-scale experiments conducted at SNLA under the present joint program, DDT was also observed in the FLAME facility in two H₂-air mixtures (24.7 and 30 percent H₂). A small fan seems to be responsible for the transition to detonation.

These large-scale experiments indicate that turbulent jet mixing is the mechanism responsible for DDT in unconfined mixtures. The critical conditions for the onset of DDT in unconfined mixtures can be formulated from the results of the flame acceleration experiments in confined

*Private Communications, I.C.T. Pförtner, Karlsruhe, West Germany.

**Private Communications, W. Geiger, Battelle Institute Frankfurt, West Germany.

⁺Private Communications, I.O. Moen, Defense Research Establishment, Suffield, Alberta, Canada.

environments reported in this Section. In confined mixtures, the onset of detonation requires flame speeds of the order of 800 m/s. This gives the minimum turbulent intensity required for mixing. The minimum size of the mixing region (tube diameter) was found to be of the order of the detonation cell size. By analogy, in unconfined mixtures, DDT will occur if a sufficiently intense turbulent mixing region of the order of a cell size can be formed. Once this initiation kernel is formed, onset of detonation will occur and subsequently a detonation may propagate throughout the unburned mixture.

7. CONCLUSIONS

The relevant dynamic detonation parameters such as the characteristic chemical length scale associated with the three-dimensional cellular structure of detonation waves (i.e., the detonation cell width), critical transmission conditions of confined detonations into unconfined environments (i.e., the critical tube diameter and critical channel width), critical initiation energy for unconfined detonations, detonability limits and critical conditions for transition from deflagration to detonation in H₂-air mixtures, pure or diluted with steam and CO₂ have been determined in small-scale as well as large-scale experiments.

For pure or diluted H₂-air mixtures, the variation of cell size as a function of hydrogen concentration exhibits a characteristic U-shape with a minimum around the stoichiometric composition (29.6 percent H₂). For H₂-air mixtures at NTP initially, the cell size increases from $\lambda = 1.5$ cm for the stoichiometric composition to $\lambda = 1.03$ m for the leanest mixture tested (13.6 percent H₂) and $\lambda = 1.35$ m for the richest mixture tested (70 percent H₂).

Increasing the initial pressure or initial temperature of H₂-air mixtures increases the detonation sensitivity (i.e., the cell size). Furthermore, preheating H₂-air mixtures to 100°C at superatmospheric initial pressures increases the detonation sensitivity from about 30 percent for fuel-lean mixtures (16.7 percent H₂) to 67 percent for stoichiometric mixtures (29.6 percent H₂) and 80 percent for fuel-rich mixtures (45.5 percent H₂).

Diluting H₂-air mixtures with CO₂ at NTP decreases the detonation sensitivity. For example, the addition of 5, 10, and 15 percent CO₂ to a stoichiometric H₂-air mixture (29.6 percent H₂) increases the cell size by factors of 1.5, 2.8, and 12.8, respectively. Similarly, diluting with steam H₂-air mixtures at 100°C and superatmospheric pressure initially causes a reduction in detonation sensitivity. For example, the addition of 10, 20, and 30 percent steam to a stoichiometric H₂-air mixture at 100°C initially increases the detonation cell size by factors of 6, 30 and 60, respectively. However, the lack of detonation studies of preheated H₂-air-CO₂ mixtures prevents any direct comparison of the efficiency of CO₂ and steam dilutions in desensitizing H₂-air detonations.

Kinetic models (developed at SNLA) to predict detonation cell size of H₂-air mixtures pure or diluted with steam and CO₂ have been reasonably successful. However, these models provide no physical understanding of the

three-dimensional structure of gaseous detonations since they do not model the nonlinear coupling mechanism between gasdynamics and chemical kinetics.

Comparison of the detonation sensitivity of H_2 -air mixtures and some mixtures of the common hydrocarbons (C_2H_2 , C_2H_4 , C_2H_6 , C_3H_8 , and C_4H_{10}) with air at NTP indicates that for all mixture compositions, C_2H_2 is the most detonation-sensitive fuel followed by H_2 , C_2H_4 and the alkanes (C_2H_6 , C_3H_8 , and C_4H_{10}). For example, for the stoichiometric composition, C_2H_2 has a cell size $\lambda \approx 0.6$ cm, $\lambda \approx 1.5$ cm for H_2 , $\lambda \approx 2.6$ cm for C_2H_4 and $\lambda \approx 5.35$ cm for the alkane group.

The present studies have demonstrated that a knowledge of the detonation cell size, λ , permits the other dynamic parameters to be determined reasonably well. The critical conditions for the transmission of confined detonations into unconfined environments have been determined for a number of confining geometries. For circular rigid tubes, the critical tube diameter d_c (i.e., the minimum diameter of a detonation tube from which a steady planar detonation wave can emerge into an unconfined volume containing the same mixture, transform into a spherical detonation and continue to propagate as a spherical detonation) obeys the empirical law of Mitrofanov and Soloukhin, $d_c \approx 13\lambda$. For H_2 -air mixtures at NTP, the critical tube diameter is of the order of 20 cm for the stoichiometric composition (29.6 percent H_2) and increases to 1.21 m for fuel-lean (18.1 percent H_2) and fuel-rich (55.5 percent H_2) mixtures, the limits of the present experiments. The same law, $d_c \approx 13\lambda$, holds for the transmission of detonations through circular orifice plates. For noncircular orifices (e.g., triangular, square, ellipse), the correlation remains valid by defining an effective critical diameter as the arithmetic mean of the diameters of the inscribed and circumscribed circles of the orifice.

