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HYDROGEN-AIR-STEAM COMBUSTION REGIMES IN LARGE VOLUMES*

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I. Abstract

Dangerous pressure waves can be generated by the combustion of H_2-air-steam mixtures if ordinary deflagrations accelerate to high speed or undergo deflagration-to-detonation transition (DDT). The purpose of this paper is to estimate the potentially dangerous mixtures in large volumes. There is a limited experimental data base for flame acceleration and DDT of H_2-air-steam mixtures in smaller geometries. There is concern about the possible explosive combustion in the Space Shuttle main engine exhaust duct at Vandenberg AFB. There are no relevant experimental data or valid theories at this large scale (duct width, W, $\simeq$ 10 m) to predict flame acceleration and DDT. We have estimated potentially dangerous mixtures by extrapolating correlations used at smaller scale based on the detonation cell width, λ. In square ducts DDTs are possible if $W/\lambda > 1$. We delineate three combustion regions: nonflammable, weakly flammable, and strongly flammable and potentially detonable. The nonflammable region is the region outside the flammability limit where self-sustaining combustion cannot occur. Flammability limits are independent of scale in large volumes. The strongly flammable region, where dangerous flame acceleration or DDT is possible, is bounded by mixtures with $\lambda = 10$ m. We estimate detonations are possible when there is less than 45% steam. The weakly flammable region, which lies between the other two, should support only slow combustion, where no significant pressure waves should be generated.

II. Introduction

There are at least two situations in which possible violent combustion of hydrogen-air-steam mixtures in large volumes are of concern. One is at the Space Shuttle launch complex on Vandenberg AFB in California, where the Space Shuttle Main Engine (SSME) exhaust is to flow through a large duct of width $\simeq$ 10 m and length $\simeq$ 100 m. Under conditions of an aborted launch or a flight readiness firing, hydrogen continues to flow from the SSMEs for about two seconds after the liquid oxygen shutoff. There is concern that hydrogen in the duct could either detonate or rapidly deflagrate giving rise to a pressure pulse that could damage the orbiter aft end. A steam-inerting system for the duct is being developed [1] to prevent any potential damage to the orbiter. However, even with the steam-inerting system, small volumes in the duct may contain flammable mixtures. Downstream of the duct exit, flammable mixtures may occur when the hot exhaust gases mix with the cool surrounding air, thus condensing the steam. There is a need to know what flammable mixtures are incapable of violent combustion and, hence, can be tolerated.

A second situation is in nuclear reactor safety. During severe accidents in which there is core damage, large amounts of

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hydrogen can be formed from the oxidation of zirconium fuel cladding and from other sources. Hydrogen-air-steam atmospheres may exist in reactor containments. Such containments are large structures with volumes of up to 8×10^4 m^3. Slow deflagration (Mach number <<1) of a flammable atmosphere will result in quasi-steady loads on the containment. A detonation or highly accelerated deflagration will cause additional impulsive loads [2].

For these applications we can divide hydrogen-air-steam mixtures into four classes:

1. Nonflammable mixtures. Such mixtures can burn near an ignition source but will not propagate a flame.

2. Weakly flammable mixtures. Such mixtures can propagate a slow deflagration but cannot propagate a fast deflagration. They cannot generate strong pressure waves.

3. Strongly flammable mixtures. Deflagrations in such mixtures can be accelerated to speeds of hundreds of meters per second. They are capable of generating shock waves.

4. Detonable mixtures. Such mixtures can undergo deflagration-to-detonation transition (DDT). These are the most dangerous mixtures.

We present the current knowledge of the boundaries of these regions and make estimates where the boundaries are not known.

III. Flammability Limits

The flammability limits of a fuel-air mixture are defined as the limiting concentrations of fuel, at a given temperature and pressure, in which a flame can be propagated indefinitely [3,4,5]. The limits are assumed independent of the method of ignition, as long as it it sufficiently strong to start the flame, and of the size of the enclosure, as long as it is much larger than the quench distance. The hydrogen flammability limits depend on buoyancy. They are wider for upward propagation than downward, particularly for lean hydrogen-air mixtures. They may also be influenced by initial turbulence.

