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CONF-850706--10

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LA-UR--85-1130

DE85 009609

TITLE: **INTERMOLECULAR EXPLOSIVES**

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MASTER

SUBMITTED TO: **8th Symposium on Detonation**
Albuquerque, NM, July 15-19, 1985.

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INTERMOLECULAR EXPLOSIVES

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The steady-state detonation characteristics of a few intermolecular explosives are examined along with results of wedge experiments on initiation and reaction growth in one system (EAK), toward a better understanding of this class of explosives. The importance of factors such as products and their states, and temperature, to performance of these systems and explosives in general is discussed.

Hydrogen compounds may be superior enough to carbon and its compounds as fuels and products for quick metal-accelerating explosives that the effect of the generally lower density is offset. High temperatures and condensation of solid products are probably the main causes of late appearance of energy in metal motion.

The unusual shock sensitiveness and explosiveness of EAK are indicated by wedge results including pseudo-Pop-plots of overtaking waves, and by plots of shock velocity versus transit distance in the wedges.

INTRODUCTION

Intermolecular explosives are the most used class, but not the most researched nor the best understood. They can be defined as a class of composites in which a significant part of the performance comes from fast reactions between mixed substances. A principal criterion and aim is synergy, performance greater than that of the individual components. The research and applications studies have as a purpose the better understanding of these systems; this might result in the development of better explosives by transcending some of the present limits on stability, sensitivity, density, performance, ease of manufacture, availability, and cost of single compounds.

The combinatorial reactions of intermoleculars are in contrast to decomposition in monomolecular (an easier term than intramolecular) explosives and the intermediate performance in mixtures of essentially noninterreactive composites such as cyclotols and monomoleculars diluted with inerts. Average and maximum distances between reactants are orders of magnitude greater in intermoleculars than in monomoleculars, resulting in much more mass transport before reaction and a broader spectrum of reaction times. Reaction orders are at least superficially different also, the principal type being

$$A + B + \ldots \rightarrow C + \ldots \quad ,$$

with the reaction type

$$A \rightarrow B + C + \ldots$$

being dominant in monomoleculars but not as important in intermoleculars. However, the orders may not be very different if one of the reactants is itself an explosive, and/or the important energy-producing reactions between oxidizer and fuel moieties are preceded by intermediate decomposition routes into those moieties in particular monomoleculars.

In addition to these principal intrinsic differences, the materials used in many intermoleculars are chemically quite different from the compounds of most common monomolecular explosives. This is no doubt a result of the former being so heavily used industrially, where cost and performance considerations are different from those of military explosives. Using the ideas of one world in the other, particularly to yield new explosives with special characteristics for military use, has resulted in some explorations which have produced practical formulations and which can be used to further the science of explosives in general.

There are still other differences, some of them, like microstructure or particle size, related to the intrinsic differences above. I would like to focus on the apparent effect of some chemical differences on steady-state detonation, and then describe some non-steady-state effects seen in one system, EAK, which is a eutectic composition (46/46/8 by weight, also with variations) of ethylenediamine dinitrate (EDD), ammonium nitrate (AN), and potassium nitrate (KN).

EXPERIMENTAL

The materials and acquisition of most of the data have been described previously (1,2,3,4,5). Material for the wedge tests was made by Group M-1 at Los Alamos. Most was made by synthesizing EDD as described in (5), melting it with AN and KN in a kettle, and then casting it into cylindrical molds from which wedges were machined in several orientations with respect to the casting geometry. Some EAK was made by synthesizing and melting under perchloroethylene, and one set of wedges was made by pressing after granulation in cold perchloroethylene.

The wedge test for quantitative determination of shock sensitivity has been described elsewhere (6) and was used with wedges not less than 76 mm in width and 150 mm long. The wedge angle was 30° for most tests with a maximum run of 55 mm; a few of the early tests used 25° and a maximum run of 30 mm. One test was carried out in double width (with a 300-mm-diameter plane-wave lens replacing the usual 200 mm one) to assure that erosion of shock strength from the sides did not occur during the free run.

