Detonation Cell Width Measurements for $H_2-N_2O-N_2-O_2-CH_4-NH_3$ Mixtures

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Introduction

Detonations of mixtures containing hydrogen and nitrous oxide were investigated in the GALCIT detonation tube (280 mm diameter, 7.3 m long). The facility and previous related studies are described in Akbar et al. (1997). We measured the detonation cell width, velocity and pressure for a range of equivalence ratios in three mixtures: 1) hydrogen-nitrous oxide; 2) hydrogen-nitrous oxide with 30% nitrogen dilution; 3) hydrogen-nitrous oxide with 30% nitrogen dilution, we investigated the influence of adding 3% methane or 3% ammonia on the detonation behavior to hydrogen-nitrous oxide mixtures. Tests were conducted at initial pressures and temperatures of 70.9 kPa and 295 K, respectively.

One-dimensional, steady, (ZND model) reaction zone calculations were performed with the modified Miller and Bowman hydrogen-nitrous oxide-methane-ammonia-oxygennitrogen mechanism (Akbar et al. 1997). These calculations were used to correlate and extrapolate the measured cell widths and also to determine the effect of initial conditions on the cell width.

Cell Width Measurements

Figures 1 and 2 show the measured cell widths for hydrogen-nitrous oxide, hydrogennitrous oxide-nitrogen, and hydrogen-nitrous oxide-nitrogen-air mixtures plotted versus the initial hydrogen concentration and the equivalence ratio, respectively. The equivalence ratios shown are based on the total amount of fuel and oxidizer, including the nitrous oxide. Although the cell width vs concentration curves appear to have the familar "U" shape obtained for hydrocarbon fuels in air, there are some significant differences when nitrous oxide is used as the oxidizer instead of oxygen. Since nitrous oxide will exothermically decompose even in the absence of fuel, the cell widths for lean mixtures are substantially smaller than in an hydrogen-oxygen-diluent mixture with the same equivalence ratio. This point is explored further in subsequent discussion.

The cell width increases with increasing dilution of either air or nitrogen at a fixed equivalence ratio. Note that the addition of air shifts the location of the stoichiometric point, making the concentration plot, Fig. 1, less useful than the equivalence ratio plot, Fig. 2 for comparing different mixtures. The trend of cell size with dilution is similar to that obtained in hydrogen-air mixtures (Berman 1986).

Note that the largest cell widths shown (100-200 mm) for each case are only indicative of the largest cell width that can be reliably be measured in our tube. Richer and leaner mixtures could potentially be detonated. In particular, there was little interest in the rich air-diluted cases and no experiments were performed for hydrogen concentrations greater than 35% for that mixture. The case of very lean hydrogen-nitrous oxide mixtures is exceptional and is discussed further below.



Figure 1: Measured cell width for hydrogen-nitrous oxide-nitrogen-air mixtures vs. hydrogen concentration. Initial conditions of 70.9 kPa and 295 K.

Adding 3% methane or ammonia significantly changes the cell widths of hydrogennitrous oxide mixtures. Figures 3 and 4 show the measured cell widths of hydrogennitrous oxide mixtures in comparison to hydrogen-nitrous oxide-methane and hydrogennitrous oxide-ammonia mixtures. For stoichiometric mixtures (overall equivalence ratio), adding 3% methane increases the measured cell width from 2.1 mm (see Fig. 16 and Table 6) to 6 mm, a factor of approximately 2.9. Adding 3% ammonia increases the measured cell width to 12 mm, a factor of approximately 5.7. Therefore, both components act as inhibitors.



Figure 2: Measured cell width for hydrogen-nitrous oxide-nitrogen-air mixtures vs. equivalence ratio. Initial conditions of 70.9 kPa and 295 K.



Figure 3: Measured cell width for hydrogen-nitrous oxide-methane-ammonia mixtures vs. hydrogen concentration. Initial conditions of 70.9 kPa and 295 K.



Figure 4: Measured cell width for hydrogen-nitrous oxide-methane-ammonia mixtures vs. equivalence ratio. Initial conditions of 70.9 kPa and 295 K.