The flammability limits of hydrogen-air mixtures at ambient temperature and pressure are [4]

Lean		Rich	
upward	4.1% H_2	upward	74% H_2
downward	9.0% H_2	downward	74% H_2

The behavior of the hydrogen-rich limits are best understood as oxygen-lean limits, where the oxygen-lean limit is 5%. The effect of the addition of a third inert diluent gas is to narrow the gap between lean and rich limits. If sufficient diluent is added, the two limits merge. If still more diluent is added, the mixture is said to be inerted, i.e., the mixture will be nonflammable for all hydrogen-air ratios.

Measurement of the hydrogen-air-steam flammability limits is made more difficult by the requirement that the apparatus be heated slightly above the saturation temperature. The experimental data available were taken at slightly superheated temperatures and near atmospheric pressures [6-14]. The results of Marshall [14] for initially quiescent atmospheres are shown in Fig. 1. Comparison of the upward flammability limits of Marshall and other researchers [7-13] in Fig. 2 [15] indicates reasonable agreement. The one area of possibly significant difference is the "nose" of the flammability limit curve, the region of nearly stoichiometric mixtures near the inerting level. Marshall obtained an inerting level of about 52% steam, while other researchers have shown various values up to 60%. For mixtures further from stoichiometric, the various flammability limits are in closer agreement.

To examine the effect of initial turbulence on the hydrogen-air-steam flammability limits, Marshall [14] used a fan to generate turbulence. There were negligible differences in flammability limits between initially quiescent and initially turbulent atmospheres. This is in accord with the results from similar tests done with fans by other researchers [7-13]. However, for weakly combustible mixtures near the flammability limits, moderate turbulence can greatly increase the completeness of combustion [5,14].

IV. Detonation Limits

Flammability limits of combustible gas mixtures are insensitive to the volume of the container, for characteristic dimensions above a few centimeters. In contrast, detonation limits continue to widen with increasing geometric scale of the surroundings. The early detonation limits of 18% and 59% hydrogen for hydrogen-air mixtures at ambient temperature and pressure, still cited in the literature [16], were based on older measurements in small-diameter tubes. More recent measurements in a larger diameter (43 cm) tube are approximately 12% to 75% hydrogen [17,18]. Detonations have been achieved with stoichiometric hydrogen-air-steam mixtures with up to 35% steam [17,18].

The detonation limit results obtained for hydrogen-air, hydrogen-air-steam, and other fuel-oxidizer mixtures can be understood in terms of the present understanding of

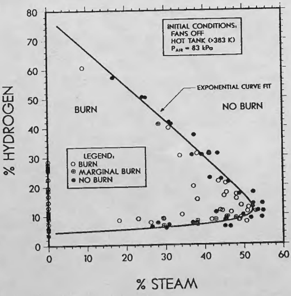

FIGURE 1. Hydrogen-Air-Steam flammability data in quiescent atmosphere from Ref. 14. The flammability limit data is fit with the following empirical equation:

$$\%Steam = 100 - \%H_2 - 37.3 \cdot exp(-0.007\%H_2) - 518.0 \cdot exp(-0.488\%H_2)$$

detonation structure, in particular, of the detonation cell width. The first comprehensive theory of detonation structure is called the Zeldovich-Von Neumann-Doering (ZND) model [19]. The ZND model consists of a chemically frozen planar shock wave followed by a chemically reacting planar deflagration zone. However, experimental evidence of gaseous detonations clearly shows the structure to consist of a more complex unsteady three-dimensional shock wave structure. Incident, transverse, and reflected shockwaves meet in Mach triple point intersections [20]. The chemical reactions take place behind the shock waves, most

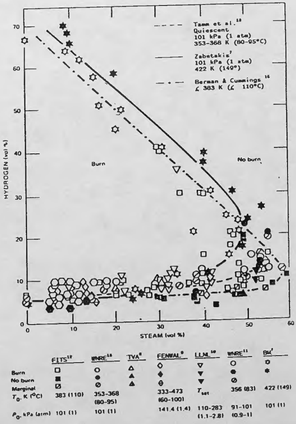

FIGURE 2. Hydrogen-Air-Steam flammability data. This figure from Ref. 15 plots data from Refs. 7-14. [FITS, Fully Instrumented Test System (Sandia National Laboratories); WNRE, Whiteshell Nuclear Research Establishment (Canada); TVA, Tennessee Valley Authority; Fenwal, Fenwal, Inc.; LLNL, Lawrence Livermore National Laboratory; and BM, Bureau of Mines.]

rapidly behind the strongest shock wave segments. The path of the Mach triple points can be recorded using their luminosity with high-speed cinematography, or from the diamond shaped tracks they leave on sooted surfaces. The width of these diamonds is called the detonation cell width of the mixture, λ. It is mainly a property of the reactivity of the detonable mixture, only slightly affected by the geometric confinement. Highly reactive mixtures give small detonation cell widths, while less reactive mixtures give larger detonation cell widths.

