# **Critical Tube Measurements at Elevated Initial Mixture Temperatures**

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## **INTRODUCTION**

The phenomenon of planar detonation wave diffraction at an abrupt area change involves the quenching of the detonation by expansion waves generated at the corner of the area change. The intrusion of the expansion waves into the detonation reaction zone results in a drop in temperature leading to the decoupling of the shock wave and reaction zone. For a given mixture initial composition, pressure, and temperature, there exists a minimum tube diameter, i.e., the critical diameter, above which a spherical detonation wave forms downstream of the area change, and below which the detonation wave decouples and fails. It has been reported in the literature that for a circular tube the minimum tube diameter required for transmission is  $d_c = 13\lambda$ , where  $\lambda$  is the average detonation cell size corresponding to the mixture condition [1,2]. Deviations from this correlation have typically been attributed to the uncertainty in the measurement of  $\lambda$ , which can be as high as  $\pm 100\%$  for off-stoichiometric mixtures with very irregular cellular structures. For mixtures with high argon dilution, critical tube measurements of  $d_c = 20-30\lambda$  have been reported [3,4,5]. This is considered a significant departure from the  $d_c = 13\lambda$  correlation. Arguments have been made that the breakdown of the  $13\lambda$  correlation observed in high-argon diluted mixtures can be attributed to the regular detonation cellular structure they display. A similar breakdown in the correlation is observed in severely overdriven detonation waves that also display a more regular cell structure [5]. A theory has been proposed to explain the difference between the critical tube experimental observations for regular and irregular detonations [6]. Critical tube measurements in fuel-air mixtures are typically limited to stoichiometric composition because of the limited size of a typical laboratory apparatus. In the present tests mixture reactivity is enhanced by testing at higher initial temperatures, and thus permitting a broader range of mixture conditions to be investigated. The objective of the present study is to determine the applicability of the  $d_c = 13\lambda$  correlation over a range of hydrogen-air mixture compositions and initial temperatures.

## **EXPERIMENTAL DETAIL**

The experiments were carried out in a heated detonation tube that is described in detail in ref. 7. The apparatus consists of a cylindrical test vessel section 27.3 cm inner-diameter by 9.1 m long, connected to a 0.89 m innerdiameter 1.22 m long receiver vessel. The detonation wave is initiated in the test vessel via an oxygenacetylene gas driver system. The mixture is produced by coaxial mixing of metered hydrogen and air flow streams piped directly into the pre-evacuated test vessel. All tests were performed at an initial pressure of 1 atm. The vessels are heated using electric resistance heaters covering the outside surface of the vessels.

The critical condition for detonation transmission is determined by fixing the tube diameter and the initial temperature and pressure and varying the mixture hydrogen composition. Three tube diameters were tested by the use of a interchangeable transfer tube positioned at the transition point between the test vessel and the larger diameter receiver vessel, as shown in Fig. 1. The core of the detonation wave propagates in the transfer tube and then transfers into the receiver vessel. The transfer tube protrudes back 15.2 cm into the last test vessel, with the tube-end machined to a sharp edge so as to minimize shock reflection effects. The outlet end of the transfer tube protrudes several centimeters into the receiver vessel so as to avoid detonation initiation by shock reflection. Tests were performed with a 10 cm and 20 cm inner-diameter transfer tube, as well as with the bare 27.3 cm vessel without a transfer tube.



Figure 1: Schematic of test vessel showing transfer tube geometry

The diagnostics used in the experiment are photo diode detectors and fast-response piezoelectric pressure transducers. The detonation wave velocity along the test vessel is measured to ensure that steady-state conditions exist at the entrance of the receiver vessel. As shown in Fig. 1, there are two pressure transducers located in the first two instrumentation ports on one side of the receiver vessel, spaced 20.3 cm apart. The pressure signals are used to discern the nature of the wave transmitted into the receiver vessel, e.g., blast wave or detonation wave. A photo diode used to measure the reaction zone time-of-arrival is located opposite the first pressure transducer in order to deduce the separation distance between the shock wave and the reaction zone. Three gas samples are taken just before the detonation is initiated. These samples are analyzed using a gas chromatograph in order to verify the hydrogen concentration. Detonation cell size measurements are made near the end of the test vessel, 15.2 cm before the leading edge of the transfer tube. The detonation cellular structure is recorded on a small 6.35 cm diameter smoked foil that is fastened to the inside surface of an extractable instrument port plug contoured to the test vessel inner-diameter.

