i.d. reactor such as was used in this study, the high surface to volume ratio of the reactor provides a very effective heat sink. Such a heat sink causes high abstraction of energy due to heat losses, and thus significantly affects the predetonation period. It is conceivable that such action would tend to magnify the effect of density on the formation of detonation and that the effect of density might be less significant in an experimental system where wall effects are minimized.

CONCLUSIONS

The experimental system and procedure used in this investigation provided an excellent simple method for determining the effect of internal properties on the formation of detonation in gaseous mixtures.

Increasing the initial mixture density significantly affected the formation of detonation in mixtures of knallgas saturated with water vapor. The induction composition or system detonation limit decreased markedly with increasing initial mixture density. It is plausible that the effect of density on the formation of detonation as reported was magnified, because of the high surface to volume ratio of the experimental reactor.

The results presented herein are characteristic of only the specific experimental system utilized. It may, however, be generalized that mixture density is an important parameter which must be considered in establishing detonation limits for gaseous mixtures. Neglecting the effect of density on such limits or on the formation process could result in serious errors of estimate, and such procedures should be regarded as an unsafe practice in the handling of combustible gases at elevated pressures.

NOMENCLATURE

P_r = peak reaction pressure, atmospheres
P_1 = initial pressure, atmospheres

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Detonation Properties of Heavy Knallgas, $2\,D_2 + O_2$

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A DETONATION study of heavy knallgas ($2D_2 + O_2$) is warranted from two basic engineering aspects: to provide valuable base data for equipment detonation design, and to determine whether such data can be adequately predicted via Chapman-Jouguet detonation theory (*7, 18*). In particular, measured and predicted detonation properties of heavy knallgas may differ significantly. Although detonation velocities in knallgas ($2H_2 + O_2$) agree well with theory (*3, 11, 12, 24, 27*), the slower reaction kinetics exhibited by deuterium-oxygen mixtures (*4, 8, 16*) suggest increased wave-front energy loss to the detonation tube. Resulting effects on the detonation velocity of heavy knallgas may thus be marked. Deviations may be more prominent, however, if consideration is also given to the detonation pressure. Agreement of theoretical and experimental velocities in itself is not evidence of strict adherence to "ideal" detonation criteria. Rather, detonation velocities may exhibit good agreement with calculated values without denoting exact or near-exact fulfillment of theoretical detonation conditions (*3, 11*). Such may not be true for the detonation pressure. This study thus considers both the stable detonation velocity and reflected, or "impact," detonation pressure in testing the experimental and theoretical detonation agreement of heavy knallgas. Conditions covered are 1 to 15 atm. initial pressure at 25° C. Since adequate comparison is necessary to the analysis, corresponding properties of knallgas have also been determined.

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APPARATUS AND PROCEDURE

A schematic of the detonation laboratory is shown in Figure 1. Control valves, gas lines, and placement of the component experimental equipment is indicated.

The detonation tube, 18 feet long, was constructed of round, straight, 1.00-inch i.d. seamless 304 steel tubing.

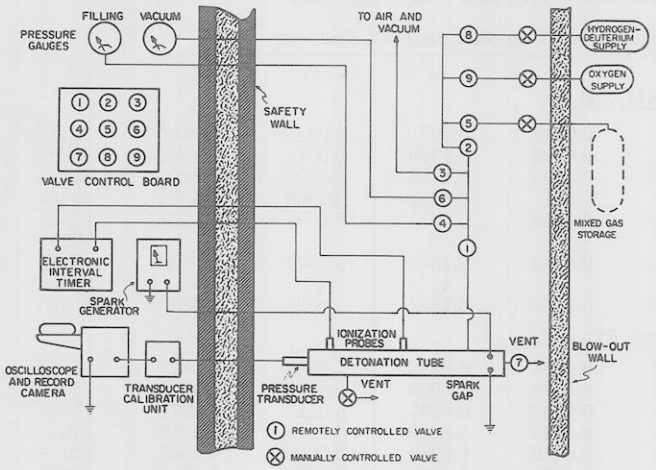

Figure 1. Experimental System

Ignition of the experimental gas was effected via condenser discharge through a spark gap. All runs were conducted at 25 ± 1° C.

