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AEC RESEARCH AND DEVELOPMENT REPORT

LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO

THE PERFORMANCE OF BORON EXPLOSIVES
(Title Unclassified)

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THE PERFORMANCE OF BORON EXPLOSIVES
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ABSTRACT

The performance of several homogeneous high-explosive systems which produce B_2O_3 and BF_3 as detonation products has been measured and calculated. The experimentally measured Chapman-Jouguet pressures and detonation velocities are consistent with those calculated using the modified Kistiakowsky-Wilson equation of state. The systems give pressures and detonation velocities similar to those of TNT. The calculations provide a plausible although tentative explanation of the behavior of these systems.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the assistance rendered by Wildon Fickett of GMX-10, Wray Garn of GMX-6, and William Rogers of GMX-2 in various phases of this investigation. We also wish to thank the Redstone Arsenal Division of the Rohm and Haas Company, the Callery Chemical Company, the Olin-Mathieson Chemical Company, and the Lawrence Radiation Laboratory for making available to us some of the materials used in the experimental program.

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I. INTRODUCTION

For at least the past fifteen years, explosives chemists have shown intermittent interest in the high heats of explosion predicted for boron-containing mixtures on the basis of elementary thermochemical considerations. Until recently, efforts to study such systems experimentally were limited, by a lack of other suitable materials, to mixtures of conventional organic explosives (generally RDX/TNT) with finely divided boron metal, the latter usually being of questionable purity. The measured performance of such mixtures was very disappointing in comparison either with expectation or with the performance of similar aluminized explosives[1,2,3]. Several explanations for this have been advanced by various people, of which the most popular seems to have been that the metallic boron was not reacting stoichiometrically because of its high melting point (~ 2000°C) and its nonhomogeneous distribution in the mixture. Whether this explanation is correct or not, it at least has the effect of leaving unsettled the question of the usefulness of boron as a high-explosive component. It further implies that definitive experiments on the performance of boron explosives must be done with homogeneous systems.

Early in 1956 a program was undertaken at Los Alamos whose immediate objectives were as follows: Obtain detonation velocity and Chapman-Jouguet pressure data on several homogeneous boron explosives and determine whether the experimental results are consistent with the best theoretical performance estimates we can make. We feel that these objectives have now been attained, and in this report we summarize the principal results of the investigation.

II. EXPERIMENTAL

A. Test Explosives

It was clear from the start that liquid mixtures of a boron compound and a suitable oxidant offered the most promising approach to the problem of obtaining a boron explosive in which the boron was dispersed on a molecular scale. At the start of the program no boron compounds containing a sufficiently high percentage of oxidizable boron were available in the quantities needed, and over a year was spent in attempting to synthesize such a compound and in studying, on a very small scale, some of the properties of possible combinations. In Table I we have summarized the considerable amount of qualitative solubility and compatibility data which was accumulated in the course of that work.

Fortunately, the ethyldecaboranes finally became available in research quantities as a product of the high-energy fuels program. Mixtures of these with tetranitromethane proved to be suitable for our purposes, and most of our experiments were done with such mixtures. Monoethyldecaborane was used in the first few shots, but was later replaced by the cheaper and more readily available mixtures of the ethyldecaboranes known as HEF-3 and Hi-Cal-3. The compositions of these materials, as supplied by the manufacturers, were as follows:

Monoethyldecaborane (EDB, $B_{10}H_{13}C_2H_5$) - Olin Mathieson. 99.8% pure

HEF-3 - Olin Mathieson

64.6%	monoethyldecaborane
28.4%	diethyldecaborane
3.9%	triethyldecaborane
3.1%	decaborane

TABLE I RESULTS OF COMPATIBILITY STUDIES

Compound Containing Boron	Compound Containing Oxygen or Halogen	Mole Ratio	Observation
Borazole	Tetranitromethane	1 - 1.25	Reacted giving yellow solid($B_3H_{14}N_4CO_{10}$)
Pentaborane	Tetranitromethane	1 - 2	Reacted giving yellow solid
Borazole	100% Nitric acid	2 - 3	Exploded upon melting
Decaborane	Carbon tetrachloride	1 - 11	Very impact-sensitive solutions
Decaborane	2,3-Dichlorohexafluorobutene-2		Less than 1% soluble
"	Kel-F Oil #1		"
"	2,2,3-Trichloroheptafluorobutane		"
"	Perfluoroacetic acid		"
"	Perfluorobutyric acid		"
"	Heptafluorobutanol		"
"	Trifluoroacetic anhydride		"
"	Trifluoroacetaldehyde hydrate		"
"	Methyl heptafluorobutyrate		"
"	1,2-Dichlorohexafluorocyclopentene-1		"
"	Ethyl trifluoroacetate		About 5% soluble but impact-insensitive
"	Trifluoroacetone		"
"	Hexafluoroacetone		"
"	Ethyl perfluorobutyrate		"
"	Difluoroethyl acetate		Soluble but impact-insensitive
"	Fluorolube HO-125 ($C_2F_3Cl)_x$		Insoluble but impact-sensitive slurries
"	Perfluorobutyric acid		"
Decaborane	Nitromethane	1 - 46	Soluble and sensitive to impact
"	Tetranitromethane		Insoluble
"	Tetranitromethane in benzene		Reacts giving white solid($B_{10}H_{13}\cdot C(NO_2)_4$?)
"	Nitromethane - Tetranitromethane	1-19-8	Reacted explosively

TABLE I - Continued

Compound Containing Boron	Compound Containing Oxygen or Halogen	Mole Ratio	Observation
Decaborane	Dinitroethane	1 - 7.5	Impact-sensitive solutions evolving H_2 and forming a solid($B_{10}H_{12}\cdot 2C_2H_4(NO_2)_2$?)
Decaborane	Dinitroethane - Trinitroethane	1-2-1	Impact-sensitive solutions evolving H_2 and forming a white solid
Decaborane	Trinitroethyl trinitrobutyrate - Nitromethane	1-1-6	Reacted giving white solid, evolving gas
Decaborane	Nitrogen dioxide	1 - 11	Insoluble and unreactive at 0°C
Pentaborane	Nitrogen trifluoride	1 - 5	Soluble between -45 and -40°C
Borazole	Nitrogen trifluoride	1 - 3	Soluble between -60 and -40°C
Decaborane	Nitrogen trifluoride	1 - 10	Insoluble between -200 and -70°C
Decaborane	Carbon tetrafluoride	1 - 10	Less than 1% soluble at -200°C
Pentaborane	Carbon tetrafluoride	1 - 3.75	Insoluble at -200°C
Decaborane	Perfluoropropane	1 - 3.75	Less than 1% soluble at -40°C
Pentaborane	Perfluoropropane	1 - 1.875	Insoluble at -40°C
Pentaborane	Tetrafluorohydrazine	1 - 3.75	Soluble between -54 and -5°C
Borazole	Tetrafluorohydrazine	1 - 2.25	Soluble between -75 and -50°C
Ethyl decaborane	Tetranitromethane	1 - 3.75	Soluble at 25°C. Very impact-sensitive
Ethyl decaborane	Kel-F Oil Grade 1	1 - 7.5	" "

TABLE I - Continued

Compound Containing Boron	Compound Containing Oxygen or Halogen	Mole Ratio	Observation
Ethyl decaborane	Perfluoro C_8 cyclic ether	1 - 3	Insoluble
Ethyl decaborane	Tri-perfluorobutyl amine	1 - 2	Insoluble
Decaborane	Tri-perfluorobutyl amine	1 - 2	Insoluble
Decaborane	Tetrafluorodinitroethane	1 - 6	Less than 1% soluble
Ethyl decaborane	Tetrafluorodinitroethane	1 - 7.5	Soluble sensitive solutions
Pentaborane	Tetrafluorodinitroethane	1 - 3.75	Soluble from -40 to 25°C
Ethyl decaborane	Perchlorofluoride	1 - 6	Less than 1% soluble -200 to 25°C
Pentaborane	Perchlorofluoride	1 - 2.2	Soluble between -100 and -68°C
HEF-3	Tetranitromethane	1 - 3.9	Soluble - very impact-sensitive solutions
Hi-Cal-3	Tetranitromethane	1 - 4.3	" "

Hi-Cal-3 - Callery Chemical Company
62.2% boron
25.0% carbon
12.8% hydrogen

All three materials were oily liquids with a density of about 0.81 gm/cc.

Tetranitromethane (TNM) was obtained from the Polynitro Chemical Company and was used as received except for a simple drying step which was necessary to remove the moisture which had condensed in some of the containers. Its density was 1.64 gm/cc and its melting point was 13.5 to 14.0°C. The IR spectrum of the material was run and failed to reveal any obvious impurities.

One EDB shot was fired using tetrafluorodinitroethane (TFDNE, $C_2F_4(NO_2)_2$) as oxidizer. This material was supplied by John Kury of the Lawrence Radiation Laboratory at Livermore. It contained about 2.7% carbon tetrachloride as an impurity. Its density was 1.60 gm/cc.

