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DETONATION PERFORMANCE CALCULATIONS USING THE
KISTIAKOWSKY-WILSON EQUATION OF STATE

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DETONATION PERFORMANCE CALCULATIONS USING THE
KISTIAKOWSKY-WILSON EQUATION OF STATE

by

Charles L. Mader

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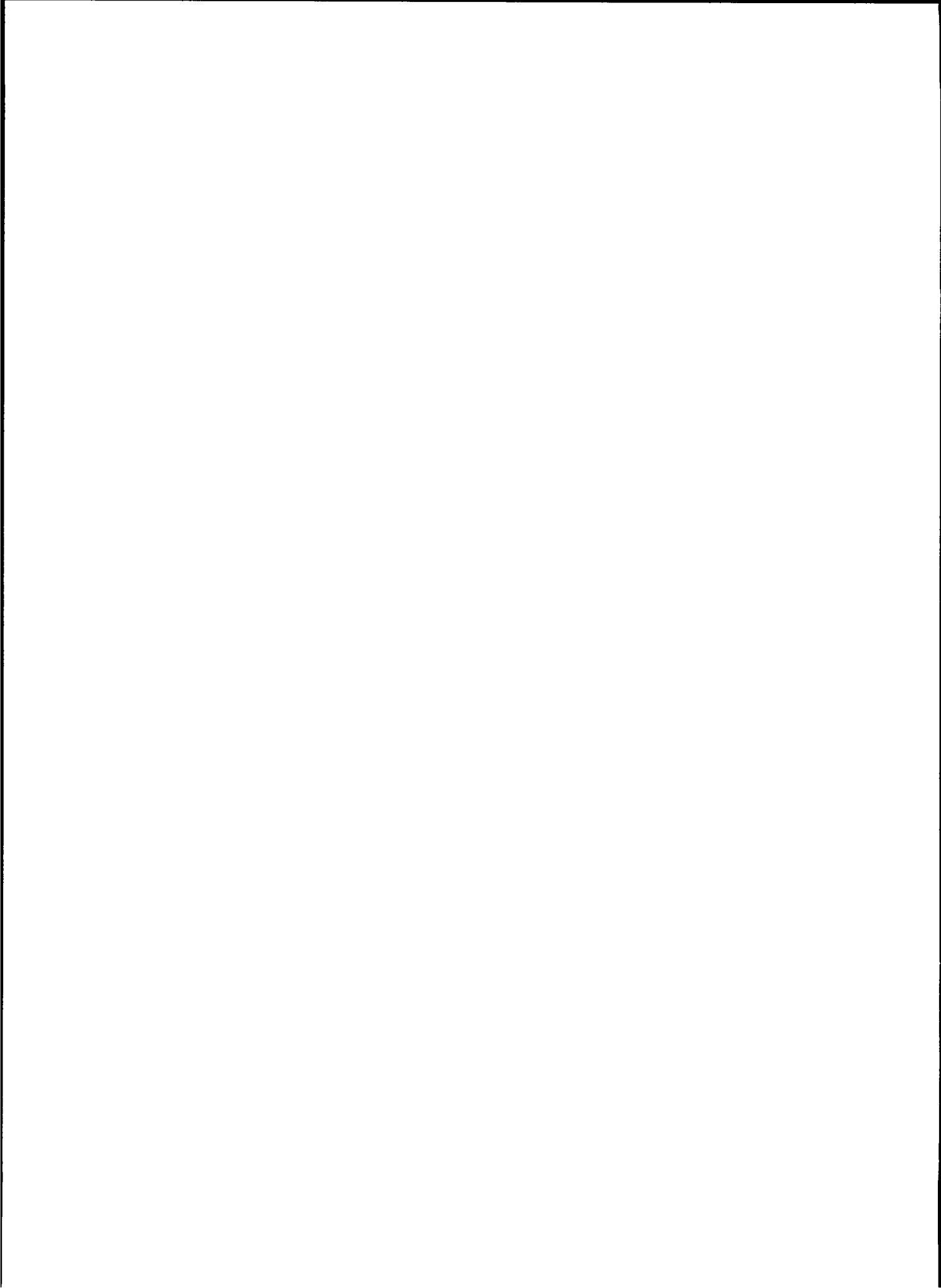


