Hydrogen Combustion Characteristics Related to Reactor Accidents

Prepared by A. L. Berlad, M. Sibulkin, C. H. Yang

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ABSTRACT

A knowledge of combustion phenomena and their characteristics is necessary in accident analyses related to the release of hydrogen. As a result of the accident at Three Mile Island, and from the results of related studies of hypothetical degraded core accidents, it is recognized that combustion of hydrogen may, under some circumstances, threaten the integrity of a reactor containment building. In general, detailed combustion information is required in order to:

(a) Understand and characterize the combustion phenomena and processes which may occur in a containment building;

(b) Identify the criticality or limiting conditions under which important combustion processes may be extinguished, initiated, or otherwise transformed;

(c) Provide data and information for analytic modeling of safety-related hypothetical accident scenarios;

(d) Allow modelers to predict with confidence, where possible, the consequences of naturally occurring and/or induced combustion processes;

(e) Guide safety-related strategies aimed at mitigation of accident-related combustion;

(f) Identify areas where inadequate understanding exists;

(g) Distinguish among the ranges of applicability of selected items and classes of combustion data, experiments, theories, and models.

In pursuit of these objectives, this report attempts to provide a perspective on combustion processes which may not otherwise be derived easily from the enormously diverse combustion literature.

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1.0 INTRODUCTION

Analyses of very low probability hypothetical degraded core accidents sequences in Zion/Indian Point reactor plants indicate that large quantities of hydrogen may be released into the containment building atmosphere[1] (Pratt and Bari, 1980) as a result of extensive metal-water reactions associated with progression of in-vessel and ex-vessel core degradation and meltdown processes. The accident sequences analyzed in Reference 1 included an extended loss of total AC power coupled with a failure in the steam-driven-turbine pump train of the auxiliary feedwater system (the so-called TMLB' scenario) and various pipe break initiators coupled with failures of core and/or containment cooling engineered safety features (e.g., S_1D, S_2D, S_2HF, etc). These sequences were analyzed with the MARCH code[2] and it was predicted that approximately 1700 lbs of hydrogen would be produced from in-vessel metal-water reactions and that the potential exists for substantially more hydrogen to be produced as a result of ex-vessel metal-water reactions. The ex-vessel metal-water reactions result from the core debris/concrete thermal interactions. The water released from the concrete interacts (is chemically reduced) on contact with any Fe, Zr, Ni, or Cr that may be present in the core debris. In addition, depending on the concrete type, carbon dioxide can also be released from (limestone) concrete and interact with the iron to produce (combustible) carbon monoxide. It was noted in Reference 1 that for limestone concrete more CO could be released than H_2 as a result of core/concrete interactions.

In a number of cases so analyzed, combustion phenomena may occur with attendant increases in pressure and temperature in the containment building and decreases in fuel concentrations. As a result of the accident at Three Mile Island, and from results of related studies of hypothetical degraded core accidents, it is recognized that combustion of hydrogen may, under some circumstances, threaten the integrity of a reactor containment building. It was emphasized in Reference 1 that there is great uncertainty in the physical phenomena associated with core meltdown processing and in the predictions (e.g., containment temperature and pressure histories) of these phenomena by the MARCH code.

Accident analyses may examine such important combustion-related questions as:

a) What combustion phenomena may occur during an accident should no attempts at mitigation be initiated?

b) During an accident, what combustion phenomena may be initiated by personnel in attempts to mitigate otherwise more severe thermal and mechanical loading conditions?

c) What are the detailed temperature, pressure, and consequential damage histories which accompany the scenarios associated with (a) and with (b)?

The combustion literature provides a large number of detailed experimental and theoretical studies which may prove helpful. These studies provide directly useful results for a number of situations involving the hydrogen-air system. Such directly useful results are generally for either quiescent or laminar, quasi-steady, premixed hydrogen-air systems which are spatially uniform. Information is also available for completely unpremixed hydrogen-air systems. For such so-called diffusion flames, fuel and air are brought together (at a flame reaction zone) from separate quasi-steady sources[2,3] (Lewis and Von Elbe (1962), Williams (1965)).

Less comprehensive combustion information is available for cases of non-laminar (turbulent, etc.) combustion processes, for spatially nonuniform fuel-air mixtures, and for time-dependent fuel-air composition fields.

A knowledge of combustion phenomena and their characteristics is necessary in accident analyses related to the release of hydrogen. There are existence limits for combustion processes and these must be considered. Combustion phenomena are studied in experimental apparatuses which are generally of much smaller scale than may be of interest for a containment building $(2.6 \times 10^6 \text{ ft}^3)$ atmosphere. The effects of scale must be considered. In some important areas of combustion science, inadequate information and uncertainties exist. These may imply uncertainties in correspondingly dependent accident analyses. In general, detailed combustion information is required in order to:

a) Understand and characterize the combustion phenomena and processes which may occur in a containment building;

b) Identify the criticality or limiting conditions under which important combustion processes may be extinguished, initiated, or otherwise transformed;

c) Provide data and information for analytic modeling of safety-related hypothetical accident scenarios;

d) Allow modelers to predict with confidence, where possible, the consequences of naturally occurring and/or induced combustion processes;

e) Guide safety-related strategies aimed at mitigation of accident-related combustion;

f) Identify areas where inadequate understanding exists;

g) Distinguish among the ranges of applicability of selected items and classes of combustion data, experiments, theories and models.

In pursuit of these objectives, this report attempts to provide a perspective on combustion processes which may not otherwise be derived easily from the enormously diverse and comprehensive combustion literature. Accordingly, some well-known, directly useful combustion processes may be dealt with here only briefly. Combustion processes which are more complex or less well understood are given more intensive scrutiny.

2.0 CLASSICALLY DEFINED HYDROGEN-AIR COMBUSTION PHENOMENA:

A BRIEF REVIEW

Combustion processes supported by premixed gaseous fuel and air have been studied largely for quiescent or nonturbulent systems defined by apparatuses of small scale (of the order of centimeters rather than meters). Experimental observations and theoretical analyses available for hydrogen and air include the following principal phenomena:

> burning velocity
> flammability limits
> pressure limits
> quenching limits
> spark ignition limits
> autoignition
> flash-back limits
> blow-off limits
> deflagration - detonation transition
> detonation velocity
> detonation limits.

Investigations reported in the literature, in some instances, also include the dependence of the above noted phenomena on initial temperature, pressure, inert concentrations, stoichiometry, apparatus size, the presence of turbulence, effects of ionizing and photochemically significant sources, and the effects of gravitational and other fields which may impose body forces.

The dominant problem in laminar flame propagation theory and experiment has classically centered on the burning velocity and its limits. A summary of the principal features of the various theoretical approaches to flame propagation (and flame extinction) theory is given in Figure 2-1. It is generally recognized that so-called "complete" and "fundamental" theories should start with a statement of the full constitutive equations (the conservation of energy, mass, atomic species, the equations of flow, and the detailed chemical kinetics). It is further known that real combustion systems are nonadiabatic and multidimensional. Accordingly, reality and completeness would require a theoretical thoroughness which none of the currently available theoretical

REFERENCES	LOSSES Non-adiabatic	MULTIDIMENSIONALITY CONSIDERED			TRANSPORT PROPERTIES CONSIDERED			FULL CONSTITUATIVE EQUATIONS
		Multi-dimensional	One-dimensional	One-dimensionalized	Free Convection	Radiation	Molecular transport of Heat and Mass	
Hirschfelder, J. (35) Curtiss, C. F., and Bird, R. B. (1954)	---	---	X	---	---	---	X	X
Spalding, D. B. (34), (1957)	X	---	---	X	---	---	X	---
Berlad, A. L. and Yang, C. H. (31) (1960)	X	---	---	X	---	X	X	---
Levy, A. (33) (1965)	---	---	X	---	X	---	---	---
Lovachev, L. A. (22), (1970)	---	---	X	---	X	---	---	---
Buckmaster, J. (32), (1976)	X	---	X	---	---	---	X	---
Mitani, T. and Williams, F. A., (4), (1980)	X	---	---	X	---	---	X	---

--- Consideration absent in theory.
X Consideration provided in theory.

Figure 2-1: Premixed Gaseous Flames: Quasi-steady flame propagation and extinction limit analyses.

structures provides. These theories, however truncated, do provide specialized insights to flame propagation and extinction. Further, the recent work of Mitani and Williams[4] is directed especially at limiting conditions for flame propagation in hydrogen-oxygen-nitrogen mixtures. Taken together with the survey on hydrogen combustion provided by Drell and Belles[5] certain general features of quasi-steady, laminar flame propagation and extinction of hydrogen-air flames may be drawn.

(1) There exist temperature-pressure-composition regimes within which autoignition (or explosion) occurs. These regimes (Figure 2-2) are functions of apparatus scale and represent parametric phase spaces within which quasi-steady flame propagation is not possible (supercritical hydrogen-air compositions follow time-dependent trajectories in their explosive behavior).

(2) Outside of the autoignition regime, there exist temperature-pressure-composition regimes where combustion occurs when a suitable ignition source is provided. Standard flammability limits have been collected by Coward and Jones[6] from data obtained in vertical, 5 cm diameter tubes. These limits depend on the direction of flame propagation because of convective effects due to gravity. For upward propagation the flammability limits (% volume) for hydrogen burning in air are about 4% (lean limit) and 74% (rich limit); for downward propagation the limits are 9% and 73%. Increasing the ambient pressure above one atmosphere tends to widen the flammability limits (for downward propagation) slightly. Increasing the ambient temperature broadens the flammability limits as shown in Figure 2-3 for downward propagation.