STEADY-STATE DETONATION

Mass transport, longer reaction time, and different reaction orders notwithstanding, the reaction front velocity (detonation speed, D) may be faster and the performance higher, both calculationally and experimentally, in particular intermolecular systems than in some detonating monomoleculars of the same density and available thermochemical energy. This can be true even when one of the intermolecular partners is itself a high-performing ideal monomolecular. Note the EA systems in Table I, especially EAK (50/42.5/7.5 by weight). Its D of 8.02 km/s in 102 mm diameter confined in 10 mm of copper is above those (7.86 and 7.73 km/s calculated by the Tiger code using the BKWR equation of state (7) or by the Kamlet-Jacobs short method (8), and above that of EDD. EDD is a powerful explosive, with both D--7.7 km/s at high density--and pressure (about 23 GPa) higher than TNT. It is not necessary, although it may be advantageous, for the intermolecularly reacting partners to be themselves detonable, as has been demonstrated in an all-solids system (4) and various ones with a liquid component, such as ANFO.

In some cases the equation of state calculational differences are greater than the difference between experiment and ideal calculation.

High pressure and relatively fast reaction are suggested by high detonation speed. Performance synergism is verified by metal propulsion as in heavily confined plate-dent tests in small diameter (1,2,5) and cylinder tests (1,2), Table I. But these tests integrate pressure-time variables, hence do not necessarily confirm high pressure or very fast reaction. In fact, very high pressures can hardly have occurred in the confined plate-dent tests, which had maximum shock (detonation) velocities of 5 or 6 km/s (1,2,4,5) because of the small diameter.

It may be noted that reaction rate or reaction zone effects may be reflected in the cylinder test by changes in D with diameter, while the cylinder wall energy may or may not be invariant over the same range of diameters: EAR (15 wt% RDX) increases greatly in D from 25.4 to 50.8 to 101.6 mm, while the cylinder energy (wall speed) changes very slightly, observable only by small changes in arrival times at particular expansion distances, which is an integration of the wall velocity. EA 50/50 increases in both D and energy with diameter, as did an aluminum/perchlorate formulation reported by Finger et al. (9).

Other systems may yield experimental speeds, pressures, and performance considerably lower than expected, even though they are or appear to be similar to well-performing intermoleculars in stoichiometries (oxygen balance), potential chemical energies, density, and detonability. They may even have the same oxidizers and calculate as well. The TNT mixtures, Amatols, in Table I are of that nature; TATB in mixture with lithium nitrate appears to be of that kind also (10). On the other hand, hydrazine/hydrazine nitrate systems perform well (11), as does the eutectic of ADNT/AN (1,12).

The differences in performance are hypothesized to be a result of different reactants--fuels, mostly, because there seems to be a better present chance to decode carbon-hydrogen differences than, for example, oxygen-fluorine differences, although there is progress there (13). Reactant differences lead to differences in products and their states, or detonation characteristics including the quantity of gas, the average molecular weight of the gases, and detonation temperature. Table II gives those parameters for some hydrogen and carbon fuel systems.

It is suggested that hydrogen is a better fuel than carbon for producing higher performance of some kinds, including metal acceleration, at least in moderate explosive mass to metal mass ratios. This thesis is supported by observations made previously (1): the speed with which energy delivery is effected is indicated by the ratios of energies at expansions of 7 and 2 times the original volume, that is $(v_7/v_2)^2$ where v is cylinder wall radial velocity. It is also supported by a paper on the chemistry of detonations (14) in which higher mole fractions of water are observed to correlate well with early cylinder energy, that is, by smaller ratios of $(v_7/v_2)^2$, while explosives having more carbon products have higher ratios; and by an observation in (15), in which it was noted that HNO explosives deviated positively from the performance predicted by an otherwise tight correlation and fitted equation (15), while CNOs deviated negatively.