ABSTRACT

The Kistiakowsky-Wilson equation of state as modified by Cowan and Fickett was used to estimate the detonation performance of explosives composed of various combinations of the elements carbon, hydrogen, nitrogen, boron, aluminum, oxygen, and fluorine. The computed velocities, pressures, and temperatures are compared with the available experimental detonation velocities, Chapman-Jouguet pressures, and brightness temperatures. Over a wide range of density and composition the computed and experimental pressures and temperatures agree to within 20%, the detonation velocities to within 10%. The interrelationships between temperature, pressure, and the particle density of the C-J products as predicted by the Kistiakowsky-Wilson equation of state are discussed.

ACKNOWLEDGMENTS

The author gratefully acknowledges the assistance rendered by Wildon Fickett, Wray Garn, William C. Davis, L. C. Smith, M. J. Urizar, E. James, W. E. Deal, B. J. Craig, E. D. Loughran, L. W. Kissinger, W. E. McQuiston, M. Schwartz, and the late William H. Rogers of the GMX Division of the Los Alamos Scientific Laboratory. He also wishes 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 some of the materials used in the experimental program.



INTRODUCTION

During the last four years the Kistiakowsky-Wilson equation of state, as modified by Cowan and Fickett, has been used to estimate the detonation performance of explosives composed of various combinations of the elements carbon, hydrogen, nitrogen, boron, aluminum, oxygen, and fluorine. The investigation was concentrated on the systems with high heats of explosion in an effort to determine if such systems are promising for use in applications for which high detonation pressures are of principal importance.

EKW CALCULATIONS

The theoretical estimation of the detonation parameters is based on the Kistiakowsky-Wilson equation of state as modified by Cowan and Fickett.¹ To make the calculations as unbiased as possible in predicting the effect of various combinations of elements, 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 all the products except the carbon-fluorine products. The 704 code was written, 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 solid (uncompressed) aluminum oxide. The Brinkley method was used for computing the equilibrium composition. This generalized version of Cowan and Fickett's technique is called the EKW calculation.

The BKW calculation computes the equilibrium composition of the explosion products at temperatures and pressures of interest, the detonation 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 the 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. The thermodynamic data used were taken from references 2 and 3. The covolumes used are given in Table I. 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 10°C . The corresponding convergence errors in pressure and detonation velocity are not the same for all systems, but are of the order of ± 5 kilobars and ± 25 meters/second, respectively.

EXPERIMENTAL C-J PERFORMANCE MEASUREMENTS

The methods used at Los Alamos to measure the C-J pressure and detonation velocity of an explosive have been described previously in the open literature and are adequately referenced in Table II. The brightness measurements of W. C. Davis of this Laboratory will be

published some time in the future. The temperatures reported are those of a black body of equivalent photographic brightness, probably with relative accuracy of 50°K, since each shot has a nitromethane internal standard, and absolute accuracy of about 200°K. The relationship between these numbers and the actual detonation temperature is not known. Since the agreement between Davis's temperatures and those of other investigators for void-free systems is rather good, we shall assume, as previous investigators have, that the temperatures we are measuring are the C-J temperatures.

The estimated errors given in Table II for some of the pressure and velocity measurements are considerably larger than normally associated with the techniques used. The accuracy of the results suffered primarily as a result of the necessity to design the shot setup in such a way as to use a minimum amount of material and the necessity of preparing and loading the hazardous mixtures by remote control.

CHNO Systems

Cowan and Fickett's version of the Kistiakowsky-Wilson equation of state was calibrated for RDX, Cyclotol, Composition B, and TNT, systems 1, 2, 3, 4, and 5 of Table II. They obtained their poorest agreement for TNT.