Comparison to Hydrogen-Air

It is of interest to compare the present results with previous data obtained on hydrogenair detonations (Berman 1986). In order to do this, we have normalized the cell width for each set of data with the minimum value of cell width λ_{min} obtained for that mixture. Figure 5 shows the ratio λ/λ_{min} for hydrogen-air and hydrogen-nitrous oxide-nitrogen-air mixtures as a function of equivalence ratio.

The minimum cell width occurs close to but not exactly at stoichoimetric, $\Phi = 1$, compositions. For hydrogen-air mixtures at 295 K and 100 kPa, $\lambda_{min} = 15.4$ mm at $\Phi = 1$ (Berman 1986). For hydrogen-nitrous oxide mixtures at 295 K and 70.9 kPa, $\lambda_{min} = 2.1$ mm at $\Phi = 1$. This estimation is based on several data points (Akbar and Shepherd 1993; Akbar et al. 1997) which have been adjusted using ZND calculations for the dependence of the reaction zone thickness Δ on the initial pressure (see Figs. 16 and 17 and Tables 6 and 7). For hydrogen-nitrous oxide-30% nitrogen mixtures at 295 K and 70.9 kPa, $\lambda_{min} = 6$ mm at $\Phi = 1$. For 50%(hydrogen-nitrous oxide-30% nitrogen)-50% air mixtures at 295 K and 70.9 kPa, $\lambda_{min} = 16$ mm at $\Phi = 1$.

For the case of rich mixtures, the ratio λ/λ_{min} appears (Fig. 5) to have the same functional dependence on equivalence ratio for all four compositions. However, for lean

mixtures the ratio is much smaller for mixtures with nitrous oxide than for hydrogen-air mixtures. This is a consequence of the exothermic nature of nitrous oxide decomposition that was mentioned earlier.



Figure 5: Normalized cell width vs. equivalence ratio for four mixtures. Hydrogen-air curve is from correlation by Berman (1986).

Comparision to ZND model

The ZND reaction zone model and a detailed chemical reaction scheme was used to estimate the reaction zone lengths Δ . The details of these computations and the validation of the reaction mechanism is given in Akbar et al. (1997). The calculations were all performed with the Miller and Bowman mechanism modified as described in Akbar et al. (1997). Following previous work, we have estimated the cell width as being linearly proportional to the reaction zone length, $\lambda = A\Delta$. For most mixtures, a value of A between 60 and 80 is found to be appropriate. This is consistent with our earlier studies and work on hydrogen-air. However, it is known that the value of A can vary significantly with composition and even the notion of a linear correlation breaks down for many mixtures (Akbar et al. 1997). In the subsequent sections, we have shown computed reaction zone lengths and associated predictions for cell width for a wider range of compositions than we have supporting data. Previous experience indicates that large extrapolations should be used with great caution, if at all. The best practice is to interpolate or just modestly extrapolate with ZND results. The extrapolations are only shown to indicate the trend of the ZND predictions.

Hydrogen-Nitrous Oxide

Figures 6 and 7 show the measured cell widths for hydrogen-nitrous oxide mixtures in comparison to calculated reaction zone thicknesses at initial conditions of 295 K and 70.9 kPa. For this mixture, a constant of proportionality A = 60 gives a reasonable fit, see Fig. 6 and 7. No detonation could be observed in the GALCIT detonation tube for lean mixtures below 4.8% hydrogen and for rich mixtures above 90% hydrogen. It is not clear if this is due to insufficient initiator energy or the intrinsic difficulty of nitrous oxide decomposition at low temperatures. The detonation velocity and cell width measurements all indicate that as long as a detonation occurs, the nitrous oxide completely decomposes. This is in contrast to the situation in flames, for which a threshold value of fuel exists. For fuel concentrations lower than the threshold (10% in the case of hydrogen), nitrous oxide decomposition does not occur in flame propagation.

The leanest mixture that could be detonated (5.7% hydrogen) had a cell width of 59 mm. This is substantially smaller than the tube diameter indicating the failure to get initiation for leaner mixtures may be a consequence of insufficient ignition energy rather than excessive cell width. However, the ZND computations discussed below indicate that the reaction zone length increases rapidly as the hydrogen concentration decreases below 5.7%, effectively making detonation initiation impractical with typical laboratory scale experiments.

