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Influence of Dense Inert Additives (W and Pb) on Detonation Conditions and Regime of Condensed Explosives

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Results of experimental and theoretical studies of the unusual detonation properties of mixtures of high explosives (HEs) with high-density inert additives W and Pb were analyzed and systematized. Typical examples of the nonideal detonation of composite explosives for which the measured detonation pressure is substantially lower and the detonation velocity is higher than the values calculated within the framework of the hydrodynamic model, with the specific heat ratio for the detonation products of ∼6–8, are presented. Mechanisms of formation of anomalous pressure and mass velocity profiles, which explain the correlation between the Chapman-Jouguet pressure for HE–W and HE–Pb mixtures, the velocity of the free surface of duralumin target, and the depth of the dent imprinted in steel witness plates, are described.

Keywords: detonation pressure, high density inert additives, high explosives, nonequilibrium, nonideal detonation, ''underdriven'' detonation

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Introduction

High explosives (HEs) and compositions the properties of which can be accurately calculated using the well-known thermodynamic and hydrodynamic codes based on the classical Chapman-Jouguet (CJ) and Zel'dovich-von Neumann-Doering (ZND) models of steady detonation are commonly called *ideal HEs* [1–4]. At the same time, in the last decades [5–11], explosive compositions with nonideal properties have been revealed, in particular powerful HEs containing inert additives, such as tungsten and lead (clearly, the boundary between ideal and nonideal HEs is a matter of convention).

The unusual detonation properties of HEs with additives of heavy inert metals first drew attention of researchers of the Los Alamos National Laboratory and other research centers of the United States and then in the USSR at the beginning of the 1950s. Although studies of the detonation of condensed HEs with high-density inert additives are fragmentary and often contradictory, being far from complete, the main features of such a system (containing the densest additives, W or Pb) have been established:

1. The measured pressure is substantially lower (by 5–15 GPa) than the hydrodynamic values calculated in the additive approximation and for the completely equilibrium conditions.
2. By contrast, the measured detonation velocity is 50–500 m/s higher than that calculated within the framework of the hydrodynamic model;
3. The specific heat ratios of the detonation products (DPs) of such mixtures are as high as ∼6–8.

This suggests that such compositions (mixtures of HEs with W and Pb additives) should belonging to a class of explosives different from that to which ordinary condensed HEs (with $\gamma \sim 3$) belong.

The most graphic examples of the nonideal behavior of such HEs are given below.

1. The experiments performed by Al'tshuler and his coworkers [5] with an HE containing W (5–10 μm fraction)

in a mass concentration β of up to 90% yielded data substantially deviating from the results of calculations in the additive approximation [5]. The pressures determined from the velocity of the free surface of an aluminum plate attached to the end face of the charge (closed circles in Fig. 1) turned out to be located substantially lower than the calculated values (open circles): by 26, 32, and nearly 50% at $\beta = 20$, 40, and 60%, respectively. By contrast, the measured detonation velocities proved to be 370–550 m/s higher than the calculated ones (the solid and dashed lines in Fig. 1 correspond to the measured and calculated detonation velocities).

2. The data obtained at the Los Alamos Laboratory [1] also demonstrated that the introduction of Pb or W powders

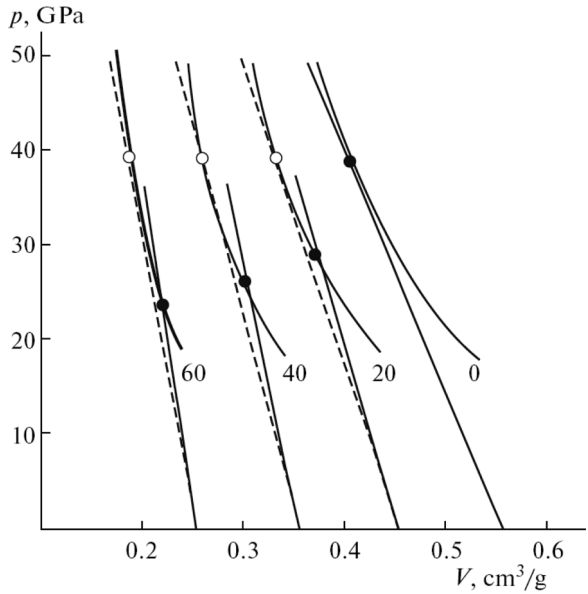

Figure 1. Pressure–volume diagram [5] for a powerful HE–tungsten mixture (0, 20, 40, and 60 wt%): (○) C–J states calculated in the additive approximation and (●) experimental parameters corresponding to the states in the detonation wave front [5].