Systems 6, 7, 8, and 9 show the effect of changes in oxygen balance. The C-J pressure and detonation velocity of the CO₂-balanced system are disappointing in comparison with what one might expect on the basis of simple heat of explosion and gamma law ($PV^\gamma = C$) predictions. If one assumes

that the detonation velocity increases with density at about 3,000 meters/second/gram/cc, then the velocity difference between the CO- and the CO₂-balanced systems may be attributed entirely to the difference in density. The temperature increases as the amount of CO₂ increases until an excess of oxygen is present, and then the temperature decreases. The observed C-J performance may be explained by the lower particle density at the C-J state for systems producing CO₂ instead of CO. The extra energy present in such a system is primarily thermal energy rather than intermolecular potential energy. Thus the temperature would be expected to increase as the amount of CO₂ formed was increased and the pressure and velocity remain relatively unchanged.

BCHNOF Systems

Systems 19, 20, and 21 are homogeneous systems which produce B₂O₃ and BF₃ as detonation products. Although the heats of explosion are almost twice that of conventional CHNO explosives, the observed C-J pressures and detonation velocities are not as high as those of the better CHNO explosives at the same densities. A possible explanation for the poor C-J pressures and velocities of the boron explosives relative to the CHNO explosives can be proposed on the basis of these calculations. Because the product molecules B₂O₃ and BF₃ are complex, the particle density at the C-J point is lower than for systems containing the product molecules CO, CO₂, H₂O, and N₂. Thus the energy is partitioned unfavorably with the intermolecular potential energy low and the thermal energy high.

At C-J densities the intermolecular potential energy is the primary pressure-determining part of the energy; thus the C-J pressures of the boron explosives are low and the C-J temperatures are high. 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 equilibrium expansion of the detonation products.

CHNOAl Systems

Systems 17 and 18 are nonhomogeneous systems, but the C-J pressures and velocities may be explained by assuming that the H.E./Al behaves as if it is a homogeneous explosive and the product molecule Al_2O_3 is formed as an equilibrium C-J product. Again the computed and experimental C-J pressures and velocities are lower than one might expect from heat of explosion considerations because of the low particle density of the detonation products. One expects that the C-J temperature would be high and that H.E./Al systems would perform well in applications which rely on the equilibrium expansion of the detonation products.

CHNOF Systems

Systems 14, 15, and 16 show the computed and experimental C-J pressures and velocities of systems containing fluorine. Although the most desirable CHNOF system would have the fluorine attached to the molecule by means of an N-F bond rather than a C-F bond, the C-J pressures and velocities of the available systems are instructive. The calculated C-J

pressures and velocities of the CHNOF systems are very sensitive to the HF, carbon, CF_4 equilibrium. If one does not consider the CF, CHF, and COF species, BKW calculations predict pressures and velocities that are higher than experimentally observed. If one includes CF_4 , the calculated pressures and velocities are somewhat lower than experimentally observed. The covolumes of the CF, CHF, and COF species were increased by a factor of 1.6 so as to cause a slight shift in the HF, carbon, CF_4 equilibrium and better agreement between experimental and calculated C-J pressures and velocities. This empirical observation may be of some value to anyone wishing to use the BKW technique for predicting the possible C-J performance of some other CHNOF explosive.

CHNOF explosives appear to form products that are energy-releasing species such as HF and CF_4 . CF_4 is less desirable than HF because of its large molecular weight and hence detrimental effect on the particle density, resulting in the energy being partitioned so as to give higher temperatures and lower pressures.

AGREEMENT BETWEEN EXPERIMENTAL AND CALCULATED C-J PARAMETERS

For the systems reported the BKW technique predicts the C-J pressure and temperature to within 20% and the detonation velocity to within 10% of the observed values. The agreement is generally poorer at lower densities. One cannot expect the BKW technique to predict the C-J performance of systems that may deviate from equilibrium. The BKW technique fails for nonhomogeneous systems loaded with large amounts of

inert metals and for systems that depend primarily upon the precipitation of a solid as the energy-releasing mechanism.

BKW ISENTROPIES

When the C-J temperature is high and the C-J pressure is low, as for BCHNOF and CHNOAL systems, much of the energy becomes available only after the products have expanded to fairly low pressures. It is this feature of the BKW results which makes the isentropes of interest.