An unsuccessful shot was fired using an EDB/Kel-F oil mixture. The Kel-F oil was obtained from the M. W. Kellogg Company and is a chlorotrifluoroethylene polymer $(C_2F_3Cl)_x$. The sample used was identified as Grade No. 1 and had a density of 1.87 gm/cc.

The chemical compatibility of the components of the mixtures which were eventually fired was investigated in the laboratory. Small quantities of the various mixtures were prepared and allowed to stand at room temperature. The samples were examined periodically, and no visible indication of a chemical reaction occurring between the components could be noted.

In most cases the density of the mixture was measured experimentally to within 0.02 gm/cc. The density of the EDB/TFDNE mixture was computed from the densities of the two components assuming no volume change on mixing. This assumption was found to hold for the other mixtures to within the accuracy of the measurements.

The stoichiometries of the simple reactions assumed for some of the mixtures and the respective heats of reaction, are as follows:

$$B_{10}H_{13}C_2H_5 + 3.75\ C(NO_2)_4 \rightarrow 5\ B_2O_3 + 9\ H_2O + 5.51\ CO + 0.24\ CO_2 + 7.5\ N_2$$

$$-\Delta H = 2090\ \text{cal/gm} = 2930\ \text{cal/cc}$$

$$B_{10}H_{13}C_2H_5 + 4.45\ C(NO_2)_4 \rightarrow 5\ B_2O_3 + 9\ H_2O + 5.16\ CO_2 + 1.29\ CO + 8.9\ N_2$$

$$-\Delta H = 2180\ \text{cal/gm} = 3120\ \text{cal/cc}$$

$$B_{10}H_{13}C_2H_5 + 12\ (CF_2CFCl) \rightarrow 7.5\ BF_3 + 2.5\ BCl_3 + 4.5\ HCl + 13.5\ HF + 26\ C$$

$$-\Delta H = 900\ \text{cal/gm} = 1490\ \text{cal/cc}$$

$$B_{10}H_{13}C_2H_5 + 7.5\ C_2F_4(NO_2)_2 \rightarrow 10\ BF_3 + 9\ H_2O + 13\ CO + 4\ CO_2 + 7.5\ N_2$$

$$-\Delta H = 1500\ \text{cal/gm} = 2200\ \text{cal/cc}$$

For comparison we have for RDX

$$C_3H_6N_6O_6 \rightarrow 3\ CO + 3\ H_2O + 3\ N_2$$

$$-\Delta H = 1230\ \text{cal/gm} = 2200\ \text{cal/cc}$$

Thus simply from the point of view of heat of reaction, some of these mixtures should be significantly better than RDX.

B. Performance Measurements

The performance data required were the C-J pressure and the detonation velocity. Since the ethyldecaboranes were available only in relatively small quantities, it was necessary to design the shot setup in such a way that both measurements could be made on a single shot with the expenditure of a minimum amount of material. In addition, the mixtures are extremely hazardous (sensitive and toxic) and had to be prepared and loaded into the shot assembly by remote control. The accuracy of the results suffered considerably as a result of the limitations imposed by these considerations, but is still good enough for our purposes.

The shot assembly used is shown in Figure 1. It was designed by Wray Garn of GMX-6 and provided for the simultaneous measurement of C-J pressure and detonation velocity. About 150 ml of explosive was needed for the 2" charge length used in most of the shots. The charge was initiated by means of a detonator, plane wave lens, and Composition B booster. The booster was isolated from the liquid explosive by means of the glass booster cup.

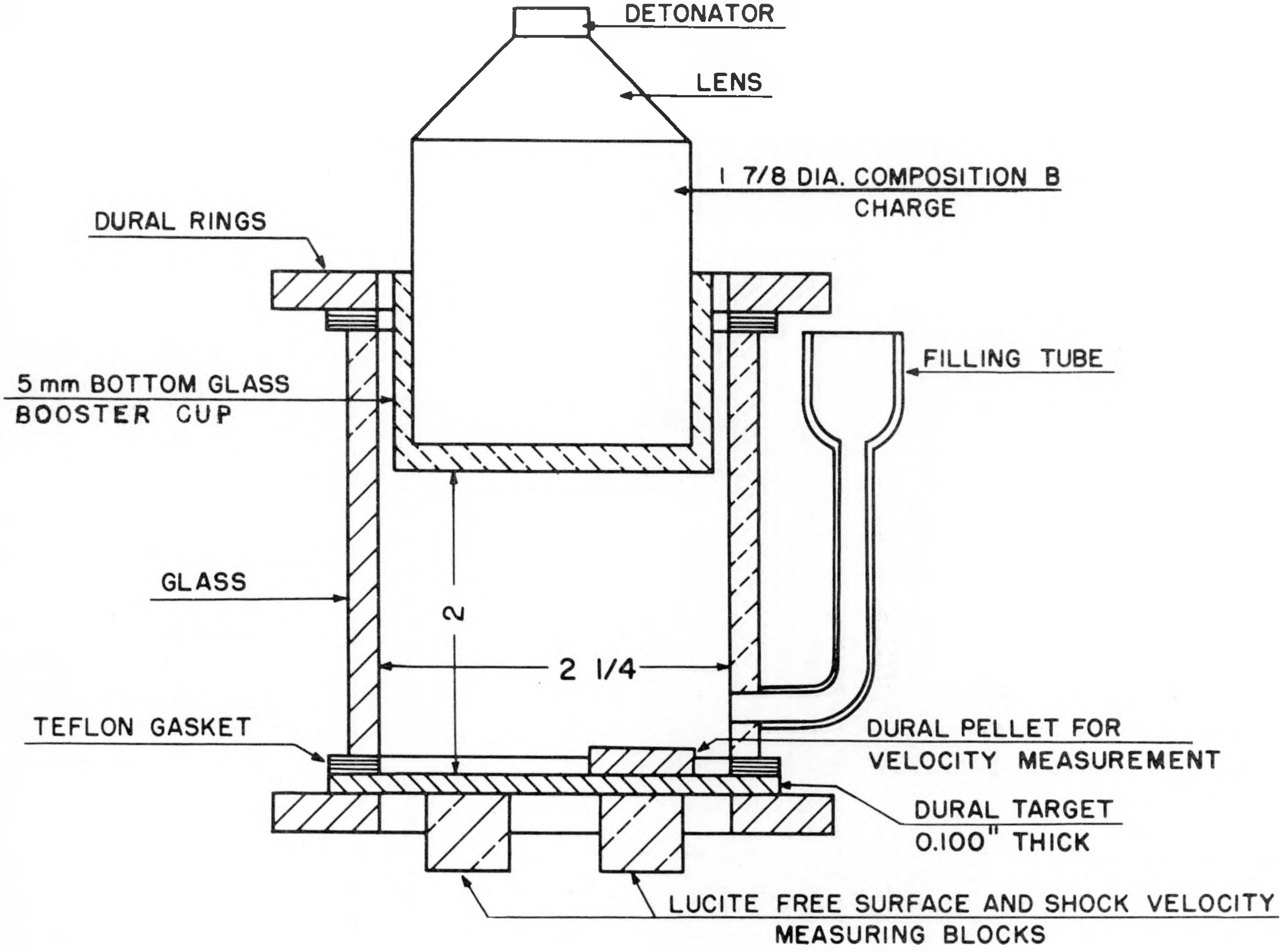

Figure 1. **Detonation Pressure and Velocity Measurement Apparatus**

The Lucite free surface velocity block on the left was used for the measurement of the C-J pressure. The principle of the method has been described in detail by Deal[4] and need not be discussed further here. A Dural pellet was placed on the plate over the other Lucite block, and the detonation velocity was determined relative to the shock velocity in Dural by the difference in transit times through the two thicknesses of metal. With this particular assembly, useful traces are obtained only if the C-J pressure exceeds about 100 kbar.

Under more favorable conditions the C-J pressure and detonation velocity can be measured with an accuracy of about one percent by this method, but in our experiments we were limited to one measurement of each type per shot by the small size of the charge, and also the observations may have been affected in some cases by wave instabilities (overdrive, curvature, etc). The indicated accuracy of our measurements is discussed later on in this report.

A few shots were fired to obtain points on the isentropes of the detonation products of two of the mixtures, using a technique which again has been described in detail by Deal[5]. The assembly (also designed by Garn) used for these shots is shown in Figure 2. It is essentially an upside-down version of Figure 1, with the Dural plate replaced by an inert material of lower shock impedance.

