

DETONATION PERFORMANCE OF ALUMINIZED COMPOSITIONS BASED ON BTNEN

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Results of experimental study of detonation performance of BTNEN (bis(2,2,2,-trinitro-ethyl)nitramin) and its mixtures with Al are presented and summarized. Influence of Al particle size including nanosize Al on detonation velocity, heat of explosion, casing ability (both for metal plate acceleration test and cylinder test), pressure and temperature profiles in and behind detonation waves in composition BTNEN/Al (85/15) are considered.

INTRODUCTION

The main goal of Al addition to HE is in enhancing of explosive performance. It is the task of applied research. However when doing this, the fundamental problem, investigation of macroscopic kinetic of Al interaction with detonation products (DP), arises. This problem is of primary scientific interest. Mainly, among aluminized HE there were studied compositions based on HE with negative oxygen balance (OB).¹⁻³ At present work, we investigated explosive compositions based on BTNEN, which is HE with positive OB. Detonation and explosive performance are differed in characteristic times in which interaction of Al with DP influences this or that parameter. This time ranges from tenths of microsecond (characteristic time for detonation velocity (DV)) to tens of microseconds (characteristic time for heat of explosion (EH)). Dispersity of Al powder was varied in

search for optimum Al particle size wanted for a given explosive action.

Following parameters were measured:

- Detonation velocity using contact gauges technique;
- Profiles of temperature and pressure using optical pyrometer;
- Velocities of expansion of a copper cylinder wall using cylinder test;
- Velocities of steel plate accelerated by DP using standard technique;
- Calorimetric heat of explosion;
- Critical pressure of explosion initiation.

MATERIALS

BTNEN ($C_4H_4O_{14}N_8$) has a few advantages: it has high density (1.96 g/cm^3), its formation enthalpy is close to zero and besides it has positive OB = +16.5% which make it interesting as a basic explosive component of aluminized HE compositions.

BTNEN particles were of needle-like shape with diameter of 15-40 μm and length of up to 500 μm . Four kinds of aluminum powder with mean grain of size 0.1, 7, 15 and 150 μm were used (below Al particle size is indicate in parenthesis next to aluminum symbol). Al content was $\sim 98 - 99\%$ in Al(7), Al(15), Al(150) powders⁴ and $\sim 87\%$ in nanosize Al(0.1).⁵

BTNEN/Al (85/15 wt.) mixtures were prepared by mixing in metal vessel with porcelain balls in inert liquid (hexane). Microscopic study of the prepared samples shows that Al particles are distributed uniformly in the bulk of the mixture and that BTNEN needles are transformed to elongated particles with transverse size approximately equal to diameter of original needles. The charges were pressed using cold unidirectional pressing up to a density of about 0.93 of TMD.

MECHANICAL SENSITIVITY

Increase of total energy stored in the explosive as well as the part of energy to be released in the immediate region (zone) which influences detonation front propagation, a priory, makes the explosive more sensitive. Experience of studies of HE with zero or positive OB shows that usually they have high mechanical sensitivity. To investigate sensitivity of BTNEN and its mixtures with Al we used the method of disintegrative tube.⁶

Table 1 lists the obtained critical pressures. For comparison, the data for primary and secondary explosives⁶ are also shown. BTNEN is more sensitive than typical secondary explosives including PETN.

BTNEN/Al mixtures, especially with Al(0.1), are even more sensitive than pure BTNEN, namely, sensitivity of these mixtures falls between that of primary and secondary explosives. Sensitivity growth when adding of Al can be attributed to two reasons. First one, according to the proposed mechanism,⁷ could be the sensitizations of HE by an additive having higher melting temperature than critical ignition temperature of potential hot spots. Second one could be the chemical interaction of BTNEN decomposition products in hot spots with Al. Al chemical activity could arise due to the destruction of the oxide film on the particle surface during plastic deformation under mechanical loading.

