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MEMORANDUM REPORT ARBRL-MR-03373

INTERCOMPARISON BETWEEN FRG & US THERMODYNAMIC CODES

Eli Freedman

August 1984

US ARMY ARMAMENT RESEARCH AND DEVELOPMENT CENTER
BALLISTIC RESEARCH LABORATORY
ABERDEEN PROVING GROUND, MARYLAND

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A round robin (ringversuch) has been conducted among various FRG thermodynamic codes, between two US codes, and between one FRG and one US code. The agreement within each country is excellent. Comparison between the two countries is hampered by the correction for non-ideal gas behavior in the only available West German results. It is probable that they would show good agreement in the limit of ideal gas behavior. The US BLAKE code gives slightly smaller co-volumes than does the West German code; the larger values

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20. Abstract (cont'd):

are probably the better ones. The definition of heat capacity of hot reacting gases is not the same in all codes and requires some attention by ballisticians.

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I. INTRODUCTION

An important parameter used in gun interior ballistic computations is the propellant's impetus, the energy contained in the hot chamber gases produced by one gram of the propellant when it is burned to completion at a specified loading density (commonly 0.2 g/cc). The computation of the impetus for the general case is straightforward only in principle; in practice many complications arise.

The most obvious one is essentially numerical. In a typical case, the chamber gas contains some 50–60 species present, only a few, usually less than 10, have any appreciable concentration. As temperatures and pressure change, the identities of these 10 species also change. The concentrations of all of the species, the temperature, and the pressure are related to one another by complex non-linear equations. Overall, the laws of conservation of mass energy must be obeyed.

This problem is complicated enough for a mixture containing only gases; the presence of one or more condensed phases (e.g., potassium hydroxide, carbon, aluminum oxide) makes it even more complicated. The concentrations of all of the gaseous species, but not the condensed phases, obey the familiar "mass action" equations, and hence, the latter must be treated separately.

The general case was not attempted until the development of electronic computers made it feasible. The need for such computations became a large national enterprise; many computer codes were written for this purpose. Since rocket motors usually operate below 35 MPa, these codes all assumed the ideal gas law, pV=nRT.

Entirely different conditions are encountered in the detonation of condensed explosives. Here, the pressures exceed hundreds of kilobars. The type of gas law that must be used under these conditions is controversial, and is outside the scope of this paper.

The initial chamber conditions for guns typically have gas densities of 0.2 g/cc or higher; at these densities, ideal gas pressures are 10 to 20% too low, so a correction must be made. A way of doing this was first pointed out by Corner.[1] His method could and was implemented using mechanical desk calculators.

The first electronic computer program embodying Corner's ideas was written circa 1950 at the Ballistic Research Laboratories (now the Ballistic Research Laboratory), but it received little notice. A report[2] containing results computed with this program did, however, receive wide circulation.

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[1] *J. Corner, Theory of the Interior Ballistics of Guns, Wiley, New York (1950).*

[2] *P. G. Baer and K. R. Bryson, "Tables of Computed Thermodynamic Properties of Military Gun Propellants," BRL Memorandum Report 1338 (1961).* AD-258-644

More recently, the present writer took advantage of the modular design of the TIGER program, which permits an almost unlimited number of different gaseous equations of state to be inserted into it with relative ease, and programmed Corner's method for it. The resulting code, called BLAKE, is now used at several US military installations, including AMCCOM-Dover, AMCCOM BRL, and the Naval Surface Weapons Laboratories at Indian Head and Dahlgren.

Even more recently, Volk and Bartheldt[3] published a description of a code that accomplishes the same task. While there are of course some differences, it appears that their approach was qualitatively similar.

As a result of Dr. F. Volk's visit to the BRL in June 1977, it was suggested that a round robin be conducted between the various available codes. Dr. Volk agreed to compare the various FRG codes, and this writer undertook to do the same for the US codes.

II. OTHER US CODES

If one thermodynamics code is to be singled out and labeled the "best," surely it must be the NASA/Lewis code, CEC71, first written almost 30 years ago and its revised and extended successor, TRAN72.[4a4b] It offers trouble-free operation, a wide range of applications, and the JANNAF library of reaction products. The only drawback to this marvelous program is that it has the ideal gas equation of state inextricably woven into it.

The calculations presented for the NASA/Lewis program were performed by R. A. Fifer, Ignition and Combustion Branch, Interior Ballistics Division, BRL.

There are, of course, a large number of thermodynamics programs available, of which three have been widely applied to propellants. These are TIGER, PEPO, AND NOTS. There was nothing to be learned by using the original TIGER program for the present exercise, since this program is incorporated into BLAKE, as already noted. The revised version of TIGER was not available for this project.