The detonation cell width is of great importance in our study for two reasons: it is a measure of the capability for detonation propagation; and, as a measure of chemical reactivity, it is an indication of the ability of a mixture to undergo a fast deflagration and DDT. The capability of a detonation to propagate down a tube depends on the ratio of the tube size to the detonation cell width [21]. For circular tubes with diameter D, propagation is possible if $D/\lambda > \sim 0.3$. The limiting mode of propagation in the circular duct is the single-head-spin mode. For rectangular ducts of large aspect ratio with width W, detonation propagation requires $W/\lambda > 1$. Although there are no data on the limiting condition for square ducts, the value of W/λ should be closer to that of the high aspect ratio rectangular duct than the circular duct because there is no spinning mode. In round ducts with annular internal obstacles, DDT was found possible only if $d/\lambda \geq 1$, where d was the diameter of the obstacle opening [22]. Similarly, in a rectangular duct with obstacles, DDT was found possible if $s/\lambda > 2$, where s was the distance between parallel obstacles [23]. Note that for DDT and detonation propagation, the ratio of characteristic length to detonation cell width is never less than unity, except for the detonation propagation through smooth round tubes. Therefore, we conservatively choose the criterion for the possibility of DDT in a large square duct of width W

$$W/\lambda \geq 1.$$

For the SSME duct application, potentially detonable mixtures would be those where $\lambda \leq 10$ m. Note that since detonation limits are wider in larger geometries, detonation limits determined in smaller apparatus are nonconservative.

Measurements have been made of the detonation cell width for hydrogen-air-steam mixtures in the Heated Detonation Tube at Sandia National Laboratories [17,18]. The results are shown in Fig. 3. Increasing steam mole fractions reduces the mixture reactivity and increases the detonation cell width. For a given initial steam mole fraction, the detonation cell width is a minimum near stoichiometric hydrogen-air ratio.

Because of apparatus limitations, the largest detonation cell widths that have been measured are about 1 m. There are no experimental data on detonations for marginally detonable mixtures with detonation cell widths $\simeq$ 10 m. The ZND model of a detonation can be considered to be correct in some average sense compared to the more complex present picture.

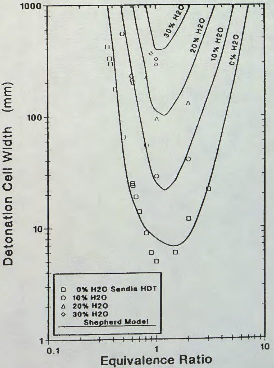

FIGURE 3. Measured detonation cell width for hydrogen-air-steam mixtures at 100° C. Air density is between 40.4 and 42.0 moles/m^3. From Ref. 17.

Westbrook [24] and Shepherd [17,25] have computed reaction zone lengths predicted by the ZND model and compared them to detonation cell width data. Shepherd [25] found that the detonation cell width for stoichiometric hydrogen-air mixtures was about 22 times the ZND chemical length. When he compared the ratio of the ZND chemical length to the experimentally determined detonation cell width for off-stoichiometric hydrogen-air mixtures, hydrogen-air-steam mixtures, and hydrogen-air-carbon dioxide mixtures, the ratio was found to hold within a factor of $\pm$ 2 while λ varied over several orders of magnitude. This indicates a ZND calculation of chemical length can be used to make a rough estimate of the detonation cell width. We have carried out calculations to estimate the detonation cell width using the Shepherd ZND code. The results are shown in Fig 4. Of importance to this study are the values of equivalence ratio and hydrogen mole fraction for λ = 10 m shown in Figs. 5 and 6 respectively. The region interior to these curves is considered potentially detonable in large 10-m width ducts.

The results in Fig. 6 indicate that detonations for stoichiometric mixtures are possible in a 10-m-square duct up to 45% steam. Detonations have been observed in the 43 cm diameter Heated Detonation Tube up to 35% steam [18]. For hydrogen-air mixtures our theoretical results indicate a lean detonation limit of about 12% hydrogen and a rich limit of about 75% hydrogen. This is in accord with Heated Detonation Tube results, and therefore may not be conservative in larger geometries.