Low pressure points on the isentropes of Systems 17 and 19 were measured by W. E. Deal and W. Garn using their techniques described in references 4 and 5. The experimental data are shown in Table III and are plotted in Figs. 1 and 2, along with the gamma law, BKW, and displaced BKW isentropes. The displaced BKW isentropes were computed using the experimental C-J pressure and the BKW Hugoniot temperature computed for that pressure as the starting condition for the computation of the displaced isentrope.

The experimental low pressure isentrope values reported by Deal⁵ for Octol, Cyclotol, and Composition B agree reasonably well with the constant gamma and with the BKW isentropes. The computed gamma law and BKW isentropes are significantly different for high energy, low particle density systems. This is because considerably more energy is available for such systems than gamma law computations would predict from the observed C-J pressure and detonation velocity. Thus the observed agreement between the BKW isentropes and the experimental values is

further evidence that the BCHNOF and CHNOAL systems have high heats of explosion and might perform well in applications which rely importantly on the equilibrium expansion of the detonation products.

CONCLUSIONS

The C-J performance of an explosive is apparently a very sensitive function of the C-J particle density. A decrease of the particle density results in an increase of the thermal energy at the expense of the intermolecular potential energy with a resultant higher C-J temperature and lower C-J pressure. The thermal energy is available only upon expansion of the detonation products to low pressures.

Table I

Covolumes Used in BKW Calculations

<u>Species</u>	<u>Covolume</u>	<u>Species</u>	<u>Covolume</u>
B ₂ O ₃	730	C	180
HBO ₂	1270	B ₂ O ₂	1740
BO	610	NO	386
B ₂	674	N	148
B	215	BN	619
BH	533	NH ₃	476
HF ₃	800	CH ₄	528
BF	685	Al ₂ O ₃	1350
F	108	Al ₂ O ₂	1800
F ₂	387	Al ₂ O	1300
HF	389	AlO	1160
CO ₂	670	AlH	948
CO	390	Al	350
H ₂ O	360	CF ₄	1330
OH	413	CF ₃	1330
H ₂	180	CF ₂	1330
O ₂	350	CHF ₃	1920
O	120	CH ₂ F ₂	1330
H	760	CH ₃ F	1920
N ₂	380	CF ₂ O	1330

Covolume = $10.46 V_1$ where V_1 has the dimensions A^3 and is the volume of the rotating molecule represented as a sphere whose radius is the maximum dimension of the molecule measured from its center of mass.

