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HEATS OF EXPLOSION AT LOW PRESSURES
II.

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I. INTRODUCTION

The work presented in this report is a continuation of a former program¹ in which the pressure dependence of heats of explosion (HEX) for RDX (secondary explosive), XM39 (RDX composite propellant) and M30 (triple base propellant) were determined. The measurements were made in calorimeter bombs which were prepressurized to various levels with inert gas (He or N₂). Low loading densities (.0002-.012 g/cc) were used in order to minimize the transient pressure increase during combustion.

The program was started to provide information which could help improve interior ballistic (IB) code predictions of gun performance at low pressures. The discrepancies between measurements and predictions of ignition delays and pressurization rate in guns and gun simulators was discussed at a recent JANNAF workshop.² It is generally believed that the discrepancies at low pressures are due to two assumptions: (1) infinite-rate kinetics (to calculate the rate of energy production) and (2) thermodynamic chemical equilibrium (to calculate the thermal energy (E) of the propellant combustion products). The validity of these assumptions becomes questionable as pressure decreases. The absence of thermochemical equilibrium among combustion products under low pressure conditions is not unexpected. It is consistent with the HEX measurements at low pressures¹ and with strand burner observations of two stage propellant flames^{1,3} where, below a critical pressure, the second stage (and thermochemical equilibrium) is inhibited. The use of finite rate kinetics will, in principle, eliminate the need for both these assumptions but acquiring the necessary information for incorporation into the codes will take time. The objective of the program was to evaluate an alternate method for estimating values of E which is independent of the thermochemical equilibrium assumption and can be incorporated into the IB codes relatively quickly. This alternate method is to substitute for the value of E at a given gun pressure the HEX value measured at the same pressure in calorimeter bombs. This implicitly assumes that combustion and cooling processes in bombs and guns are similar. If this assumption is valid then the HEX measurements at various pressurization levels are expected to be better estimates of E than the calculated thermochemical equilibrium values that are now being used in IB codes. There is the additional requirement (for use in the IB codes) that these processes depend primarily on total pressure (i.e., are not strongly dependent on gas composition).

The earlier work¹ showed that as initial (inert) pressures (P₀) and loading densities (LD) decreased there was a fall off in measured HEX and ignition became more difficult. The goal of this work is to extend the HEX measurements to lower LD in order to minimize uncertainties in the pressurization level in the calorimeter. At low LD the ignition energy can be comparable to the combustion energy and must be subtracted from the (total) thermal energy of the calorimeter to obtain HEX values.

II. EXPERIMENTAL TECHNIQUE

The experimental technique has been described previously.¹ HEX were measured with a Parr Calorimeter System. The bombs (volume = 340 cc) were modified to provide for transient pressure measurements and sampling of combustion products. The data

reported here are for perforated propellant grains except for a few XM39 data which were taken with propellant slabs (solid).

It was possible to obtain data for M30 at lower LD (and Po) by using larger diameter (.016" vs .006" Ni) ignition wire and increasing the wire area in contact with the propellant, i.e., increasing ignition power and energy to the propellant. These data are denoted in Table 1 as enhanced ignition. For XM39, it was also necessary to place Zirconia Felt (Zicar Products), a thermal insulator, at the bottom of the capsule which supports the grains in the calorimeter during ignition and flamespreading (i.e., decrease the the heat transferred from the propellant) in order to obtain data (denoted in Table 2 as thermal insulation) at lower LD.

Po (psia)	Gas	m (g)	<HEX> (cal/g)	Nh	Sh (cal/g)	<p*> (psi)	Np	Sp (psi)
1015	He	1	963	2	9	476	2	196
1015	N ₂	1	977	2	9	329	2	49
1015	He	.28	943	5	7	131	5	55
1015	N ₂	.28	945	2	10	108	2	5
1015	He	.16	893	2	46	103	2	5
1015	He	.08	838	4	32	60	2	1
465	He	1	958	2	20	628	2	52
465	He	.28	956	3	19	89	3	43
165	He	1	943	4	8	352	2	81
165	N ₂	1	913	2	4	301	1	--
165	He	.28	923	3	6	57	3	28
165	N ₂	.28	870	1	--	93	1	--
65	He	1	927	3	11	200	2	56
65	He	.28	864	2	32	--	0	--
65	N ₂	.28	777	2	12	61	2	1
45	He	1	918	5	17	171	4	39
45	He	.28	818	4	11	34	4	13
25	He	1	911	3	34	190	1	--
25	N ₂	1	928	2	13	--	0	--
25*	He	.28	693	6	81	17	5	9
25*	N ₂	.28	723	2	29	25	2	18
25*	He	.16	825	2	119	12	2	1
25*	He	.08	587	2	176	13	2	1

* = Enhanced ignition.

TABLE 2. XM39

Po (psia)	Gas	m (g)	<HEX> (cal/g)	Nh	Sh (cal/g)	<P*> (psi)	Np	Sp (psi)
1045 ⁺	He	4.4	829	1	--	---	0	--
940	He	2.2	830	2	0	(600)	1	--
940	N ₂	2.2	835	1	--	1072	1	--
940	He	.52	810	3	12	272	1	--
940	N ₂	.52	802	2	5	216	1	--
940	He	.24	794	4	23	123	4	8
940	He	.13	731	2	4	80	2	10
940	He	.07	661	2	90	38	2	12
795 ⁺	He	2.2	822	1	--	---	0	--
545 ⁺	He	4.4	833	1	--	---	0	--
465	He	2.2	825	2	3	(600)	1	--
465	N ₂	2.2	823	1	--	880	1	--
465	He	.52	810	2	7	182	2	4
465	N ₂	.52	798	2	27	163	2	4
295 ⁺	He	4.4	830	1	--	---	0	--
245 ⁺	He	4.4	832	1	--	---	0	--
245 ⁺	He	2.2	807	1	--	---	0	--
240	He	2.2	825	2	4	452	1	--
240	N ₂	2.2	819	2	2	594	2	24
240	He	.52	769	2	36	123	2	4
240	N ₂	.52	759	2	11	128	2	7
165	He	2.2	812	2	14	312	2	78
165	N ₂	2.2	804	1	--	480	1	--
165	He	.52	739	5	51	71	5	15
165	N ₂	.52	712	2	14	86	2	7
45	He	2.2	818	2	25	108	1	--
45	He	.52	711	2	29	29	2	1
45\$*	He	.24	578	3	82	7	2	1
45\$*	He	.14	581	3	57	7	2	1
45\$*	He	.07	529	2	83	7	2	4