TABLE 1. CRITICAL PRESSURES OF EXPLOSION

| Explosive | P_{cr} , GPa |
|-------------------|-----------------|
| Mercury fulminate | 0.20 ± 0.01 |
| Lead azide | 0.38 ± 0.03 |
| BTNEN/Al(0.1) | 0.55 ± 0.02 |
| BTNEN/Al(7) | 0.74 ± 0.02 |
| HMX/Al(0.1) | 0.67 ± 0.03 |
| BTNEN | 0.79 ± 0.03 |
| PETN | 0.93 ± 0.03 |
| HMX/Al(7) | 0.95 ± 0.03 |
| HMX | 1.00 ± 0.03 |
| RDX | 1.15 ± 0.03 |
| TNT | 1.35 ± 0.03 |

DETONATION VELOCITY

Table 2 reports the data on DV. DV in pure BTNEN was measured at two initial densities (ρ_0). In Table 2, it is also given detonation velocity (D^*) estimated by extrapolation of measured values (D_{ex}) to a unique density of BTNEN in pressed aluminized charges equal to 1.90 g/cm^3 . Cal-

culated density of aluminized composition is than 1.99 g/cm^3 . Relative density referred to TMD (η) is given in brackets. Densities of the explosive in the tested charges (ρ_{0HE}) as well as charge diameter (d) are also given in the Table.

TABLE 2. DETONATION VELOCITY.

| Al, μm | $\rho_0, \text{g/cm}^3$ (η) | $\rho_{0HE}, \text{g/cm}^3$ | $D_{ex}, \text{km/s}$ | $D^*, \text{km/s}$ | d, mm |
|-------------------|---------------------------------------|-----------------------------|-----------------------|--------------------|----------------|
| - | 1.870 (0.954) | - | 8.50 | 8.62 | 20* |
| | 1.909 (0.974) | | 8.66 | | |
| 150 | 1.965 (0.961) | 1.875 | 8.38 | 8.48 | |
| 15 | 1.955 (0.956) | 1.864 | 8.30 | 8.44 | |
| 7 | 1.955 (0.956) | 1.864 | 8.28 | 8.42 | |
| 0.1 | 1.910 (0.934) | 1.815 | 8.07 | 8.40 | |
| 0.1 | 1.900 (0.930) | 1.805 | 7.92 | 8.29 | 20 |
| 0.1 | 1.914 (0.936) | 1.820 | 8.04 | 8.35 | 40 |
| 0.1 | 1.830 (0.895) | 1.731 | 7.66 | 8.32 | |

* - In copper casing (Cylinder test).

The tested compositions have high detonation ability and the experiments were carried out at a charge diameter of 20 mm. However for a mixture with fine Al(0.1), DV has been additionally determined at charge diameter of 40 mm. D^* is close to that measured in 20-mm diameter charge. Figure 1 shows dependence of D^* on Al particle size for mixtures based on BTNEN along with similar dependence for HMX-based mixtures¹ shown for comparison. It is seen that Al addition decreases detonation velocity and its value is minimal for the smallest 0.1- μm Al for both compositions. However drop of DV for a mixture

BTNEN/Al(0.1) is only near a half of that for a similar composition containing HMX. This fact, possibly, reflects both high chemical activity of free oxygen released under BTNEN decomposition and large heat effect of Al interaction with free oxygen.

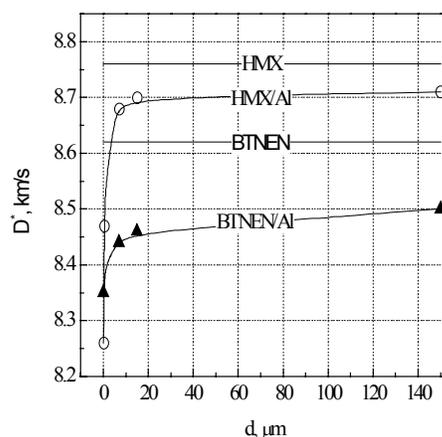


FIGURE 1. DETONATION VELOCITY (D^*) VERSUS Al PARTICLES' SIZE: \circ - HMX/Al; \blacktriangle - BTNEN/Al; SOLID LINES SHOW DV OF PURE HE.

PRESSURE PROFILES

In the pyrometric studies described in this and next section tested charges were 40 mm in diameter and 45 mm in length. The technique of measuring pressure and temperature profiles was described earlier.¹

Pressure profiles were measured using indicator technique with bromoform utilized as the indicator. Usually, the surface of tested materials is covered by a thin layer of a black varnish or Al foil which allows one to cut-off luminosity of the detonation front until it arrives to a contact boundary with

an indicator.⁸ Al foil was used in first experiments. The records (see Fig. 2) show existence of little “step-wise” part at the front of the pressure profile.