The remote mixing and loading apparatus, designed by W. H. Rogers of GMX-2, is shown in Figure 3. Bulbs A and B were loaded separately with the boron compound and oxidizer, respectively, and taken to the firing mound. The mixing apparatus and shot assembly were set up in a large

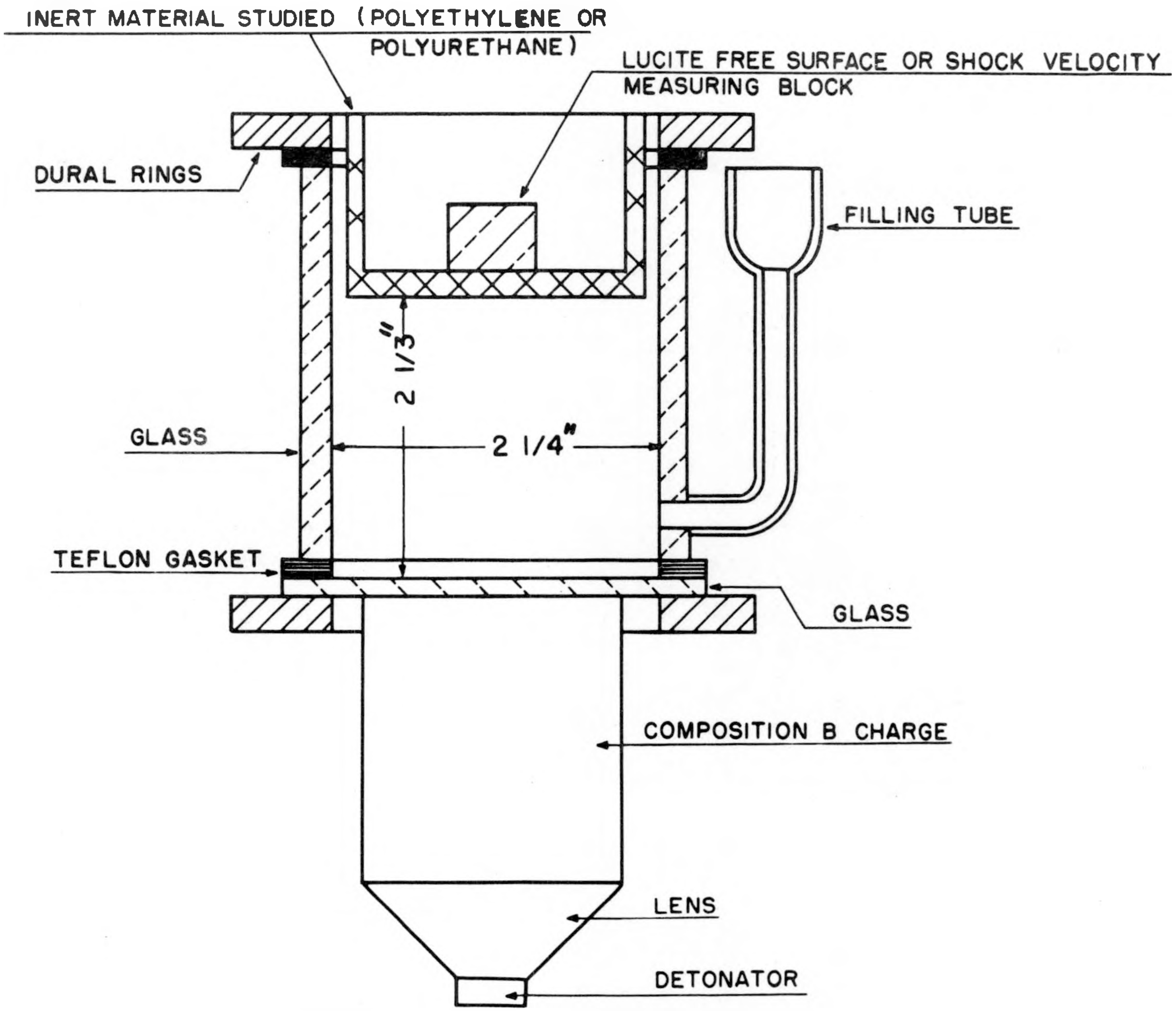

Figure 2. Isentrope Measuring Apparatus

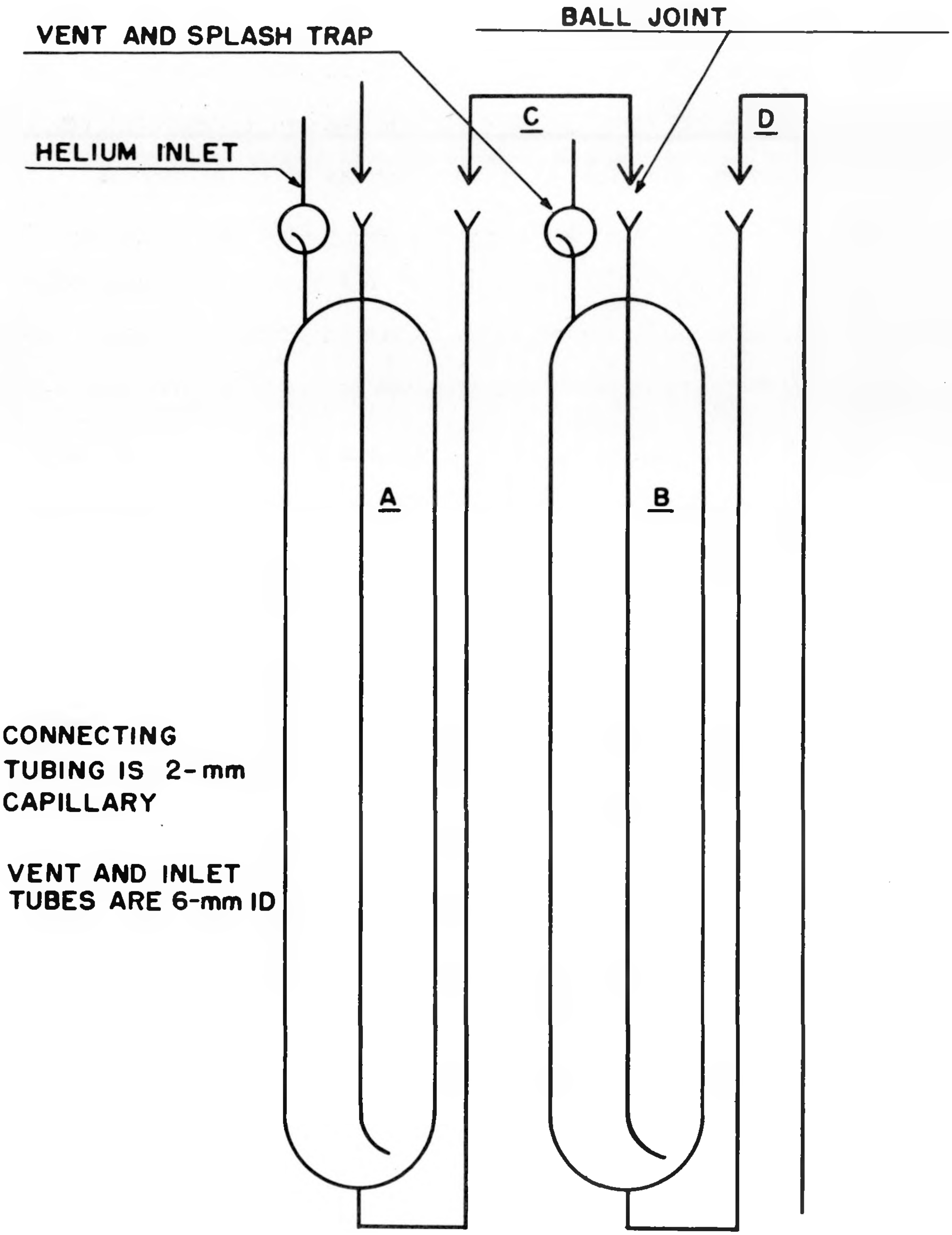

Figure 3. Remote Mixing Apparatus

plywood box, with the exit tube D of the mixing apparatus inserted in the filling tube of the shot assembly. The plywood box had a removable top, a transparent front wall through which the progress of the operation could be observed from the personnel shelter, and a hair dryer and thermometer for adjustment of the temperature in the box. A rubber tube from a helium tank in the control building was attached to the inlet tube on bulb A, and a remotely operated solenoid valve was attached to the vent tube on bulb B.

After all connections to the mixing apparatus and shot assembly were made, all personnel retired to the control building. The temperature in the box was brought to 25°C by means of the hair dryer, and the contents of bulb A were transferred to bulb B by admitting helium to the system with the solenoid open. Helium was bubbled through the mixture for several minutes to insure that mixing was complete. The solenoid was then closed and the helium forced the mixture out of bulb B into the shot assembly. The camera was then started and the shot fired.

C. Data

A summary of the velocity and pressure measurements is given in Table II, and the isentrope data are given in Table III.

Shots 5, 4, and 10 were fired to determine how serious overdrive was at the 2" charge length. It appears that the 1" charge is overdriven, while the agreement between the results for the 2" and 3" charges is fortuitously good. Shots 6, 7, and 8 were fired to get some idea of how the free surface velocity depends on plate thickness. That objective was lost in the scatter of the data.