+ = Slab

\$ = Thermal insulation

* = Enhanced ignition

The ignition energy was obtained by integrating (over time) the product of the instantaneous ignition voltage and current. The Parr Calorimeter System uses a transformer to provide a 60 Hz ignition voltage. This was measured. The ignition current was measured using a Pearson current monitor (Model #411) (Pearson Electronics, Inc.).

III. EXPERIMENTAL RESULTS

Figure 1 shows the instantaneous ignition voltage, ignition current and calculated ignition energy using .016" Ni ignition wire for an experiment with 1.03 g M30 propellant in N_2 , $P_o = 465$ psia. The ignition energy is 18.3 cal. which is about 2% of the total heat measured in the calorimeter. Figure 2 shows the corresponding pressure increment which is obtained from the piezoelectric transducer signal. The pressure measurement is subject to error due to heating of the piezoelectric transducer during the experiment. Efforts were made to minimize this heating by shielding with vacuum grease but were not entirely successful. At present, it has been assumed that the maximum pressure increments (P^*) are not greatly affected by changes in heating due to differences in LD.

The data reported earlier¹ did not include corrections for ignition energy. These corrections have been made for the data presented in Tables 1 and 2. Table 1 summarizes the HEX and P^* measurements for M30. m is the sample mass ($\pm 10\%$), $\langle \rangle$ signifies the mean of the measurements, N_h and N_p are the number of HEX and P^* measurements, respectively, S_h and S_p are the corresponding standard deviations. The corresponding information for XM39 is given in Table 2.

Various plots of the data in Tables 1 and 2 are shown in Figures 3 through 8. Figure 3 is a plot of HEX vs. P_o for M30 which shows the effects of changes in mass and ambient gas on the measurements. At $P_o \geq 465$ psia, for both $m = 1$ g and $m = .28$ g, HEX values are relatively constant (~ 950 cal/g) and (at $P_o = 1015$) equal in He and N_2 . At $P_o \leq 165$ psia, for $m = 1$ g in He, there is some indication of a fall off in HEX values. At $P_o \leq 165$ psia and $m = .28$ g, the fall off is obvious for both gases and HEX appear to be smaller in N_2 than in He (data at $P_o = 25$ psia have large scatter).

Figure 4 is the corresponding plot for XM39. HEX appear to be equal for grains (perforated) and slabs (solid) and slightly less in N_2 than in He. At $P_o \geq 465$ psia, the HEX values for $m \geq 2.2$ g are relatively constant (~ 820 cal/g) and those for $m = .52$ g are slightly smaller but also appear to be constant (~ 800 cal/g). At $P_o \leq 240$ psia there is little change in HEX with $m \geq 2.2$ g but there is an obvious fall off in HEX for $m = .52$ g (in both He and N_2).

Figure 5 is a plot of HEX vs. m for M30. This figure shows the fall off in HEX with m (or LD) at two different initial pressures. The fall off for $P_o = 1015$ psia is noticeable at $m < .28$ g. At $m = .08$ g, HEX = 838 cal/g and the corresponding operating pressure range ($P_o \rightarrow (P_o + P^*)$) for values of P_o and P^* listed in Table 1) is 1015-1075 psia. For $P_o = 25$ psia the fall off in HEX with m is noticeable at $m = 1$ g. The fall off appears to be interrupted between $m = .08$ g and $m = .28$ g, data taken under "enhanced ignition" conditions. These data have large scatter and the change in behavior may not be real.