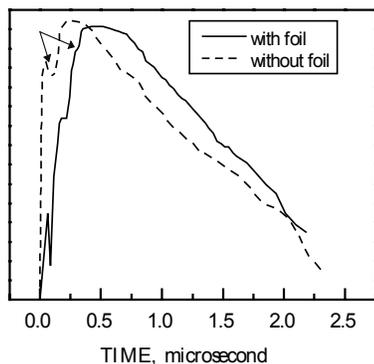


FIGURE 2. TYPICAL LUMINOSITY TIME HISTORIES AT THE SHOCK FRONT IN BROMOFORM.

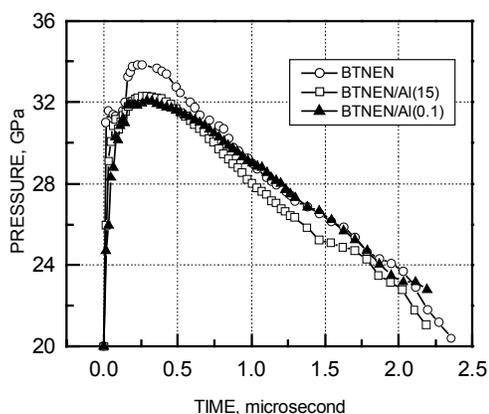


FIGURE 3. PRESSURE TIME HISTORIES AT THE SHOCK FRONT IN THE BROMOFORM.

At first we interpreted these steps as the result of shock reverberations in the foil. In next tests the surface of samples was covered by a special opaque organic compound with thickness below 20 μm . In this case

one can also see some steps at the leading part of pressure records (these steps are marked in the Figure 1 by arrows). We think that these steps can reflect peculiarity of BTNEN detonation decomposition - namely, it goes into two stages. Possibly first one is slightly endothermic. Recently such complicated pressure histories have been observed for ADN under certain conditions.⁹ But there is still no proper explanation of the effect.

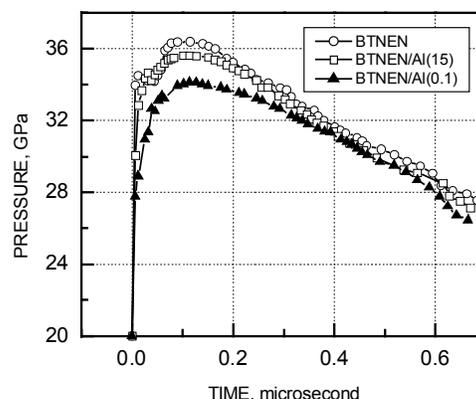


FIGURE 4. PRESSURE TIME HISTORIES IN DETONATION PRODUCT.

Pressure histories for BTNEN and BTNEN/Al mixtures measured at the front of the shock wave propagating in bromoform are given in Fig. 3. BTNEN-containing compositions exhibit the common features in pressure profiles. First, both BTNEN/Al and pure BTNEN demonstrate "step-wise" character of the front of pressure profile. However, these “steps” are smoothed out with decrease of Al particle size, and pressure rise at the leading front becomes slower. Second, introduction of finer Al results in the greater flattening out of pressure profile (see Fig. 3). For BTNEN/Al in contrast to HMX/Al there is

no two-peak pattern of the pressure profile.¹ The features of pressure profiles mentioned above demonstrate that Al is involved into reaction with HE detonation products immediately behind the wave front.

For comparison, bearing in mind that these pressure profiles could not be described accurately in terms of ZND theory, one can recalculate pressure histories from the front of the shock wave propagating in bromoform into the DP according the procedure,⁸ see Fig. 4. Values of DV (D_{ex}) wanted for recalculation were taken from Table 2. Furthermore, the obtained maximum pressures (P_{ex}) corresponding to the initial charge density (ρ_0), see Table 3, were transformed to P^* basing on a unique BTNEN density in charges (1.90 g/cm^3) and values D^* (see Table 2).

TABLE 3. MAXIMUM PRESSURES.

| Al, μm | ρ_0 , g/cm^3 | P_{ex} , GPa | p^* , GPa |
|-------------------|----------------------------|----------------|-------------|
| - | 1.88 | 36.4 | 37.4 |
| 15 | 1.97 | 35.6 | 36.6 |
| 0.1 | 1.92 | 34.1 | 38.4 |

Pressure profiles in BTNEN/Al mixture show that Al addition results in small drop of maximal pressure in the wave not taking into account the differences in the charge density. Some benefits of Al(0.1) can be seen when comparison is performed at the same charge density (see data in Table 3). This effect can be attributed to the larger characteristic times of Al interaction in comparison with time duration of detonation zone influencing DV.