The effects on the flammability limits of adding an inert gas to a hydrogen-air mixture is shown in Figure 2-4 for N_2 and CO_2 at normal temperature and pressure. An upper limit of additive concentration exists above which the mixture is inert. Similar results are obtained for hydrogen-air-water vapor mixtures. At a pressure of one atmosphere, the inertion limit for saturated water vapor addition is 60% at 86°C[6,7]. In order to safely satisfy steam inertion criteria under the variety of possible containment building conditions, even higher temperature (than 86°C) saturation conditions should be established.
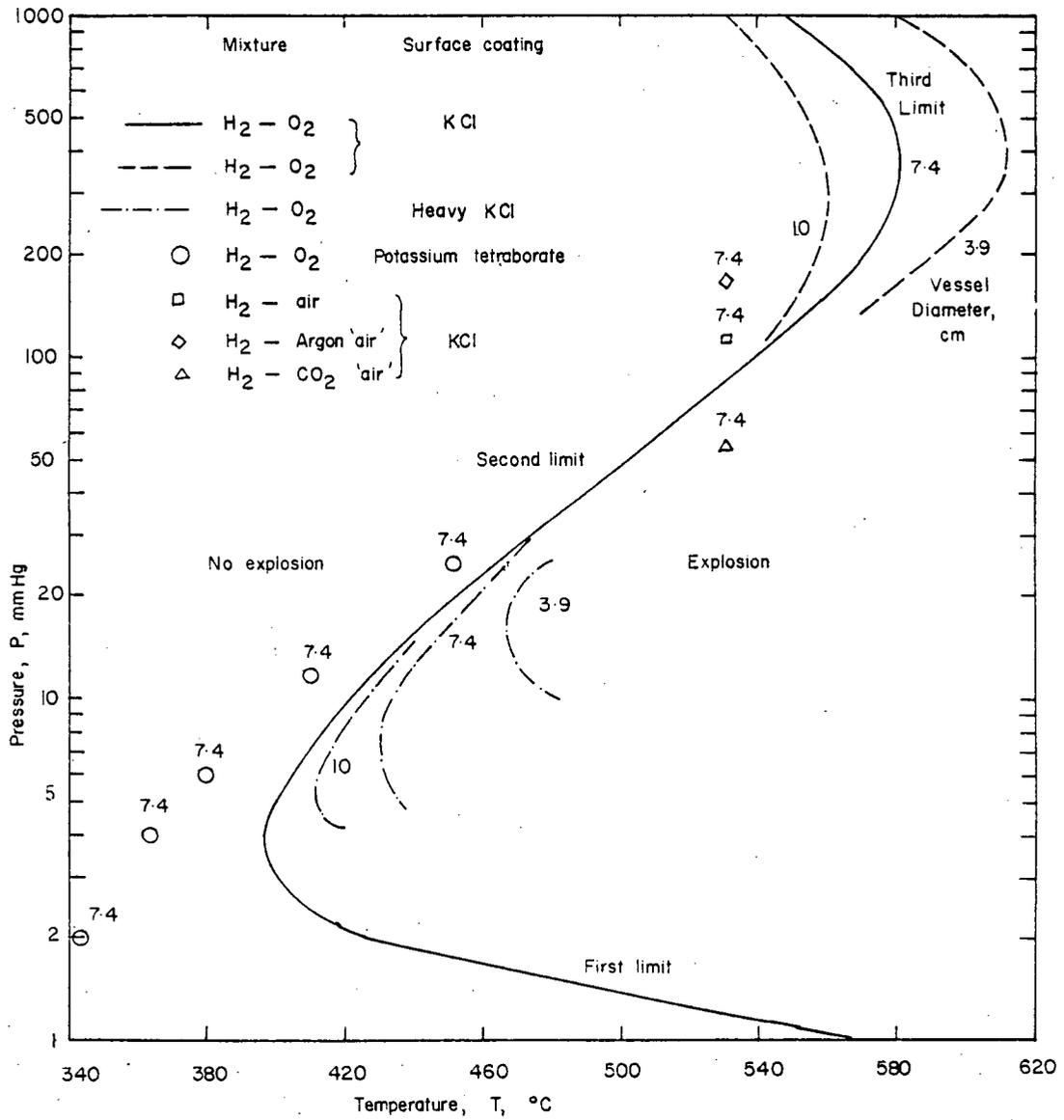
Figure 2-2: Explosion limits of stoichiometric hydrogen mixtures
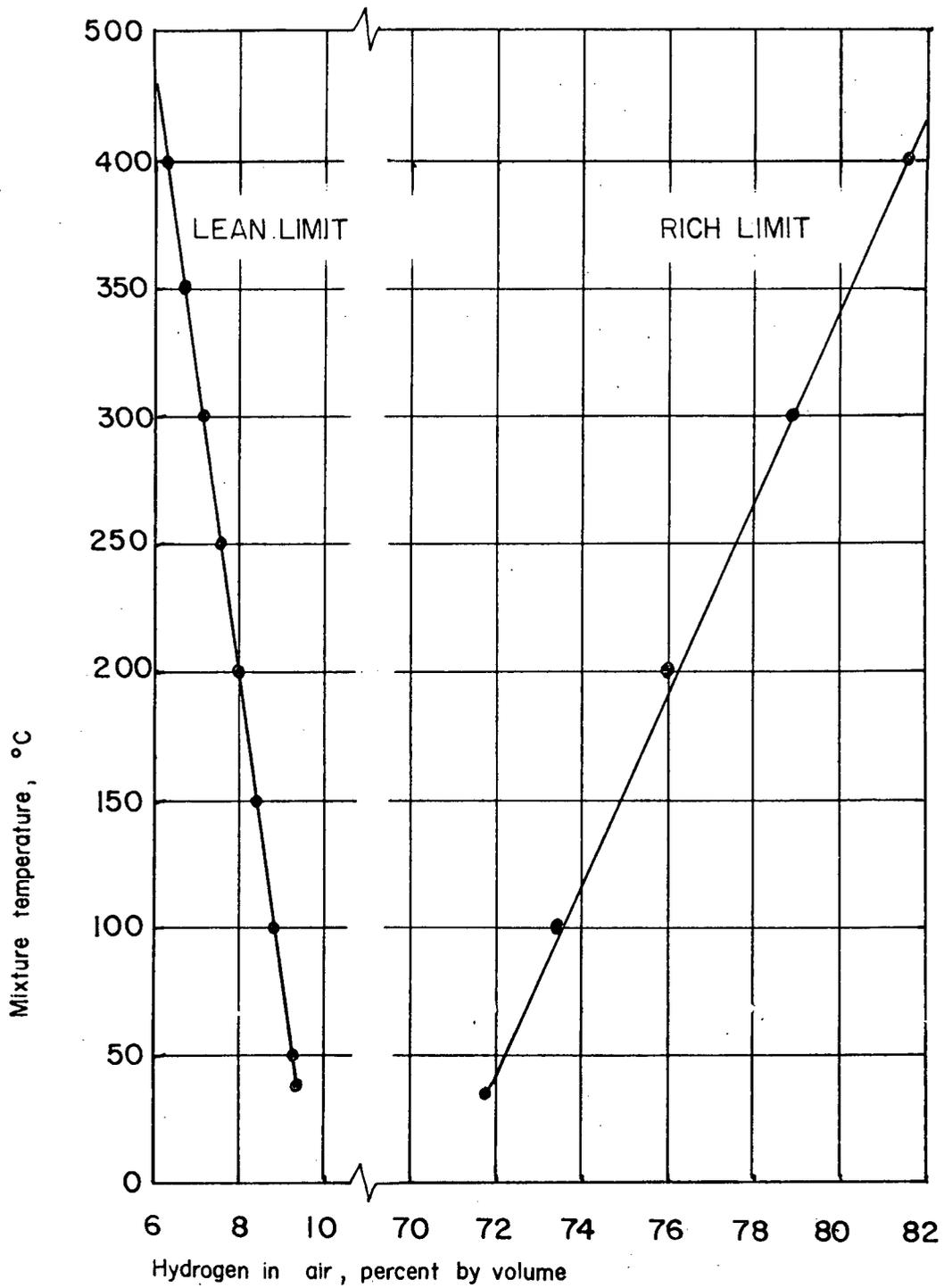(Ref. Drell and Belles).