Four hypotheses about hydrogen and its products being better than carbon in this performance connection are:

(1) More gas is produced. Assuming that an explosive charge is volume-limited and that the hydrogenous explosive has density comparable to carbonaceous explosive (although on the whole it is lower, in practice it need not be greatly so), then there are more moles of hydrogen than carbon, and there can be more moles of water than carbon oxides by 1.56-2.44 to 1 ($H_2O/CO-H_2O/CO_2$). On average, including lower density, the ratio of H/C oxide gases predicted by simple stoichiometry will be somewhere near 2. In thermohydrodynamic code calculations and experiment, the ratios of the total gases are nearer 4/3. See Table II for some numbers.

(2) The average molecular weight of the gases will be lower. If there are no solid products, of course the molecular weight is the reciprocal of the number of moles of gas produced, so the more gas, the lower the MW. In reaction motors or in Defourneaux's thesis on propulsion by explosives (16), the velocity of the rocket or the metal, respectively, is inversely proportional to the molecular weight of the working fluid gas.

(3) The temperature of the fluid is lower. The various thermohydrodynamic codes calculate detonation temperatures of water-rich systems perhaps 500°K or more lower than CO_2-rich systems. The total thermochemical energies of the two kinds of systems can easily be about the same; EA systems heats of detonation are close to those of TNT and Amatols, both calculationally and by calorimetry (17,1) on both weight and volume bases. If total energies are equal, then partition to lower rotational and vibrational energies means greater translational energy.

(4) Transitional states of carbon during explosive decomposition may produce thermic events of a nature not usually considered. It has been reported (18) that at CJ all molecular bonds are broken. If that occurs, or to the extent that occurs, carbon not yet reacted nor condensed to solids or quasi solids is effectively a gas (if not ionized), which has the high positive heat of formation of +172 kcal/mol or over 14 kcal/gm. The heat sink of unreacted, uncondensed carbon would reduce the system's immediately available energy, while the carbon would not be a good low-molecular-weight gas because of its elevated thermal state and the ensuing condensation.

Aluminum is a fuel of interest and pragmatic importance, differing from carbon in practice because it is usually in elemental particles in formulations. If small particles were

TABLE I

PERFORMANCE IN CYLINDER TESTS

Explosive	Diameter mm	Density gm/cc	TMD %	D km/s	Wall Velocity km/s $V/V_0=2$	$V/V_0=7$	$V/V_0=11$
TNT	25.4	1.63	98.5	6.99	1.23	1.40	1.46
EDD	24.4	1.55	97.2	7.55	1.29	1.44	1.46
Comp B 64 RDX	25.4	1.72	98.5	7.99	1.44	1.63	1.68
EA 50/50	25.4	1.60	96.5	6+	1.06	1.24	1.33
	50.8	1.59	95.9	7.67	1.28	1.45	1.50
	101.6	1.61	97.1	7.37	1.34	1.49	1.53
EAK 50/42.5/7.5	101.6	1.64	98.5	8.02	1.33	1.47	1.52
EAK 42.5/42.5/15	25.4	1.62	96.6	6.93	1.35	1.52	1.57
	50.8	1.60	95.4	7.70	1.36	1.50	1.56
	101.6	1.63	97.2	8.17	1.36	1.50	1.55
EARL-1, 5.2 Al	50.8	1.65	96.4	7.46	1.34	1.51	1.52
	101.6	1.66	97.0	8.02	1.37	1.54	1.61
EARL-2, 14.8 Al	50.8	1.72	96.8	7.23	1.30	1.46	1.49
	101.6	1.71	96.2	7.70	1.39	1.61	1.69
Amatol 57AN 5μm	25.4	1.61	95.1	6.07	1.01	1.18	1.23
54AN 25μm	50.8	1.61	95.2	6.05	1.01	1.18	1.24

Compositions are weight percent. Cylinder wall thickness was 0.1 x I.D. (diameter shown). Amatol data courtesy F. Helm , D. Breithaupt, R. McGuire, Lawrence Livermore National Laboratory.