DETONATION PRODUCT TEMPERATURE

Brightness temperatures were measured at the LiF/explosive charge interface at two wavelengths ($\lambda=627$ and 420 nm). To increase the registration time, glycerol was poured on LiF crystal (its thickness was about 8 mm), since glycerol also does not lose its transparency in the studied pressure range. Figure 5 shows temperature profiles for DP of BTNEN and BTNEN/Al mixtures. For BTNEN, as well as in major pressed HE, one can see that luminosity of detonation wave becomes detectable before detonation front arrives to the interface. Sharp peak of luminosity at the instant when interaction of detonation front with LiF begins could be due to a possible luminescence (observed also in HMX and RDX).¹⁰ Non-thermal luminosity can be caused both by fracture of HE crystals at the shock front prior to subsequent detonation decomposition of HE grains and by HE decomposition itself since it can be accompanied by non-equilibrium luminosity. These reasons are likely to be responsible for a large temperature difference between the two wave bands arising when detonation front impacts LiF crystal. As well as majority of other HE studied at the same experimental conditions,¹ the temperature profile for BTNEN demonstrates a falling down nature.

For BTNEN/Al, one can see a little plateau region at the initial part of temperature record after which temperature increases again, namely in $0.25 \mu\text{s}$ for BTNEN/Al(15) and in $0.4 \mu\text{s}$ for BTNEN/Al(0.1). Similar feature was observed in pressure records, but it is necessary to keep in mind that the pressure and

temperature profiles cannot be directly compared.

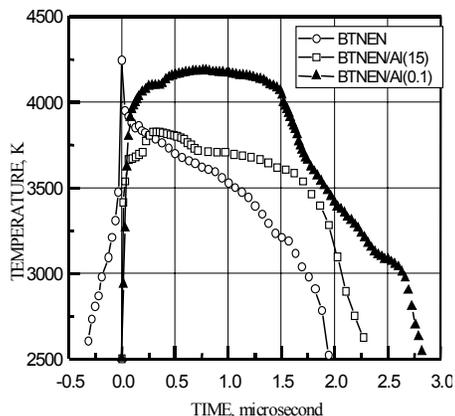


FIGURE 5. BRIGHTNESS TEMPERATURE TIME HISTORIES.

After a small interval of time behind the detonation front, a temperature of aluminized explosives becomes higher than that of pure BTNEN (this interval equals $0.25 \mu\text{s}$ both for BTNEN/Al(15) and BTNEN/Al(0.1)). Temperature profiles in BTNEN/Al compositions demonstrate a few more features. First one is the character of temperature profile itself - "plateau-wise" in contrary to the monotonously falling down one as observed in majority of high explosives including pure BTNEN. As has been stated previously for RDX/Al compositions¹¹ the presence of the "plateau" at the temperature profile is a bright evidence of chemical reaction. The flattering out of the temperature profile is observed for some aluminized compositions.¹ However this effect has been observed in temperature range, which is significantly below the detonation temperature of pure HE. The second feature of the tested compositions is rather high level of temperatures, for example, above 4000 K for

BTNEN/Al(0.1). The drop of temperature observed at the records for BTNEN/Al compositions after nearly $1.5 \mu\text{s}$ is due to arrival of the rarefaction wave coming from the LiF/glycerol interface.

Thus, temperature profiles for BTNEN/Al mixture confirm that intense reaction between metal particles and products of decomposition of BTNEN takes place. The temperature for BTNEN/Al(0.1) exceeds that of BTNEN/Al(15). It means that nanosize Al undergoes more intensive oxidation in immediate proximity behind the wave front.

METAL PLATE ACCELERATION

Table 4 lists experimental values of velocity of steel plate (W_1). They were recorded at different distances (ΔL) along its trajectory according to the appropriate technique¹² for tested BTNEN-based compositions having slightly different values of initial density (ρ_0). For BTNEN/Al, density of the explosive in the charge is given in brackets. To facilitate comparison of results obtained for different compositions Fig. 6 demonstrates dependence of W_1 on distance (ΔL) for extrapolated values corresponding to the same density of BTNEN in compositions with density equal to 1.90 g/cm^3 (see last column of the Table).

Comparison shows that BTNEN/Al(7) and BTNEN/Al(15) mixtures provide higher gain in W_1 than do the mixtures with negative OB such as HMX.¹ Namely, addition of 15 wt. % of Al(7) to HMX resulted in 3% increase of final plate velocity,¹³ and here in similar mixture with BTNEN the plate velocity augments by nearly 6%. It

indicates that Al oxidation in DP of BTNEN is more intense and more exothermic than in case of decomposition products of HE with negative OB. Though plate acceleration ability of pure BTNEN is lower than that of pure HMX, the acceleration ability of aluminized BTNEN approaches that of HMX/Al(7)¹³ at the same porosity of HE in the considered charges.