into RDX- or HMX-based HEs resulted in a systematic lowering of the C–J pressure measured by the free surface method (by 5–10 GPa) compared to that predicted by thermodynamic calculations (Fig. 2 shows the results of the calculations performed by Mader with the use of the Becker-Kistiakowsky-Wilson (BKW) equation of state of DPs [1] and by the present author within the framework of the BKW–RR equation of state with the set of parameters proposed by Orlenko [4]). The measured detonation velocities turned out to be close to the hydrodynamic detonation velocities calculated by using various thermodynamic codes under conditions of equilibrium between the DPs and metal additive in mass velocity, pressure, and temperature (Tables 1 and 2).

3. Craig and Urizar's experiments described in Mader [1] involved measuring the velocity of the free surface of aluminum plates attached to a 55:10:35 vol% HMX–Exon–W charge with a density of 7.9 g/cm^3. The results

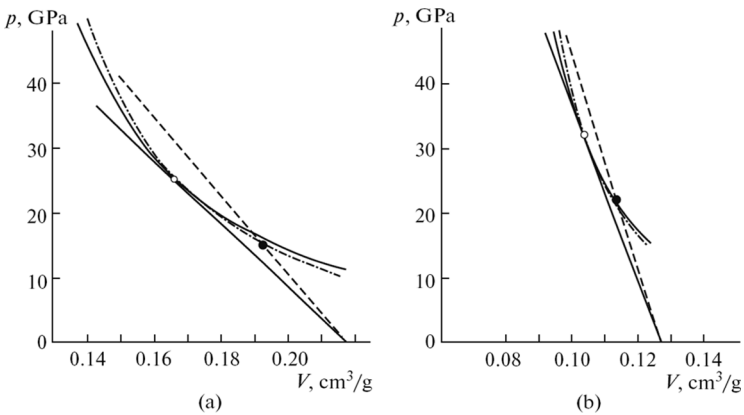

Figure 2. Hugoniot curves calculated using the (–○–) BKW [1] and (– · – · –) BKW-RR equations of state and (–●–) the experimentally measured detonation parameters for (a) 60:10:30 RDX–Exon–Pb ($\rho_0 = 4.6$ g/cm^3) and (b) 55:10:35 HMX–Viton–W ($\rho_0 = 7.9$ g/cm^3) compositions.

Table 1
Experimental [1] and calculated parameters of detonation of RDX–Exon and RDX–Exon–Pb composite explosives

Explosive composition (vol %)	Experiment				Calculation				
	ρ_0 (g/cm^3)	D_{C-J} (m/s)	p_{C-J} (GPa)	γ	D_{C-J} (m/s)	p_{C-J} (GPa)	T_{C-J} (K)	γ_{C-J}	Equation of state of the DPs
RDX–Exon (90:10) ($\alpha = 0$ and $\beta = 0$)	1.787	8,404	32	2.93	8,403	31.7	2,468	2.98	BKW[a]
					8,438	31.4	2,461	3.06	BKW[b]
					8,502	31.2	3,279	3.14	BKW-RR[b]
RDX–Exon–Pb (80:10:10) ($\alpha = 0.1$ and $\beta = 0.42$)	2.700	6,734	—	—	6,692	29.6	2,447	3.08	BKW[a]
					6,620	28.2	2,508	3.19	BKW[b]
					6,656	28.1	3,237	3.26	BKW-RR[c]
					6,669	28.4	3,270	3.23	BKW-RR[d]
					6,683	28.7	3,302	3.21	BKW-RR[e]
RDX–Exon–Pb (70:10:20) ($\alpha = 0.2$ and $\beta = 0.62$)	3.650	5,709	—	—	5,748	28.5	2,352	3.23	BKW[a]
					5,648	26.5	2,459	3.40	BKW[b]
					5,655	26.2	3,129	3.46	BKW-RR[c]

					5,682	26.8	3,204	3.40	BKW-RR[d]
					5,710	27.1	3,282	3.39	BKW-RR[e]
RDX–Exon–Pb	4.600	5,012	15	6.70	4,887	24.9	2,368	3.41	BKW[a]
(60:10:30)($\alpha = 0.3$					4,926	24.1	2,396	3.63	BKW[b]
and $\beta = 0.74$)					4,910	23.7	2,994	3.68	BKW-RR[c]
					4,954	24.6	3,130	3.58	BKW-RR[d]
					5,001	25.6	3,276	3.49	BKW-RR[e]

Note: α and β are the volumetric and mass fraction of metals in the mixture.