TABLE II

SOME CALCULATED PRODUCT AND DETONATION PARAMETERS

Explosive	Fuel Ratios Atoms H/C	WT% H/(H+C)	KSM[a] N mols/kg	MW gm/mol	C mols/kg	BKWR[b] N mols/kg	T °K	D km/s	P GPa	BKW N mols/kg	T °K	D km/s	P GPa
TNT	0.71	5.6	25.3	28.5	23.1	27.9	2711	7.18	20.4	28.5	2905	6.99	20.8
Amatol (60AN)	3.14	20.8	36.3	25.7	5.5	36.4	2421	8.37	28.0	36.3	1616	8.45	29.9
Amatol (79AN)	6.71	35.9	39.8	25.1	0	40.0	2265	8.68	29.5	40.0	1292	8.74	31.7
TATB	1.00	7.7	29.1	27.2	17.4	29.0	2177	8.39	31.3	29.1	2128	7.85	29.7
HMX	2.00	14.3	33.8	27.2	6.8	33.8	3093	9.38	40.2	33.8	2264	9.01	39.4
EDD	5.00	29.4	40.3	22.4	8.1	40.3	2140	7.78	24.1	40.3	1337	8.49	27.6
EA 44/56	11.00	47.8	42.2	23.7	0	42.2	2176	8.68	28.3	42.2	1196	8.85	30.8

a Kamlet-Jacob H_2O-CO_2 "arbitrary" (8); MW is average of gases; C is carbon

b BKW with modified parameters (7)

Some calculations (a and b) at TMD, others at 95 to 98 TMD converted to a gas by the detonation or by liberation from a compound, as suggested for carbon, the effects might be quite similar to those of carbon: Upon forming individual molecules of oxides, those too would be what is normally a solid in the gaseous state, with very high positive heat of formation and a hard-to-predict thermic situation dependent on condensation states and rates.

The effects of aluminum on performance in moderate time frames such as in the cylinder test or plate dent may change considerably with variations in the amount in the formulation: In relatively small quantities in fine particulate form it may help by both burning and condensing quickly enough to raise the ambient temperature of the fluid and thereby increase the overall reaction rate; the influence of temperature on the reaction rate is discussed by McGuire in this symposium (10). In larger amounts the times might be too long. The formulations EARL-1 and EARL-2 have approximately 5 and 15 percent by weight aluminum (5 to 20 micrometers diameter), and as may be seen from the cylinder data of Table I, the lesser amount of aluminum depressed the detonation velocity of the nonaluminized EAR only slightly in either 50 or 100 mm diameter, with D over 8 km/s in the larger size. At the same time it had little if any effect on the energy in 50 mm diameter but increased it in 100 mm diameter at expansions greater than V/Vo = 2. The greater amount of aluminum reduced the detonation velocity in both sizes and reduced the cylinder energy in 50 mm diameter but increased it significantly at all expansions in 100 mm size. Such crossovers point toward nonlossy ways to tailor energetics effects.

WEDGE TESTS OF SHOCK INITIATION

One of the effects of plane shocks of moderate duration in EAK was a rather linear increase in shock speed through the wedge thickness (maximum 55 mm) over a wide range of input pressures. Slopes and intercepts are shown in Fig. 1, which illustrates the effects of input pressure and composition proportions. The eutectic EAK composition, 46/46/8 by weight, is apparently more sensitive than 50/42/8, even though there is less of the detonable component, EDD. It is not unreasonable to expect the sensitivity to be near the maximum at eutectic proportions because the intimacy of mixing should be greatest and the effect of particle size smallest. Reducing the EDD to 42/50/8 depressed sensitivity and produced almost the only failing waves observed, in 4 out of 5 tests.

Some batches appeared to have a low maximum velocity. This could mask classical transitions to high detonation velocity and can even give the impression of failing, if the input shock is very strong, overdriving the materials, as in Batch 334, Fig. 2. The low performance is probably a result of coarse microstructure, as seen, along with considerable anisotropy, in Figs. 3 through 6. These scanning electron micrographs are from three mutually orthogonal cuts of the same sample, the fourth picture identifying the particles which are AN.

There were a few occurrences of the classical wedge-test result, rapid transition to high-speed detonation. One of these is seen in Fig. 7, where there is also evidence of overshoot to higher-than-normal D.