TABLE 4. PLATE VELOCITY VERSUS DISTANCE

| Al, μm | ρ_0 , g/cm^3 | ΔL , mm | | | | | |
|-------------------|----------------------------|-----------------|------|------|------|------|------|
| | | 8 | 16 | 24 | 32 | 40 | 40* |
| | | W_1 , km/s | | | | | |
| - | 1.86 | 1.74 | 1.86 | 1.95 | 2.01 | 2.03 | 2.07 |
| | 1.92 | 1.80 | 1.91 | 2.00 | 2.06 | 2.08 | |
| 0.1 | 1.91 (1.81) | 1.71 | 1.87 | 1.99 | 2.06 | 2.08 | 2.15 |
| 7 | 1.97 (1.88) | 1.74 | 1.93 | 2.07 | 2.15 | 2.17 | 2.19 |
| 15 | 1.99 (1.90) | 1.79 | 1.95 | 2.06 | 2.13 | 2.16 | 2.16 |
| 150 | 1.99 (1.90) | 1.69 | 1.84 | 1.95 | 2.01 | 2.05 | 2.05 |

* Extrapolated values.

The shape of $W_1(\Delta L)$ curves also indicates that Al oxidation takes place in course of plate acceleration. Indeed, aluminized mixtures provide higher acceleration of metal plate than pure BTNEN during initial stage of DP expansion. The curvature of final parts of trajectories becomes nearly the same.

As anticipated, plate acceleration ability depends on Al particle size. W_1 obtained, e.g. for BTNEN/Al(150) is always lower than in case of BTNEN. In case of Al(7) initial W_1 is slightly lower than in case of pure HE but later reaction of Al provides

significant gain in plate velocity. $W_1(\Delta L)$ curve for a mixture BTNEN/Al(0.1) lies near that for BTNEN/Al(15). In another words, the composition containing Al(0.1) provides practically the same plate velocity along the major distance of plate acceleration as the mixture with Al(15). This fact indicates that during time interval of plate acceleration Al(0.1) as well as Al(7) demonstrating the highest effect are deeply consumed by DP of BTNEN, and hence, the difference in the acceleration effect between compositions with Al (0.1) and Al(7) is mainly due to the content of active Al.

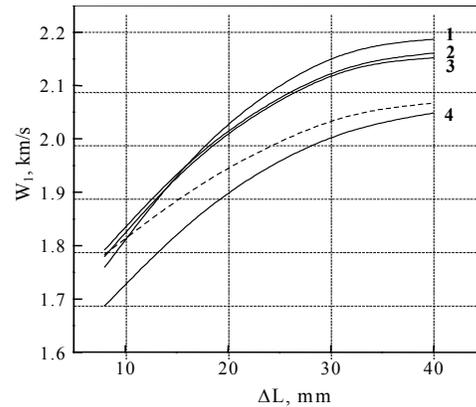


FIGURE 6. STEEL PLATE VELOCITIES VERSUS DISTANCE: 1 - Al(7); 2 - Al(15); 3 - Al(0.1); 4 - Al(150); DASH LINE CORRESPONDS TO PURE BTNEN.

CYLINDER TEST

The velocities of expansion of a copper cylinder (W_2) for given values of the increment of external radius of the cylinder presented in Table 5 were measured according to the technique.¹² Initial densities of the charges (ρ_0) BTNEN/Al slightly differed. Figure 7 illustrates the cylinder expansion curves ($W_2(\Delta R)$) corrected for unique density of BTNEN in the charge

(1.90 g/cm³). As well as in previous sections, for carrying out this correction the experimental data obtained for BTNEN at two different densities were utilized (see Table 5). For aluminized compositions, density of the explosive in the charge is given in brackets.