Mixture composition, equivalence ratio Φ , cell width λ , calculated (Reynolds 1986) Chapman-Jouguet wave speed V_{CJ} , measured wave speed V_{exp} , and reaction zone thickness Δ , are given in Table 1.



Figure 6: Measured cell width and calculated reaction zone thickness for hydrogen-nitrous oxide mixtures at initial conditions of 70.9 kPa and 295 K.



Figure 7: Measured cell width and calculated reaction zone thickness for hydrogen-nitrous oxide mixtures at initial conditions of 70.9 kPa and 295 K.

H_2	N_2O	Φ	λ	$\lambda \pm$	V_{CJ}	V_{exp}	Δ
%	%		mm	mm	m/s	m/s	mm
0	100	0.000	-	-	1698	610.9	16.49
4.8	95.2	0.050	-	-	1759	644.8	1.389
5.7	94.3	0.060	128	48	1771	1647	1.077
6.5	93.5	0.070	91	28	1782	1650	0.873
7.4	92.6	0.080	71	17	1793	1665	0.701
9.1	90.9	0.100	48	10	1815	1711	0.399
11	89	0.124	25	7.0	1840	1777	0.241
13	87	0.149	13	2.0	1865	1839	0.179
20	80	0.250	6.5	2.5	1953	1943	0.106
40	60	0.667	2.8	0.8	2223	2216	0.0761
50	50	1.000	2.1^{*}	0.8^{*}	2379	-	0.0847
60	40	1.500	2.8	0.8	2554	2545	0.119
70	30	2.333	8.0	2.0	2749	2737	0.470
80	20	4.000	34	9.0	2961	2937	1.215
85	15	5.667	110	20	3056	3023	5.880
90	10	9.000	-	-	3087	968.1	137.0

Table 1: Data summary of hydrogen-nitrous oxide detonation experiments and related calculations at initial conditions of 295 K and 70.9 kPa.

* estimated - see Fig. 16 and text

Hydrogen-Nitrous Oxide-Nitrogen

Figures 8 and 9 show the measured cell widths for hydrogen-nitrous oxide-nitrogen mixtures in comparison to the calculated reaction zone thicknesses at initial conditions of 295 K and 70.9 kPa. A fixed nitrogen dilution of 30% was used for all equivalence ratios. For this mixture, a constant of proportionality A = 60 gives a reasonable fit, similar to hydrogen-nitrous oxide mixtures. Mixture composition, equivalence ratio Φ , cell width λ , calculated (Reynolds 1986) Chapman-Jouguet detonation wave speed V_{CJ} , measured wave speed V_{exp} , and reaction zone thickness Δ , are given in Table 2.

Table 2: Data summary of hydrogen-nitrous oxide-nitrogen detonation experiments and related calculations at initial conditions of 295 K and 70.9 kPa.

H_2	N_2O	N_2	Φ	λ	$\lambda \pm$	V_{CJ}	V_{exp}	Δ
%	%	%		mm	mm	m/s	m/s	mm
7	63	30	0.111	210	50	1693	1576	0.799
14	56	30	0.250	35	13.0	1812	1745	0.383
28	42	30	0.667	7.5	2.5	2037	2033	0.213
42	28	30	1.500	9.0	3.0	2218	2210	0.323
49	21	30	2.333	21.0	6.0	2238	2224	0.892
56	14	30	4.000	165	35	2160	2141	9.041



Figure 8: Measured cell width and calculated reaction zone thickness for hydrogen-nitrous oxide-nitrogen mixtures at initial conditions of 70.9 kPa and 295 K.



Figure 9: Measured cell width and calculated reaction zone thickness for hydrogen-nitrous oxide-nitrogen mixtures at initial conditions of 70.9 kPa and 295 K.

Hydrogen-Nitrous Oxide-Nitrogen-Air

Figures 10 and 11 show the measured cell widths for hydrogen-nitrous oxide-nitrogen-air mixtures in comparison to calculated reaction zone thicknesses. The hydrogen-nitrous oxide mixture is first diluted with 30% nitrogen, this mixture is then further diluted with 50% air. The final oxygen and nitrogen concentrations are 10.5% and 54.5%, respectively. For this mixture, the constant of proportionality A = 80 instead of 60 as in the prevous cases. Mixture composition, equivalence ratio Φ , cell width λ , calculated (Reynolds 1986) Chapman-Jouguet detonation wave speed V_{CJ} , measured wave speed V_{exp} , and reaction zone thickness Δ , are given in Table 3.