Figure 2-3: Effect of temperature on flammability limits of hydrogen in air for downward propagation (Ref. Coward and Jones).
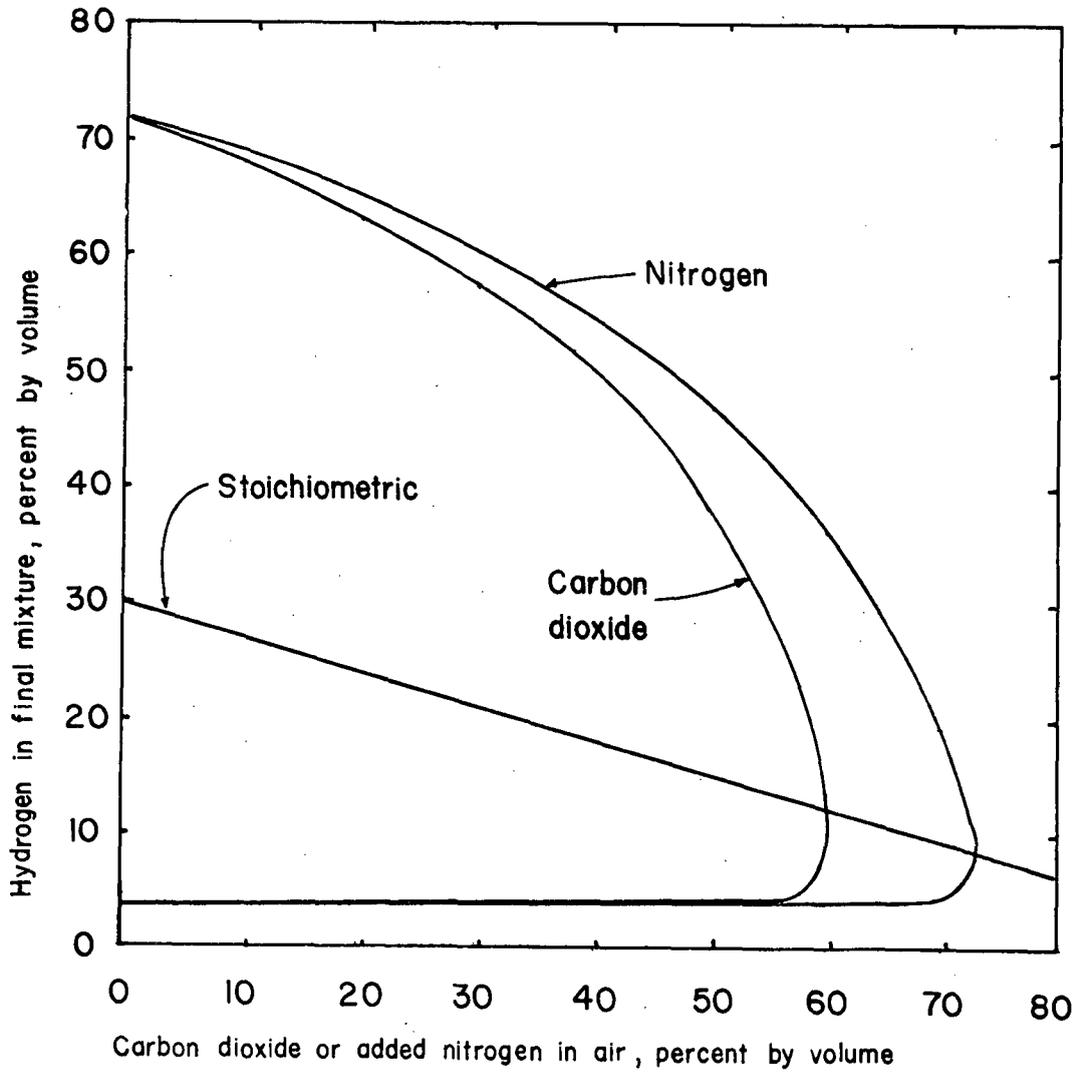
Figure 2-4: Flammability limits of hydrogen in air diluted with nitrogen or carbon dioxide (Ref. Coward and Jones).

(3) The so-called "flammable regime" of autoignition diagrams is further restricted by a number of limiting extinction conditions. Flames which are caused to propagate through narrow tubes sustain heat losses. For some tube diameter smaller than a critical value "flame quenching" occurs. These limits are frequently referred to as "quenching limits". For a given size of apparatus (e.g., tube diameter) it is found that there exists a critical pressure below which steady state flame propagation does not occur. Such limits are frequently referred to as "pressure limits".

Drell and Belles[5] recognized at an early time that "flammability limits", "quenching limits", and "pressure limits" as defined by classical experiment are related quantities. These experimental facts are clearly demonstrated through the sets of <u>smooth</u> "extinction limit" curves given by Drell and Belles (1958) (Figure 2-5).

(4) Flames can be stabilized at the mouth of a tube through which premixed fuel and air flows. There exist, nevertheless critical flow conditions for "blow-off" and for "flash back" of the flame. Experimentally, it is observed that for a given size duct, a given laminar hydrogen-air flame will "blow off" (move away, downstream of the flame holder) at flow rates higher than some critical value. It is also observed that for a given size duct,.the same laminar hydrogen-air flame will "flash back" (propagate upstream, into the fuel-air mixture in the duct) at flow rates that are smaller than some critical value. Lewis and Von Elbe[2] have characterized these critical conditions in terms of the duct's boundary velocity gradient. These critical duct boundary velocity gradients have been termed "boundary velocity gradient for blow-off" and "boundary velocity gradient for flash back". The data of Fine[8,9] (1956 and 1957) are used to correlate boundary velocity gradient for flashback for laminar and turbulent hydrogen-air flames (Figure 2-6 and 2-7). In Figures 2-6 and 2-7, ϕ represents the "equivalence ratio" for the combustible mixture (volumetric fuel-air ratio divided by the stoichiometric fuel-air ratio). The ordinate g_b is the velocity gradient at the tube wall in units of sec^{-1}. These data describe
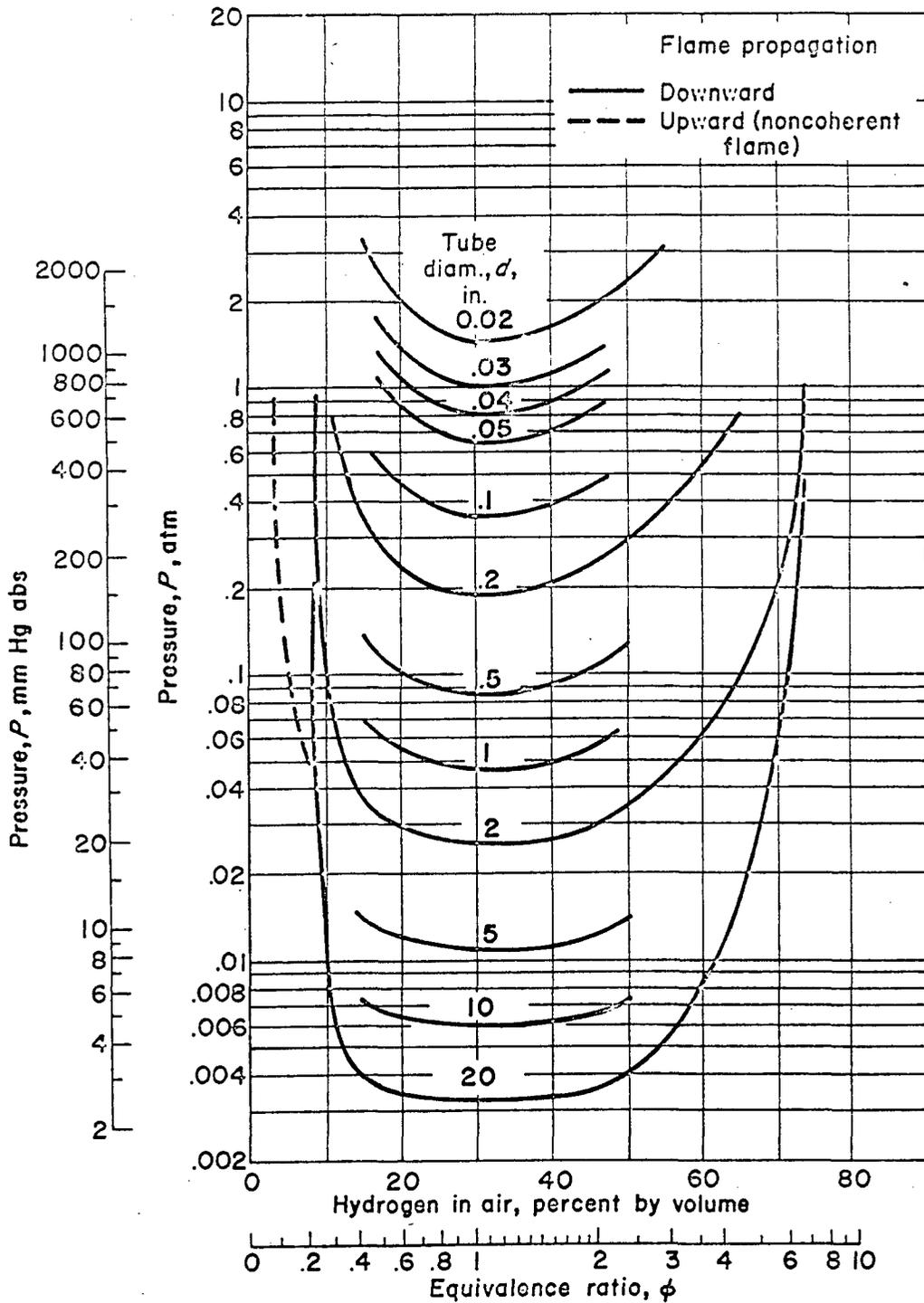
Figure 2-5: Estimated pressure limits of flame propagation for hydrogen-air mixtures with various tube diameters (Ref. Drell & Belles) based on quenching data of Potter & Berlad.
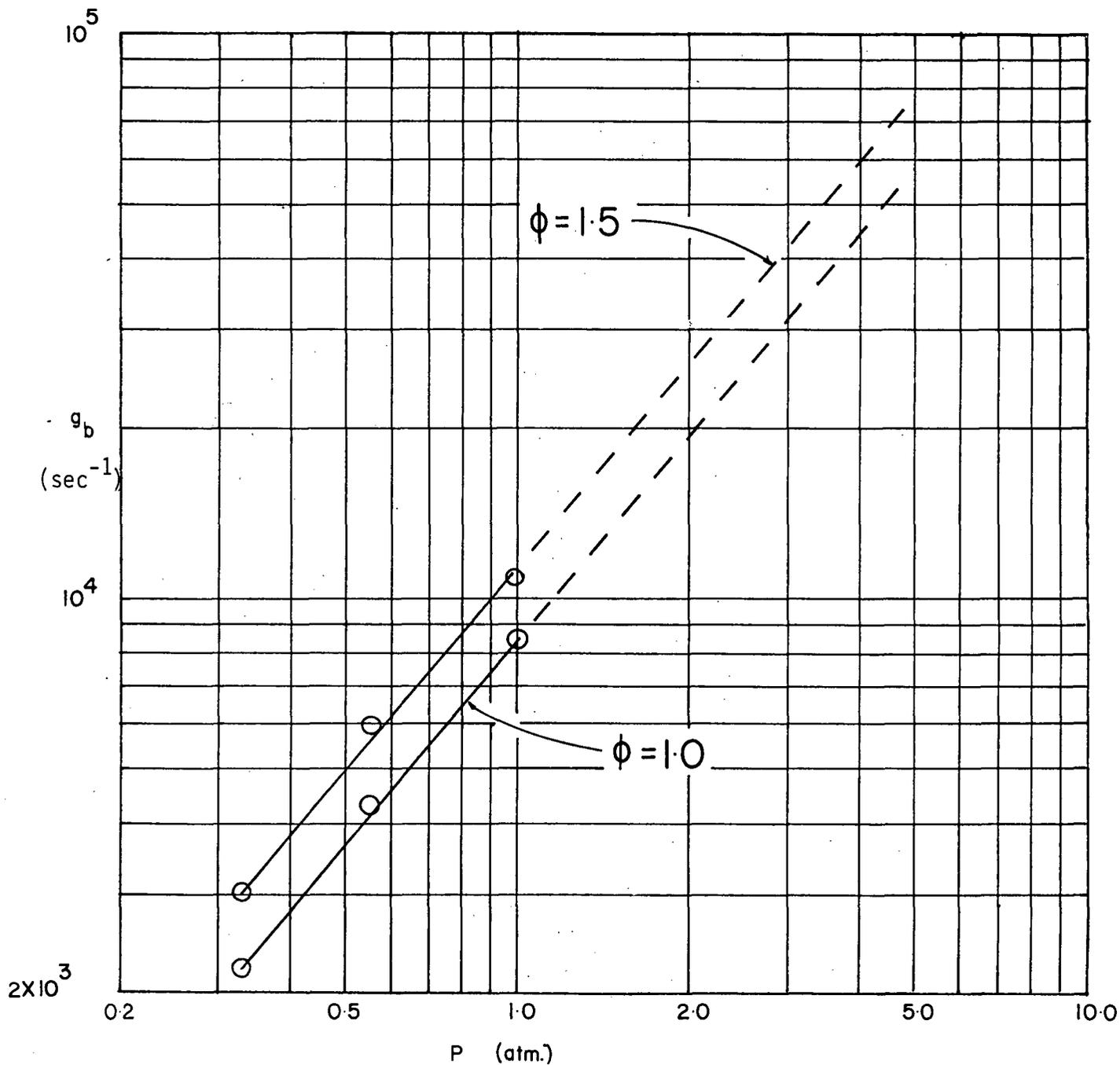
Figure 2-6: Effect of pressure on critical boundary velocity gradient for
Flashback of Laminar H_2-Air Flames (T_0 = 300°K)
(Refs.:   Fine, B. NACA-TN 3833 (1956).
Von Elbe, G. & Mentser, M. J. Chem. Phys. 13,
(February 1945)).
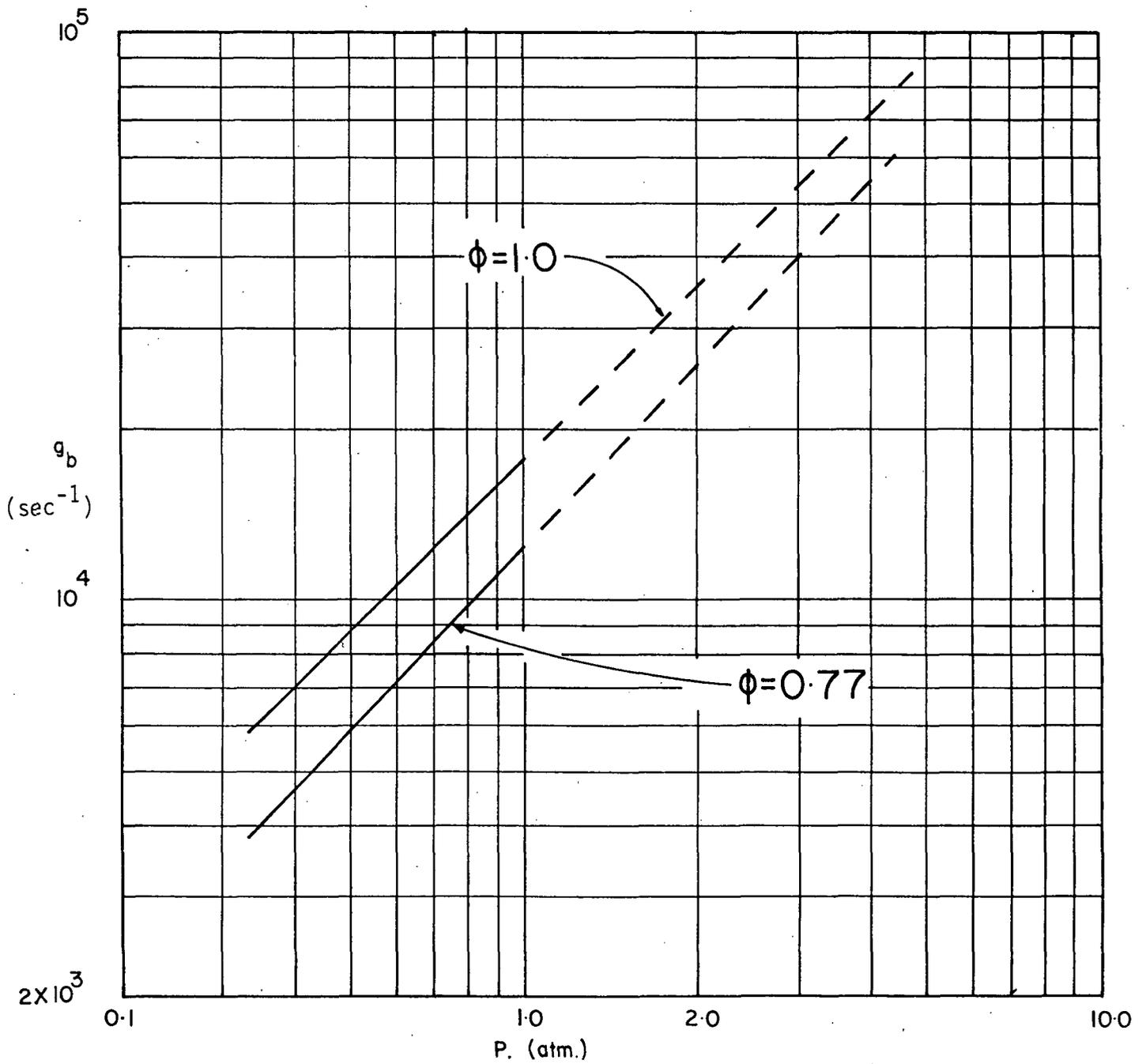
Figure 2-7: Effect of pressure on critical boundary velocity gradient for Flashback of Turbulent H_2-Air Flames ($T_0 = 300°K$) (Ref: Fine, B. NACA-TN 3977 (1957)).