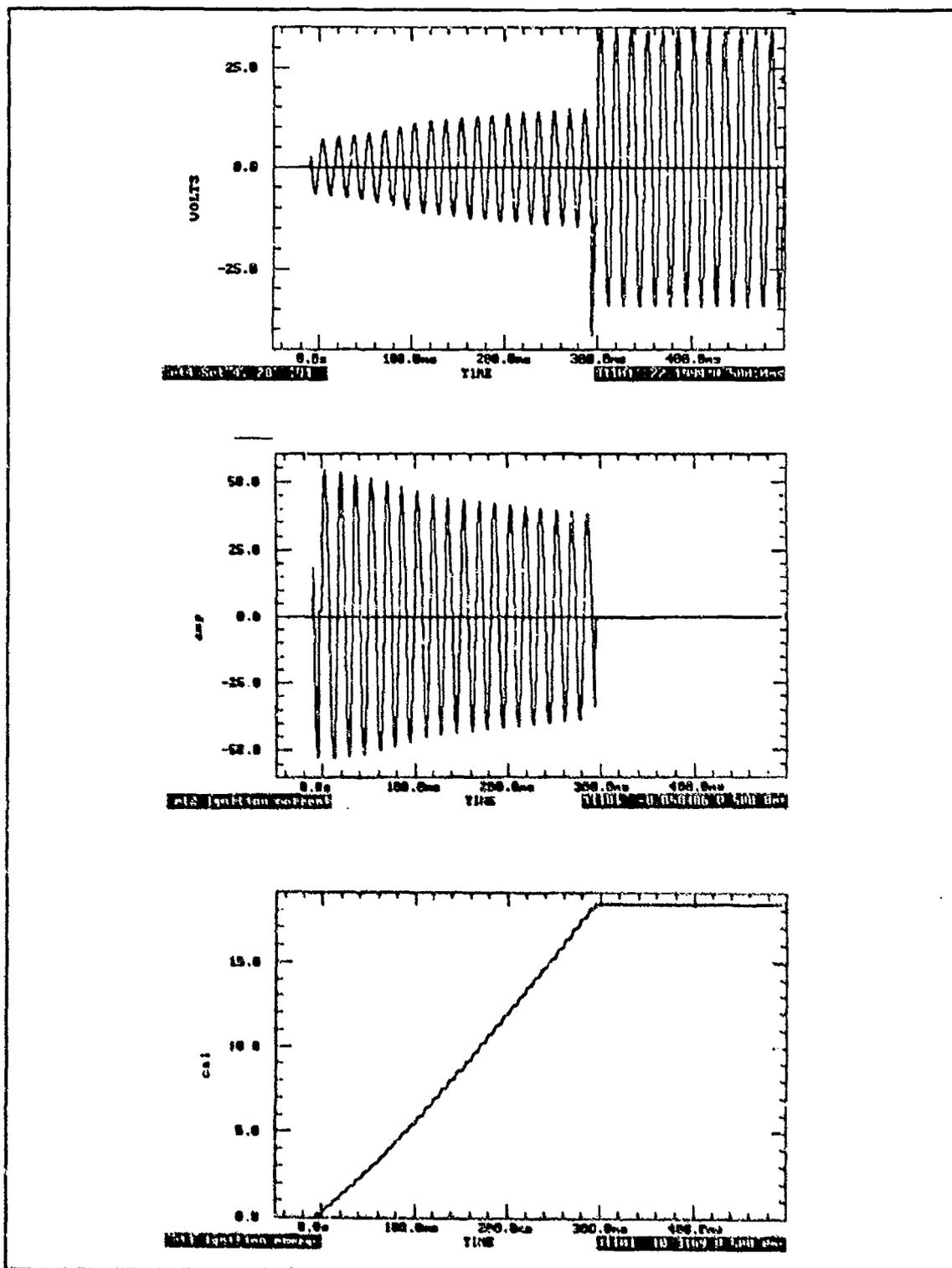


Figure 1. Ignition Characteristics of Parr Calorimeter System with .016" Ni Fuse Wire .
 (a) Ignition voltage (60 Hz): increase in voltage at Time = 294 ms is due to cessation of current flow. (b) Ignition current (60 Hz). (c) Ignition energy: calculated from integration (overtime) of the product of instantaneous voltage and current shown in (a) and (b).

At $m = .08$ g, HEX = 587 cal/g and the corresponding operating pressure range is 25-38 psia.

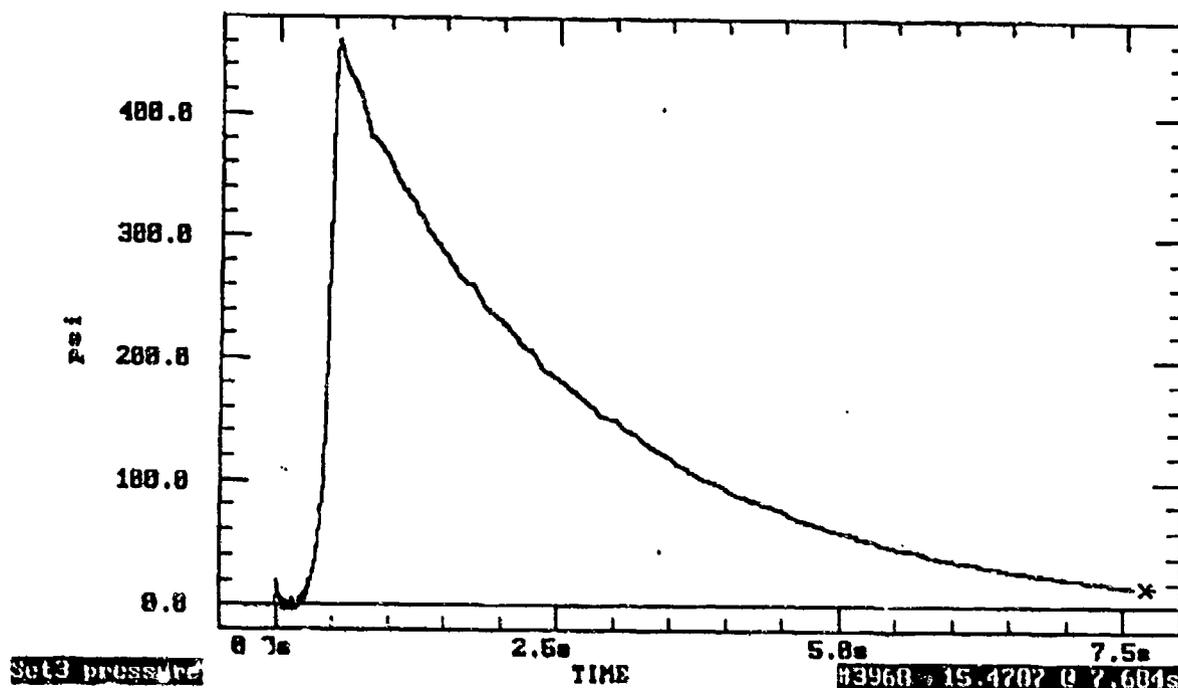


Figure 2. Piezoelectric Transducer Signal: Transient Pressure During Combustion and Cooling in the Calorimeter. Signal was processed by a 30 Hz electronic filter to reduce noise during ignition.

Figure 6 is the corresponding plot (to Fig. 3) for XM39 data. The fall off in HEX with m for $P_0 = 940$ psia is noticeable at $m < .52$ g. At $m = .07$ g, HEX = 661 cal/g and the corresponding operating pressure range is 940-978 psia. For $P_0 = 45$ psia, the fall off starts at $m < 2.2$ g. At $m = .07$ g, HEX = 529 cal/g and the corresponding operating pressure range is 45-52 psia.

The fall off in HEX with m at fixed P_0 and especially for small values of m suggest that HEX are not solely dependent on the operating pressure level in the calorimeter. This will make it difficult to use measured HEX for estimating values for E.