The transmission of a planar detonation either through a rectangular orifice or from a rectangular channel of large aspect ratio ($L/W > 7$) and its transformation into a cylindrically unconfined detonation requires a critical width $W_c \approx 3\lambda$. As the aspect ratio is decreased, W_c/λ increases and in the limit $L/W \rightarrow 1$ (i.e., a square orifice or channel), the critical width obeys the empirical law of Mitrofanov and Soloukhin $W_c \approx 10\lambda$. Criteria for the transmission phenomenon under different boundary conditions formulated by Lee using the concept of critical wave curvature recover the empirical laws of Mitrofanov and Soloukhin.

The critical energies for the direct initiation of spherical (i.e., unconfined) detonations in H_2 -air mixtures at NTP initially range from 1 g of tetryl (4.27 kJ) for the stoichiometric composition (29.6 percent H_2) to about 462 g of Comp. C-4 (2250 kJ) for the leanest (17.4 percent H_2) and richest (60 percent H_2) mixtures successfully detonated in large-scale experiments. The surface energy model developed by Lee et al. indicates a cubic dependence of the critical initiation energy E_c on the detonation cell size, λ . The model predicts quite well the critical initiation energy data of H_2 -air mixtures and has been used to estimate critical initiation energies of H_2 -air- CO_2 mixtures. For the stoichiometric composition (29.6 percent H_2), the model predicts that the addition of 5, 10, and 15 percent CO_2 increases the critical initiation energy of the

undiluted mixture by factors of 3, 22 and 2100, respectively, due to the cubic dependence of E_c on λ . The surface energy model also predicts quite well the experimental results for the critical initiation energy of the alkane-air mixtures and lean ethylene-air mixtures. For the stoichiometric composition, the critical initiation energy ranges from 4.75 g of tetryl for C_2H_4 -air mixtures to 55 g of tetryl for the alkane-air mixtures.

The relative detonation sensitivity D_H of various fuels (defined as the ratio of the critical initiation energy of any fuel-oxidizer mixture to that of the most detonable common gaseous mixture, i.e., the stoichiometric $C_2H_2-O_2$ mixture) has been estimated using the surface energy model. For fuel-air mixtures, the values of D_H increases from about $D_H = 10^5$ for C_2H_2 to $D_H \approx 8 \times 10^7$ for the alkanes (C_2H_6 , C_3H_8 , and C_4H_{10}). With a value of $D_H \approx 10^6$, H_2 is approximately 10 times less sensitive than C_2H_2 but 10 times more sensitive than C_2H_4 . However, a stoichiometric H_2 -air mixture diluted with 5 percent CO_2 is only 2 times more sensitive than a stoichiometric C_2H_4 -air mixture. With 10 percent CO_2 dilution, stoichiometric H_2 -air mixtures become slightly less sensitive than stoichiometric C_2H_4 -air mixtures. The addition of 15 percent CO_2 makes H_2 -air mixtures 40 times less sensitive than the stoichiometric alkane-air mixtures.

The detonability limits of a reactive mixture have been defined as the critical conditions for the propagation of a self-sustained detonation. For confined mixtures, the detonability limits depend on both initial and boundary conditions. The onset of single-head spinning detonation which defines the boundary between the "self-sustained" detonation regime and the "detonation-like" regime (in which the detonation wave is unstable to finite perturbations) has been chosen to define detonability limits. The present experiments indicate that the detonability limits of a reactive mixture initiated by a strong initiation source in a smooth rigid round tube occur when the tube perimeter is of the order of the detonation cell size, i.e., $\lambda = \pi D$. For mixtures less sensitive than that specified by the criterion $\lambda = \pi D$, the detonation is highly unstable to finite perturbations with velocity fluctuations in excess of 10 percent from the Chapman-Jouguet velocity. From the limit criterion, the limiting tube diameter D^* for stable propagation of a detonation initiated by a strong source in a smooth rigid round tube has been estimated using detonation cell size data viz., $D^* = \lambda/\pi$. For stoichiometric H_2 -air mixtures (29.6 percent H_2) at NTP, the minimum tube diameter is $D^* \approx 0.5$ cm. With 15 percent CO_2 dilution, this value increases by one order of magnitude ($D^* \approx 6.1$ cm). For hot H_2 -air mixtures (at $100^\circ C$ initially), the minimum tube diameter for the stoichiometric composition $D^* \approx 0.16$ cm. With 30 percent steam dilution, this value increases by a factor of 60 to $D^* \approx 9.6$ cm. For the hydrocarbon-air mixtures at the stoichiometric composition, the minimum tube diameters are $D^* \approx 0.19$ cm for C_2H_2 , $D^* \approx 0.83$ cm for C_2H_4 and $D^* \approx 1.7$ cm for the alkanes.