the conditions under which flames in the neighborhood of a fuel-air filled duct may "strike-back" into the duct (against the flow), or remain stable, or "blow away".

(5) Flames, shock waves, or other igniters may initiate transients which may lead to a reaction wave which is supersonic (based on the speed of sound in the cold reactants). Such quasi-steady reaction waves are called detonations. Experiments in tubes are usually used as the bases for determination of observed "detonation velocities"[3,10,11]. It is also observed, for a given experimental apparatus, that there are hydrogen concentration limits above which and below which detonations do not propagate. These observations are usually used as the bases for determination of observed "detonation limits". There is evidence that "detonation limits" are functions of apparatus and that "detonation limits" observed for cylindrical tubes are not necessarily those which limit the existence of a detonation wave in other geometries and scales.[12]

(6) The quasi-steady laminar flame or the quasi-steady detonation wave do not occur without some source of ignition. For electrically-energized spark igniters, the minimum spark energy required for initiation of a flame is called the "minimum ignition energy"[2,5]. Where ignition of deflagration is caused by a more complex source (e.g., a glow plug) characterization of the critical conditions for ignition are more complex.

(7) The properties (composition, pressure, temperature and species fields) of a quasi-steady deflagration or detonation wave are independent of the source(s) used to initiate the wave. Accordingly, an ignition source which fails to successfully ignite a hydrogen-air mixture within the appropriately defined deflagration or detonation limits is (at least in part) a deficient ignition source. "Ignition limits" which are defined by such an ignition source are device-specific "ignition limits" and are not to be confused with extinction limits for flames or detonations.

(8) Combustion behavior of hydrogen-air (or other) flame systems under non-laminar flow conditions have also been studied experimentally.

In general flames which are supported by turbulent or other non-laminar transport properties give rise to higher deflagration speeds (than for the laminar case) and narrower extinction limits. Minimum spark ignition energies are higher and different values for such other parameters as boundary velocity gradients for "blow-off" or "flash back" are observed. Drell and Belles have included, in their survey, some of the observed turbulent hydrogen-air combustion properties. Unfortunately, turbulent and other nonlaminar conditions of gaseous fuel-air mixtures cannot be simply characterized. One cannot simply tabulate observed values of flame-speeds, etc. for nonlaminar conditions in a manner analogous to that for premixed laminar conditions. Libby and Bray[13] have recently discussed these difficulties. Experimental observations of complex nonlaminar apparatus-specific combustion phenomena cannot therefore be used as bases for predicting the detailed behavior of different combustion apparatuses or experimental arrangements. Thus it is difficult to generalize from the interesting observations of Moen, et.al.[14] regarding the acceleration of flame propagation rates by obstacles.

3.0 SOME CURRENT ASPECTS OF COMBUSTION

3.1 Deflagration Processes in Large Volumes

The classical determination of flammability limits in vertical tubes described in Section 2 is adequate for many technical purposes. However, whether a fundamental flammability limit exists for a given mixture of gases independent of the effects of gravity and the specific method of testing is still an open scientific question (see Reference 15). Attempts to determine fundamental flammability limits using a complete set of hydrodynamical equations and detailed chemical kinetics for hydrogen-oxygen systems are currently in progress[16,17] (Oran et al., 1979; Jones, 1980).