TABLE II

DETONATION PRESSURE AND VELOCITY MEASUREMENTS

Shot No.	Mixture	Density (gm/cc)	Charge Length	Al Plate Thickness	Free Surface Vel(mm/μsec)	Det Vel (m/sec)	C-J Pressure (kbars)
1	$B_{10}H_{13}C_2H_5 + 3.75\ C(NO_2)_4$	1.40	2"	0.1"	2.40	6740	172
2	$B_{10}H_{13}C_2H_5 + 4.45\ C(NO_2)_4$	1.43	2"	0.1"	2.29	6820	167
3	$B_{10}H_{13}C_2H_5 + 12(C_2F_3Cl)_x$	1.66	2"	0.1"	no record		< 100
5	$B_{10}H_{22.3}C_{3.62}$(Hi-Cal-3) + 4.3 $C(NO_2)_4$	1.40	1"	0.1"	2.40	6770	171
4	"	1.40	2"	0.1"	2.20	6660	157
10	"	1.40	3"	0.1"	2.21	6700	157
8	"	1.40	2"	0.1"	2.14		
6	"	1.40	2"	0.2"	2.29		
7	"	1.40	2"	0.4"	2.08		
9	$B_{10}H_{13}C_2H_5 + 7.5\ C_2F_4(NO_2)_2$	1.47	1"	0.1"	2.93 2.73*	7020 6910*	220 206*

* Corrected from 1" to 2" charge length using overdrive observed in Shots 5 and 4.

TABLE III

EXPERIMENTAL ISENTROPE STATES

Shot No.	Material	Density (gm/cc)	Pressure (kbars)	Particle Velocity (mm/μsec)	Mixture
11	Polyethylene	0.91	119.1	2.164	Ethyl decaborane-Tetranitromethane (Balanced to CO)
12	Polyurethane	0.356	52.5	3.40	Hi-Cal-3 - Tetranitromethane (Balanced to CO)
13	Polyurethane	0.108	20.9	4.49	Hi-Cal-3 - Tetranitromethane (Balanced to CO)

The total number of shots is small, and only shots 4 and 8 were made with the same mixture and shot assembly. The difference in U_{fs} is about 3%. To get some idea of the experimental error, in the absence of any straightforward way of doing so, we corrected the U_{fs} data for shots 6 and 7 back to 0.1" plate thickness using a slope of 0.01 mm/μsec/mm of plate thickness (see reference 4), so that these could be combined with the other 0.1" plate data for this mixture. We also added shot 10 to the collection. We then have the following five values for U_{fs} for the same mixture with about the same charge geometry: 2.20, 2.21, 2.14, 2.31, 2.16. The average of these is 2.20, and the standard deviation is about 3%.

The C-J pressure is obtained from U_{fs} (with plate thickness and impedance corrections) by means of the known Hugoniot for Dural[6]. In the region of interest to us, the dependence of P on U_{fs} is such that if the $\pm 2\sigma$ range for U_{fs} is ± 6%, the $\pm 2\sigma$ range for P_{CJ} is ± 7%. This is the only basis we have for assigning an error estimate to P_{CJ}. It is to be interpreted as meaning that the chances are 95 in 100 that the pressure determined in a single shot is within about 7% of the true value.

We can say essentially nothing about the probable accuracy of the velocity data other than to express our opinion that they are probably good to within 100 m/sec. The nearest thing we have to duplicate shots in this case is shots 4 and 10. These two velocity measurements differ by 40 m/sec.

In any case, no reasonable assignment of errors to the pressure and velocity measurements can affect the conclusion that the performance of these mixtures, as judged from their detonation velocities and C-J pressures, is about the same as that of TNT (189 kbar, 6940 m/sec at a density of 1.637 gm/cc; see ref 4).

III. THEORETICAL

In the absence of an obviously better choice, it was decided to base the theoretical estimation of the detonation parameters on the Kistiakowsky-Wilson equation of state, as modified by Cowan and Fickett[7]. To make the calculations as unbiased as possible in predicting the effect of boron, the Cowan and Fickett treatment was taken unchanged as the starting point, and the new product species were incorporated in it without adding any adjustable parameters. This was done by using geometrical covolumes for the new species and the same covolume scaling factor as was used by Cowan and Fickett for the products of a CHON explosive. The 704 code was rewritten with Fickett's assistance so that it would handle mixtures containing up to five elements and fifteen components, one of which may be solid carbon or boron nitride. This generalized version of Cowan and Fickett's technique is called the BKW calculation.

The BKW calculation computes the equilibrium composition of the explosion products at temperatures and pressures of interest, the Hugoniot, and the values of the hydrodynamic and thermodynamic variables at the

C-J point. The isentrope of the reaction products also can be obtained in either the pressure-volume or pressure-particle velocity plane. As input data the calculation requires, for the explosive, its elemental composition, heat of formation, density, and molecular weight; and for the explosion products, their elemental compositions, heats of formation, covolumes, and cubic fits of their ideal gas free energies, enthalpies, and entropy values as a function of temperature. A summary of the thermodynamic data used is given in Table IV. The equation of state of solid boron nitride was assumed to be the same as that of solid carbon[7]. The constants used in the K-W equation of state are $\theta = 400$, $\alpha = 0.5$, $\beta = 0.09$, and $K = 11.85$.

The C-J state was computed by an iteration procedure which was terminated when the convergence error in temperature was less than $\pm$ 10°C. The corresponding convergence errors in P and D are not the same for all systems, but are of the order of $\pm$ 5 kbar and $\pm$ 25 m/sec, respectively.

Using the above equation of state, equilibrium calculations were first made on two hypothetical mixtures, at pressures and temperatures in the range of C-J states, for the purpose of exploring the probable product compositions. The results of these calculations are given in Table V. In Table VI are summarized the results of the complete BKW detonation calculations. The computed equilibrium compositions correspond very closely to what one would write down on the basis of elementary thermochemical considerations. In particular, such species as HBO_2, BO, and BF appear in insignificant amounts over the interesting ranges of temperature

TABLE IV

THERMODYNAMIC FUNCTIONS USED IN BKW CALCULATIONS

A. $$\frac{F^o - H_o^o}{T} = A + BT + CT^2 + DT^3 \quad \text{cal/mole}$$

A	B	C	D	ΔH_f^o(cal/mole)	CoVolume	Comp
-4.6175941 01	-2.3242597 -02	3.9420140 -06	-2.7950911 -10	-2.1600000 05	7.3000000 02	B_2O_3
-4.4961337 01	-2.2038564 -02	4.6758886 -06	-4.3077710 -10	-1.2500000 05	1.2700000 03	HBO_2
-3.9766972 01	-1.2228144 -02	2.5230951 -06	-1.9876849 -10	-1.3500000 04	6.1000000 02	BO
-3.9544840 01	-1.2900944 -02	2.6710943 -06	-2.0975776 -10	2.0500000 05	6.7400000 02	B_2
-3.0355121 01	-8.6132032 -03	1.8716478 -06	-1.4993168 -10	1.3700000 05	2.1500000 02	B
-3.2264965 01	-1.2061703 -02	2.4997218 -06	-1.9732940 -10	1.1900000 05	5.3300000 02	BH
-4.7015899 01	-2.1361678 -02	3.9887672 -06	-2.9885974 -10	-2.6540000 05	8.0000000 02	BF_3
-3.8892234 01	-1.2598552 -02	2.6047180 -06	-2.0461300 -10	-4.1000000 04	6.8500000 02	BF
-3.1406387 01	-9.0184776 -03	1.9732591 -06	-1.5836815 -10	1.7800000 04	1.0800000 02	F
-3.8687314 01	-1.4303212 -02	3.3040978 -06	-2.9771648 -10	0.0000000 00	3.8700000 02	F_2
-3.2443138 01	-1.2866897 -02	3.0560516 -06	-2.8116360 -10	-6.4200000 04	3.8900000 02	HF
-3.9865955 01	-1.7239855 -02	3.7256316 -06	-3.3145889 -10	-9.3968600 04	6.7000000 02	CO_2
-3.7846828 01	-1.3549372 -02	3.2586333 -06	-3.0346358 -10	-2.7201000 04	3.9000000 02	CO
-3.4275974 01	-1.5515916 -02	3.5913964 -06	-3.3442338 -10	-5.7107000 04	3.6000000 02	H_2O
-3.4381767 01	-1.3568660 -02	3.3231040 -06	-3.0976957 -10	0.3560000 04	4.1300000 02	OH
-2.2030133 01	-1.3200634 -02	3.2328952 -06	-3.0484668 -10	0.0000000 00	1.8000000 02	H_2
-3.9455340 01	-1.3819999 -02	3.2965518 -06	-3.0646792 -10	0.0000000 00	3.5000000 02	O_2
-3.1845495 01	-8.9554835 -03	1.9610073 -06	-1.5761807 -10	5.8586000 04	1.2000000 02	O
-2.1199476 01	-8.5188205 -03	1.8441029 -06	-1.4747150 -10	5.1620000 04	7.6000000 01	H
-3.6339823 01	-1.3494712 -02	3.2540765 -06	-3.0356132 -10	0.0000000 00	3.8000000 02	N_2
-3.0775740 01	-9.8937231 -03	2.5040278 -06	-2.3512957 -10	1.7039000 05	1.8000000 02	C_{gas}
-5.1460249 01	-2.9074957 -02	7.2300918 -06	-7.3550121 -10	-9.4500000 04	1.8800000 03	BCl_3
-4.1281894 01	-2.7594993 -02	6.1904193 -06	-5.4899036 -10	-1.0700000 05	1.7400000 03	B_2O_2
-4.1214478 01	-1.2480916 -02	2.5876096 -06	-2.0422183 -10	2.1477000 04	3.8600000 02	NO
-2.9898633 01	-9.6433717 -03	2.4230184 -06	-2.2644542 -10	1.1320000 05	1.4800000 02	N
-4.3143356 01	-1.5649299 -02	4.2908723 -06	-4.4598778 -10	1.5800000 05	6.1900000 02	BN_{gas}
-3.3299359 01	-8.5759733 -03	1.7876378 -06	-1.3893633 -10	2.8610000 04	2.5500000 02	Cl
-4.4036909 01	-1.3177905 -02	2.6476910 -06	-2.0345443 -10	0.0000000 00	9.5600000 02	Cl_2
-4.2552618 01	-1.3255273 -02	2.7100678 -06	-2.1046991 -10	-1.3200000 04	6.1000000 02	ClF
-3.6240431 01	-1.1369114 -02	2.2672961 -06	-1.7488606 -10	-2.2019000 04	6.3700000 02	HCl
6.8400312 -01	-3.9691768 -03	5.4320370 -07	-3.5436910 -11	0.0000000 00		C_{solid}
9.7547996 -01	-8.6061024 -03	1.1290291 -06	-8.2084012 -11	-5.9614000 04		BN Solid
-3.5026940 +01	-1.4878630 -02	2.5258870 -06	-1.8353660 -10	-9.3680000 03	4.7600000 02	NH_3