Figures 8 and 9 show two other responses often seen. One (Fig. 8) shows fairly steady shock velocity increases through the wedge after a period of 20 mm or so of no acceleration, implying slow or late growth in reaction behind the front. There is a suggestion of an overtaking wave at 35 mm, followed by further linear increase in velocity to near-normal D. In Fig. 9 there is evidence of instability, possibly reflecting irregularity and anisotropy in the microstructure; the structure of the shock progress is very complex, with, possibly, more than one transition.

Figure 10 shows shock velocity decreasing, taken to be incipient failure in EAK 42.

Because of the dearth of clear, quick transitions to known high detonation velocity, true Pop-plots could not be fitted; but nonlinear increases in velocity of the shock suggested overtaking waves often enough to try fitting distances to these "transitions" to input pressure in log-log space. In Fig. 11, which also includes Pop-plots of several standard materials for comparison, the right ends of these plots occupy positions near those of insensitive explosives, but with relatively long run even at high pressures. The slopes are much flatter, putting the left ends nearer the region of more sensitive explosives, with only moderately longer runs at lower pressures than at high pressures.

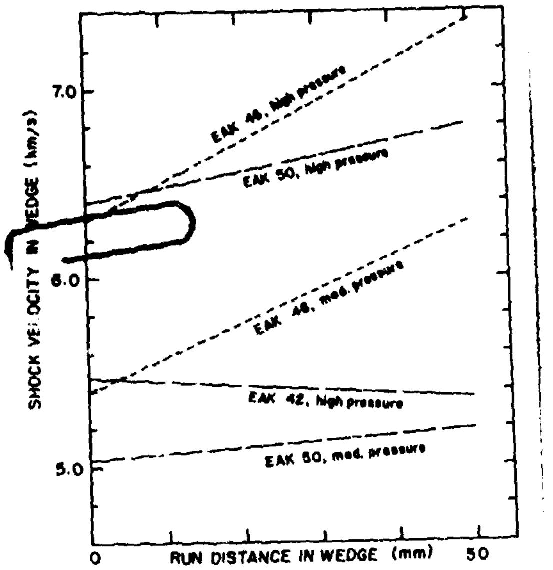


Fig. 1. Slopes and intercepts, or initial and final velocities. The eutectic composition (EAK 46) is apparently more reactive than one with more of the detonable component, EDD (EAK 50). EAK 42 displayed incipient failure.

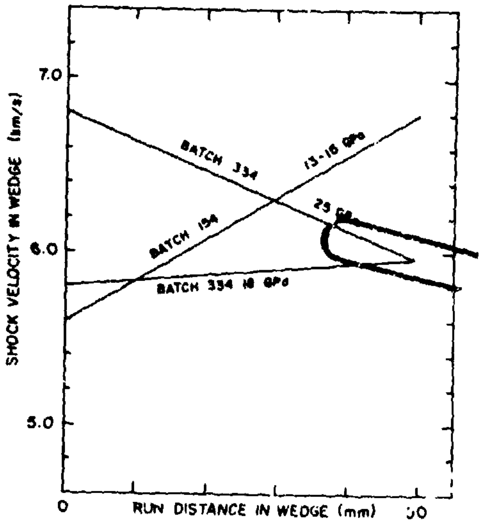


Fig. 2. Slopes and intercepts, or initial and final velocities. Batch 334 apparently has a maximum D of about 6 km/s, even when directly initiated by Comp. B (upper line) and not too high density, with 6% voids. Batch 154 shots had only 1% voids.

Fig. 3.