[a]Calculated by using the BKW equation of state with the set of parameters for RDX [1].

[b]Calculated by using the BKW equation of state with the set of parameters for RDX, present work.

[c]Calculated by using the BKW-RR equation of state with the metal additive (Pb or W) considered inert, compressible, and completely heated.

[d]Calculated by using the BKW-RR equation of state with the metal additive considered inert, compressible, and heated by 50% (by mass).

[e]Calculated by using the BKW-RR equation of state with the metal additive (Pb or W) being considered inert, compressible, and heated according to its Hugoniot curve.

Table 2
Experimental [1] and calculated parameters of detonation of HMX–Exon and HMX–Exon–W composite explosives

	Experiment				Calculation					
Explosive composition (vol %)	ρ_0 (g/cm^3)	p_{C-J} (GPa)	D_{C-J} (m/s)	γ	p_{C-J} (GPa)	D_{C-J} (m/s)	T_{C-J} (K)	γ	Equation of state of the DPs	Description of the behavior of tungsten
HMX–Exon (90:10) ($\alpha = 0$ and $\beta = 0$)	1.833	34.3	8,665	3.01	34.0	8,625	2,387	3.01	BKW[a]	Without W
					33.5	8,666	2,371	3.10	BKW[b]	
					33.1	8,724	3,232	3.21	BKW-RR[b]	
HMX–Exon–W (55:10:35) ($\alpha = 0.35$ and $\beta = 0.74$)	7.90	23.0	4,900	7.25	32.8	4,807	1,755	4.56	BKW[a]	Compressible W in equilibrium with the DPs in p and T
					29.9	4,630	1,820	4.67	BKW[b]	
					27.2	4,490	2,424	4.87	BKW-RR[c]	
					29.2	4,600	2,746	4.72	BKW-RR[d]	
					31.8	4,649	2,165	4.36	BKW[a]	Compressible W in equilibrium with the DPs in p but E and T are determined from its Hugoniot curve
					32.3	4,716	2,382	4.43	BKW[b]	
					32.1	4,742	3,226	4.54	BKW-RR[e]	

31.1	5,024	1,692	5.41	BKW[a]	Incompressible W
30.4	5,016	1,708	5.55	BKW[b]	
27.5	4,825	2,349	5.69	BKW-RR[c]	
29.7	4,969	2,700	5.57	BKW-RR[d]	
32.8	5,162	3,242	5.42	BKW-RR[e]	

[a]Calculated by using the BKW equation of state with the set of parameters for RDX [1].

[b]Calculated by using the BKW equation of state with the set of parameters for RDX, present work.

[c]Calculated by using the BKW-RR equation of state with the metal additive (Pb or W) considered inert, compressible, and completely heated.

[d]Calculated by using the BKW-RR equation of state with the metal additive considered inert, compressible, and heated by 50% (by mass).

[e]Calculated by using the BKW-RR equation of state with the metal additive (Pb or W) being considered inert, compressible, and heated according to its Hugoniot curve.

of these experiments could be reproduced in hydrodynamic calculations only with the use of a DP isentrope with $\gamma = 7.25$ passing through an experimentally determined C–J point corresponding to a pressure of 23 GPa, ~10 GPa lower than the value calculated for this composition by using the BKW equation.

4. This idea is confirmed by Smith tests [7] in making dents in steel plates (witnesses) with explosives in contact. These tests are very useful and sufficiently simple, because they can be easily reproduced in laboratory conditions and then used not only for direct comparison of properties of brizant charges of single and mix explosives but for quite satisfactory assessments of their C–J pressures (see Fig. 3). As shown in the literature [1, 7, 8], the dent depth linearly correlates with test C–J pressures for most explosives with ideal behavior. However, as Fig. 1 shows, for explosives with dense metal additives the correlation is not valid. Moreover, there is practically doubled difference in assessment of C–J pressures from the correlation ''C–J pressure-dent depth'' (C–J pressure of 29–34 GPa) and from the free surface method (C–J pressure of 15–16 GPa).

   C–J pressure for composition with Pb, calculated by Mader according to BKW EOS, appeared to be 27 GPa, with detonation velocity of 5,096 m/s [1]. Apparently, in this calculation Pb is considered an incompressible material.