TABLE IV - Continued

B. $H^o - H^o_o = E + FT + GT^2 + HT^3$ kcal/mole

E	F	G	H	Comp
-7.5311403 00	2.3501528 -02	5.0078832 -07	-3.7061636 -11	B_2O_3
-1.5194107 00	1.2819217 -02	1.8939762 -06	-1.7341590 -10	HBO_2
-3.7703777 -01	7.4744433 -03	3.7497569 -07	-2.7574505 -11	BO
-5.7108593 -01	8.2962363 -03	1.6292012 -07	-1.3166030 -11	B_2
8.4079326 -01	3.4606143 -03	5.1363168 -07	-4.8099577 -11	B
-2.8786683 -01	7.2181232 -03	4.0318854 -07	-3.2090681 -11	BH
-2.9916057 00	1.6516358 -02	8.5849355 -07	-7.1685729 -11	BF_3
-5.0730324 -01	7.9414833 -03	2.5368681 -07	-2.1063694 -11	BF
6.5200688 -02	5.1873716 -03	-5.5932327 -08	4.6651210 -12	F
-5.0902677 -01	8.3756700 -03	1.5041495 -07	-1.2587522 -11	F_2
1.0560298 -01	6.4134383 -03	4.9843289 -07	-2.7044019 -11	HF
-1.7520862 00	1.1324844 -02	9.3943765 -07	-6.9022596 -11	CO_2
-2.8805780 -01	7.2392401 -03	4.2348950 -07	-3.1952568 -11	CO
-3.1040336 -01	7.8668458 -03	1.2521248 -06	-9.1715274 -11	H_2O
2.0349765 -01	6.3385812 -03	6.0057014 -07	-4.0202533 -11	OH
1.2571716 -01	6.3156688 -03	5.3753858 -07	-2.5869179 -11	H_2
-2.6812553 -01	7.3342769 -03	5.1272257 -07	-3.1081972 -11	O_2
1.2460506 -01	5.0825015 -03	-5.2444136 -08	8.6921368 -12	O
-3.9557220 -03	4.9706877 -03	-6.3061052 -10	4.7101173 -14	H
-2.3173451 -01	7.1023081 -03	4.4577141 -07	-3.3338623 -11	N_2
1.5242314 -01	4.8289840 -03	4.8509021 -08	1.9523957 -12	C_{gas}
-6.4316249 -01	1.3725061 -02	2.8122757 -06	-3.0445101 -10	BCl_3
-1.7132206 00	1.5867957 -02	1.4274445 -06	-1.3544221 -10	B_2O_2
-2.9580690 -01	7.5421514 -03	3.6031572 -07	-2.6057759 -11	NO
-9.1004850 -02	5.1855190 -03	-1.3189395 -07	2.3067381 -11	N
-1.3814640 -01	6.9941684 -03	8.1456142 -07	-1.0560044 -10	BN_{gas}
-4.8996546 -01	8.7012293 -03	1.2807482 -07	-5.4518577 -12	Cl
-1.0939097 -01	5.4786457 -03	-1.1382814 -07	8.7553181 -12	Cl_2
-7.1510673 -01	8.9344785 -03	-1.4181942 -07	3.5493056 -11	ClF
-7.7426673 -02	6.7454587 -03	5.1749092 -07	-3.5747382 -11	HCl
-9.6113438 -01	3.1036394 -03	1.1108129 -06	-1.2080233 -10	C_{solid}
-2.6159186 00	8.2425876 -03	1.3040688 -06	-9.4143886 -11	Solid BN
-1.3538320 00	-9.6928240 -03	2.2455920 -06	-1.7166100 -10	NH_3

TABLE IV - Continued

C. $S^{o} = I + JT + KT^{2} + LT^{3}$ cal/deg - mole

I	J	K	L	Comp
5.9813394 01	2.7045000 -02	-4.3370370 -06	2.8516278 -10	B_2O_3
5.1988193 01	2.9032767 -02	-6.4045530 -06	5.5585372 -10	HBO_2
4.6361125 01	1.3154985 -02	-2.6749591 -06	2.0775673 -10	BO
4.5852910 01	1.4214440 -02	-2.9280442 -06	2.2716433 -10	B_2
3.5426967 01	8.5134851 -03	-1.8397128 -06	1.4673010 -10	B
3.9000068 01	1.2581781 -02	-2.4386612 -06	1.8070420 -10	BH
5.5077063 01	2.8561902 -02	-5.7378235 -06	4.4062451 -10	BF_3
4.5448140 01	1.3909949 -02	-2.9008867 -06	2.2705562 -10	BF
3.6736610 01	8.8742542 -03	-1.9468930 -06	1.5642193 -10	F
4.6098419 01	1.4414513 -02	-3.0117631 -06	2.3636263 -10	F_2
3.9588333 01	1.1947426 -02	-2.3801665 -06	1.8567894 -10	HF
4.7088296 01	2.0192047 -02	-3.9224113 -06	2.9761140 -10	CO_2
4.5124549 01	1.2893814 -02	-2.6088139 -06	2.0215143 -10	CO
4.2400094 01	1.5265242 -02	-2.8158015 -06	2.1041134 -10	H_2O
4.2069383 01	1.2123368 -02	-2.4434109 -06	1.9276978 -10	OH
2.9392185 01	1.2000980 -02	-2.4252063 -06	1.9178467 -10	H_2
4.6724952 01	1.3410523 -02	-2.7020135 -06	2.1078870 -10	O_2
3.7276429 01	8.6409084 -03	-1.8808859 -06	1.5121595 -10	O
2.6167783 01	8.5165526 -03	-1.8411183 -06	1.4686805 -10	H
4.3649305 01	1.2724816 -02	-2.5671316 -06	1.9879985 -10	N_2
3.7223696 01	7.8336751 -03	-1.6379070 -06	1.3018974 -10	C_{gas}
-5.3540075 -01	7.5076291 -03	-1.3805167 -06	1.0545415 -10	C_{solid}

The format of each number is ± X.XXXXXXX E ± XX where E is the exponent to the base 10.

TABLE V

EQUILIBRIUM COMPOSITION (MOLE FRACTIONS) COMPUTED BY THE BKW CALCULATIONS

SYSTEM: $B_3N_6F_9 \rightarrow 3\ BF_3 + 3\ N_2$

Temp (°K)	Pressure (kbar)	BF_3	BF	B_2	B	N_2	F_2	F	Volume (cc/mole gas)
3000	300	0.500	3×10^{-5}	$< 1 \times 10^{-8}$	8×10^{-5}	0.500	$< 1 \times 10^{-8}$	3×10^{-4}	14.80
	400	0.500	4×10^{-6}	$< 1 \times 10^{-8}$	3×10^{-5}	0.500	$< 1 \times 10^{-8}$	9.6×10^{-5}	13.29
5000	300	0.496	5×10^{-4}	5×10^{-5}	1×10^{-3}	0.497	1.7×10^{-4}	4.6×10^{-3}	15.72
	400	0.498	6×10^{-5}	3×10^{-5}	7×10^{-4}	0.499	1×10^{-4}	2.4×10^{-3}	13.98