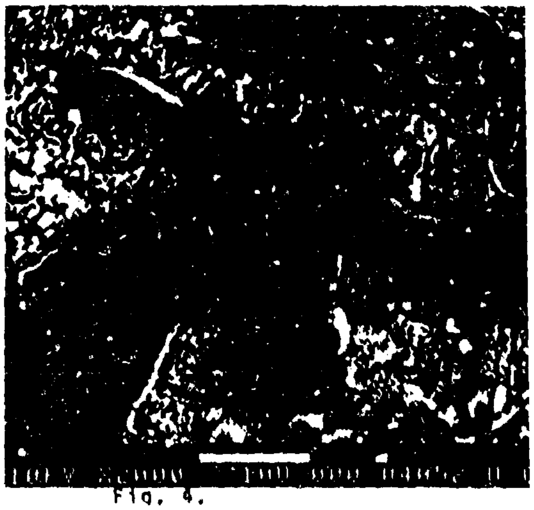


Fig. 4.

See next page for description.

Fig. 5.

Fig. 6.

Figs. 3. - 6. Scanning electron micrographs of a sample of cast EAK 46 cut in three mutually orthogonal planes. Fig. 5 is along the cleavage plane. Fig. 6 is a potassium x-ray distribution map of the field in Fig. 5.

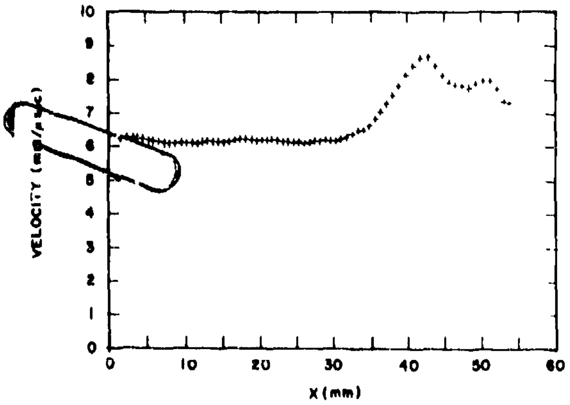


Fig. 7. Cast EAK-46 at 1.657 gm/cc, .992 of TMD, pressure of 21.4 GPa in the wedge. A true transition to detonation occurred about 35 mm free run.

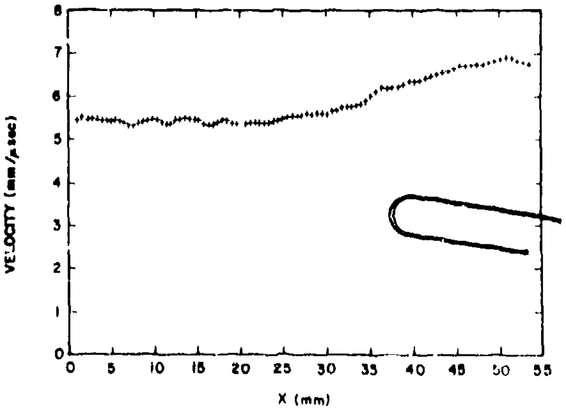
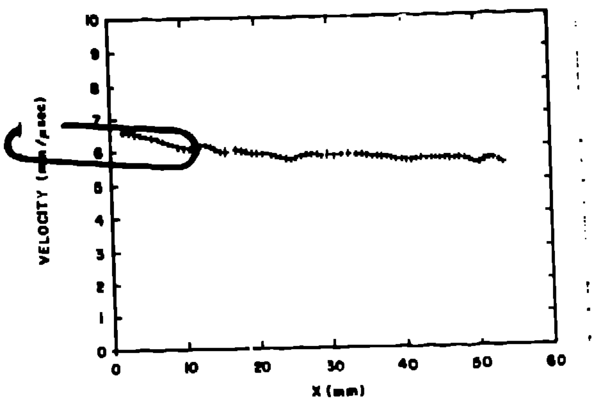


Fig. 8. Cast EAK-46 at 1.65 gm/cc, .99 of TMD, pressure of 13.0 GPa in the wedge.