Table 3: Data summary of hydrogen-nitrous oxide-nitrogen-air detonation experiments and related calculations at initial conditions of 295 K and 70.9 kPa.

H_2	N_2O	N_2	O_2	Φ	λ	$\lambda \pm$	V_{CJ}	V_{exp}	Δ
%	%	%	%		mm	mm	m/s	m/s	mm
10.5	24.5	54.5	10.5	0.231	250	70	1563	1528	1.094
14	21	54.5	10.5	0.333	60	16	1650	1631	0.533
21	14	54.5	10.5	0.600	17.0	4.0	1814	1805	0.262
24.5	10.5	54.5	10.5	0.778	14.5	4.5	1890	1886	0.221
28	7	54.5	10.5	1.000	16.0	4.0	1954	1945	0.212
31.5	3.5	54.5	10.5	1.286	21.0	5.0	1975	1966	0.271
35	0	54.5	10.5	1.667	28.0	8.0	1939	1941	0.556



Figure 10: Measured cell width and calculated reaction zone thickness for hydrogennitrous oxide-nitrogen-air mixtures at initial conditions of 70.9 kPa and 295 K.



Figure 11: Measured cell width and calculated reaction zone thickness for hydrogennitrous oxide-nitrogen-air mixtures at initial conditions of 70.9 kPa and 295 K.

Hydrogen-Nitrous Oxide-Methane

Figures 12 and 13 show the measured cell widths for hydrogen-nitrous oxide-3% methane mixtures in comparison to calculated reaction zone thicknesses at initial conditions of 295 K and 70.9 kPa. The constant A = 60 is consistent with the very limited amount of data obtained for this mixture. No detonation could be observed in the GALCIT detonation tube for lean mixtures below 5% initial hydrogen and for rich mixtures above 75% hydrogen. However, only a limited number of experiments were carried out with this mixture. Further work would be needed to better quantify this inhibition effect. Mixture composition, equivalence ratio Φ , cell width λ , calculated (Reynolds 1986) Chapman-Jouguet detonation wave speed V_{CJ} , measured wave speed V_{exp} , and reaction zone thickness Δ , are given in Table 4.

Table 4: Data summary of hydrogen-nitrous oxide-methane detonation experiments and related calculations at initial conditions of 295 K and 70.9 kPa.

H_2	N_2O	CH_4	Φ	λ	$\lambda \pm$	V_{CJ}	V_{exp}	Δ
%	%	%		mm	mm	m/s	m/s	mm
5	92	3	0.185	-	-	1866	526.3	0.340
10	87	3	0.253	16.0	4.0	1923	1923	0.203
42.5	54.5	3	1.000	6.0	2.0	2337	2333	0.100
75	22	3	3.955	-	-	2850	810.3	1.671



Figure 12: Measured cell width and calculated reaction zone thickness for hydrogennitrous oxide-methane mixtures at initial conditions of 70.9 kPa and 295 K.



Figure 13: Measured cell width and calculated reaction zone thickness for hydrogennitrous oxide-methane mixtures at initial conditions of 70.9 kPa and 295 K.

Hydrogen-Nitrous Oxide-Ammonia

One experiment was carried out using ammonia dilution. Figures 14 and 15 show the measured cell width for a stoichiometric hydrogen-nitrous oxide-3% ammonia mixture in comparison to calculated reaction zone thicknesses at initial conditions of 295 K and 70.9 kPa. A constant A = 80 was used to predict the cell width. Obviously, the choice of A is highly suspect when there is only a single datum for cell width. A value of 80 actually appears to be too low for this case. Previous experience (Akbar, Kaneshige, Schultz, and Shepherd 1997) with mixtures containing ammonia suggest that these mixtures are atypical and extreme caution should be used in interpreting the cell width-reaction zone length correlations. Although the presence of ammonia clearly has an inhibiting effect on stoichiometric mixtures, the ZND model predictions for off-stoichiometric cases cannot be viewed as reliable until more experimental data is obtained. In particular, predictions for near-limit mixtures may be totally erroneous. The data are given in Table 5.