SYSTEM: $B_3N_3O_6H_3 \rightarrow 1.5\ B_2O_3 + 1.5\ N_2 + 1.5\ H_2O$

Temp (°K)	Pressure (kbar)	B_2O_3	HBO_2	B_2O_2	BO	N_2	H_2O	H_2	O_2	Volume (cc/mole gas)
3000	300	0.332	1×10^{-5}	$< 1 \times 10^{-8}$	$< 1 \times 10^{-8}$	0.332	0.326	6.6×10^{-3}	3.3×10^{-3}	13.3
	400	0.332	1×10^{-6}	$< 1 \times 10^{-8}$	$< 1 \times 10^{-8}$	0.332	0.328	4.3×10^{-3}	2.2×10^{-3}	11.9
4000	300	0.325	1.7×10^{-3}	1×10^{-6}	2×10^{-5}	0.326	0.281	4.4×10^{-2}	2.2×10^{-2}	14.1
	400	0.327	1.7×10^{-4}	3×10^{-8}	9×10^{-6}	0.327	0.291	3.6×10^{-2}	1.8×10^{-2}	12.5

TABLE VI. TYPICAL RESULTS OF BKW CALCULATIONS

Compound or Mixture	ΔH_f (kcal/mole)	ρ_o (gm/cc)	P_{CJ} (kbars)	T_{CJ} (°K)	V_{CJ} (cc/mole gas)	Det Vel (m/sec)	No. moles gas/mole expl.	No. mole Sol. carb.	γ	MOLE FRACTION OF GASEOUS DETONATION PRODUCTS																						No. moles Solid BN
										BF_3	BF	B_2O_3	HBO_2	NH_3	BO	B_2	B	H_2O	OH	H_2	O_2	CO_2	CO	HF	F_2	N_2	C_{gas}	H	O	F	BH	
$B_{10}H_{13}C_2H_5$ + 3.75 $C(NO_2)_4$ SHOT #1	+ 40	1.40	191	4900	16.5	6890	27.3	0	2.50			0.178	.0072	.023	3.8×10^{-3}	6×10^{-8}	6×10^{-8}	0.219	0.017	0.065	0.013	0.075	0.136			0.264	1×10^{-4}					0
$B_{10}H_{13}C_2H_5$ + 4.45 $C(NO_2)_4$ SHOT #2	+ 46	1.43	190	4810	16.6	6850	30.8	0	2.50			0.158	0.006	.010	.002	4×10^{-9}	2×10^{-5}	0.230	0.023	0.032	0.046	0.111	0.098			0.284	3×10^{-4}				2×10^{-5}	0
$B_{10}H_{13}C_2H_5$ + $C_2F_4(NO_2)_2$ SHOT #9	-1290	1.467	185	4500	17.2	6620	44.9	0	2.47	0.198	1.4×10^{-4}	0.012				2×10^{-7}	8×10^{-5}	0.119	7×10^{-3}	0.041	3.5×10^{-3}	0.120	0.259	0.071	3.4×10^{-4}	0.167						
$B_{10}H_{13}C_2H_5$ + 12 C_2F_3Cl SHOT #3*	-1793	1.66	108	2710	22.4	5870	28.6	26	4.26	0.348	1.7×10^{-6}						1×10^{-8}			.0595				0.207	1.8×10^{-5}		3×10^{-7}	1×10^{-3}		1×10^{-3}		
N-Trinitroborazole $B_3N_6H_3O_6$	- 116	1.70	207	3340	14.9	6530	6.06		2.50			0.247	2×10^{-4}		6×10^{-7}	1×10^{-9}	1×10^{-6}	0.231	0.003	0.014	0.006					0.494		1×10^{-3}	1×10^{-3}			0
N-tri(difluoroamino) B-trifluoroborazole $B_3N_6F_9$	- 435	1.70	146	4120	20.7	5730	6.02		2.83	0.497	0.001					1.4×10^{-5}	5.6×10^{-4}								5.5×10^{-5}	0.498				3.4×10^{-3}		
B-tri(difluoroamino) N-trifluoroborazole $B_3N_6F_9$	- 184	1.70	212	6380	18.4	6400	6.35		2.28	0.442	0.005					1.5×10^{-4}	.0046								0.014	0.462				0.057		0.04
N or B-tri(difluoroamino) borazole $B_3N_6H_3F_6$	- 135	1.70	203	4040	15.9	6530	6.18		2.59	0.317	0.006					0.050	0.035			0.218				0.013	1×10^{-6}	0.323		0.010		5×10^{-4}	0.025	
RDX	14.7	1.80	346	2567	11.1	8610	7.83	1.17	2.85									.374	1.4×10^{-4}	0.009	3×10^{-4}	0.157	0.076			0.383	1×10^{-8}	1×10^{-4}	3.5×10^{-5}			

* Also 1.4×10^{-3} BCl_3, 3.4×10^{-2} Cl_2, 3.9×10^{-2} Cl,
5.6×10^{-3} ClF, 0.302 HCl.

and pressure. Thus the heats of explosion computed for these systems by this method are, indeed, high. On the other hand, the predicted pressures and velocities are low.

The experimental and calculated values of P and D are compared in Table VII. The agreement is sufficiently good for our purposes, although the differences are generally larger than would be expected on the basis of the assigned errors.

A possible explanation for the relatively poor performance of the boron mixtures can be proposed on the basis of the results of the calculations. Because the product molecules B_2O_3 and BF_3 are complex, the particle density at the C-J point is low (V_{CJ} is high -- see Table VI). As a result, the temperature is high, the pressure is low, and hence much of the energy becomes available only through an adiabatic expansion of the products to fairly low pressures. It is this feature of the results which makes the isentropes of interest.

It was mentioned previously that the BKW code can be used to compute the isentrope of the reaction products, and it has also been shown[8] that a simple γ-law equation of state reproduces the experimentally determined isentrope of Composition B reasonably well over the range studied. In Fig 4 we have plotted the BKW and γ-law isentropes for EDB/TNM, using for the latter the γ computed from the experimental P and D by means of the relation

$$\gamma = \frac{\rho_0 D^2}{P} - 1.$$

TABLE VII

COMPARISON OF EXPERIMENTAL AND COMPUTED DETONATION VELOCITIES AND PRESSURES

Mixture or Compound	Density (gm/cc)	C-J Pressure (kbar) Exper	C-J Pressure (kbar) Calc	D (m/sec) Exper	D (m/sec) Calc
$B_{10}H_{13}C_2H_5 + 3.75\ C(NO_2)_4$	1.40	172	191	6740	6890
$B_{10}H_{13}C_2H_5 + 4.45\ C(NO_2)_4$	1.43	167	190	6820	6850
$B_{10}H_{13}C_2H_5 + 12\ C_2F_3Cl$	1.66	< 100[a]	108		5870
$B_{10}H_{13}C_2H_5 + 7.5\ C_2F_4(NO_2)_2$	1.47	206[b]	185	6910[b]	6620
RDX	1.767	338[c]	334	8640[c]	8500

[a] No trace obtained. Estimated pressure < 100 kbar.

[b] Corrected for overdrive. See Table II.

[c] Experimental data from reference 4.