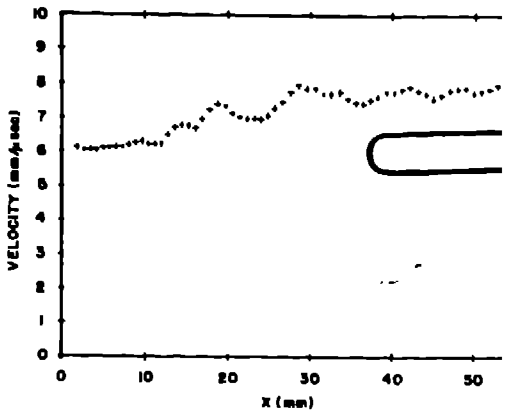


Fig. 9. Pressed EAK-46 at 1.664 m/oc, .995 of TMD, pressure of 20 GPa in the wedge. Transition at 14 mm is followed by instability, perhaps another transition or growth, finally reaching the known high D.

Fig. 10 Cast EAK-42 at 1.65 gm/cc, .984 of TMD, pressure in wedge directly from 12.7 mm thick Comp. B (no attenuator). The plot is interpreted as a failing wave.

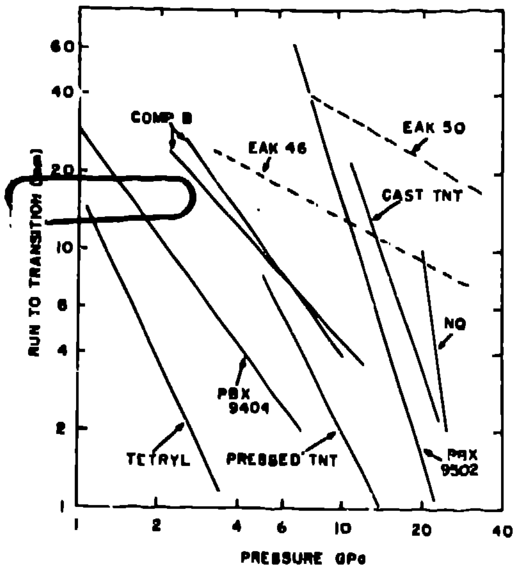


Fig. 11. Pop-plots (solid lines) of some-well known explosives, and runs to transitions versus pressure in EAKs.

In the context of duality in the nature of sensitivity, these materials might be said to exhibit low explosiveness--likelihood of escalation to very violent reactions--but not low sensitiveness. The results of Foster and Craig (19) using a large-scale (200 mm diameter confined, 200 mm long run) "gap test" are not antithetical to such a description.

The effect of void space or its lack was variable: densities were mostly quite high, and density was often overridden by other strong variables. For example, fast-frozen, fine-microstructure EAK pressed to high density was as insensitive as any of the other batches, yet one of the few fast transitions to nigh D occurred in this material at over .99 of theoretical maximum density.

High-energy composite propellants can sometimes exhibit reaction structure similar to EAK in wedge tests, as observed from the work of Dick (20). The slow growth of reaction in all these materials, with failure of EAK--most of the time--to transit in these experiments to known steady-state detonation velocities of 7.5 km/s and higher, is, we suppose, idiocratic to intermolecular systems. The events in the early states of initiation and growth of reaction are made clearer by the wedge method, while the test's constraints on run time and distance to maintain one-dimensionality and minimize confusion with diameter effects limit definition of the final stages before steady state.

CONCLUSIONS

Explosives physicists should find it worthwhile to study detonation products and their states more comprehensively. Explosives chemists might look for intermolecular systems and monomolecular compounds with more hydrogen fuel and less carbon fuel even at some cost in density, a parameter skewing the search for new compounds toward carbon. Such new intermoleculars and compounds are reasonably sure to be good and useful explosives in some way, if only to advance detonation science.

The wedge test or some other noninvasive experiment with internal shock following, plus longer runs, could provide basic data on reaction buildup in intermoleculars and other composite systems. The data might also be useful in the safety contexts of sensitiveness versus explosiveness.

ACKNOWLEDGEMENTS

The work discussed was supported by Eglin Air Force Base (Armament Laboratory), Air Force Office of Scientific Research, Picatinny Arsenal, and the Army Research Office, over a period of time.

Many people have helped carry out the research and provided guidance and explication. While it is not possible to recognize everyone by name, I do thank you all, and to the lists of those previously mentioned, I would like to add, with special thanks, W. C. Davis, G. H. Parsons, A. P. Torres, and J. R. Travis.

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