The standard flammability test referred to in Section 2.0 is made in a tube having a diameter of 5 cm. This dimension was chosen because larger diameters did not appear to appreciably alter the limit for upward flame propagation. The fraction of combustible material consumed, however, does depend on the tube diameter. The effect of tube diameter on the upward burning of methane-air mixtures[18] (Babin and V'yun, 1972) is shown in Figure 3-1 for methane concentrations from 5.6 to 6.0 percent (volume). It was found that while burning is nearly complete in a 5 cm tube, the fraction burned in a 24 cm. diameter tube dropped from 0.8 for 6.0% CH_4 to 0.3 for 5.6% CH_4. The proposed explanation of these results[19] (Lovachev, 1973) is shown in Figure 3-2, and is based on the inability of these fuel mixtures to support downward flame propagation. For a given composition, fuel is consumed within a cone of fixed angle; thus larger tube diameters leave more unburned fuel.

In large volumes the mode of burning is strongly affected by the mixture composition. Tests in a $70M^3$ spherical rubber confinement for 10% methane burning in air[20] (Ivashenko and Rumyantsev, 1978) show the development of a nearly spherical "fireball" which also rises due to convection. For mixtures closer to the lean flammability limit burning is quite different. A photographic sequence of burning in a $8M^3$ chamber[22] (Lovachev, 1979) is shown in Figure 3-3 for spark ignition near the chamber ceiling. A spherical flame kernel forms shortly after ignition; as the kernel rises and expands it is deformed into a mushroom shape; upon reaching the ceiling the flame front
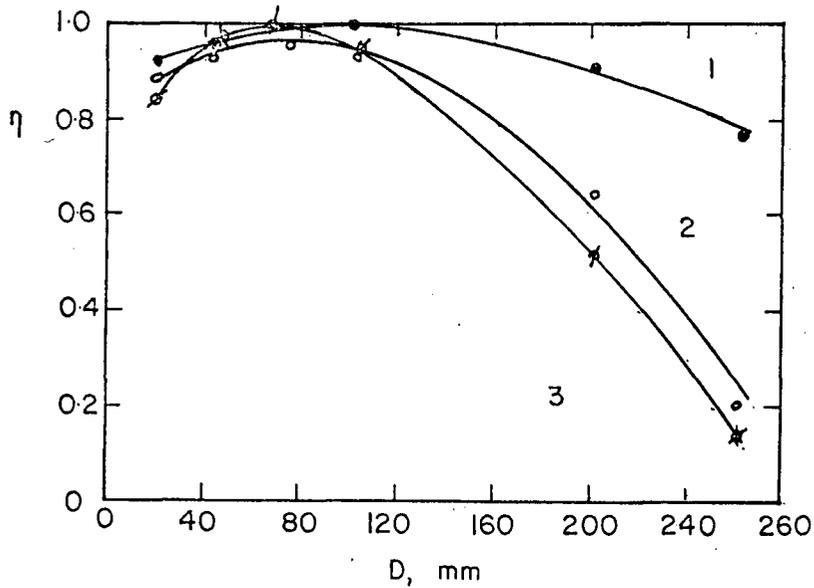
Figure 3-1:   The degree of combustion of methane-air
mixtures in tubes of different diameters.
1-6.0%, 2-5.7%, 3-5.6% of CH_4.   Tube length
26 cm; P_i = 5 atm, T_i = 294°K, ignition at
the base [16]. (Ref. L. A. Lovachev et al.,
Flammability Limits: An Invited Review,
Combustion and Flame, 20, 259, (1973)).
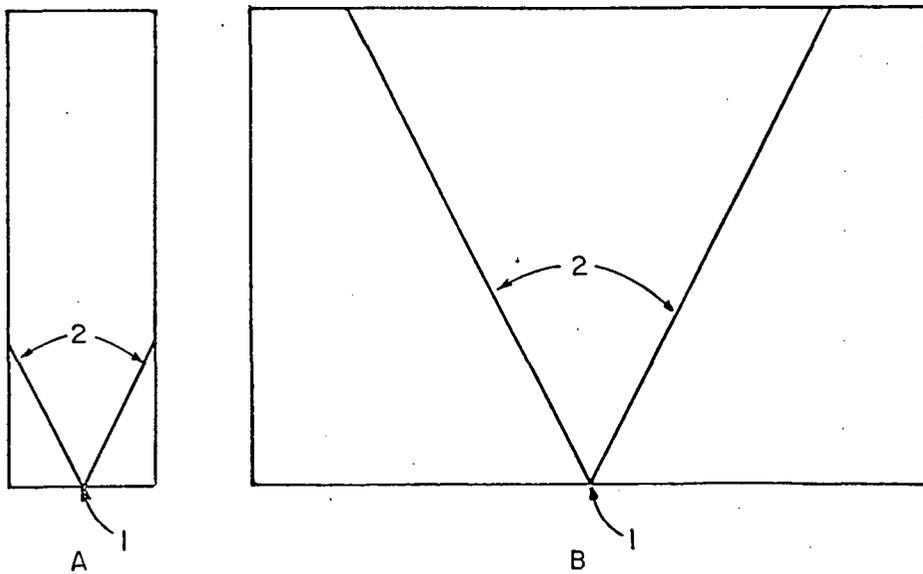
Figure 3-2:   Flame propagation near the limits in a narrow (A) and a
large (B) tube; 1-ignition source, 2-trace of convective
upward flame propagation. (Ref. L. A. Lovachev et al.,
Flammability Limits: An Invited Review, Combustion and
Flame, 20, 259, (1973)).

| 0.12 sec | 0.37 sec | 0.71 sec | 1.00 sec |
| 1.77 sec | 2.33 sec | 3.42 sec | 4.46 sec |
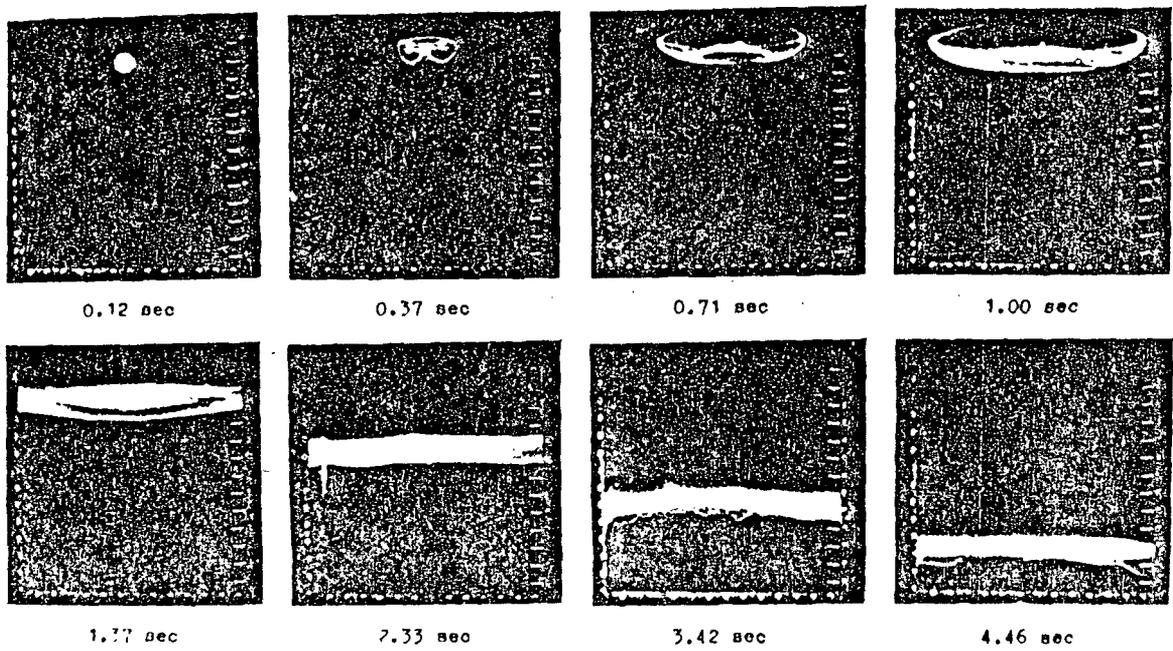
Figure 3-3: The spreading of flame beneath the ceiling is shown in the four upper frames, and the downward flame propagation in the four lower frames for an 8 m^3 chamber (2.1 percent NH_3 in air) (Ref. L. A. Lovachev, Flammability Limits - A Review, Combustion Science and Technology, 20, 209-224 (1979)).

spreads laterally until the flame reaches the wall; the flame front then prop-
agates downward as a nearly planar wave. For leaner mixtures downward flame
propagation did not occur; for still leaner mixtures, extinction occurred when
the flame kernel reached the ceiling. Thus the combustion is incomplete close
to the flammability limit.

Specific results for hydrogen burning in air have been obtained by Furno
et al.[23] (1971) for spark ignition in a 12 ft diameter sphere. The meas-
ured pressure rise as a function of hydrogen concentration is shown in Figure
3-4. For H_2 concentrations between 5 and 8.5%, only upward propagation
occurs and the pressure rise is negligible. Between 8.5 and 12% both upward
and downward propagation occurs and the pressure rises rapidly and approaches
the calculated thermodynamic pressure rise.

Lovachev[21] has reported on the flammability conditions under which
large volumes of hydrogen-air (or other fuel-air) mixtures, ignited from be-
low, may burn only partially. Data on extent of burning are based on short
duration ignition sources. No comparable experimental results for continuous
ignition sources have been found in the literature. Where natural convection
causes an unreacted gas mixture to flow past a continuous ignition source,
greater combustion completeness may be expected.

The results presented in this section were all obtained for mixtures
initially at room temperature and pressure. It is expected that at moderately
elevated temperatures similar qualitative results would be obtained, but at
fuel compositions shifted in the lean direction.

3.2 Flame Spreading Rates

For laminar burning, the rate of flame spreading depends upon the mixture
composition and initial conditions[5] (Drell and Belles, 1958). For hydro-
gen burning in air at standard initial conditions the maximum velocity is about
3 m/s. This velocity increases to 10 m/s at a temperature of 700K. The effect
of pressure is not well documented but is believed to be small.