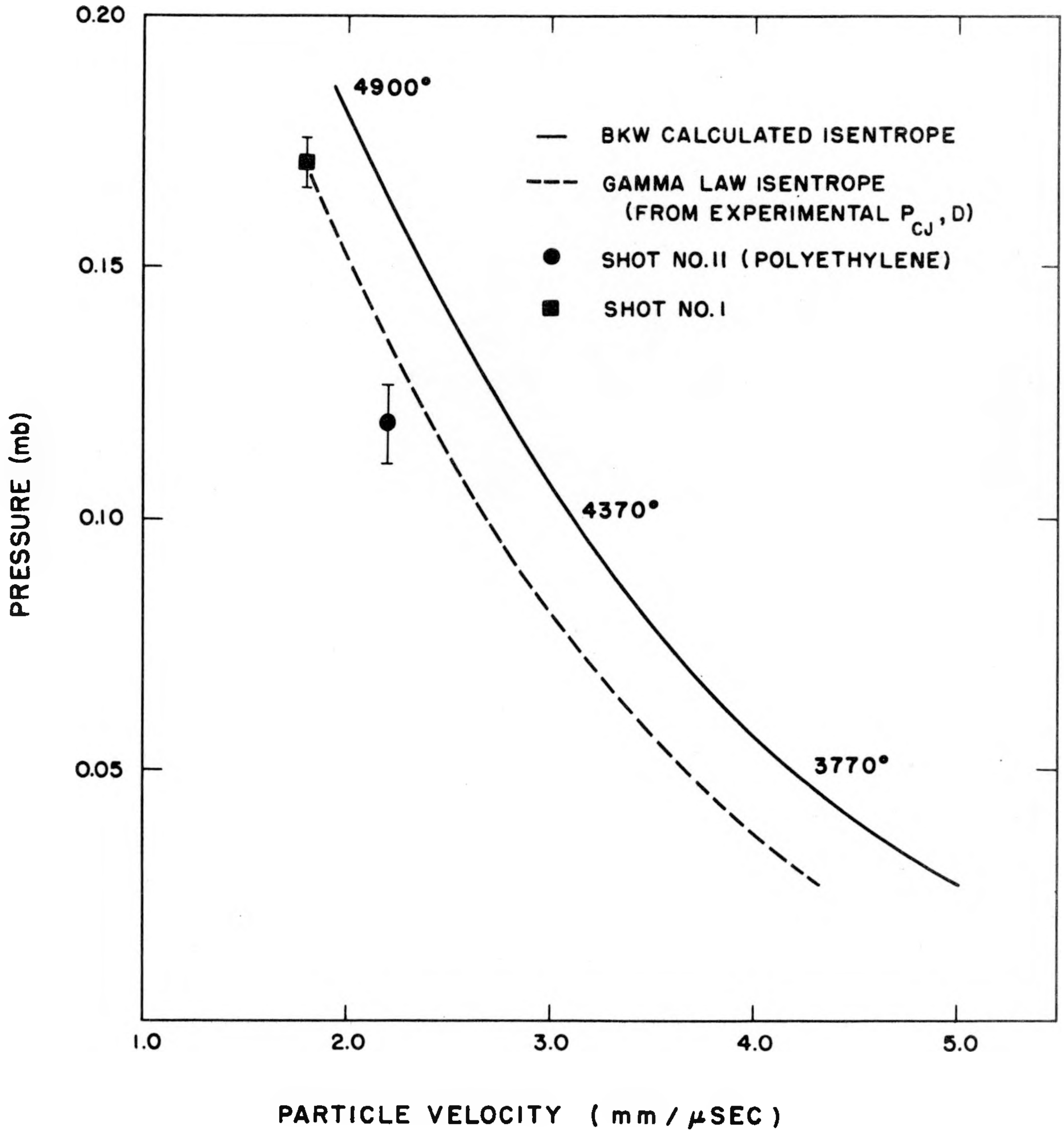

Figure 4. Isentrope for Ethyl Decaborane-Tetranitromethane Mixture

The two experimental values also are shown. The two isentropes do not differ greatly in shape over the experimental range, but the γ-law isentrope is, of course, forced to go through the experimental C-J point and therefore appears to represent the data better.

In Fig 5 the γ-law isentrope and experimental points are shown for the Hi-Cal-3/TNM mixture. The BKW calculations were not done for this system since the heat of formation of Hi-Cal-3 is not known.

By considering the trajectory in the P-V plane of an element of volume of a detonating explosive one can deduce that the net work available from the adiabatic expansion of the products from the C-J state is given by

$$E = \int_{V_{CJ}}^{V} P\, dV - 1/2\, P_{CJ}\, (V_o - V_{CJ}), \qquad (1)$$

where the integral is the area under the isentrope, and the second term on the right is the work done in the shock compression from the initial state to the C-J state. As $V \rightarrow \infty$, this becomes approximately equal to the constant volume heat of explosion per gram of explosive. In the special case of the γ-law equation of state, the integral is evaluated along the path

$$PV^{\gamma} = P_{CJ}\, V_{CJ}^{\gamma} \qquad (\gamma \text{ constant})$$

and, for the upper limit $V = \infty$, the equation reduces readily to

$$E = P_{CJ}/2\rho_o(\gamma - 1) \ ,$$

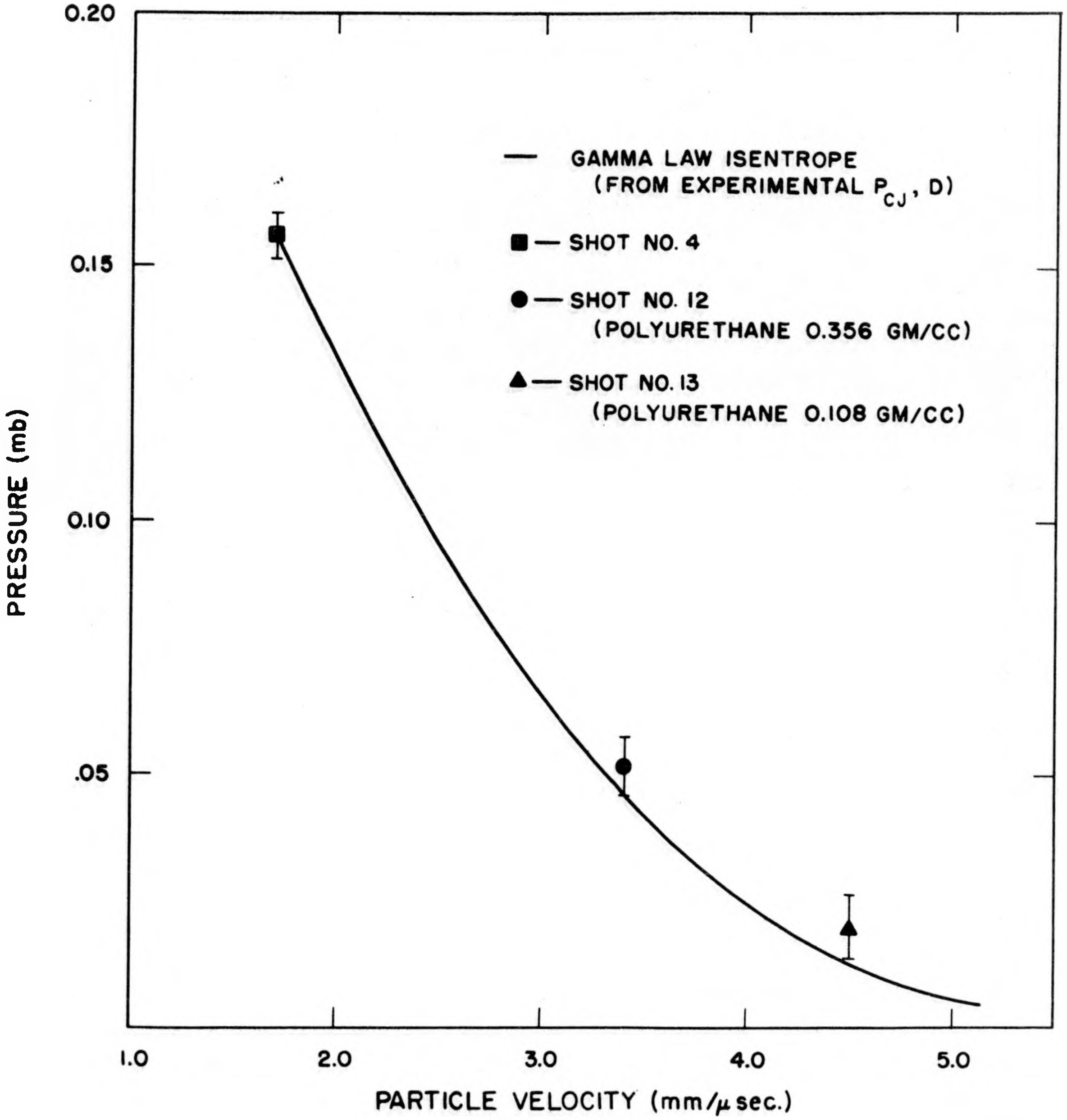

Figure 5. Isentrope for Hi-Cal 3-Tetranitromethane Mixture

a result obtained by Jacobs[9] on the basis of more detailed considerations. This equation works reasonably well for many organic explosives, but in the case of the boron mixtures studied by us, the heats of explosion computed in this way are only about half the heats of explosions corresponding to the reactions involved. Presumably the principal reason for this is failure of the γ law to hold in the region of low pressures.

We have evaluated the integral along the BKW isentropes for several systems. The computed isentropes for RDX at an initial density of 1.80 gm/cc, and for EDB/TNM at an initial density of 1.40 gm/cc, cross at a pressure of about 10 kbar (expansion ratio, V/V_{CJ}, about 4), but the energies for the two systems do not become equal until the pressure is below 5 kbar. For RDX at the same initial density as the EDB/TNM mixture (1.40 gm/cc), the isentropes cross at an expansion ratio of about 1.4, while the energies become equal at a pressure of about 35 kbar (expansion ratio about 2).

To compare the two systems on an equal volume basis we have to multiply E by the initial density. The integral then becomes

$$\frac{1}{V_o} \int_{V_{CJ}}^{V} P\,dV = \int_{V_{CJ}}^{V} P\,d\left(\frac{V}{V_o}\right) = \int_{V_{CJ}}^{V} P\,d\left(\frac{V-V_{CJ}}{V_o}\right) .$$

The BKW isentropes for RDX at 1.80 gm/cc and for EDB/TNM at 1.40 gm/cc are plotted in this form in Fig 6, together with the rays representing the second term in equation (1). E as a function of V is then given by the difference between the area under the isentrope and the (constant) area under the triangle.