Table II

Experimental and BKW Calculated C-J Performance of Explosives

System No.	Empirical Formula of System	Composition	Solid (S) or Liquid (L)	ΔH_x^0 (kcal/mole EE)	Reference to ΔH_x^0	Density (grams/cc)	BKW Calculated				
							CJ Gas Volume (cc/mole gas)	Moles Gas/ Mole EE	CJ Solid Carbon Volume (cc/mole)	Moles Solid Carbon/ Mole EE	Gamma
CHNO SYSTEMS											
1	$C_3H_6O_6$	RDX	(S)	14.71	(1)	1.80	11.47	7.55	3.77	1.42	2.92
2	$C_{5.045}H_{7.461}O_{6.876}$	Cyclotol (6)	(S) mixture	9.5	(21)	1.755	11.95	9.26	3.87	2.91	2.87
3	$C_{6.851}H_{8.750}O_{7.650}$	Comp. B (7)	(S) mixture	4.9	(21)	1.715	12.43	10.78	3.97	4.20	2.92
4	$C_7H_5N_3O_6$	TNT	(S)	-17.81	(1)	1.640	13.98	5.87	4.21	5.10	2.97
5	$C_7H_5N_3O_6$	TNT	(L)	-12.95	(22)	1.447	15.54	6.04	4.46	4.91	2.81
6	CH_3NO_2	Nitromethane	(L)	-21.28	(2)	1.128	15.33	2.35	4.56	0.60	2.53
7	$C_{1.071}H_{1.3}N_{1.284}O_{2.568}$	HM/TM (8)	(L) solution	-20.66	(21)	1.197	15.28	2.85	4.50	0.32	2.54
8	$C_{1.25}H_{1.2}N_{1.4}O_4$	HM/TM (9)	(L) solution	-19.08	(21)	1.310	15.78	3.78	--	0	2.46
9	$C_{1.5}H_{1.3}N_{1.5}O_5$	HM/TM (10)	(L) solution	-16.88	(21)	1.397	15.38	4.76	--	0	2.52
10	$C_{7.29}H_{5.16}N_{10.32}$	C_6H_6 /TM (11)	(L) solution	+22.98	(21)	1.362	15.86	10.39	4.46	2.39	2.60
11	$C_3H_5N_3O_9$	Nitroglycerine	(L)	-84.6	(24)	1.590	14.22	7.26	--	0	2.61
12	$C_4H_8N_4O_8$	Dina	(L)	-70.0	(25)	1.430	13.96	8.24	4.26	1.68	2.68
13	$C_{4.25}H_{1.6}N_{6.10}$	AM/TM (12)	(L) solution	+55.0	(21)	1.380	17.05	9.01	--	--	2.46
CHNOF SYSTEMS											
14	$C_5H_7N_4O_6F_3$	TFPA (13)	(S)	-180	(23)	1.692	14.13	7.61	4.08	2.93	2.74
15	$C_2H_3N_2O_2F_3$	TFPA (14)	(L)	-162	(23)	1.523	17.58	3.62	4.43	0.97	2.55
16	$C_{3.7}H_{6.35}N_{5.3}O_{6.1}F_{1.05}$	RDX/TFPA (15)	(S) mixture	-53.4	(21)	1.754	12.42	7.60	3.91	1.95	2.88
CHNOAL SYSTEMS											
17	$C_{1.873}H_{2.469}N_{1.613}O_{2.039}Al_{0.7338}$	Alex 20 (16)	(S) mixture	-1.0	(21)	1.801	13.20	1.97	4.24	1.66	3.40
18	$C_{1.647}H_{2.093}N_{1.365}O_{1.744}Al_{1.142}$	Alex 32 (17)	(S) mixture	-1.1	(21)	1.880	14.58	1.30	4.52	1.64	3.33
BECHNOF SYSTEMS											
19	$H_{10}B_3C_5.75N_{15}O_{30}$	KDB/TM (18)	(L) solution	+40	(26)	1.40	16.51	27.07	--	--	2.42
20	$H_{10}B_3C_6.45N_{17.8}O_{35.6}$	KDB/TM (19)	(L) solution	+46	(26)	1.427	16.94	29.94	--	--	2.42
21	$H_{10}B_3C_{17}N_{30}O_{30}N_{15}$	KDB/TFMEX (20)	(L) solution	-1290	(26)	1.467	17.2	44.9	--	--	2.47

HKJ Calculated (Continued)