Higher flame speeds are observed when the flow is non-laminar. Work on
flames propagating in homogeneous turbulent gas mixtures has been correlated by
Abdel-Sayed and Bradley[24] (1977). Ratios of turbulent-to-laminar flame
speeds for hydrogen-air mixtures up to a value of 10 have been observed in

Figure 3-4: Pressure rise ΔP for combustion of near-limit hydrogen-air mixtures [44]. (Ref. L. A. Lovachev et al., Flammability Limits: An Invited Review, *Combustion and Flame*, 20, 259, (1973)).

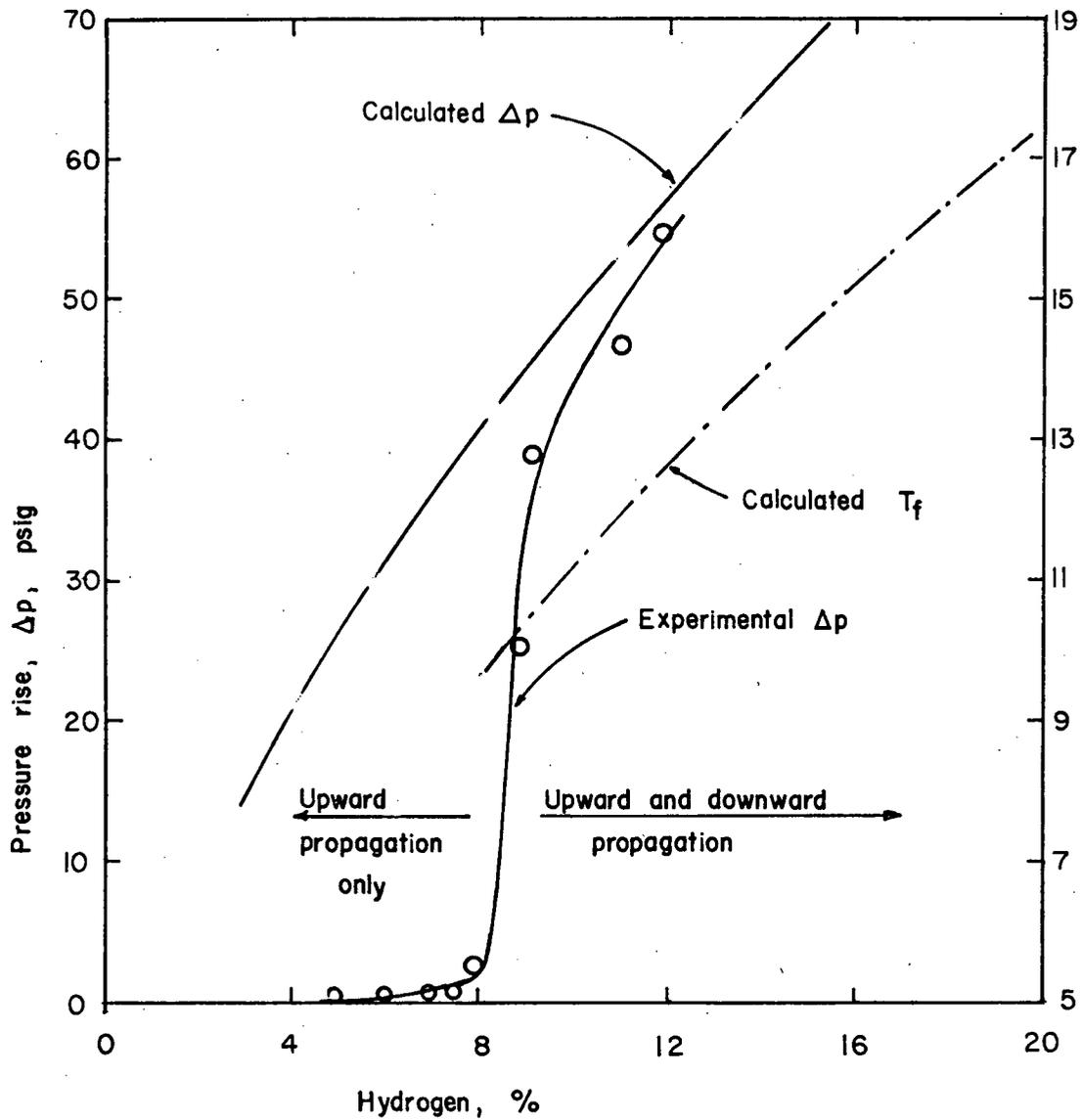slot burners and ratios up to 5 have been observed in confined vessels. Even higher flame speeds have been observed when initially laminar flames propagate past obstacles in the flow field which leads to recirculation and turbulence. For a specially constructed cylindrical chamber (Moen et al., 1980, Reference 14) flame speeds up to 130 m/s have been observed.

3.3 Detonation Processes

A detonation wave may be regarded as a combustion wave that is preceeded by a shock wave in a reactive medium. When the post-shock reaction rates are high enough to prevent the shock wave's decay, the detonation wave becomes a steady-state phenomenon. There are composition limits (Atkinson, et. al. Reference 12) outside of which detonation waves are not observed. These are called limits of detonability. Within the central portions of the detonability range the pressure-temperature-velocity characteristics are calculable from thermodynamic considerations alone. Such a detonation wave is called a Chapman-Jouget (C-J) detonation. In the neighborhood of the detonability limits the C-J calculation overestimates peak P-T values and detonation velocities. The C-J theory is incapable of prescribing limits of detonability.

If a C-J detonation is initiated by a shock wave which is much stronger than that of a C-J detonation, the detonation is said to be "overdriven". The overdriven wave will ultimately decay to a C-J detonation. If the mixture composition is outside of the detonability limits, no shock strength will be adequate to initiate a steady state detonation wave. Unlike the initiation of a deflagration wave (where a very small energy source may be sufficient to initiate the steady state wave) the initiation of a detonation wave usually requires much larger energy sources. The rate of energy input and the spatial distribution of the input energy also affects the required initiation conditions.

Detonations may be initiated most readily in long narrow ducts of various geometries and sizes. In such apparatuses, C-J detonations are frequently observed and appear to be plane waves. For an approximate "point source" initiator, centrally located in a large volume, Atkinson, et. al.[12] have reported the observation of spherical detonations. The preponderant fraction of reported detonation experiment and theory is concerned with planar detonation wave phenomena. Much of the information derived from the study of detonations

in tubes has not been demonstrated to be directly applicable to the character-
ization of spherical detonation waves. Limits of detonability for spherical
detonation waves have not been demonstrated to be the same as those for planar
waves. No widely accepted theory of detonability limits is available.

Some of the important characteristics of detonations are discussed be-
low:

3.3.1 Detonation Initiation

Detonation waves may be initiated in a gas mixture by strong pressure
waves of strengths greater than threshold levels (Zeldovich and Razier, Ref-
erence 10). Initiating shock waves may be generated by mechanical means, or
by an accelerating deflagration wave. Shock tube apparatuses are frequently
used to initiate planar detonation waves in long tubes. An accelerating de-
flagration wave in a closed tube emits pressure waves which may coalesce
(Lewis and Von Elbe, 1962, Reference 2) to provide C-J detonations. For the
spherical configuration, mechanical shock wave generation for initiation is
generally provided by a blast wave (Atkinson, et. al 1980, Reference 12). The
centrally initiated blast wave is, in turn, created by the detonation of a
small solid explosive charge.

The overpressure emitted by an accelerating chemical reaction in a re-
circulating turbulent deflagration wave may induce an asymmetric spherical de-
tonation wave (Knystantas, et. al 1979, Reference 25). Various obstacles and
obstacle patterns may create such recirculating turbulent deflagration waves.
Where detonable mixture ratios exist, such turbulent deflagration waves may
serve as detonation initiators.

In general, any high power density source may ultimately lead to detona-
tion ignition of a detonable mixture. Bach, et. al[26] demonstrated that
intense Laser energy sources could initiate spherical detonation waves in both
solid explosives and in gaseous mixtures.

3.3.2 Detonability Limits

The lower detonability limit for hydrogen-air mixtures in long tubes has
been observed by Zeldovich[11] and others to occur at about 18 percent hy-
drogen. For spherical waves, Atkinson et. al[12] has reported a lean deton-
ability limit to occur at 13 percent hydrogen. The observations reported by

Atkinson et al., (Figure 3-5) are limited to steady state waves which traveled a distance of only 30 cm. without appreciable decay. This distance is too short to establish the steady state nature of such waves. Demonstration of a steady-state C-J detonation in tubes usually requires a much larger spatial traverse. Furthermore, a precise definition of a spherical C-J detonation wave is not agreed upon (Jouget[27], Taylor[28], Zeldovich and Kompaneets[11], Strehlow[29] and Bach[26]). Thus, detonability limits for spherical detonation waves will be difficult to establish by either theoretical or experimental means.

Just outside detonability limits, a decaying detonation wave may still derive substantial support from a post-shock reaction rate. Accordingly, when a propagating detonation wave suddenly enters a spatial regime that is just outside the detonability limits, only slow decay of the shock strength may occur. Therefore, a wall (or other barrier) which is contiguous to a non-detonable mixture may still sustain substantial damage.

3.4 Combustion Generated Pressure Effects

Combustion of a flammable gas mixture in a confined volume leads to an increase in pressure. When the velocity of the combustion process is much less than the speed of sound, the pressure will rise uniformly throughout the volume. For a uniform gas mixture the pressure rise is given by an equilibrium calculation which depends only on the mixture composition and initial conditions (Lewis and Von Elbe, 1962, Reference 2, pp 644 ff). An estimate of pressure rise effects due to combustion of a gas pocket in a confined volume is given by Sibulkin[30].