Table 5: Data summary of hydrogen-nitrous oxide-ammonia detonation experiment and related calculations at initial conditions of 295 K and 70.9 kPa.

H_2	N_2O	NH_3	Φ	λ	$\lambda \pm$	V_{CJ}	V_{exp}	Δ
%	%	%		mm	mm	m/s	m/s	mm
46.25	50.75	3	1.000	12.0	4.0	2365	2357	0.0858



Figure 14: Measured cell width and calculated reaction zone thickness for hydrogennitrous oxide-ammonia mixtures at initial conditions of 70.9 kPa and 295 K.



Figure 15: Measured cell width and calculated reaction zone thickness for hydrogennitrous oxide-ammonia mixtures at initial conditions of 70.9 kPa and 295 K.

Dependence on Initial Conditions

Figure 16 shows measured cell widths for stoichiometric hydrogen-nitrous oxide mixtures at an initial temperature of 295 K and initial pressures from 10.1 to 100 kPa. These data were obtained by Akbar and Shepherd (1993) and Akbar et. al (1997). Note that the cell width was not actually measured at the initial pressure of 70.9 kPa used in the present experiments. In order to obtain this value, we have interpolated the existing data (see Table 6) by making ZND reaction zone thickness calculations (see Fig. 17 and Table 7). In this fashion, we infer a value of $\lambda_{min} = 2.1 \pm 0.8$ mm at $\Phi=1.0$, 70.9 kPa, 295 K. This value is also consistent with measured cell widths at neighboring equivalence ratios and 70.9 kPa initial pressure (see Fig. 2 and Table 1).



Figure 16: Measured cell widths vs. initial pressure for stoichiometric hydrogen-nitrous oxide mixtures (Akbar and Shepherd 1993; Akbar et al. 1997), initial temperature = 295 K.

Figs. 17 and 18 show calculated reaction zone thicknesses for hydrogen-nitrous oxide mixtures at initial pressures from 70.9 to 100 kPa and initial temperatures from 295 to 340 K for various equivalence ratios. For all mixtures, lean to rich, the reaction zone thicknesses slightly decrease with increasing initial pressure. The ratio of reaction zone thickness at 100 kPa to reaction zone thickness at 70.9 kPa decreases slightly from 0.69 for lean mixtures ($\Phi = 0.1$) to 0.66 for rich mixtures ($\Phi = 1.5$). The reaction zone thicknesses are almost independent of the initial temperature, except for very lean mixtures ($\Phi =$ 0.1). For all but the leanest mixture, Δ decreases with increasing initial temperature. The ratio of reaction zone thickness at 340 K to reaction zone thickness at 295 K increases slightly from 1.06 for lean mixtures ($\Phi = 0.5$) to 1.07 for rich mixtures ($\Phi = 1.5$). All data are given in Tables 7 and 8.

Table 6: Summary of measured cell widths vs. initial pressure for stoichiometric hydrogennitrous oxide mixtures (Akbar and Shepherd 1993; Akbar et al. 1997), initial temperature = 295 K.

H_2	N_2O	Φ	р	measured λ	source
%	%		10^5 Pa	mm	
50	50	1.0	0.101	2 - 12	(Akbar and Shepherd 1993)
50	50	1.0	0.202	3 - 10	(Akbar and Shepherd 1993)
50	50	1.0	0.403	2 - 4	(Akbar and Shepherd 1993)
50	50	1.0	0.602	1 - 2	(Akbar and Shepherd 1993)
50	50	1.0	0.709	1.3 - 2.9	estimated
50	50	1.0	0.729	1 - 3	(Akbar et al. 1997)
50	50	1.0	0.8	1 - 1.75	(Akbar and Shepherd 1993)
50	50	1.0	1.0	1 - 1.5	(Akbar and Shepherd 1993)



Figure 17: Calculated reaction zone thickness for hydrogen-nitrous oxide mixtures vs. initial pressure (70.9 - 100 kPa), initial temperature = 295 K.



Figure 18: Calculated reaction zone thickness for hydrogen-nitrous oxide mixtures vs. initial temperature (295 - 340 K), initial pressure = 100 kPa.