The data in Figure 7 demonstrates that HEX for M30 is not solely dependent on pressure levels in the bomb during burning, but can depend on loading density. The data for .28 and 1.0 g (light points) indicate that HEX increase slightly with maximum total pressure (initial pressure + measured maximum pressure increment) but are not greatly dependent on mass. The data for .08 and .16 g (dark points) have initial pressures (i.e., minimum bomb pressures) = 1030 psia. HEX for these data are lower than HEX for the data obtained at higher loading densities (seven points) although five of the latter data have maximum bomb pressures < 600 psia. For these experiments, it appears that HEX depend more strongly on loading density (i.e., the concentration of combustion products in the bomb) than on pressure levels.

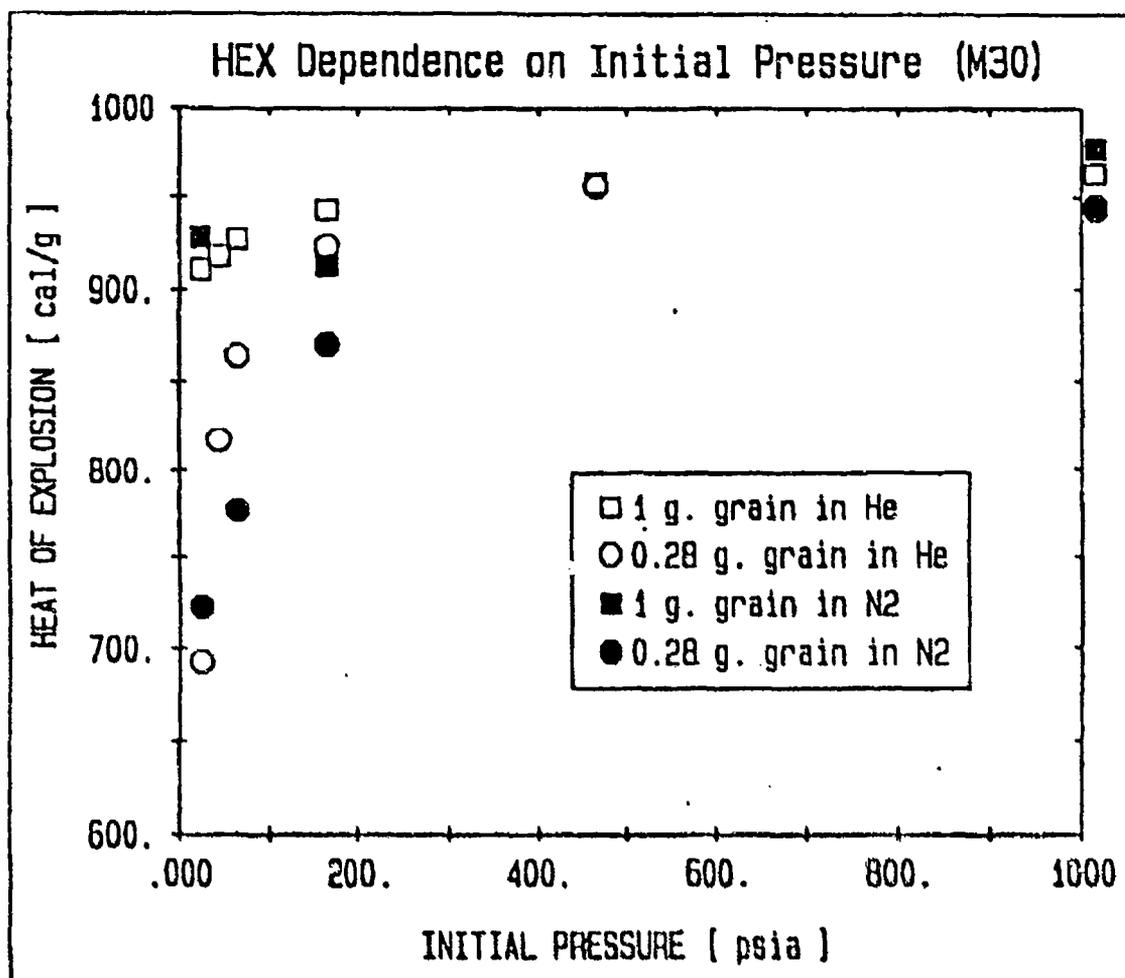


Figure 3. Dependence of Experimental Heats of Explosion on Initial Pressure for M30

Figure 8 is the corresponding plot (to Figure 7) for XM39 data. All data with total maximum pressures > 800 psia have $P_o = 940$ psia. The other data have maximum total pressure < 800 psia. Despite lower operating pressures in the calorimeter, HEX values for $m = 2.2$ g with maximum total pressures < 700 psia appear to be greater than those those for $m = .07, .13,$ and $.24$ g. This stronger dependence on loading density than on pressure is similar to the M30 behavior shown in Figure 7.

IV. DISCUSSION OF RESULTS

The data in Figure 3 for 1 g M30 suggests that HEX are not greatly dependent on P_o for $P_o \geq 25$ psia and on pressurizing gas composition. The differences between measured HEX (911-977 cal/g) and those calculated (-955 cal/g) assuming a (generally accepted) freeze-out temperature greater than 1500 K is not great. This suggests that for $P_o > 25$ psia combustion in the calorimeter is almost complete (i.e., combustion products correspond to a thermochemical equilibrium mixture) for 1 g samples. Comparison of