When the velocity of the combustion process becomes comparable to the speed of sound, dynamical effects occur which lead to pressures exceeding the equilibrium pressure rise. The damage done by a blast or detonation wave to any structure is a function of both the magnitude of the peak pressure and the impulse of the wave (Strehlow, R. A., Reference 29). No damage can be inflicted to a structure if the impulse is below a certain threshold value irrespective of the magnitude of the peak pressure. For a blast wave these two factors are proportional to the energy input and inversely proportional to the distance to the blast center. A local detonation confined in a limited
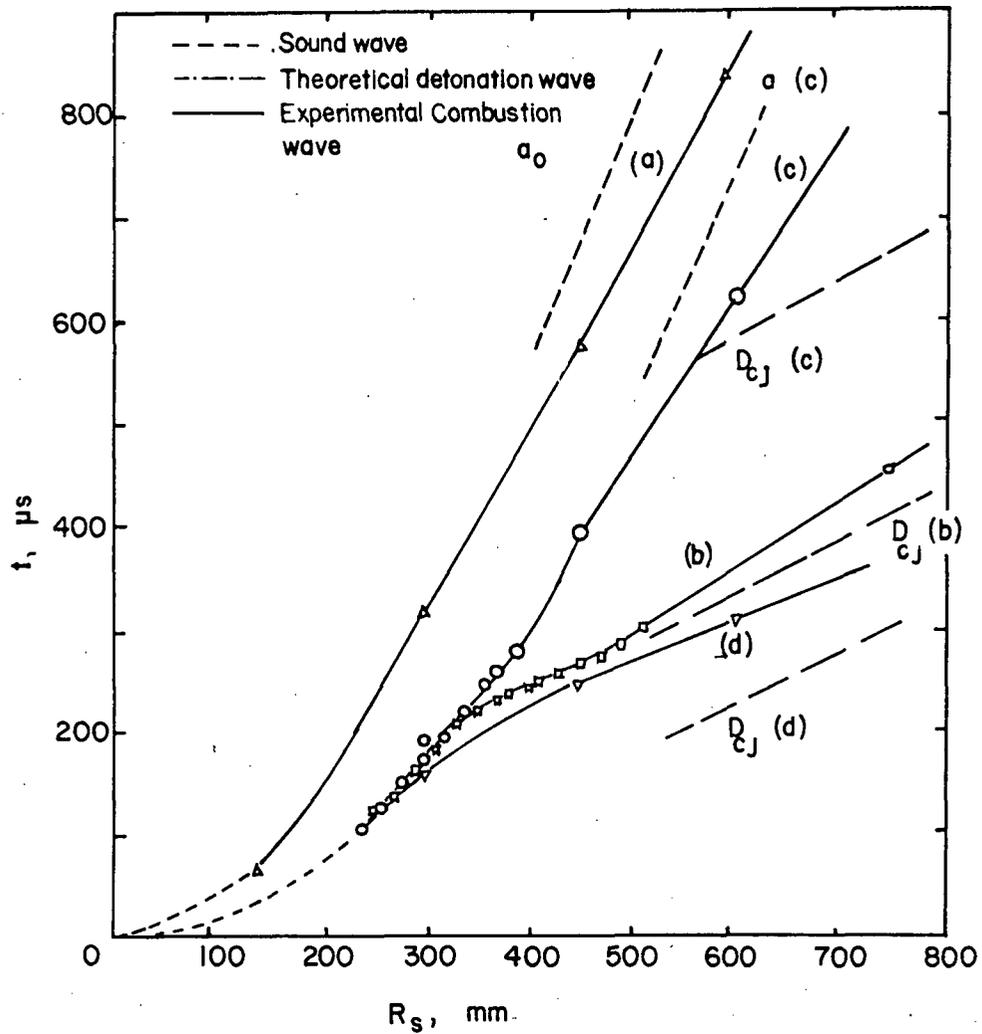
Figure 3-5: Typical radius time plot for combustion and blast waves initiated at 1 atm initial pressure by 1 g Tetryl. (Ref. R. Atkinson et al., Initiation of Spherical Detonation in Hydrogen-Air, Combustion and Flame, 39, 287 (1980).

space in the center of a volume will probably not seriously threaten the integrity of the walls while inflicted damage may be serious if a detonation occurs near the walls.

3.4.1 Effects of Water Sprays

Water droplet spray systems are available in reactor containment buildings to reduce pressure build up by the condensation of steam. Such water sprays may affect combustion of hydrogen in a number of ways.

Where use of a water spray system leads to a large steam component in the gaseous mixture, flame propagation may be completely suppressed. (As noted in Section 2.0, complete inertion of hydrogen-air mixtures occurs for saturated water vapor addition at temperatures above 86°C[6,7].)

Water in the form of liquid droplets, when present in sufficient quantity, will also suppress flame propagation by acting as a heat sink. (The use of liquid water and solid rock dust as flame suppressants is a standard practice in mine safety technology[39].)

At lower concentrations which permit flame propagation, the effect of water droplets will be to lower the equilibrium pressure after burning. The rate of evaporation of water droplets depends inversely on the square of the droplet diameter. Thus, for large droplets, the peak transient pressure associated with a hydrogen burn may exceed the final equilibrium pressure.

4.0 HYDROGEN COMBUSTION CHARACTERISTICS RELATED TO

REACTOR ACCIDENTS

There are a number of proposals which emphasize the "preventive burning of hydrogen" to mitigate the hazards which may derive from degraded core accidents. It is intended that released hydrogen be burned in a predictable and well-controlled manner. These proposals generally assume that:

(a) Mixing processes are adequate to assure "reasonably uniform hydrogen concentrations" within a containment building (or subcompartment);

(b) Hydrogen generation rates are such that "reasonably uniform" hydrogen concentrations increase monotonically in time, until a combustion event causes a reduction;

(c) Combustion initiation can be effected for hydrogen concentrations which are somewhat larger than those for the nominal lean flammability limit;

(d) After such a "preventive burn" the subsequent low hydrogen concentration may then again increase in a "reasonably uniform" manner and the "preventive burn" tactic repeated;

(e) This strategy is intended to provide an acceptable pressure history for the containment building.

If events develop in such a way as to make these assumptions questionable, the success of the "preventive burn" strategy may be in doubt. Clearly, accidental and/or planned hydrogen burns need not necessarily imply catastrophic consequences. The Three Mile Island burn, in itself, did not appear to be an intolerable event.

4.1 Uniformity of Hydrogen Concentrations

Accidental release of hydrogen into an initially normal (preaccident compositions) containment atmosphere invariably leads to nonuniform hydrogen concentrations. Molecular transport and free convective processes tend to suppress these nonuniformities. Forced convective processes (jets and ducted flows) may also be effective in suppressing nonuniform concentrations of hydrogen.

Several processes tend to accentuate the hydrogen concentration nonuniformities. One, of course, is the existence of a localized, continuous

source of hydrogen. In the spatial neighborhood of such a source, hydrogen concentrations are necessarily high, as long as the hydrogen source is significantly active. Where the localized hydrogen source is also a source of steam, the maximum gaseous concentration of hydrogen is limited by the local (gaseous) hydrogen/steam ratio. However, should a local condensation of steam occur, the corresponding hydrogen/steam ratio may increase rapidly. In general, a rapid condensation event (e.g., restoration of containment cooling capability) may correspond to the rapid transformation of an inert hydrogen-air-steam mixture to a flammable hydrogen-air-steam mixture.

Also to be noted is the fact that the low molecular weight of hydrogen acts as an inhibitor to mixing through hydrogen's tendency to stratify in the upper portions of a large volume. The low molecular weight also results in the very high molecular diffusivity of hydrogen which tends to promote mixing.

The question of the spatial and temporal hydrogen concentrations associated with an accident is important. The existence and characteristic properties of combustion phenomena are sensitive functions of composition. The current limited state of knowledge of possible reactor accident scenarios and of condensation and mixing processes limits our ability to predict the spatial and temporal containment building history of hydrogen concentrations. These limitations cast serious doubt on the validity of the previously cited assumptions, (a) - (e).

4.2 Significance of Various Combustion Characteristics

The body of known combustion characteristics may be employed to help assess the combustion-related hazards as well as mitigating strategies that are possible[30] for containment building atmospheres.

4.2.1 Minimum Ignition Energies

Spark ignition of flammable hydrogen-air mixtures can be achieved with very small spark energies. Minimum ignition energies of the order of millijoule or less are typical. The precise values which appear in the literature (e.g., Lewis and Von Elbe, 1962, Reference 2) appear not to be very significant in our deliberations. If, spark igniters are employed, ignition energies required are small. If nonspark ignition occurs (due to operator and/or accident-initiated processes), these data are not applicable as quantitative measures of the ease of ignition.

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4.2.2 Flame Spreading Rates

Flame or deflagration spreading rates have a lower limit specified by the laminar burning velocity. Scenario analyses which involve tabulated laminar flame speeds for hydrogen-air mixtures will generally give rise to minimal rates of hydrogen burning and minimum rates of pressure rise.

Turbulent-burning velocities may correspond to a broad range of experimental conditions and spreading rates. Observed spreading rates may be an order of magnitude or more greater than those tabulated for laminar flames. Scenario analyses which involve such experimental results implicitly assume levels and characteristics of turbulence which may or may not realistically apply to containment building atmospheres. These assumptions could result in an order of magnitude increase in pressure rise rates.

Nonlaminar flame spreading may also occur due to the presence of obstacles or other recirculation-causing conditions. These flame spreading rates also may be an order of magnitude or more. (Moen, et. al)[14] greater than those for laminar flame propagation.

4.2.3 Flame Quenching

Where a containment building consists of more than one compartment (e.g., an ice condenser plant) the question arises as to whether or not a flame in one compartment can propagate through a small opening into another. Figure 4-1 provides quenching distance versus pressure data for hydrogen and air. Openings between compartments (even cracks) as small as 0.01 cm. may, under some circumstances,[5,31] permit flame propagation. Accident analyses and scenarios for mitigation of hazards in multicompartment containment building may need to consider the significance of these very small quenching distances on flame propagation between compartments.