Detonation velocities were determined by measurement of the time interval required for an established wave to traverse a distance of 8.984 feet measured over the far end of the detonation tube length. Timing siginals were generated by two ionization-type (*6, 17, 27*) velocity probes mounted flush with the inner tube wall at each end of the distance traverse. Time intervals were recorded by a Berkeley Model 5120 time interval meter accurate to ± 1 μsec. Care was taken to ensure stability for each of the experimental waves. This was indicated for the knallgas experiments by comparison of the stabilization distance reported by Laffitte and Dumanois (*22*) for the "weakest" (1 atm.) knallgas wave, 70 cm., to the approximate 9-foot stabilization length in the experimental tube. Stability for both mixtures was checked experimentally, however, via an auxiliary ionization probe which divided the normal velocity distance traverse into approximate 3- and 6-foot intervals. Velocities measured for 1-atm. waves over each of these intervals were the same (within experimental error), thus indicating stability.

The experimental pressure measurements consisted of reflected, or impact, detonation pressure signals generated by a Kistler PZ-6 quartz pressure transducer flush-fit with the tube end. Signal amplification and calibration were provided by a Kistler PT-6 transistorized amplifier-calibrator standardized against a Crosby Style CD-210 dead-weight tester. Pressure and calibration traces were displayed and recorded via a Tektronix Type 531 CRO equipped with a Type 53/54D differential high-gain direct current pre-amplifier and DuMont Type 302 Polaroid land camera.

The knallgas mixtures were prepared by premixing and storing (in and underground, remote location) the proper amounts of hydrogen or deuterium and oxygen. Component gas specifications were: hydrogen, electrolytic (Matheson), 99.9+ % purity; deuterium (Oak Ridge National Laboratory), 99.9+ % purity; oxygen, U.S.P. (Matheson), 99.6% purity. Mixing times of 3 to 5 days, the longer for the heavy knallgas, were allowed before use of the freshly prepared gas. Stoichiometry of these mixtures, checked via Orsat methods, deviated from the 2 to 1 mole composition by no more than 0.5 mole %.

Run procedure involved initial evacuation of the detonation tube prior to desired pressurization with the gas under study. As a safety precaution, voltage was not applied to the ionization probes until such procedure had been completed and the system isolated via shut-off valves. A spark was then passed through the tube. The velocity and reflected pressure of the resulting detonation wave were recorded. Preparation for a succeeding run involved cooling and drying the detonation tube by sweeping it with compressed air.

In general, two and in several cases, three experimental runs were made at each condition studied, with the exception of the higher pressures. The ignition spark failed to fire in several instances at these conditions, thus necessitating venting of the gas for cleaning and, if needed, readjustment of the spark probes. Since the capacity of the mixture tank was relatively small and limited by safety considerations to 20 atm. maximum pressure, the obtainment high-pressure runs was difficult without time-consuming mixture re-preparation. These were not reproduced, once obtained.

THEORETICAL CALCULATIONS

The theoretical detonation calculations were based on Chapman-Jouguet theory (*7, 18*) with the assumption of complete reaction to chemical equilibrium in all cases. Incident, or normal, stable wave velocities were based on frozen sound speed. Reflected wave pressures were based on normal shock reflection from a plane, rigid boundary, with chemical equilibrium achieved at the reflected wave conditions. Species assumed present at equilibrium were, for the hydrogen system, H_2O, H_2, O_2, OH, H, and O; for the deuterium system, D_2O, D_2, O_2, OD, D, and O. Thermodynamic data for the former species were taken from the work of Luker, McGill and Alder (*25, 26*); for D_2O, D_2, OD, and D from the recent data of Ryan (*30*). Solution of the detonation equations was obtained numerically with the aid of an IBM 650 digital computer. A convergence of approximately 1 part in 1,000 was achieved. The base temperature considered in all calculations was 25° C.