PEPO (an acronym derived from Propellant Evaluation Program) has been extensively used in many US Navy laboratories. It is limited to ideal gases and cannot do constant volume calculations and so was not further considered.

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[3] *F. Volk and H. Bartheldt, Propellants and Explosives, Vol 1, 7- 14, (1976).*

[4] *(a) S. Gordon and F. Zeleznik, "A General IBM 704 or 7090 Computer Program for Computation of Chemical Equilibrium Compositions...." NASA TN D-1454 (October 1962).*

(b) R. Svehla and B. McBride, "Fortran IV Computer Program for Calculation of Thermodynamic and Transport Properties of Complex Chemical Systems," NASA TN D-7056. (January 1973).

NOTS (which was first written at the US Naval Ordnance Test Station, hence its name) is a descendant of PEPO. It, too, is limited to the ideal gas state, but it can do constant-volume calculations. NOTS is extensively used at the Naval Weapons Center at China Lake, California. Previous informal comparisons between it and BLAKE indicated that the two codes agreed well for the ideal-gas case. Efforts at including NOTS in the present study were unsuccessful owing to a high priority project at China Lake. The BLAKE code has been described elsewhere.[5]

III. INPUT

The well-known Encyclopedia of Propellants[6a] was used as the source for composition data on five different US military powders. Their compositions were normalized so as to add to 100 percent, including residual solvent and additives, if any. (Bear in mind that US military powder description sheets do not usually present component information directly as percentages of the final mix.) The values chosen are given in Table 1.

These compositions were chosen to satisfy several requirements. Obviously, they should be typical of modern gun propellants. Additionally, it is desirable that they cover a range of temperatures and energies. For double base propellants of the types used, energies and flame temperatures are usually closely correlated. If further testing of the codes is required, a nitramine composition for which this correlation does not hold should be included.

Compared to either NASA/Lewis or the NOTS codes, BLAKE runs more slowly with compositions for which solid products are formed at chamber conditions. Also, convergence problems arise in such cases. Composition M15 was picked to represent this class of propellants. The BLAKE calculation for it converged readily as soon as the list of possible gaseous products was trimmed to 29, the maximum permitted at any one time.

Dr. Volk and this writer had earlier agreed that the BRL would furnish all of the heats of formation for the initial ingredients. These values are given in Table 2. Both groups rely strongly on the various standard compilations of thermochemical data, especially the books by Cox and Pilcher,[6b] and by Stull, Westrum, and Sinke,[7] so it is unlikely that there would have been serious disagreements on any ingredient other than

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[5] *E. Freedman, "BLAKE - A Thermodynamics Code Based on TIGER: Users' Guide and Manual," ARBRL-TR-2411 (July 1982). ADA-121-259.*

[6a] *Basil T. Federoff and Oliver E. Sheffield, Encyclopedia of Explosives and Related Items, PATR 2700, Vol 2, pp t-34,35 (1962).*

[6b] *J. D. Cox and G. Pilcher, Thermochemistry of Organic and Organo-metallic Compounds, Academic Press, London (1970).*

[7] *D. R. Stull, E. F. Westrum, Jr., and G. C. Sinke, The Chemical Thermodynamics of Organic Compounds, Wiley, New York (1969).*

TABLE 1. SOME TYPICAL US MILITARY POWDERS

DESIGNATION	M1A1	M15	T29	M9	M18
Ingredient Abbreviation					
NC	83.173	19.940	45.954	58.330	79.600
(% N in NC)	(12.60)	(13.15)	(13.15)	(13.25)	(13.15)
NG	--	18.943	21.478	40.400	9.950
NGU	--	54.536	30.669	--	--
DNT	9.843	--	--	--	--
DBT	4.429	--	--	--	8.955
KCRY	--	0.299	--	--	--
DPA	0.984	--	--	--	0.995
EC	--	5.982	1.50	0.760	--
ET	0.98	0.300	0.30	0.510	0.500
H_2O	0.59	0.0	0.0	0.0	0.0
C	--	--	0.100	--	--

All ingredients are assumed to be distributed homogeneously for the purposes of computation.