Experimental Data

Mole Fraction of Gaseous Products for Species Present More than 1×10^{-3}								CJ Pressure (mb)	CJ Temp. ($^{\circ}$ K)	Det. Vel. (cm/ μ sec)	CJ Pressure (mb)	Detonation Velocity (cm/ μ sec)	Brightness Temperature ($^{\circ}$ K)
H ₂ O	H ₂	O ₂	CO ₂	CO	NH ₃	N ₂							
.391	--	--	.195	.014	.005	.395		.338	2677	.8584	.341 \pm .005 (27)	.859 \pm .005 (27)	
.395	--	--	.211	.019	.005	.369		.311	2730	.8281	.313 \pm .005 (27)	.829 \pm .002 (27)	
.396	--	--	.220	.025	.006	.352		.284	2760	.8068	.292 \pm .005 (27)	.802 \pm .002 (27)	
.417	--	--	.281	.043	.005	.253		.225	2757	.7361	.190 \pm .005 (27)	.695 \pm .002 (27)	
.399	.002	--	.248	.098	.008	.244		.173	2960	.6747	.170 \pm .002 (28)	.658 \pm .001 (28)	3030 (33)
.596	.009	--	.086	.084	.023	.202		.151	2960	.6871	.130 \pm .001 (32)	.629 \pm .001 (32)	3380 (33)
.494	.007	--	.144	.120	.017	.217		.168	3200	.7045	.138 \pm .010 (29)	.657 \pm .010 (29)	3480 (33)
.395	--	.009	.312	.018	.003	.264		.195	3850	.7177	.156 \pm .010 (29)	.688 \pm .010 (29)	3750 (33)
.315	--	.054	.311	.004	--	.315		.200	3570	.7099	.168 \pm .010 (29)	.678 \pm .015 (34)	3580 (33)
.270	.005	--	.251	.221	.009	.244		.182	3700	.6948		.685 \pm .015 (34)	3520 (33)
.344	--	.035	.412	.002	--	.207		.256	3270	.7630		.758 \pm .010 (34)	3470 (33)
.470	.001	--	.219	.063	.009	.238		.217	2960	.7469			3480 (33)
H ₂ O	H ₂	O ₂	CO ₂	CO	NH ₃	N ₂	NO						
.165	--	.018	.415	.056	--	.323	.020	.191	4520	.6930	.156 \pm .010 (29)	.671 \pm .015 (34)	4000 (33)
HF	CO ₂	CO	H ₂ O	NH ₃	N ₂	CF ₄							
.026	.173	.007	.436	.007	.259	.092		.247	2300	.7387	.249 \pm .010 (31)	.740 \pm .005 (31)	
.068	.092	.003	.365	.010	.271	.190		.164	1950	.6176	.174 \pm .010 (31)	.665 \pm .005 (31)	
.016	.188	.012	.402	.005	.346	.030		.298	2530	.8120	.324 \pm .010 (31)	.822 \pm .005 (31)	
Solid Al ₂ O ₃ moles	H ₂ O	H ₂	CO ₂	CO	NH ₃	N ₂							
.3669	.354	.051	.012	.098	.148	.336		.251	4860	.7671	.230 \pm .005 (35)	.753 \pm .002 (35)	
.5708	.015	.271	--	.009	.343	.353		.211	6340	.6971	.215 \pm .005 (35)	.730 \pm .002 (35)	
Solid H ₂ O ₃ not present in equilibrium composition													
H ₂ O	H ₂	O ₂	CO ₂	CO	NH ₃	N ₂	O ₂						
.184	.0007	.292	.007	.047	.166	.028	.273	.192	5180	.6849	.172 \pm .015 (30)	.674 \pm .01 (30)	4460 (33)
.166	.0010	.293	.0004	.138	.077	.005	.297	.194	5240	.6825	.167 \pm .015 (30)	.682 \pm .01 (30)	
*.012	--	.119	--	.120	.259	.004	.167	.185	4500	.6620	.206 \pm .015 (30)	.691 \pm .01 (30)	

*Also 0.198 HF₃ and 0.071 HF

Table III

Experimental Isentrope States

<u>System No.</u>	<u>Composition</u>	<u>Material</u>	<u>Pressure (kbs)</u>	<u>Particle Velocity (cm/μsec)</u>
19	EDB/TNM	Polyethylene ($\rho_0 = 0.91$ g/cc)	119.1	0.216
19	EDB/TNM	Air	0.570	0.810
17	Alex 20	Air	0.682	0.715