RESULTS AND DISCUSSION

Velocity Measurement. The experimental and theoretical velocity data are presented in Tables I and II. In Figure 2, the effect of initial gas pressure on the stable detonation velocity is shown. Average values are plotted because of excellent data consistency: Mean velocity deviation, based on all experimental points, is ± 0.19%. Maximum deviation is less than 0.7%. The lower detonation velocities in heavy knallgas primarily reflect lower sound speed in the denser deuterium mixture.

Consideration of the plotted velocity values reveals that detonation velocities of both heavy knallgas and knallgas

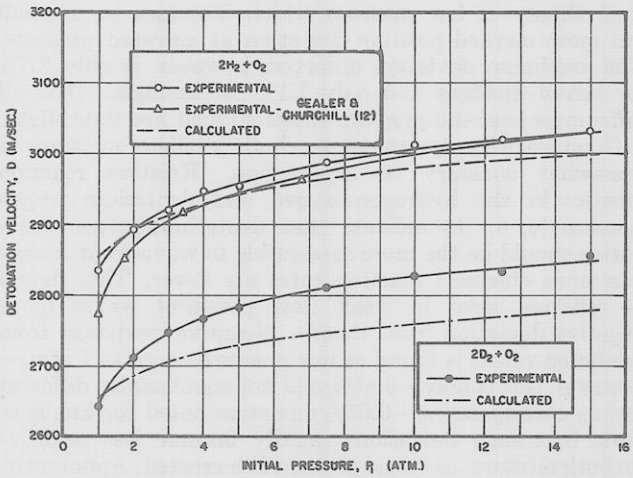

Figure 2. Detonation velocities of knallgas and heavy knallgas

Table I. Calculated Detonation Properties of Knallgas and Heavy Knallgas

P_1	T_2	P_2	D	Equilibrium Wave Composition, Mole Fraction						P_3
				y_a	y_b	y_c	y_d	y_e	y_f	
				Knallgas, $2H_2 + O_2$						
1	3675	18.12	2852	0.5298	0.1671	0.0512	0.1281	0.0821	0.0417	43.00
2	3810	37.24	2890	0.5434	0.1646	0.0492	0.1300	0.0744	0.0384	88.65
3	3892	56.69	2912	0.5520	0.1628	0.0480	0.1308	0.0700	0.0364	135.2
4	3951	76.36	2928	0.5585	0.1614	0.0471	0.1312	0.0668	0.0350	182.3
5	3997	96.17	2941	0.5637	0.1602	0.0464	0.1314	0.0644	0.0339	229.9
7.5	...	146[a]	2963[a]	...	...	...	...	...	...	352[a]
10	4143	196.4	2978	0.5808	0.1560	0.0442	0.1315	0.0570	0.0305	470.8
12.5	...	246[a]	2990	...	...	...	...	...	...	600[a]
15	4230	297.3	2999	0.5916	0.1532	0.0429	0.1312	0.0528	0.0283	713.7
				Heavy Knallgas, $2D_2 + O_2$						
1	3552	17.56	2653	0.4678	0.2819	0.0994	0.0923	0.0144	0.0442	41.76
2	3670	35.98	2686	0.4840	0.2731	0.0962	0.0939	0.0127	0.0401	85.83
3	3741	54.64	2705	0.4941	0.2676	0.0942	0.0947	0.0117	0.0377	130.5
4	3791	73.59	2717	0.5021	0.2633	0.0927	0.0949	0.0110	0.0360	176.2
5	3832	92.54	2729	0.5074	0.2603	0.0916	0.0953	0.0106	0.0348	221.5
7.5	3906	140.4	2747	0.5185	0.2542	0.0894	0.0957	0.0097	0.0325	336.7
10	3959	188.6	2761	0.5266	0.2498	0.0878	0.0958	0.0091	0.0309	452.6
12.5	4000	237.0	2771	0.5331	0.2463	0.0865	0.0957	0.0087	0.0297	569.2
15	4034	285.3	2779	0.5385	0.2433	0.0855	0.0957	0.0083	0.0287	685.8

[a] Interpolated value.