TABLE 2. NAMES, FORMULAS, AND HEATS OF FORMATION

ABBREVIATIONS	COMMONLY-USED NAME	FORMULA	Δh°_f (kcal/mole)
NC	nitrocellulose	(see note)	(see note)
NG	nitroglycerin	$C_3H_5O_9N_3$	- 88.6
NGU	nitroguanidine	$CH_4O_2N_4$	- 22.1
DNT	dinitrotoluene	$C_7H_6O_4N_2$	- 17.1
DBT	dibutylphthalate	$C_{16}H_{22}O_4$	-208.07
KCRY	potassium cryolite	$K_3A\ell F_6$	-744
DPA	diphenylamine	$C_{12}H_{11}N$	31.07
EC	ethyl centralite	$C_{17}H_{20}ON_2$	- 25.1
ET	ethanol	C_2H_6O	- 66.42
H_2O	water (liquid)	H_2O	- 68.315
C	graphite	C	0.0

Note: For nitrocellulose containing N % nitrogen, the formula is

$$C_6H_{10-x}O_{5+2x}N_x$$

where $x = \frac{1.62143\ N}{14.0067-0.44998\ N}$;

$\Delta h^\circ_f = -1406.22 + 62.3186\ N$ cal/g;

and molecular weight = $162.143 + 44.998\ x$

nitrocellulose. For this substance, the BRL relies on the values published by Jessup and Prosen,[8] while ICT relies on Medard.[9]

IV. THE WEST GERMAN ROUND ROBIN

Details of the West German Round Robin will be handled by Dr. Volk,[10] who may publish the results separately.

V. THE BLAKE NASA/LEWIS COMPARISON

This comparison was made on the basis of ideal gas behavior only, for the reason noted above. The results are shown in Table 3. They are obviously in excellent agreement. An additional indication of the close agreement occurs with M15, where both codes show the formation of liquid Al_2O_3. BLAKE gives the mole fraction of this product as 0.000124; NASA/Lewis, 0.0001236.

The only significant difference between the two sets of computations occurs with the ratio of heat capacities, gamma, and the heat capacity at constant pressure, C_p. This will be discussed in detail below.

VI. COMPARISON BETWEEN BLAKE AND THE ICT CODE

Unfortunately, a comparison between the two codes for the ideal gas case cannot be made, since Dr. Volk[11] sent results only for the real gas case.

The results for the two codes are shown in Table 4.

The agreement between the calculated temperatures is such that it appears probable that the two codes would agree well for the ideal gas case; still, it would be desirable to make the comparison under those conditions.

Overall, the agreement is good. At higher loading densities, the BLAKE results are lower than the ICT results (or the latter are higher). The former case is the more likely, since BLAKE computes the third virial coefficient on the assumption of hard spheres with a radius 0.81 times the Lennard-Jones radius, as suggested by Hirschfelder.[12] The ICT code calculated both second and third virial coefficients from either the Stockmayer or the Lennard-Jones potentials, depending on whether the molecule is polar or not.

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[8] *R. S. Jessup and E. J. Prosen, J. Research NBS 44, 387 (1950).*

[9] *L. Medard, Mem. Artillerie Francaise (1954).*

[10] *P. Volk, private communication to E. Freedman, dated 22 December 1977.*

[11] *F. Volk, private communication to E. Freedman, dated 19 January 1978.*

[12] *J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, Molecular Theory of Fluids, John Wiley, New York, p 157 (1954).*

TABLE 3. COMPARISON OF BLAKE WITH NASA/LEWIS

IDEAL GAS						
LOADING DENSITY (g/cc^3)	BLAKE		NASA-Lewis		% Differences	
	T (K)	P (10^5 Pa)	T (K)	P (10^5 Pa)	T	P
		M1A1				
0.2	2266.	1736.9	2265.	1735.6	0.04	0.07
0.4	2285.	3479.2	--	--	--	--
0.5	2295.	4352.7	2295.	4348.2	0.0	0.10
0.6	2306.	5227.7	--	--	--	--
		M15				
0.2	2559.	1975.3	2558.	1974.1	0.04	0.06
0.4	2563.	3948.3	--	--	--	--
0.5	2565.	4933.9	2565.	4929.9	0.0	0.08
0.6	2567.	5919.0	--	--	--	--
		T29				
0.2	3221.	2218.4	3220.	2216.9	0.03	0.07
0.4	3230.	4444.7	--	--	--	--
0.5	3232.	5558.2	3230.	5552.8	0.06	0.10
0.6	3234.	6671.6	--	--	--	--
		M9				
0.2	3815.	2356.5	3813.	2355.0	0.05	0.06
0.4	3865.	4756.8	--	--	--	--
0.5	3880.	5961.5	3877.	5955.5	0.08	0.10
0.6	3891.	7168.1	--	--	--	--
		M18				
0.2	2602.	1925.7	2600.	1923.8	0.08	0.10
0.4	2604.	2888.4	--	--	--	--
0.5	2608.	4812.8	2606.	4806.3	0.08	0.14
0.6	2610.	7697.8	--	--	--	--