We have calculated for the three different models the following values: $p_{C-J} = 23.7$ GPa and $D_{C-J} = 4{,}910$ m/s (Pb is an inert compressible additive, completely heated to detonation product temperature); $p_{C-J} = 24.6$ GPa and $D_{C-J} = 4{,}954$ m/s (Pb is a partially heated compressible additive); $p_{C-J} = 25.6$ GPa and $D_{C-J} = 5{,}001$ m/s (Pb is an unheated shock-compressible additive).

Although the results, calculated within the frames of thermodynamic approach, have an intermediate position between two groups of test data (see Fig. 3), the thermodynamic

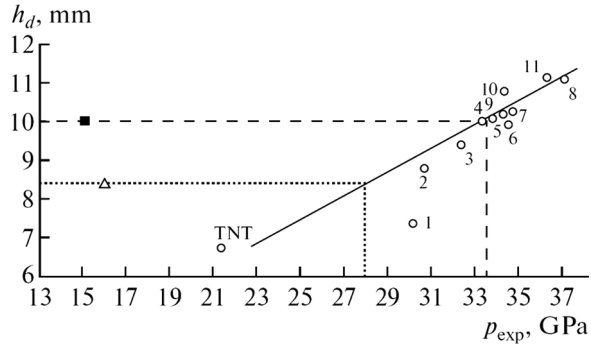

Figure 3. Correlation between the dent depth and detonation pressure: (○) compositions 1–11 are given in Smith [7] and Orlova et al. [8] (wt%): (1) HMX–nitroguanidine–polyesterurethane plasticizer (PUP; 29.7:64.4:5.4); (2) HMX-PUP (86.4:13.6); (3) HMX-PUP (90.0:10.0); (4) HMX-PUP (93.4:6.6); (5) HMX–diaminotrinitrobenzene–PUP (85.6:9.2:5.2); (6) HMX–nitroguanidine–Kel-F 3700 (chlorotrifluoroethylene, $\rho = 2.02$ g/cm^3; 65.7:26.4:7.9); (7) HMX–perfluoro-propylene-vinylidene fluoride copolymer (85:15); (8, 11) HMX–nitrocellulose–tris(β-chloroethyl)phosphate (94:3:3; PBX-9404 composition); (9) HMX-TNT (76.3:23.7); (10) HMX–Exon (chlorofluorocarbon polymer, $\rho = 1.7$ g/cm^3)-tris(β-chloroethyl)phosphate (92:6:2). (■) 60:10:30 (vol %) RDX–Exon–Pb composition with $\rho_0 = 4.6$ g/cm^3 [7]; (Δ) 34:11:55 (vol %) HMX–Exon–W composition with $\rho_0 = 7.47$ g/cm^3 [9].

calculation itself does not allow explaining systematic (in ~ 10 GPa) underestimation of C–J pressures, obtained from tests according to free surface method, or the fact the dent depth for HE with Pb and W additives is considerably greater than predicted from test (and calculation) C–J pressures. The reasons are of complex character and, on one hand, are the increase (or, at least, conservation) of impulse of contact effect from HE charge explosion in the axial direction when adding metals (especially dense, serving as an "internal constraint or shell," preventing free radical expansion of the detonation products) and, on the other hand, the realization of self-sustaining

''underdriven'' detonation in such explosives with thermal and velocity relaxation following the detonation front (the velocity relaxation of phases plays the major role). The mathematical models, which allow qualitative and quantitative description of the features of detonation of mixtures like HE–W, are presented in the literature [9–18]. Some results, also presented in the literature [13–16], show that the very inclusion of the delayed acceleration of W particles results in the considerable change in main detonation conditions and appearance of abnormal pressure and particle velocity profiles behind the detonation front and are presented in Fig. 4.

In this model, the chemical reaction zone is treated as a narrow region behind the shock wave front in which, along with HE decomposition, rapid relaxation processes occur that tend to establish equilibrium between the temperatures and velocities of the DPs and particles. The model allows for the situation where the specified parameters fail to equilibrate within the characteristic time of HE decomposition so that, at the end of the reaction zone, the DPs and particles can have different temperatures and velocities. In this case, the relaxation is completed in the flow behind the wave front. The intensity of temperature and velocity relaxation is characterized by the parameters F and

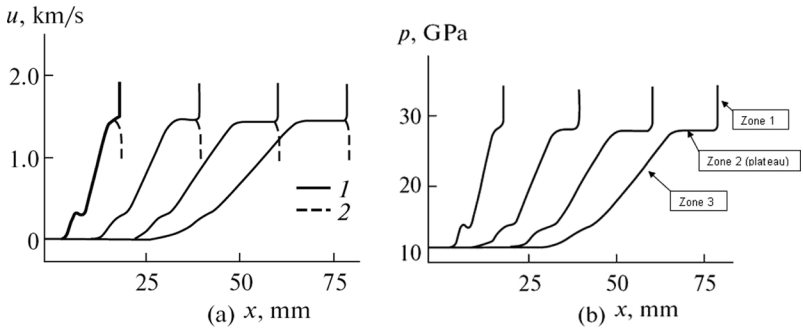

Figure 4. (a) Distribution of the velocities of detonation products and tungsten particles and (b) the pressure profiles behind the detonation wave front at various instants of time: (*1*) detonation products and (*2*) tungsten particles (20 wt% W).