Table II. Measured Detonation Properties of Knallgas and Heavy Knallgas

P_1	D, Meters/Sec.				P_3, Atm.			
	Run 1	Run 2	Run 3	Av.	Run 1	Run 2	Run 3	Av.
				Knallgas, $2H_2 + O_2$				
1.00	2832	2835	...	2834	44.7	44.4	...	44.6
2.00	2895	2889	...	2892	106	105	...	106
3.00	2919	2919	...	2919	154	150	...	152
4.00	2948	2941	...	2945	203	193	...	198
5.00	2951	2954	...	2953	230	240	...	235
7.50	2986	...	...	2986	358	...	...	358
10.0	3013	3009	...	3011	405	481	...	443
12.5	3019	...	...	3019	...	...	...	...
15.0	3032	...	...	3032	...	...	...	...
				Heavy Knallgas, $2D_2 + O_2$				
1.00	2641	2633	2648	2641	23.6	22.1	25.0	23.6
2.00	2703	2711	2719	2711	69.4	64.0	83.0	72.1
3.00	2727	2749	2763	2746	130	125	120	125
4.00	2769	2763	...	2766	196	180	...	188
5.00	2789	2774	...	2782	247	233	...	240
7.50	2809	2811	...	2810	440	440	...	440
10.0	2829	2823	...	2826	581	486	...	534
12.5	2832	...	...	2832	...	...	...	...
15.0	2855	...	...	2855	...	...	...	...

are well predicted by theory. Experimental and theoretical agreement for both mixtures is essentially the same. Both data curves exhibit a slight negative deviation of experiment from theory at low pressure which increases to a small but more marked positive deviation at elevated pressure. The maximum deviation observed, however, is only 2.7% for heavy knallgas and only 1.1% for knallgas. Over-all differences over the pressure range studied are thus slight.

Good velocity agreement for both knallgas mixtures is somewhat contrary to expectation. Relative reaction kinetics in the hydrogen-oxygen and deuterium-oxygen systems (*4, 8, 16*) indicate that deuterium-oxygen detonation should be the more susceptible to wave-front energy loss since chemical reaction rates are lower. This should be reflected most in weak (low pressure) waves by a negative deviation from theory. Negative deviation from predicted values is found at low pressure—e.g., at 1 atm.—however, this is only −0.45% and not significantly different from a corresponding −0.63% deviation noted for knallgas. Note that such deviations rapidly become less negative for both mixtures as initial pressure is increased. Apparently, wall effects lose significance quickly in both cases as mixture "strength" is raised. The further trend towards marked positive velocity deviation noted with increasing pressure is assumably related to inadequacy of the predicted velocity values due to gas-law assumptions in theoretical calculation (*2*).

The general nature of the observed velocity deviations can be examined more fully. Valuable comparison is made to knallgas velocity data interpolated from the experimental values of Gealer and Churchill (*12*; cf. Figure 2). These data (Off-scale high pressure points used in determining this interpolated velocity curve are not shown.) were obtained in a detonation tube of twice the surface to volume ratio of the present one. As such, velocities should be lower at corresponding initial pressures than those obtained herein due to more significant tube-wall energy losses. This is clearly the case as is evidenced by the plotted values. In addition, the lessening of wall effects on stronger detonation waves is indicated by the merger of the two experimental knallgas curves at elevated pressure. This merger tends to support the view that positive velocity deviations are attributable to gas-law inadequacy in theoretical calculation. It is thought that ideal gas

assumption become increasingly inadequate with pressure in representing detonation-front burned-gas behavior (*12*). The fact that the present experimental curve and that of Gealer and Churchill join above the theoretical essentially precludes the possibility of "high" experimental values. Thus, assumption of gas-law inadequacy is strengthened. The extent of this inadequacy is apparently greater for heavy knallgas than for knallgas. Why this is so is not clear, since the deuterium system exhibits greater relative dissociation of the primary reaction product than does the hydrogen system (*30*) and hence should obey the ideal gas law more closely. The differences shown however, would have to be more marked to warrant further investigation.