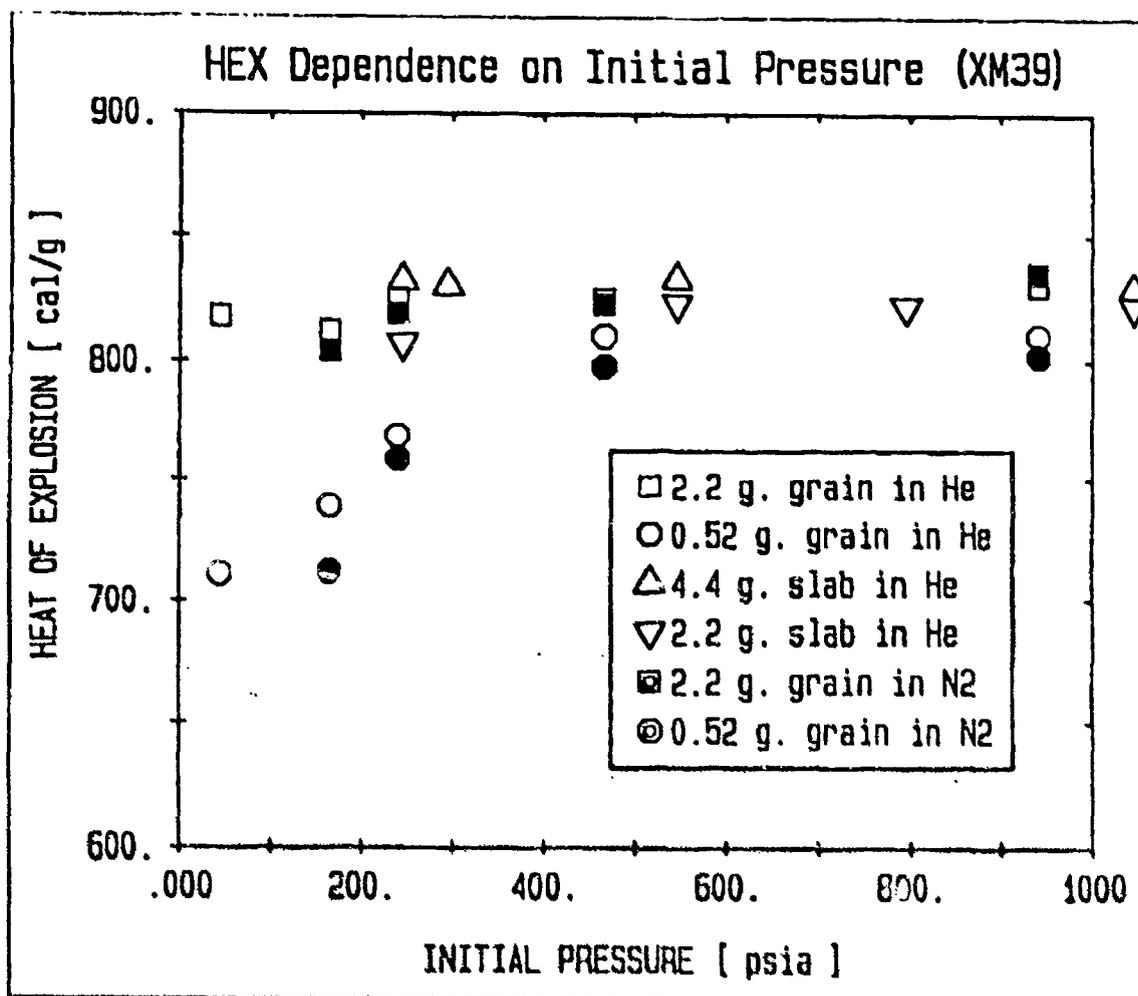


Figure 4. Dependence of Experimental Heats of Explosion on Initial Pressure for XM39

measured and calculated HEX values for .28 g M30, at $P_o = 465$ and 1015 psia also indicate that combustion is almost complete. At lower P_o (<465 psia) there is a fall off in HEX from the calculated values (the calculated values are not greatly dependent on P_o or LD) for the .28 g samples. This fall off suggests that changes occur in the combustion mechanism (or extent of reaction) at low pressures which inhibit the establishment of thermochemical equilibrium among combustion products.

The data in Figure 4 for 2.2 g and 4.4 g XM39 indicate that differences between measured HEX (800-835 cal/g) and calculated values (~810 cal/g) are small suggesting that combustion is complete. Comparisons of the HEX values (798-810 cal/g) for .52 g XM39 with calculated values indicate that combustion is complete at $P_o > 465$ psia. At lower P_o there is fall off in HEX values from calculated values, similar to the behavior with M30, indicative of changes in the combustion mechanism at low pressures and deviations of combustion products from thermochemical equilibrium.