H_2	N_2O	Φ	р	V_{CJ}	Δ	$\Delta/\Delta_{70.9\ kPa}$
%	%		10^5 Pa	m/s	mm	
9.091	90.909	0.1	0.709	1815	0.482	1.0
9.091	90.909	0.1	0.8	1817	0.422	0.88
9.091	90.909	0.1	0.9	1818	0.371	0.77
9.091	90.909	0.1	1.0	1819	0.334	0.69
33.33	66.67	0.5	0.709	2128	0.0783	1.0
33.33	66.67	0.5	0.8	2131	0.0684	0.87
33.33	66.67	0.5	0.9	2135	0.0595	0.76
33.33	66.67	0.5	1.0	2138	0.0527	0.67
50	50	1.0	0.101	2295	0.930	11.0
50	50	1.0	0.202	2325	0.393	4.64
50	50	1.0	0.403	2355	0.168	1.98
50	50	1.0	0.602	2373	0.102	1.20
50	50	1.0	0.709	2379	0.0847	1.0
50	50	1.0	0.729	2379	0.0847	1.0
50	50	1.0	0.8	2384	0.0732	0.86
50	50	1.0	0.9	2390	0.0633	0.75
50	50	1.0	1.0	2394	0.0559	0.66
60	40	1.5	0.709	2554	0.119	1.0
60	40	1.5	0.8	2560	0.103	0.87
60	40	1.5	0.9	2565	0.0896	0.75
60	40	1.5	1.0	2570	0.0787	0.66

Table 7: Data summary of hydrogen-nitrous oxide reaction zone thickness vs. initial pressure calculations, initial temperature = 295 K.

H_2	N_2O	Φ	Т	V_{CJ}	Δ	$\Delta/\Delta_{295 \ K}$
%	%		Κ	m/s	mm	
9.091	90.909	0.1	295	1819	0.334	1.0
9.091	90.909	0.1	310	1819	0.191^{*}	0.57
9.091	90.909	0.1	325	1819	0.139	0.42
9.091	90.909	0.1	340	1820	0.135	0.40
33.33	66.67	0.5	295	2138	0.0527	1.0
33.33	66.67	0.5	310	2136	0.0539	1.02
33.33	66.67	0.5	325	2135	0.0548	1.04
33.33	66.67	0.5	340	2133	0.0560	1.06
50	50	1.0	295	2394	0.0559	1.0
50	50	1.0	310	2391	0.0576	1.03
50	50	1.0	325	2388	0.0591	1.06
50	50	1.0	340	2386	0.0603	1.08
60	40	1.5	295	2570	0.0787	1.0
60	40	1.5	310	2566	0.0813	1.03
60	40	1.5	325	2563	0.0831	1.06
60	40	1.5	340	2561	0.0844	1.07

Table 8: Data summary of hydrogen-nitrous oxide reaction zone thickness vs. initial temperature calculations, initial pressure $= 1 \times 10^5$ Pa.

* estimated - computation did not converge.

Conclusions

Detonation cell width measurements for hydrogen-nitrous oxide-diluent mixtures have been carried out. Although the familar U-shaped curves are obtained for cell width vs. equivalence ratio, the normalized cell width λ/λ_{min} is substantially lower for lean hydrogen-nitrous oxide mixtures than for hydrogen-air mixtures at the same equivalence ratio. For rich mixtures, the normalized cell widths are comparable at the same equivalence ratio. Dilution with nitrogen and air both increase the cell width when the comparison is carried out at a fixed equivalence ratio. The addition of modest (3%) amounts of ammonia or methane act as inhibitors for stoichiometric hydrogen-nitrous oxide mixtures. A summary of the cell width data for stoichiometric mixtures is given in Table 9. The cell widths have been corrected to atmospheric pressure using the factor of 0.66 obtained from the ZND pressure effect analysis. Table 9: Summary of cell width data for stoichioimetric mixtures at 295 K, corrected to a standard pressure of 100 kPa.

Mixture	λ
	(mm)
H_2-N_2O	1.4
$H_2-N_2O + 30\% N_2$	4
$(H_2-N_2O+30\% N_2) + 50\%$ air	11
$H_2-N_2O + 3\% CH_4$	4
$H_2-N_2O + 3\% NH_3$	8

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