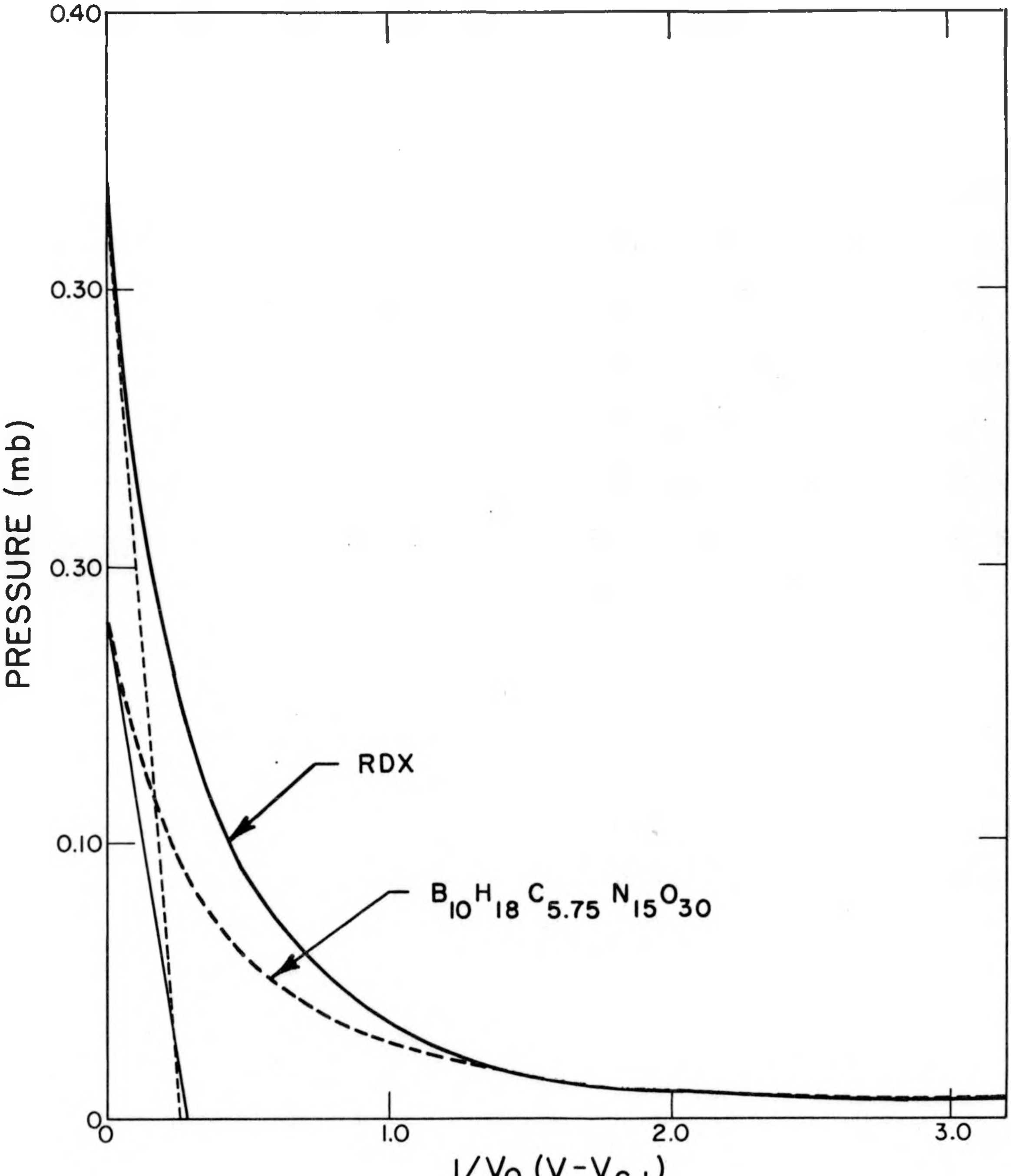

Figure 6. Calculated Isentropes

The computed and experimental pressures and velocities are in sufficiently good agreement to make it of some interest to use the BKW code to estimate the performance of various hypothetical boron systems. For example, we might inquire what the performance of a CO-balanced boron explosive would be if it were possible to obtain such a mixture at a higher density. The results for the EDB/TNM mixture are as follows:

ρ_o	P_{CJ}(kbar)	D(m/sec)	T(°K)
1.40	191	6890	4900
1.80	313	7920	4510
RDX-1.80	347	8610	2567

This suggests that even at much increased densities, the detonation pressure of a boron/oxygen explosive of this type would not equal that of the common organic explosive RDX.

Calculations also have been done for mixtures of a hypothetical boron explosive with RDX. The idea here is to use the energy released in the formation of B_2O_3 to further heat the detonation products of the organic explosive. The boron explosive was assumed to have a zero heat of formation and B_2O_3 as its only product. The calculations were done for mixtures of the two materials in various proportions at a constant assumed density of 1.70 gm/cc. The calculated maximum P_{CJ} was about 10% higher than that of pure RDX at this same density and was obtained for a composition corresponding to three moles of RDX per mole of B_2O_3 formed. Hence systems of this sort also appear unpromising.

Along this same line, calculations were performed for a boron system ($\rho_o = 1.40$, $E^o = 109.2$) in which the number of moles of gas per gram of explosive was increased so as to partition the energy available more favorably. A marked increase in performance was predicted as follows:

System	T_{CJ}(°K)	V_{CJ}	P_{CJ}(kb)	D(m/sec)
$B_{10}H_{18}C_{5.75}N_{15}O_{30}$	4900	16.5	191	6890
$B_{10}H_{50}$ "	3710	13.4	256	7980
$B_{10}H_{100}$ "	2430	10.3	368	10050

However, there appears to be little prospect of obtaining actual systems of this type.

The most interesting systems for which the calculations have been done are those producing boron nitride and hydrogen as the only products. Here the boron appears as a diatomic molecule, and the average molecular weight of the products is low. The computed results for decaborane/hydrazine and pentaborane/hydrazine mixtures are given in Table VIII. The first three lines of the table were obtained for realistic densities, which are quite low. The C-J pressures also are low, but the velocities are surprisingly high. Furthermore, as the last three lines of the table indicate, both the pressure and velocity are very strong functions of the density, so that the computations offer considerable encouragement for a search for a higher density BNH system.

The $B_{10}H_{12} \cdot 2\ N_2H_4 + 8\ N_2H_4$ mixture has been fired, and a C-J pressure of 145 kbar and a detonation velocity of 6625 m/sec were obtained. Hence

TABLE VIII

BKW RESULTS FOR DECABORANE/HYDRAZINE AND PENTABORANE/HYDRAZINE

	ΔH_f (kcal/ mole)	E^o (kcal/ mole)	ρ_o (gm/cc)	P_{CJ} (kilo-bars)	T_{CJ} (°K)	V_{CJ} (cc/ mole gas)	Det Vel (m/sec)	n_{gas}	$n_{(BN)}$ solid	γ	MOLE FRACTION							
											BN	B_2	B	BH	N_2	H_2	H	NH_3
$B_{10}H_{12} \cdot 2\ N_2H_4$ + 3 N_2H_4	86.15	131.9	1.00	242	2970	8.04	9800	16.1	10.0	2.98	1×10^{-8}	6×10^{-7}	2×10^{-4}	3×10^{-5}	1×10^{-8}	0.989	9×10^{-3}	3×10^{-4}
$B_{10}H_{12} \cdot 2\ N_2H_4$ + 8 N_2H_4	146	221.4	1.00	201	2430	11.58	8930	21.2	10.0	2.95	1×10^{-8}	1×10^{-8}	1×10^{-8}	1×10^{-8}	5×10^{-3}	0.532	1×10^{-3}	0.461
B_5H_9 + 2.5 N_2H_4		+63	0.78	137	2400	9.94	8560	9.52	5.00	3.1	1×10^{-8}	1×10^{-8}	1×10^{-8}	1×10^{-8}	2×10^{-5}	0.996	4×10^{-3}	1×10^{-4}
			1.20	448	2170	5.88	12,620	9.50	5.00	3.27	"	"	"	"	8×10^{-8}	0.999	8×10^{-4}	1×10^{-8}
			1.70	1160	985	3.92	17,790	9.50	5.00	3.63	"	"	"	"	1×10^{-8}	0.999	1×10^{-8}	1×10^{-8}

in this case there is a significant disagreement between calculation and experiment. Considering the manner in which the equation of state was originally calibrated, we can hardly be surprised at this, and it would certainly be premature to write these systems off on the basis of the results presently available.

IV. SUMMARY AND CONCLUDING DISCUSSION

As matters now stand, we feel that we have a satisfactorily consistent experimental and theoretical picture of the behavior of boron/oxygen explosive systems, and it is our conclusion that such systems are unpromising for use in applications for which high detonation pressures are of principal importance. On the other hand, we have no reason to doubt that the heats of explosion of these mixtures are high, and the possibility remains that they would perform well in applications which rely importantly on the expansion of the detonation products, as, for example, in underwater explosions.

Admittedly, these statements are based on rather scanty experimental data, and on a theoretical treatment which is open to some criticism. Our confidence in the theory would be strengthened considerably if we could obtain experimental verification of an additional computed hydrodynamic or thermodynamic variable at the C-J state, such as the temperature. The computed C-J temperatures of the boron/oxygen systems are considerably higher than those of the usual organic explosives, and the difference should be experimentally detectable. Experiments along these lines are in progress using a technique being developed by W. C. Davis of GMX-8.

Thus far the experimental results are in qualitative agreement with the computed temperatures, but it would be premature to claim any more than this at the present time.

As to the BNH systems, the situation, while much less clear, remains sufficiently interesting in several respects that we feel that further work, both experimental and theoretical, is clearly justified.

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