## **EXPERIMENTAL RESULTS**

Tests performed using the 10 cm transfer tube with a mixture of stoichiometric hydrogen-air at both 300K and 500K resulted in detonation failure, and therefore a critical composition for this combination of tube diameter and initial temperature cannot be measured. In these tests, detonation failure in the receiver vessel is characterized by a pressure pulse at the receiver vessel pressure transducers with a magnitude much lower than that of a CJ detonation wave. Concurrently, the receiver vessel photo diode measures a very slow rise in light intensity. The wave velocity between the two pressure transducers is close to the mixture speed of sound, characteristic of a weak blast wave. For tests done with the 10 cm transfer tube at 650K, mixtures between 26.3% and 30% hydrogen resulted in detonation transmission. The measured peak overpressure at the first pressure transducer is 17.7 atm which is just over the 15.7 atm calculated normal reflected CJ pressure. The receiver vessel photo diode indicates an initial small increase in light intensity associated with the detonation emerging from the transfer tube, followed immediately by a very sharp rise in the light intensity marking the arrival of the detonation wave at the photo diode detector. For mixtures with less than 26.3% hydrogen the detonation fails while entering the receiver vessel but re-initiates at the rear elliptical-head cover of the receiver vessel as a result of shock focussing of the reflected blast wave. This type of initiation was also observed by Krok [8] in hot jet experiments performed in a similar receiver vessel. Since the time required for this process to occur is much longer than the transit time for the decoupled shock to travel from the tube exit to the first pressure transducer, it does not interfere in the assessment of detonation transmission.

In general, the phenomenon of transmission and failure of the detonation wave observed in the 20 cm and 27.3 cm diameter tube is very similar to that just described for the 10 cm tube. The measured minimum hydrogen concentrations required for detonation transmission for the three tube diameters and the three initial temperatures are given in Table 1. The critical hydrogen concentration of 29.1% at 300K for the 20 cm tube is in excellent agreement with the results reported in ref. 2. In the 20 cm diameter tube, tests performed at 650K in mixtures near the critical condition resulted in detonation initiation via reflection of the decoupled shock wave from the receiver vessel side-wall. For mixtures with more than 20.0% hydrogen, detonation transmission was assessed based on the pressure trace displaying a detonation-like profile with a peak pressure consistent with the mixture reflected CJ detonation pressure. For mixtures with less than 19.0% hydrogen, detonation failure was assessed based on a pressure signal characteristic of a weak blast wave, followed by detonation initiation due to shock focusing from the rear vessel cover. In two tests with 19.6% and 20% hydrogen, the pressure transducer signals indicate that detonation initiation resulted from shock reflection of the decoupled shock wave from the receiver vessel side-wall. In these tests the first pressure transducer signal shows the arrival of a blast wave (typically 2.8 atm overpressure) followed shortly after by a much stronger detonation wave like signal with a peak pressure of 20 atm relative to the initial pressure. The signal recorded on the second pressure transducer is characteristic of a detonation wave, indicating that transition to detonation occurred in the near vicinity of the first pressure transducer.

When analyzing detonation cellular structure from a smoked foil one typically measures the average cell size. The average cell size is obtained by identifying dominant diagonal bands of similarly shaped cells and measuring the average soot track line spacing. One also characterizes the regularity of the cellular structure in terms of the distribution in the measured line spacing. A regular structure is one where the distribution is small and an irregular structure is one that has a large distribution in line spacing. Typically within the dominant bands, finer track lines appear and disappear, lines merge and branch, and long lasting lines tend to drift between the two dominant lines. One can include the presence of this substructure as part of the characterization of cell regularity even though one typically does not consider these lines in the measurement of the average cell size.

Tube	Temperature	Hydrogen	Cell Size(cm)			$d/\lambda_c$		
Dia. (mm)	(K)	(%)	Min.	Ave.	Max.	Min.	Ave.	Max.
	300	26.5	11.0	13.5	16.0	17.0	20.2	24.8
273	500	23.3	12.0	14.6	17.0	16.1	18.7	22.7
	300	29.1	7.0	10.3	15.0	13.3	19.4	28.6
200	500	24.6	9.0	10.5	14.0	14.2	19.0	22.2
	650	20.7	7.0	8.5	10.0	20.0	23.5	28.6
100	650	26.3	4.0	5.5	6.5	15.4	18.2	25.0

Table 1: Critical mixture condition for each tube diameter and corresponding cell size measurement

The critical mixtures for the different tube diameters and initial temperatures are listed in Table. Displayed in Fig. 2 are three smoked foil samples taken for each of the 20 cm tube critical mixtures, one for each initial temperature. The diameter of the circular foil, i.e., 6.2 cm, can be used as a reference scale. The cellular structure observed in the 29% hydrogen mixture at 300K is typical for hydrogen-air mixtures at 300K. The dominant track lines are fairly well defined with finer-scale substructure evidenced by varying shades of gray

between the dominant lines. The lines defining the substructure are not very pronounced. For the 19% hydrogen mixture at 650K, the dominant track lines are very well defined and no significant substructure is detectable. The dominant line spacing regularity in the 650K foil is *comparable* to that of the 300K foil. In the 650K foils the apex of the cell can be identified by a local bright spot, or removal of soot on the foil. This soot dispersal is caused by the "local explosion" produced by the collision of transverse waves. These bright spots prove very useful in selecting the dominant lines. The lines that make up the cellular substructure, which is difficult to see in the 300K foil, are more apparent in the 500K foil corresponding to a 24.5% hydrogen mixture. The substructure in the 300K and 500K foils makes the selection of the dominant lines, that are subsequently used in the determination of the average cell size, more subjective. More so for the 500K foil since the finer lines are more defined. In the 650K there is little substructure present and thus the measurement of the average is less subjective.