Good prediction of heavy knallgas detonation velocities is thus afforded by Chapman-Jouguet detonation theory. From a fundamental viewpoint, however, the observed agreement, although good, does not necessarily denote near-exact or exact fulfillment of Chapman-Jouguet detonation criteria. It is thought that highly-dissociated chemical systems, such as those in the present study, display a "buffering" action on the detonation velocity (*3, 11*). Such may compensate for significant deviation from theoretically ideal conditions within the detonation reaction zone. This may not be true, however, for other detonation parameters; deviations may be more marked. The fact that the detonation pressure, for example, may be a more sensitive criterion of ideal detonation conditions is indicated by simple investigation of a typical detonation Hugoniot diagram (*23*). Apparently, small deviation from complete (equilibrium) reaction in the detonation front can result in considerably more significant change in pressure than in velocity. Consideration of the detonation pressure measurements may thus be more effective than the detonation velocities in denoting subideal conditions, if such exist, for heavy knallgas.

Pressure Measurement. The experimental and theoretical pressure data are presented in Tables I and II. In Figure 3 the effect of initial gas pressure on the stable reflected detonation pressure is shown with all experimental values plotted, since the deviations observed are relatively significant. Reproducibility in all cases, however, is within ± 15%.

Actual pressure values were determined from the experimental oscillograms by taking a peak reading of the pressure-time record. The measured pressures may thus be subject to some experimental interpretation. High-frequency pressure-measuring instruments in general lack the extreme transient response needed to record accurately detonation pressures. Based on a reaction zone width of 0.1 mm. (*19*), an instrument response of 0.03 μsec. is neccessary to measure adequately the true Chapman-Jouguet pressure of a 1-atm. knallgas detonation. Such response is only approached by the pressure transducer employed herein (rise time approximately 2 μsec.); certainly, it has not been achieved by any of the various pressure-measuring devices previously employed in detonation studies—e.g. rupture disks (*5, 13, 31*), penetration gages (*15, 29*), piezoelectric gages (*1, 2, 14, 26, 32, 33*), strain gages (*15, 26, 28*), and pressure-bar gages (*9, 10, 11*) although a recent pressure-bar study (*11*) does give relatively constant pressure-time records. These are presumably due to lessened rarefaction effects in a large (10 cm.) detonation tube. The present experimental pressures are thus subject to limitation in an absolute sense: therefore, no representation as to the accuracy of such values will be made. However, it is believed that the reported peak pressures will be subject to a minimum of error in analysis, and that these will provide as adequate a comparison of the heavy knallgas and knallgas systems as will the true pressures.

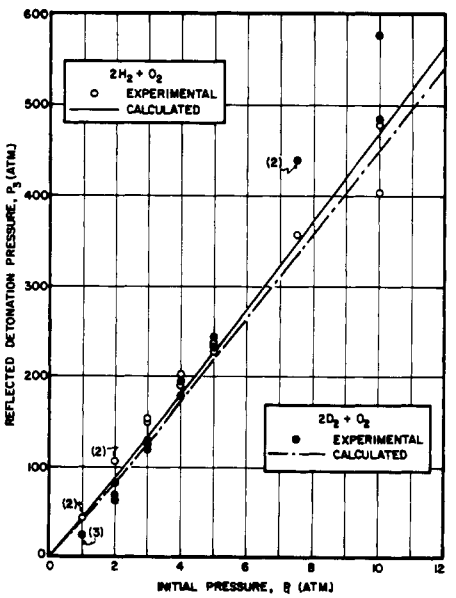

Figure 3. Reflected detonation pressures of knallgas and heavy knallgas

Numbers in parentheses refer to the number of experimental values

Consideration of Figure 3 reveals general adequate agreement between the experimental pressures obtained for heavy knallgas and knallgas and those predicted by theory. The knallgas pressures appear to exhibit relatively consistent agreement, while those for heavy knallgas vary more from the theoretical, particularly at very low and very high initial pressures. It is felt that experimental values obtained at the high pressures are least reliable, since these correspond to measured pressures in excess of the rated transducer maximum. The low-pressure deviations, however, are significant as regards heavy knallgas detonation. These are best noted if percentage deviations are considered; comparison of these deviations is made in Table III.