TABLE 4. COMPARISON OF 'BLAKE' WITH THE ICT CODE

Loading Density (g/cm^3)	BLAKE T (K)	BLAKE P (MPa)	BLAKE Co-Vol (cm^3/g)	ICT T (K)	ICT P (MPa)	ICT Co-Vol (cm^3/g)	% Differences T	% Differences P	% Differences Co-Vol
				REAL GAS					
				M1A1					
0.2	2279.	224.55	1.123	2288.	227.50	1.163	-0.39	-1.31	-3.56
0.4	2342.	584.71	1.002	2348.	600.89	1.044	-0.26	-2.77	-4.19
0.6	2421.	1131.75	0.889	2402.	1153.73	0.914	0.78	-1.94	-2.81
				M15					
0.2	2564.	254.82	1.121	2573.	254.94	1.110	-0.35	-0.05	0.98
0.4	2581.	659.12	1.002	2590.	664.42	1.012	-0.35	-0.80	-1.00
0.6	2614.	1263.16	0.887	2613.	1266.79	0.895	0.04	-0.29	-0.90
				T29					
0.2	3229.	279.64	1.026	3238.	279.05	1.006	-0.28	0.21	1.95
0.4	3243.	709.61	0.930	3251.	713.65	0.935	-0.25	-0.57	-0.54
0.6	3253.	1339.41	0.834	3247.	1355.38	0.846	0.18	-1.19	-1.44
				M9					
0.2	3843.	292.47	0.949	3852.	294.10	0.962	-0.23	-0.56	-1.37
0.4	3911.	734.83	0.867	3928.	750.86	0.898	-0.43	-2.18	-3.58
0.6	3950.	1371.32	0.786	3948.	1424.05	0.819	0.05	-3.85	-4.20
				M18					
0.2	2608.	246.35	1.086	2612.	248.38	1.113	-0.15	-0.82	-2.49
0.4	2626.	632.59	0.975	2624.	648.01	1.015	0.08	-2.44	-4.10
0.6	2663.	1206.62	0.867	2641.	1239.66	0.898	0.83	-2.74	-3.58

TABLE 5. HEAT CAPACITIES AND GAMMAS CALCULATED BY THREE DIFFERENT CODES

	NASA/Lewis (Equi)	NASA/Lewis (Frozen)	BLAKE	ICT
	M1A1 at 0.2 g/cm^3			
Cp Joule/mole	41.84	39.57	40.17	41.09
γ	1.2602	-----	1.2614	1.2691
	M9 at 0.2 g/cm^3			
Cp Joule/mole	73.26	48.41	49.10	49.39
γ	1.1687	-----	1.2098	1.2120

A mixture of reacting gases has two heat capacities. The first of these is merely the mole-fraction-weighted sum of the individual heat capacities of the components of the mixture. The other heat capacity consists of the frozen value plus an additional term that arises from the contribution of the shifting equilibrium. This additional term is roughly proportional to the square of the enthalpy of reaction. The matter has been discussed by a number of authors; see, for example, Hirschfelder et al.[12] For an application to propellants, see Fifer and Cohen.[13]

The "transport" option of the modified NASA/Lewis program[4b] computes both heat capacities. The results for a cool propellant (M1A1) and a hot one (M9) are shown in Table 5.

The agreement among the various heat capacities and hence the gammas for M1A1 is good for all of the codes. With M9, however, only the frozen heat capacities agree; the equilibrium value is about 50% larger than the frozen one.

The heat capacity itself is not an extremely important quantity in interior ballistics, but gamma is. As C_p increases, C_v also increases, so gamma is not a trivial matter. Apparently, there are enough undetermined factors remaining in the treatments of interior ballistics that this effect is not yet significant. Nevertheless, it deserves explicit treatment.

VII. CONCLUSIONS

THE NASA/Lewis code and the BLAKE code agree virtually perfectly on the calculation of temperature, pressure, and related quantities (except heat capacity and gamma) for hot propellant gases considered to be ideal gases.

The BLAKE code and the ICT code are in very good, but not perfect agreement.

The area of disagreement between BLAKE and the ICT code is very likely the differences in the correction for non-ideal gas effects.

It is not obvious yet that these differences are great enough to require further work.

The use of frozen heat capacities (and hence gammas) in gun codes should be closely examined.

---

[13] *R. A. Fifer and A. Cohen, "High Temperature Thermal Conductivity and Heat Transfer in the Gun Propellant and Primer Combustion Gases," Proceedings of the 14th JANNAF Combustion Meeting CPIA Publication 292, December 1977, Volume II, p 363.*

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