Q, the momentum and energy exchanged between the DPs and particles during HE decomposition. The parameters F and Q can vary from zero (frozen regime) to values corresponding to the complete equilibration of the mixture at the end of the chemical reaction zone.

To estimate the influence of the relaxation processes on the parameters of the detonation wave, we calculated the characteristics of the detonation of mixtures of powerful HEs with tungsten for different limiting regimes of detonation and different mass concentrations of tungsten. The calculation results showed that the main factor is the velocity relaxation of the system. The acceleration of particles is the process that strongly changes the basic detonation parameters with increasing additive concentration. In contrast to the predictions of the additive approximation, calculations within the framework of the proposed model are in close agreement with the available experimental data in both detonation velocity and pressure. For mixtures containing 20, 40, and 60% tungsten, the maximum discrepancy between the calculation and experimental results was found to be within 5% [5].

Some of the calculation results presented in the literature [13–16] are displayed in Fig. 4, which shows the spatial distributions of the velocities of the DPs and particles and of the pressure profiles behind the detonation wave front at distances of 20, 40, 60, and 80 mm from the initiation plane ($\beta = 20\%$).

As can be seen from Fig. 4, the flow behind the detonation wave front consists of three characteristic zones. The first, adjacent to the front, is a narrow (almost vertically in picture) zone within which velocity relaxation is completed; in this zone, an intense equilibration of the velocities of the DPs and particles occurs while the pressure drops sharply to a certain equilibrium level. The width of zone 1 is 20–30 diameters of the particles of tungsten, which is about 1–2 mm. The next zone (zone 2) features a plateau that expands with time and corresponds to constant flow parameters (these parameters that are recorded in free-surface measurements). The self-similarly expanding plateau is followed by the Taylor rarefaction wave (zone 3). The structure of the detonation wave

qualitatively corresponds to the classical regime of self-sustained underdriven detonation [4].

Three-Dimensional Hydrodynamic Model

The hydrodynamics of the interaction of the detonation wave with the matrix of tungsten particles in an HMX-based HE charge was simulated using a three-dimensional Eulerian (3DE) hydrodynamic code [9,17,18]. Simulations without regard for the HE decomposition kinetics yielded a velocity and pressure of the detonation wave propagating through the mixture substantially higher than those observed experimentally. When the kinetics of HMX decomposition was set identical to that of shock-initiated forest fire heterogeneous HE, it turned out that part of elementary weak detonation waves (wavelets, according to Mader's terminology) damped while traveling between tungsten particles. Upon shock impact, HE material continued to decompose and release energy within a wide zone behind the shock wave front. Thus, the nonideal behavior of HEs with metallic filler was explained by the weakening of some of the weak detonation wave between metal particles and the subsequent decomposition of HE behind the detonation wave front. The qualitative character of the influence of the particle size was simulated and pressure profiles with a flat truncated tops were obtained, which are characteristic of underdriven (weak, according to Mader's terminology [1,9]) detonation. Later [9,17,18], the model was experimentally tested by measuring the detonation wave velocity *Db* in an aqueous medium (an aquarium) above the upper end face of a cylindrical HE–tungsten charge as a function of the distance traveled by the wave and comparing the result with the calculated detonation wave velocity. Generally, a satisfactory agreement was observed; when recalculated to the pressure in the front of the detonation wave, which had a Taylor wave profile with truncated top (plateau), a value of 16 GPa was obtained.

Thus, the theoretical models from the literature [9–18] considered in the present work provide a pictorial and comprehensive explanation of the characteristics of HE–high-density inert

additive composite systems, including the phenomenology of formation of anomalous pressure and mass velocity profiles behind the detonation wave front (such profiles are indicative of self-sustained underdriven detonation) and explain why there is no correlation between the C–J pressure and the depth of the dent in a steel target for mixtures of powerful HEs with tungsten and lead.

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