4.2.4 Combustion and Pressure Suppression By Liquid Water Sprays

Liquid sprays may be used to abstract heat from combustion gases, thereby reducing the adiabatic pressure rise. Such procedures are critically dependent upon droplet diameters, droplet number density, the initial thermodynamic state of the containment gases, and the timing and duration of the water spray relative to the combustion event. A combination of large number density and
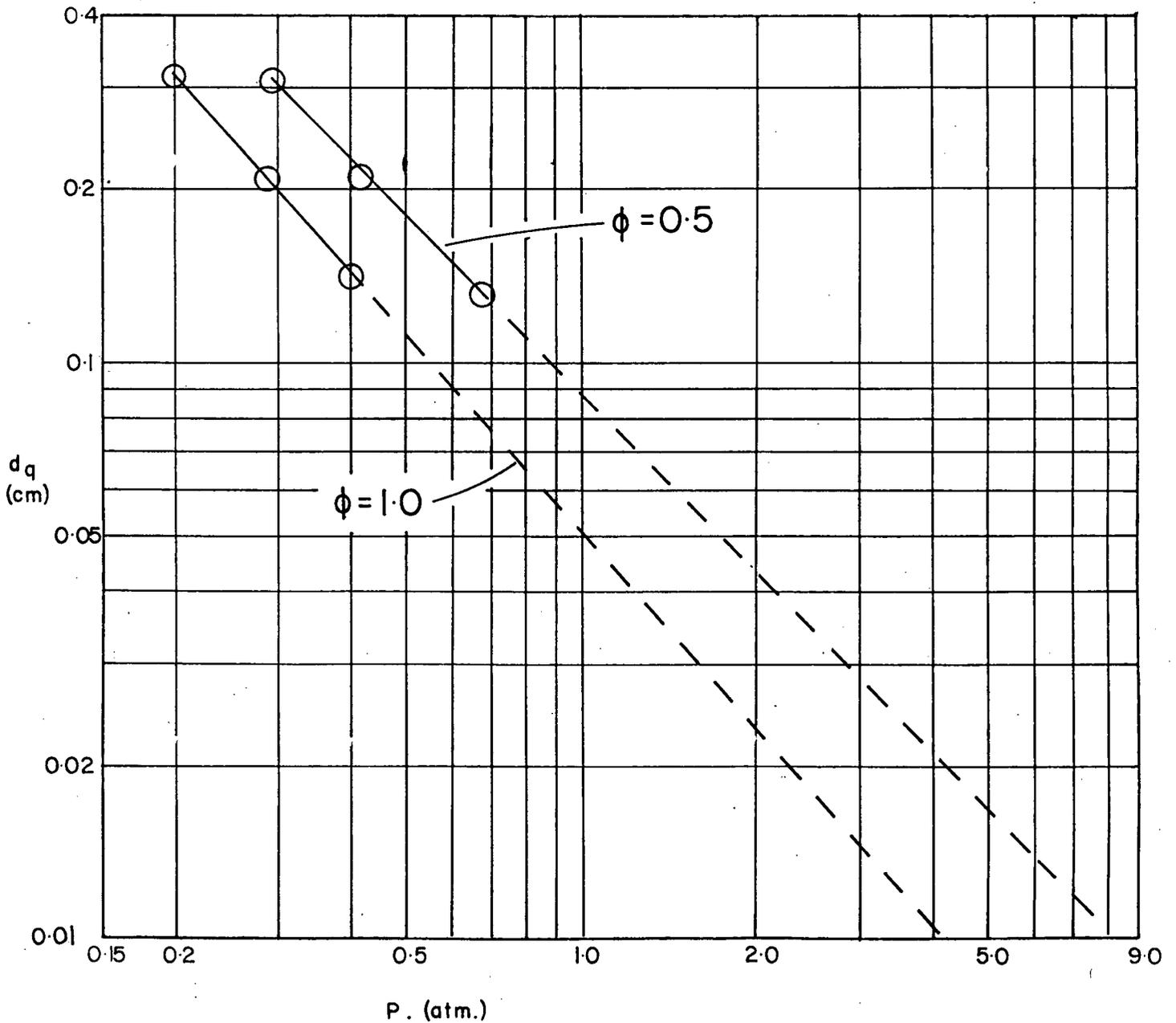
Figure 4-1: Effect of pressure on Quenching Distance of Hydrogen-Air Mixtures. (Ref. A. E. Potter, Jr. and A. L. Berlad, The Effect of Fuel Type and Pressure on Flame Quenching, VI Symposium (1957)).

small drop sizes may prevent a planned combustion event from occurring. Mechanisms of flame extinction include the abstraction of flame reaction zone heat by droplet vaporization, and reduction of the flame reaction zone kinetic rates by inert water vapor addition. Given sufficient time, any continuous spray will lead to a saturated atmosphere. At ambient temperatures above a critical value (e.g., 86°C at 1 atmosphere pressure) such a continuous spray will prevent burning. Under accident conditions, containment pressures may vary, implying varying saturation temperatures. The structuring of water spray/combustion timing to suppress the (transient) peak pressure rise may be ineffective if drop sizes are too large. Large drops may evaporate too slowly to be effective.

4.2.5 Flame Flash Back

Techniques and apparatuses in place to promote mixing of atmospheric constituents include ducts used for forced convective flow. Such ducts may also serve to separate/connect different containment building compartments (e.g., an ice condenser plant). The question as to whether or not a flame in one compartment may "flash back" (against the forced convective flow) into the duct may be addressed through examination of the critical "boundary velocity gradient for flash back". Figures (2-6) and (2-7) provide flash back data for laminar and selected turbulent conditions, respectively. Given a flammable hydrogen-air mixture in a duct which issues into an ignited space, downstream, the very large "boundary velocity gradients for flash back" (of Figures 2-6 and 2-7) show that the potential for flash-back under such circumstances must be considered.

4.2.6 Flame Composition Limits

Composition limits for propagation of a hydrogen-air deflagration are illustrated in Figure (2-5). For hydrogen and air at standard conditions, upward flame propagation at some 4.5 percent hydrogen in air and some 9.0 percent for downward propagation represent limiting conditions on the ability to initiate quasi-steady flame propagation by a localized ignition source. For spatially uniform mixtures, these limits also apply to non-continually operating ignition sources. The composition limits given in the reviews of Drell and Belles[5] and by Lovachev[15,19,21,22] are apparatus dependent.

Accordingly, spatially distributed, continuous ignition sources (in large closed volumes) may correspond to a somewhat different experimental definition of flammability limits than those given by Lovachev. Degree of reactant consumption may be different. As a practical matter, "preventive burn" strategies generally require igniters whose operating limits are characterized as "ignition limits" and are device specific. Device specific "ignition limits" are narrower than the flammability limits defined in either the classic way (for a 5 cm. tube) or by the methods described by Lovachev. For deliberate ignition attempts, the most successful igniters are those which closely approximate "flammability limit" performance with only a small (< 1 sec) ignition delay.

For the assumption of uniform composition in the containment building, literature values of flammability limits provide the approximate boundaries within which (non-catalytically-assisted) combustion processes may occur. Analysts who employ correct values of spatially-averaged hydrogen-air-steam compositions cannot determine, however, the flammability characteristics of various subvolumes of the containment building atmosphere. Depending on the diversity of possible conditions, these may range from "completely inert" to "readily detonable", even for a spatially averaged composition which corresponds to neither of these extremes.

4.2.7 Detonations and Detonation Limits

The concern that detonations may occur in containment buildings stems from the fear of possible damage to the building shell by very high pressure loadings. The current state of knowledge about detonations in uniform mixtures (Section 3.3) is incomplete. Further, the best understood aspects of detonations are for planar waves that may be characterized in terms of the highly simplified Chapman-Jouget theory. Local ignition of spherical detonations in the laboratory normally requires high power density sources (compared to those required to initiate ordinary burning). However, transition from ordinary burning to detonation-like combustion may be possible in large chambers with internal obstructions[25,29]. The limitations of current detonation theory and experiment for central (and noncentral) ignition of detonations in large volumes correspondingly limits our ability to accurately

predict pressure and impulse loading histories for containment building walls. This makes the accurate assessment of possible damage to building walls difficult.

For the case of nonuniform accident-derived hydrogen concentrations, detonable mixtures may exist locally even where spatially averaged hydrogen concentrations are considered nondetonable. Local detonations (or other destructive combustion processes) may damage and incapacitate ducts, ventilators, coolers, recombiners, heat exchangers or other devices whose unimpaired operation during the period of an accident may be considered necessary. Nondetonable mixtures in the immediate vicinity of a wall may not necessarily imply that wall damage by detonations cannot occur. Detonations initiated in spatial regimes which are remote from walls, and sustained by large volumes of detonable gaseous mixtures, may decay only slowly, when propagating through a localized "nondetonable' mixture near a wall. Depending on conditions (where the detonation is initiated and how much detonable gas exists) wall pressure and impulse loading histories may vary substantially.

Although definitive conclusions regarding anticipated damage from detonations cannot be provided, one cannot conclude that containment building walls and building contents are immune to unacceptable detonation damage.

ACKNOWLEDGMENTS

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16. ABSTRACT *(200 words or less)*

A knowledge of combustion phenomena and their characteristics is necessary in accident analyses related to the release of hydrogen. As a result of the accident at Three Mile Island, and from the results of related studies of hypothetical degraded core accidents, it is recognized that combustion of hydrogen may, under some circumstances, threaten the integrity of a reactor containment building. In general, detailed combustion information is required in order to understand and characterize the combustion phenomena and processes which may occur in a containment building. It is also important to identify the criticality or limiting conditions under which important combustion processes may be extinguished, initiated, or otherwise transformed. Data and information is also required for analytic modeling of safety-related hypothetical accident scenarios and to allow modelers to predict with confidence the consequences of combustion processes. Safety-related strategies aimed at mitigation of accident-related combustion also require a knowledge of combustion phenomena. The report identifies areas where inadequate understanding exists and distinguishes among the ranges of applicability of selected items and classes of combustion data, experiments, theories, and models. In pursuit of these objectives, this report attempts to provide a perspective on combustion processes which may not otherwise be derived easily from the enormously diverse combustion literature.

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