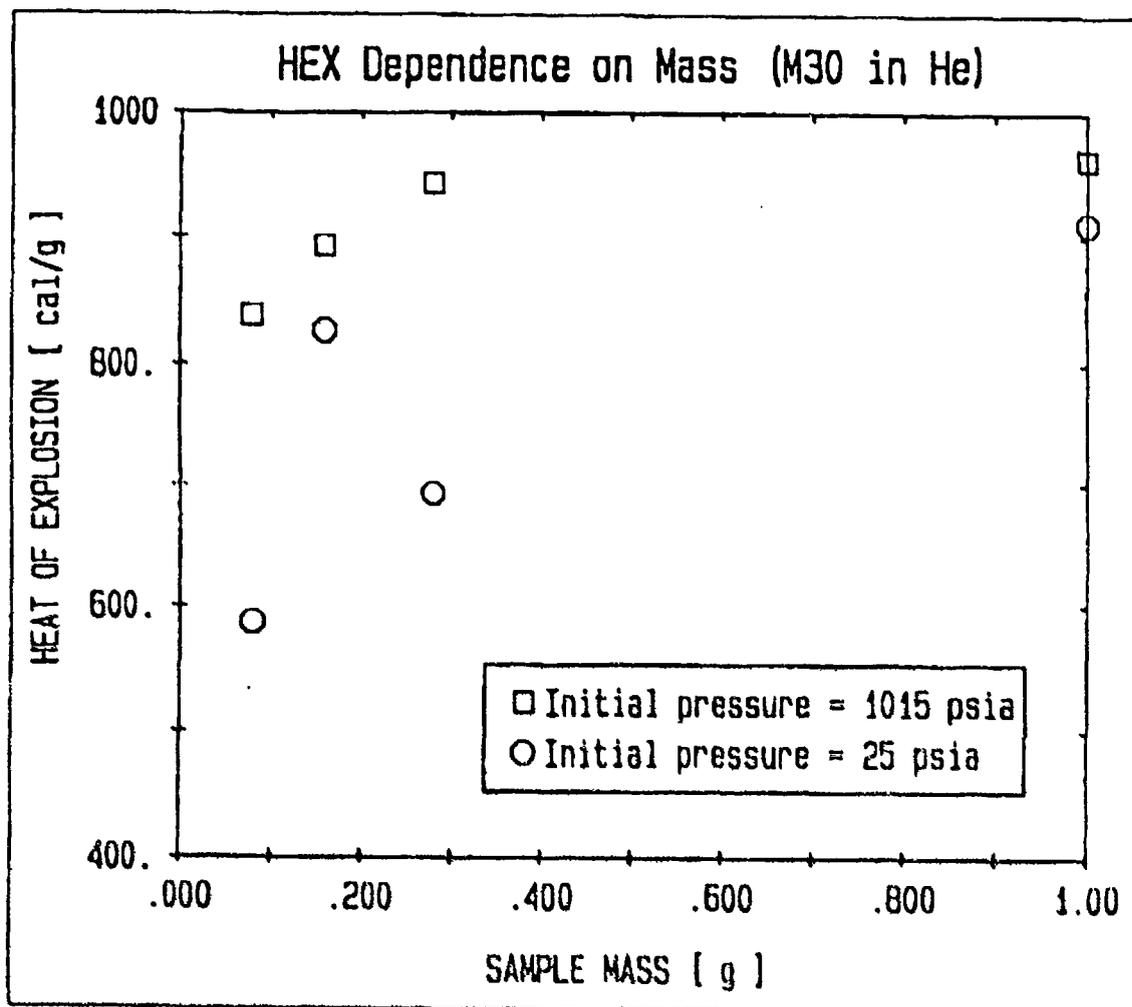


Figure 8. Dependence of Experimental Heats of Explosion on Loading Density for M30

The HEX calculations and reasons for the HEX dependence on initial pressures and loading densities has been discussed previously.¹ The transition at lower P_o and LD from complete to incomplete combustion is attributed to a decrease in chemical kinetic rates and the subsequent increased importance of transport rates (diffusion and heat transfer) in the combustion process. This increased dependence on transport processes under these conditions is thought to be responsible for the increase in variability of the data at low P_o and low LD which can be seen from the values of the standard deviations (s) in Tables 1 and 2. The small decrease in HEX when N_2 is substituted for He at low P_o and low LD may be due to differences in transport properties of the gases.

The thermochemical properties of the propellant combustion products presently used in IB codes are those calculated for closed bomb combustion products in thermochemical equilibrium at loading densities between .2 - .33 g/cc. Blake⁴ calculations for M30 under these conditions give values for E (which do not greatly depend on LD) near 1025 cal/g. If the alternate method for estimating E which is now under consideration (i.e., for a given pressure measured HEX and E values are equal) is valid then the HEX

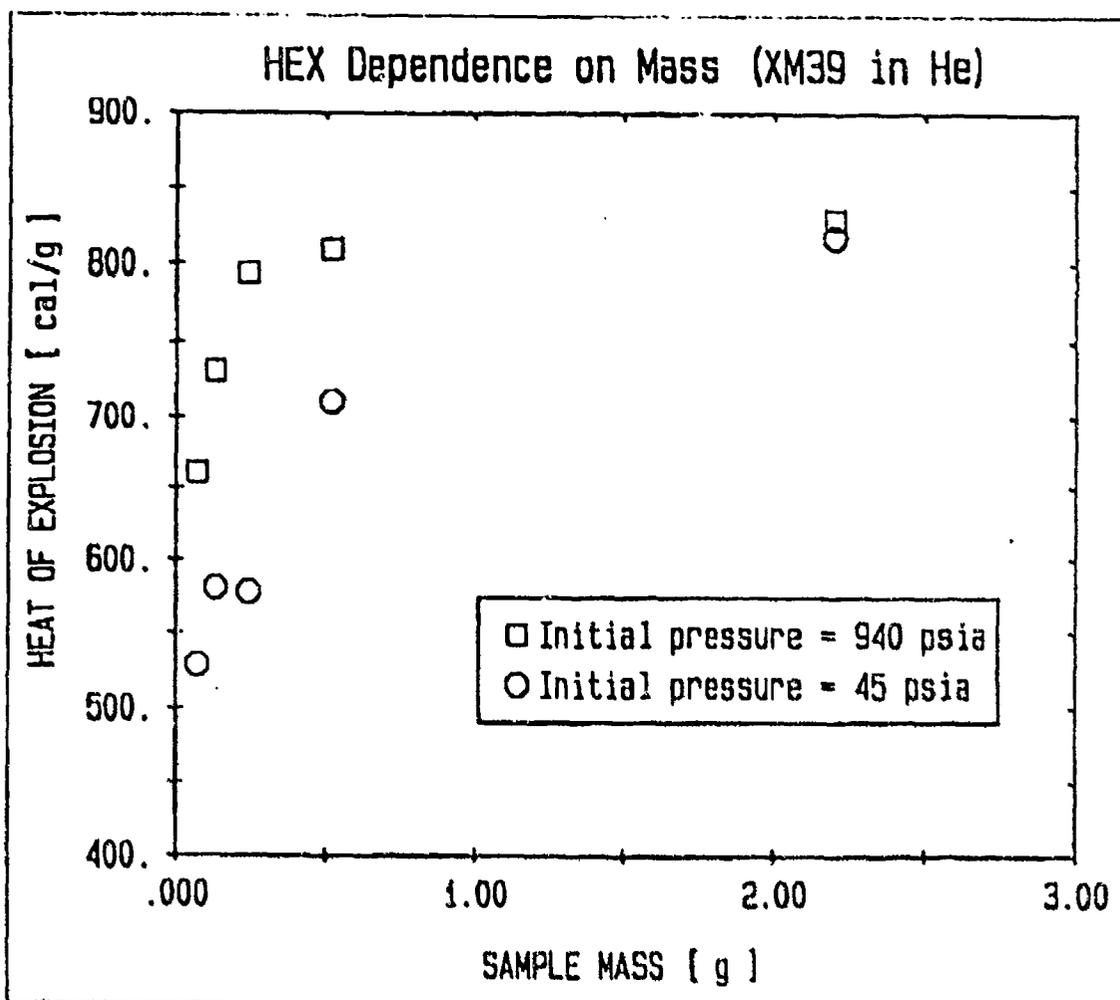


Figure 6. Dependence of Experimental Heats of Explosion on Loading Density for XM39