The criterion for the detonability limits of mixtures confined in two-dimensional channels of large aspect ratios formulated by Vasiliev is $W = \lambda$ where W is the channel width. Limit criteria for other tube geometries have yet to be established.

For unconfined detonation waves, there exists no quantitative theory or experimental method to predict detonability limits. Since for the stable propagation of a diverging detonation wave, the average cell size must remain constant, the rate of generation of detonation cells must be the same as the rate of increase of the surface area. Therefore, failure of the cells to multiply at the critical rate might be used to define the composition limits.

Experimental studies on turbulent flame acceleration in rough and smooth-walled long linear tubes in the context of the phenomenon of deflagration to detonation transition (DDT) have revealed the existence of several regimes of flame propagation. In tubes roughened by obstacles (e.g., orifice plates, wire spirals), four flame propagation regimes have been identified following an initial flame acceleration: the self-quenching regime, the weak deflagration regime, the choking regime and the quasi-detonation regime. For the most insensitive mixtures, the flame extinguishes itself within the obstacle field. With mixtures of increasing sensitivity, the flame reaches a steady-state velocity within the obstacle field of the order of a few tens of meters per second in the weak deflagration regime, 800 to 1000 m/s in the choking regime and in excess of 1100 m/s in the quasi-detonation regime. For a given tube diameter and obstacle configuration, not all regimes can be observed in a given fuel. For all mixtures studied, the criterion for transition to detonation in a rough tube is that $\lambda/d \lesssim 1$, where λ is the detonation cell width and d is the orifice opening diameter. For H₂-air mixtures, DDT was observed in rough tubes (5, 15, and 30 cm in diameter and blockage ratio BR = 0.43) at 22, 18, and 16 percent H₂ on the fuel-lean side and 47.5, 57, and 60 percent H₂ on the fuel-rich side, respectively. When a supersonic flame propagating in a rough tube in the choking regime transmits into a smooth tube, the flame either decays to a slow steady mode of propagation or reaccelerates and undergoes DDT. The criterion for DDT in a smooth tube is that $\lambda/D \lesssim 1$ where D is the inner diameter of the smooth tube. For H₂-air mixtures, DDT occurs at 20 and 51 percent H₂ in a smooth tube (5 cm in diameter).

The different criteria on detonability limits ($\lambda/D \approx \pi$) and DDT ($\lambda/D \lesssim 1$) derived in the present studies demonstrate the influence of the initiation source strength on detonation propagation in smooth rigid round tubes. These criteria indicate that the composition range for stable propagation of a detonation increases with increasing strength of the initiation source.

Whereas for confined mixtures, DDT may result from flame acceleration due to turbulence generated by obstacles, large-scale experiments indicate that turbulent jet mixing is the mechanism responsible for DDT in unconfined mixtures.

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5. AUTHOR(S) C. M. Guirao, R. Knystautas, J. H. Lee		4. DATE REPORT COMPLETED <table border="1"> <tr> <td>MONTH</td> <td>YEAR</td> </tr> <tr> <td>June</td> <td>1987</td> </tr> </table>		MONTH	YEAR	June	1987
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MONTH	YEAR						
May	1989						
10. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code) Division of Systems Research Office of Nuclear Regulatory Research U.S. Nuclear Regulatory Commission Washington, DC 20555		8. PROJECT/TASK/WORK UNIT NUMBER 9. FUNDING OR GRANT NUMBER A1246					
		11a. TYPE OF REPORT Technical					
12. SUPPLEMENTARY NOTES		b. PERIOD COVERED (Inclusive dates)					
13. ABSTRACT (200 words or less) Dynamic detonation parameters are reviewed for hydrogen-air-diluent detonations and deflagration-to-detonation transitions (DDT). These parameters include the characteristic chemical length scale, such as the detonation cell width, associated with the three-dimensional cellular structure of detonation waves, critical transmission conditions of confined detonations into unconfined environments, critical initiation energy for unconfined detonations, detonability limits, and critical conditions for DDT. The detonation cell width, which depends on hydrogen and diluent concentrations, pressure, and temperature, is an important parameter in the prediction of critical geometry-dependent conditions for the transmission of confined detonations into unconfined environments and the critical energies for the direct initiation of unconfined detonations. Detonability limits depend on both initial and boundary conditions and the limit has been defined as the onset of single head spin. Four flame propagation regimes have been identified and the criterion for DDT in a smooth tube is discussed.							
14. DOCUMENT ANALYSIS -- a. KEYWORDS/DESCRIPTORS Detonation, reactor safety		15. AVAILABILITY STATEMENT Unlimited					
b. IDENTIFIERS/OPEN-ENDED TERMS		16. SECURITY CLASSIFICATION (This page) Unclassified (This report) Unclassified					
		17. NUMBER OF PAGES					
		18. PRICE					