Table III characterizes the nature of the experimental and theoretical pressure agreement well. Knallgas agreement is adequate over the pressure range studied; deviations in no case exceed 20%. The heavy knallgas agreement, however, exhibits an increasing negative deviation of experiment from theory as initial pressure is lowered, with the 1-atm. value 44% below that predicted by theory. It is believed that this indicates a marked subideal behavior for heavy knallgas detonation waves evidenced by experimentally-low heavy knallgas pressures. The trend toward greater negative deviation at low pressure is as would be expected: At low pressures, the detonation reaction zone is wider (*20, 21*) and the wave slower-moving. Consequently, energy losses to the detonation tube which affect the detonation pressure are more effective. Note

Table III. Comparison of Reflected Detonation Pressures

Initial Pressure, Atm.	Average Pressure Deviation, %: $2H_2 + O_2$, exptl. from theoret.	$2D_2 + O_2$, exptl. from theoret.	$2D_2 + O_2$ from $2H_2 + O_2$, exptl.
1	4.0	−44.0	−47.0
2	20.0	−25.0	−32.0
3	14.0	−8.4	−18.0
4	16.0	8.5	−5.0
5	4.4	10.0	2.1
7.5	1.7	31.0	23.0
10	−14.0	28.0	21.0

that, as discussed, the experimental pressure values may not be too meaningful in an absolute sense. A relative comparison of the heavy knallgas and knallgas pressures, however, can be equally informative (Table III). Here, an increasing negative deviation between the two systems is even more evident with lowered initial pressure. Consider now that the heavy knallgas and knallgas theoretical curves deviate from one another over the pressure range studied by only a maximum 3.8%. Then since the stable knallgas pressures can assumably, at best not exceed their ideal values, corroboration of a subideal trend of heavy knallgas detonation pressure is indicated. A low chemical reaction rate in heavy knallgas detonation (*8*) apparently contributes most to the observed differences; evidently, the differences are reflected more strongly by pressure agreement than by detonation velocity agreement. This is of considerable note, since detonation studies of this type of necessity normally consider velocity agreement only. Based on reproduciblilty of the pressure data, the subideal detonation behavior of heavy knallgas noted herein is significant at initial gas pressure less than 2 to 3 atm.

Studies of the present type are warranted in further cases, particularly under conditions where it is possible to obtain more reliable absolute pressure measurements. Work at lower (subatmospheric) pressures should thus be informative. Reaction zone lengths are longer and, perhaps, ordinary high-frequency pressure devices more suitable. The techniques involved in obtaining stable detonation waves or detonation at very low pressure, however, may be difficult.

Brief note should be made that stable detonation properties, particularly the theoretical reflected pressures reported herein, do, in some cases, constitute adequate criteria for engineering design (*26, 27*). However, situations exist during the detonation formation process wherein unstable detonation pressures are developed (*1, 2, 15, 31, 32, 33*). These unstable pressures may exceed the stable (Chapman-Jouguet) pressures by as much as 3 to 4 times. It cannot be stressed too strongly that these unstable "over-properties" must be taken into account in any adequate design study for detonation containment.

CONCLUSIONS

Stable detonation velocities in heavy knallgas at 25° C. agree closely with those predicted by Chapman-Jouguet detonation theory over an initial pressure range of 1 to 15 atm. The slight deviations noted are assumably due to, at low pressure, energy losses to the detonation tube. At elevated pressure, deviations presumably relate to inadequate ideal gas assumption in theoretical calculation, as suggested by Gealer and Churchill (*12*). Similar behavior is noted for knallgas.

Stable detonation in heavy knallgas at 25° C. is subideal at low pressure (less than 2 to 3 atm.). Conclusion is based primarily on the relative agreement with theory of both heavy knallgas and knallgas reflected detonation pressures. This fact is not evident from the detonation velocity agreement. Pressure agreement is, apparently, a more effective indication of detonation ideality. Deviation is probably due in largest extent to a low chemical reaction rate in heavy knallgas detonation (*8*).

NOMENCLATURE

D = stable detonation velocity, meters/second
M = meters
P = pressure in atmospheres
T = temperature ° K.
y = mole fraction

Subscripts

1 = initial gas condition
2 = stable incident detonation
3 = stable reflected detonation
a = H_2O or D_2O
b = H_2 or D_2
c = O_2
d = OH or OD
e = H or D
f = O

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