values for M30 in Figure 5 at $m = .08$ g indicate that in the pressure range 1015-1075 psia, $E \sim 838$ cal/mole and in the pressure range 25-38 psia, $E \sim 587$ cal/g. Although the corresponding standard deviations for these measurements are quite large, 32 and 176 cal/g, the difference between the calculated E and measured HEX are statistically significant for both the high and low pressure ranges.

For XM39, the calculated $E \sim 895$ cal/g. The HEX values for XM39 in Figure 6 at $m = .07$ g indicate that in the pressure range 940-978 psia, $E \sim 731$ cal/g and in the range 45-52 psia, $E \sim 529$ cal/g. The corresponding standard deviations (4 and 83 cal/g) indicate that the differences between calculated E and those based on the HEX measurements are significant.

Lower values for E may improve IB code predictions at low pressures but the substitution of measured HEX values for E in IB calculations requires that they be independent of LD (i.e., dependent only on total pressure in the calorimeter). It is necessary to determine if values for HEX measured at the lowest LD ($m = .08$ and $.07$ g

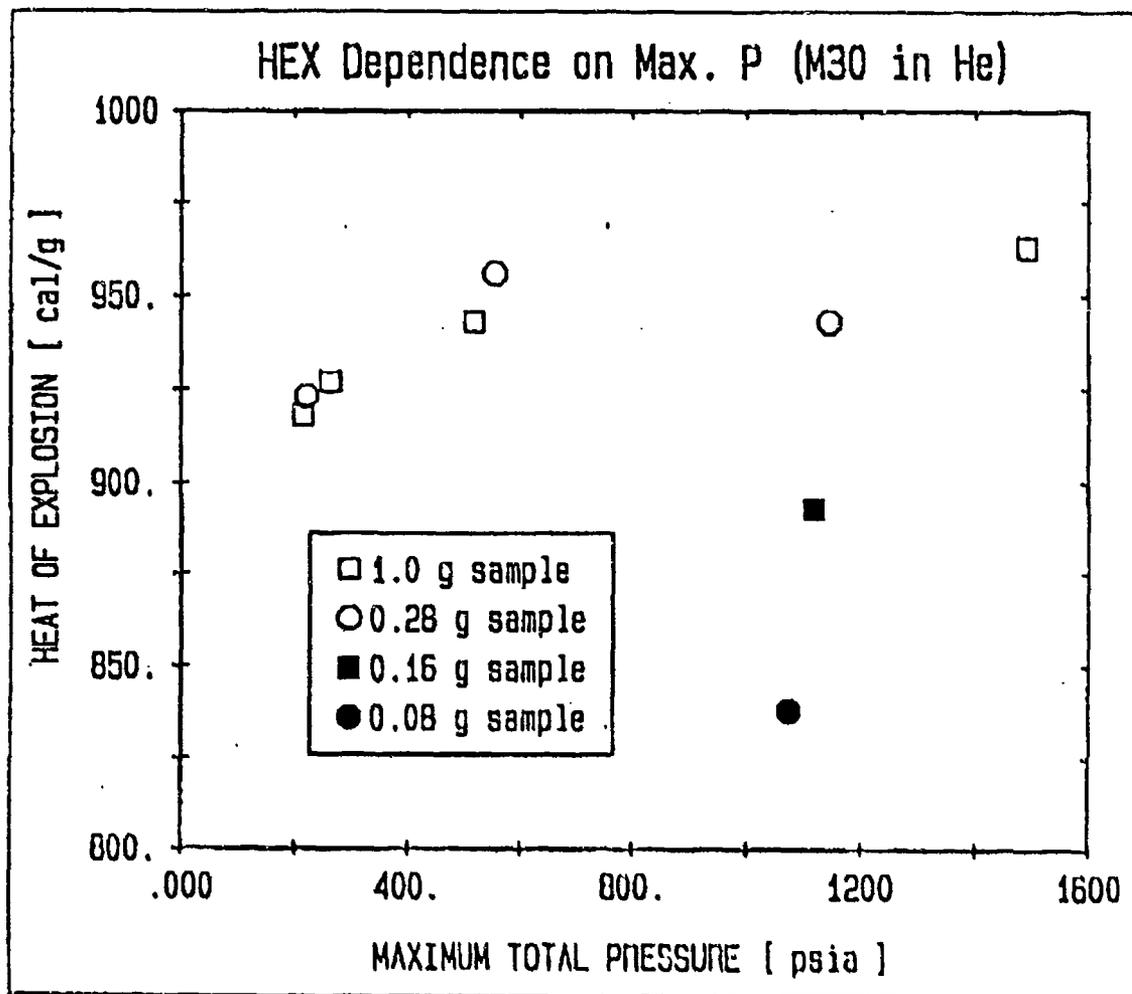


Figure 7. Maximum Total Pressure and Heats of Explosion: Selected M30 Data to Show Pressure and Loading Density Effects on Experimental Heats of Explosion

in Figures 5 and 6, respectively) which have been used to make estimates of E, are consistent with this requirement. The data in Figures 7 and 8 indicate that HEX values are affected by changes in LD which suggests that the HEX have some dependence on the concentration of combustion products in addition to the operating pressure levels in the calorimeter.