V. Strong Deflagration Limits

For highly reactive mixtures, slowly moving deflagrations can be accelerated to high speeds by any of several mechanisms such as obstacles in the flow path, initial turbulence, flow-combustion instabilities, etc. Flame acceleration in hydrogen-air and other fuel-air mixtures was observed in tests using a long tube, closed at the ignition end and open on the far end, with numerous obstacles in the tube to accelerate deflagrations to a terminal flame

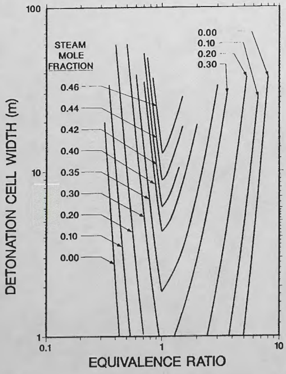

FIGURE 4. Estimated detonation cell width for hydrogen-air-steam mixtures at 1 atm pressure and saturation temperature.

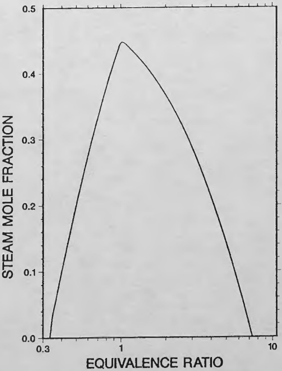

FIGURE 5. Estimated steam mole fraction for 10 meter detonation cell width.

velocity [22,26]. It was found that the deflagration of very lean and very rich mixtures would be quenched as shown in Fig. 7. The quench limit of 10% hydrogen appeared to be independent of the tube size up to the largest tube used, 30 cm diameter. For mixtures near the stoichiometric ratio, DDT was observed. The region of transition to detonation was wider in the larger diameter tubes. On both lean and rich ends, a narrow region between quench and DDT was observed where the deflagration accelerated to the maximum deflagration speed, that corresponding to choked conditions behind the deflagration. This "highly accelerated deflagration" region became narrower as the tube diameter became larger. The only data on flame acceleration in hydrogen-air-steam mixtures we have obtained are those of Brehm [27] in a 6.7 cm diameter tube. He obtained a highly accelerated deflagration for stoichiometric mixtures with up to 35% steam. With the present understanding of the strong deflagration limit, it seems that at large scale it will be close to the detonation limit. We assume them to be identical.

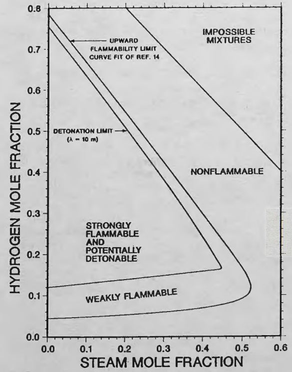

FIGURE 6. Hydrogen-Air-Steam combustion regimes.

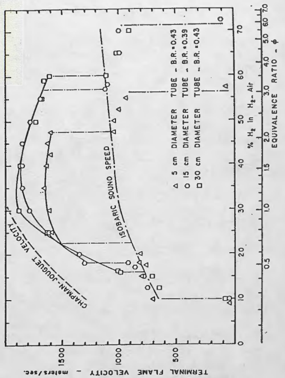

FIGURE 7. Asymptotic flame speed observed in 5, 15, and 30 cm dia. tubes with annular obstacles. From Ref. 22.

VI. Conclusions

For stoichiometric hydrogen-air-steam mixtures, experimental results indicate mixtures with up to 35% steam can detonate. The inerting mole fraction of steam measured in moderate-sized apparatus is about 55% and is believed independent of scale. There is a lack of experimental data on detonation and highly accelerated deflagrations of hydrogen-air-steam mixtures at large scale. Strong deflagration limits are considered to be very close to detonation limits at large scale. For the 10-m-square duct at Vandenberg AFB, our estimate of the potentially dangerous mixtures are shown in Fig. 6. This figure shows that detonations may be possible in stoichiometric mixtures with up to 45%steam. Lesser mole fractions of steam are required to prevent violent combustion in off-stoichiometric mixtures. Figure 6 has become a valuable tool for the design of the SSME steam inerting system by showing the region of tolerable combustible mixtures.

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