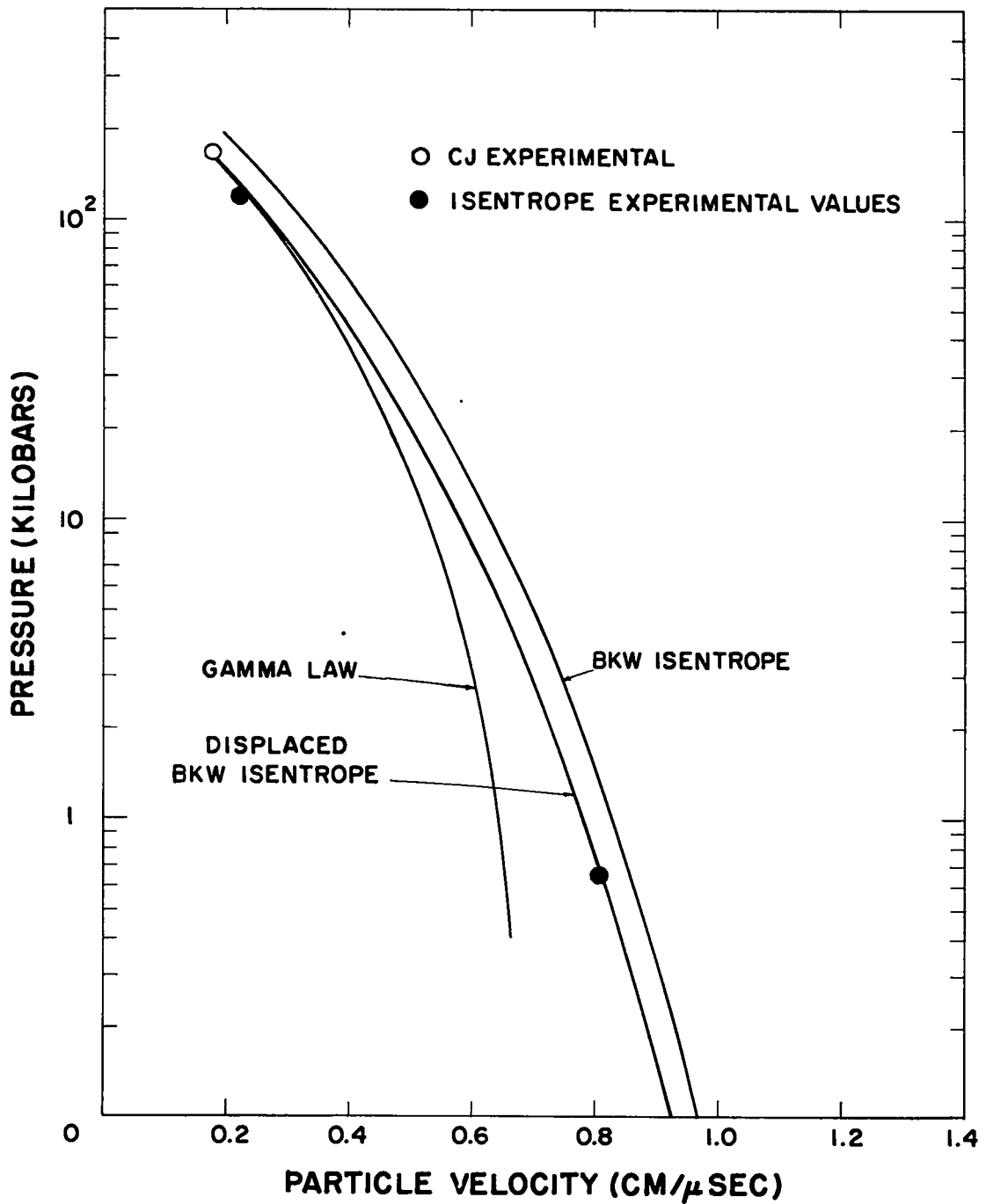


Figure 1. Semilogarithmic plot of BKW and gamma law computed isentropes and experimental values for a solution of 1 mole ethyldecaborane and 3.75 moles of tetranitromethane.