It is noteworthy to emphasize that the effect of systematic errors in the calorimeter determination of thermal energy can greatly affect HEX values. Determination of the calorimeter energy equivalent which is used to convert the calorimeter temperature increase into energy is obtained by a standardization method in which increases in the calorimeter temperature are approximately 3°C. At low loading densities ($m = .07$ g), the increases in temperature are approximately .03°C. Conclusions based on the low loading density HEX values remain questionable without determining the validity of using the calorimeter energy equivalent for such small increases in temperature.

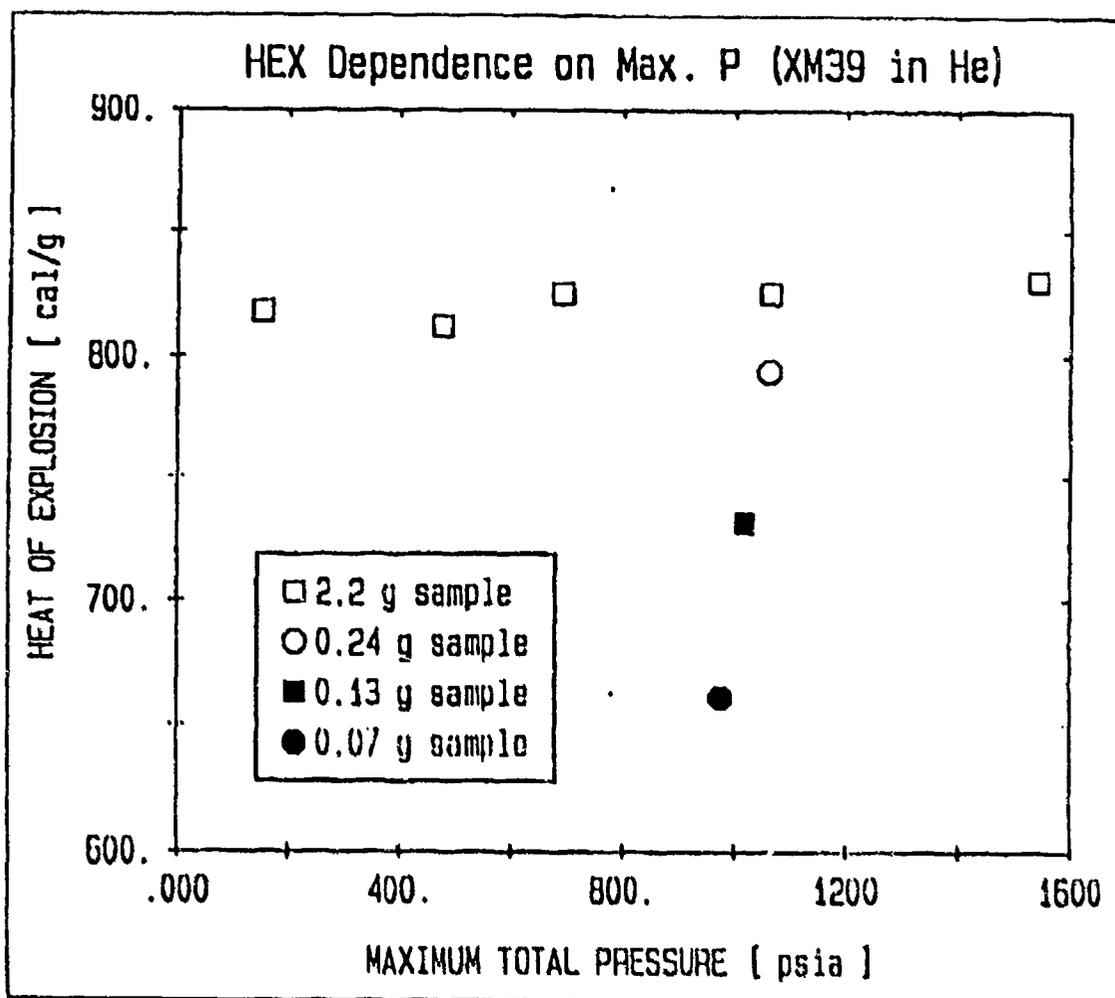


Figure 8. Maximum Total Pressure and Heats of Explosion: Selected XM39 Data to Show Pressure and Loading Effects on Experimental Heats of Explosion

V. SUMMARY

An alternate method for estimating the thermal energy (E) of propellant combustion products for calculating gun performance at low pressures has been investigated. The method assumes that at a given gun pressure, E is equal to the measured heat of explosion (HEX). HEX have been measured in calorimeters for M30 and XM39 at low initial pressures (P_o) (25-1015 psia). Low loading densities (LD) (.012 - .0002 g/cc) were used to minimize the uncertainty in the calorimeter pressure during combustion. At the higher LD, HEX values for M30 and XM39 were constant and not much different from HEX values calculated for an equilibrium combustion process. As P_o and LD decrease, a fall off in HEX from the values obtained at higher LD and higher P_o is observed. The HEX values in this low pressure fall off region are much less than calculated HEX and also the calculated E values now used in gun calculations. HEX values in the fall off region may

also depend on the concentration of combustion products in the calorimeter. This latter dependence will increase the difficulties of using the HEX measurements in IB codes.

The HEX measurements in the fall off region are also dependent on the physical environment. The addition of thermal insulation to the capsule which support the propellant grains and changes in its location relative to the grains which affect transport processes during combustion also affect HEX values.¹ This implies that some knowledge of the physical processes which occur during combustion at low pressures in calorimeter bombs (and guns) will be required to assess the usefulness of estimating values for E from the HEX measurements.

Future work to determine the magnitude of the calorimeter errors in measuring small thermal inputs is planned and an effort will be made to obtain information on the sensitivity of HEX measurements to the presence of combustion product gases and to simulated (inert) propellant grains.

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