Figure 2: Smoked foils for critical mixtures in 20 cm tube

The measured average detonation cell size for the various critical mixtures at the three initial temperatures are given in Table 1. Also given are the smallest and largest cell size measured on each foil. In Fig. 3 the average cell size measurements for the 300K and 650K tests are plotted in conjunction with previously reported data [9]. Also shown in Figure 3 are cell size predictions using the calculated one-dimensional ZND reaction zone length multiplied by a proportionality constant "A". The agreement between the present measurements and those made in the previous tests are very good, and satisfactorily reproduced by the ZND predictions. Based on the values given in Table 1 the average deviation from the average cell size is  $\pm 22\%$ . This is in very good agreement with the results found for the cell size data from ref. 9 where an average deviation of  $\pm 25\%$  was reported. In the composition range of interest to the present critical tube tests, the cell size deviation per foil is larger than the shot-to-shot variation in the measured average cell size given in Fig. 3. If one defines cell regularity based on the cell size deviations one sees that there is no effect of initial temperature on cell regularity for these mixtures and temperatures tested.



Figure 3: Average cell size measurements

The value of  $d/\lambda$  based on the average, minimum, and maximum cell size for each of the critical conditions is given in the last three columns of Table 1. The average  $d/\lambda_c$  values range from a low of 18.2 to a high value of 23.5. Based on the minimum and maximum cell sizes reported in Table 3, the value of  $d/\lambda_c$  range from 13 to 29.

## DISCUSSION AND CONCLUSIONS

The determination of the critical mixture composition for a given tube diameter is nonsubjective and very repeatable. For example, in the present tests the threshold between transmission and failure could be identified down to a fraction of a percentage of hydrogen. Any uncertainty in assigning a value for  $d/\lambda_c$  rests on the measurement of the critical mixture average detonation cell size. It has been argued that for heavy argon diluted mixtures displaying a very regular cellular structure  $d/\lambda_c$  takes on a value between 20 and 30 and therefore the  $d/\lambda_c = 13$  criterion breaks down [3,4,5]. This implies that applicability of  $d/\lambda_c = 13$  is restricted to irregular cell structure detonations. In the present tests performed over a range of tube diameters and mixture compositions the critical mixture condition was found to have an average value of  $d/\lambda_c$  between 18 and 24, with no correlation with initial temperature. For the measured foil minimum and maximum cell size, the value of  $d/\lambda_c$  lies between 13 and 29 which covers the entire range of values reported for high argon diluted mixtures. These high value measurements of  $d/\lambda_c$  are corroborated by recent critical tube experiments performed in hydrogen-oxygen-nitrogen mixtures that resulted in a value of  $d/\lambda_c = 21$  [10]. In the same study tests performed using methane and propane in oxygen and nitrogen mixtures resulted in  $d\lambda_c$  of 24 and 21, respectively, demonstrating the universality of this finding. The measured uncertainty in the average cell size measurement of  $\pm 25\%$  is not sufficient to explain these significant departures from  $d/\lambda_c = 13$ . This leads one to the conclusion that the  $d/\lambda_c = 13$  is not a unique correlation that breaks down only for special classes of detonations. The exception made in the past for high argon diluted mixtures, or regular structure detonations in general, is not warranted and should instead be considered an indication that the correlation is not sufficient to describe the complexity of the detonation diffraction phenomenon.

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### REFERENCES

- 1. Mitrofanov, V.V. and Soloukhin, R.I., Soviet Phys. Dokl. 9:1055 (1964).
- 2. Knystautas, R, Lee, J.H., and Guirao, C.M., Combust. Flame 48:63-83 (1982).
- 3. Moen, I.O., Sulmistras, A., Thomas, G.O., et al., Prog. Astronaut. Aeronaut. 106:220-243 (1986).
- 4. Shepherd, J., Moen, I.O, Murray, S., Thibault, P., *21st Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, PA, p.1649-1658, 1986.
- 5. Desbordes, D., Guerraud, C., Hamada, L., and Presles, H.N., *Prog. in Astronaut. Aeronaut* 153:347-359 (1993).
- 6. Lee, J.H., Dynamics of Exothermicity, Combustion Science and Technology Book Series, ISSN 0883-5519, Vol. 2, R. Bowen ed., Gordon Breach Publ., NY, 1996.
- 7. Ciccarelli, G., Ginsberg, T., Boccio, J., et al., "Detonation Cell Size Measurements in High Temperature Hydrogen-Air-Steam Mixtures at the BNL High-Temperature Combustion Facility," NRC Report, NUREG/CR-6391, 1997.
- 8. Krok, C., "Jet Initiation of Deflagration and Detonation," California Institute of Technology, Explosion Dynamics Laboratory Report FM97-11, 1997.
- 9. G. Ciccarelli, T. Ginsberg, J. Boccio, Combust. Science Tech. 128:181-196, 1997.
- 10. Shultz, E. and Shepherd, J., Detonation Diffraction Through a Mixture Gradient", Cal. Inst. Tech., GALCIT, EDL Report FM00-1, February 8, 2000.