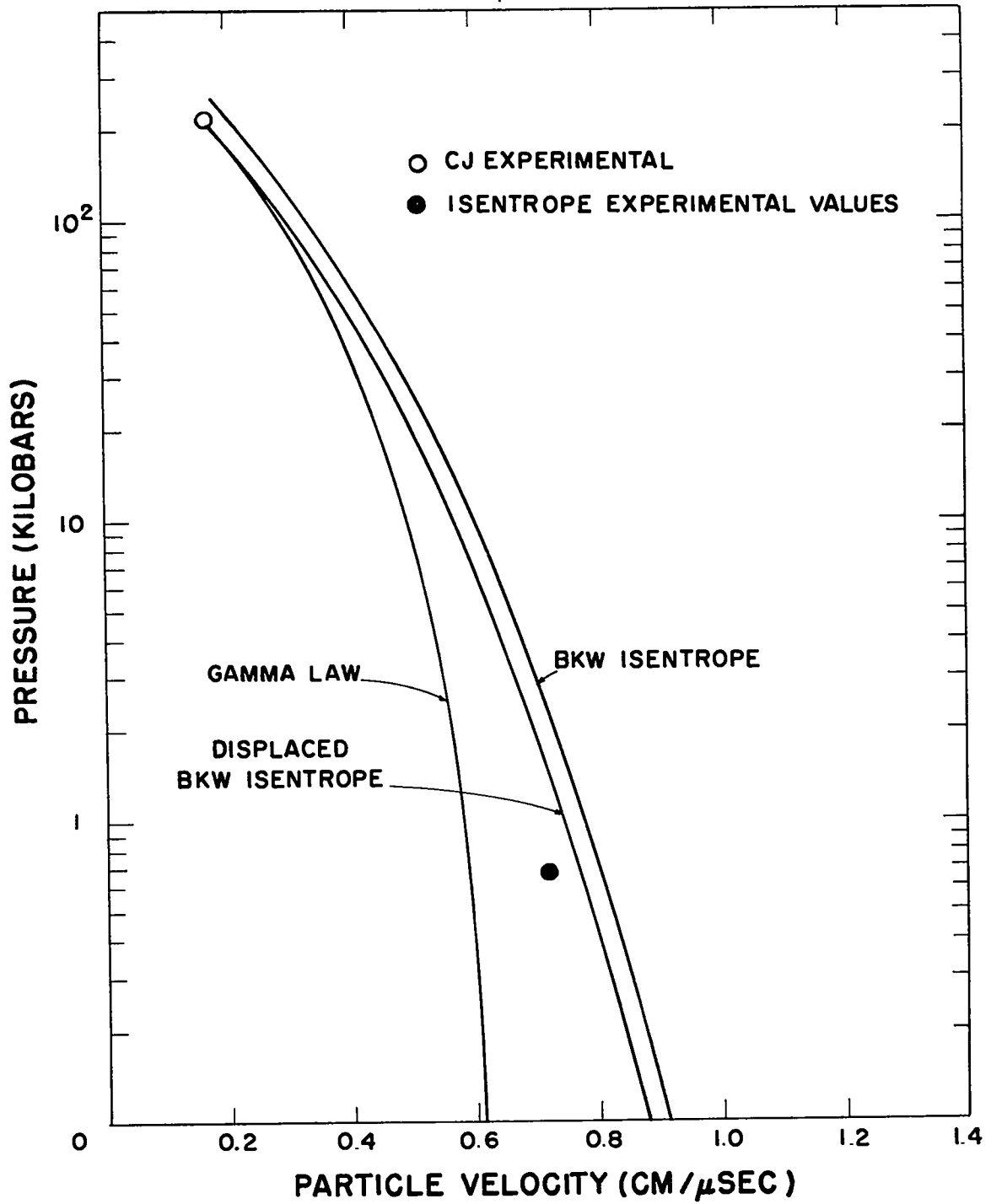


Figure 2. Semilogarithmic plot of BKW and gamma law computed isentropes and experimental values of Alex 20, a RDX/TNT/Al/wax mixture.

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4. W. E. Deal, Phys. of Fluids, 1, 523 (1958).
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6. Composition is 77/23 wt. % RDX/TNT.
7. Composition is 64/36 wt. % RDX/TNT.
8. Composition is 1 mole nitromethane to 0.071 mole tetranitromethane.
9. Composition is 1 mole nitromethane to 0.25 mole tetranitromethane.
10. Composition is 1 mole nitromethane to 0.50 mole tetranitromethane.
11. Composition is 1 mole benzene to 1.29 moles tetranitromethane.
12. Composition is 1 mole acrylonitrile to 1.25 moles tetranitromethane.
13. Composition is 1,1,1-trifluoro-3,5,5-trinitro-3-azahexane.
14. Composition is trifluoroethyl nitramine.
15. Composition is 65/35 wt. % RDX/TFNA.
16. Composition is 44/32.2/19.8/4.0 wt. % RDX/TNT/Al/wax.
17. Composition is 37.4/27.8/39.8/4.0 wt. % RDX/TNT/Al/wax.
18. Composition is 1 mole ethyldecaborane to 3.75 moles tetranitromethane.
19. Composition is 1 mole ethyldecaborane to 4.45 moles tetranitromethane.
20. Composition is 1 mole ethyldecaborane to 7.50 moles tetrafluorodinitroethane.

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