TABLE 5. EXPANSION VELOCITIES OF A COPPER CYLINDER WALL.

| Al, μm | ρ_0 , g/cm ³ | ΔR , mm | | | | | | |
|-------------------|------------------------------|-----------------|------|------|------|------|------|------|
| | | 4 | 5 | 7 | 10 | 12 | 15 | 15* |
| | | W_2 , km/s | | | | | | |
| - | 1.85 | 1.48 | 1.53 | 1.61 | 1.68 | 1.71 | 1.74 | 1.78 |
| | 1.90 | 1.52 | 1.57 | 1.65 | 1.72 | 1.75 | 1.78 | |
| 0.1 | 1.94 (1.85) | 1.49 | 1.57 | 1.67 | 1.76 | 1.80 | 1.83 | 1.88 |
| 7 | 1.98 (1.89) | 1.52 | 1.61 | 1.73 | 1.82 | 1.86 | 1.89 | 1.90 |
| 15 | 1.99 (1.90) | 1.52 | 1.60 | 1.70 | 1.80 | 1.84 | 1.88 | 1.88 |
| 150 | 1.99 (1.90) | 1.42 | 1.49 | 1.59 | 1.68 | 1.72 | 1.76 | 1.76 |

* Extrapolated values.

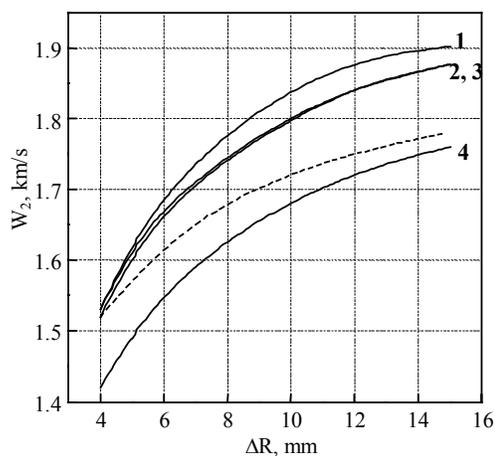


FIGURE 7. COPPER CYLINDER VELOCITY VS. RADIAL EXPANSION: 1 - Al(7); 2 - Al(15); 3 - Al(0.1); 4 - Al(150); DASH LINE CORRESPONDS TO BTNEN.

The data for the cylinder test are in qualitative agreement with results on plate acceleration tests. So, the highest W_2 has been obtained for BTNEN/Al(7). Compositions with Al(0.1) and Al(15) provide almost identical acceleration but smaller than BTNEN/Al(7). $W_2(\Delta R)$ curve for BTNEN/Al(150) mixture (see Fig. 7) lies below. With correction for unique density of BTNEN in the charge, the final velocity registered for BTNEN/Al(7) composition exceeds that for BTNEN by 7% and is close to the value registered for the mixture HMX/Al(7).¹³

EXPLOSION HEAT

Results of measurements of EH are summarized in Table 6. One can see that the measured values of EH demonstrate a high degree of Al oxidation by DP under the considered conditions providing large time for Al oxidation. EH of a mixture with Al(7) exceeds that of pure BTNEN by 3.37 MJ/kg or by 64%. As shown earlier,¹³ addition of 15% Al(7) to HMX resulted in increase of EH at 1.39 MJ/kg. And although BTNEN is inferior to HMX in EH, that of BTNEN/Al composition exceeds EH of similar composition with HMX in average by 24%.

TABLE 6. EXPLOSION HEAT.

| Al | - | Al(0.1) | Al(7) | Al(15) | Al(150) |
|------------------------------|------|---------|-------|--------|---------|
| ρ_0 , g/cm ³ | 1.89 | 1.90 | 1.94 | 1.95 | 1.94 |
| Q, kJ/kg | 5230 | 8350 | 8600 | 8500 | 8280 |

In general, the smaller Al particle size the higher is EH except for Al(0.1) which shows smaller EH than Al(7). This can be attributed to lesser content of Al in Al(0.1).

CONCLUSIONS

As a whole, our investigations into detonation performances of aluminized compositions based on HE of different classes have shown that Al influence depends mainly on HE nature and Al particle size. The positive effect is observed either for HE with positive OB or for those with negative OB, but with high content of hydrogen and low content of carbon in a molecule of high explosive.¹⁴ It was expected that the decrease of Al particle size would gain in the positive effect. Benefits of nanosize Al are obvious only when characteristic times of Al interaction with DP are rather small, and the leading role is of specific surface area (see the data on DV pressure profiles and temperature ones). However when the characteristic time is larger, as for acceleration ability, it was found to be the largest for particles with size about several micrometers. It can be explained by the fact that the decrease of Al particle size up to sub-micron and nanometric size is accompanied with augmentation of the fraction of Al oxide film, and, hence, with a drop of activity of Al. Besides, particles of tested high explosives have the average size of about tens of micrometers. Hence, reduction of Al particle size only cannot completely solve the problem of augmentation of heat release rate in the course of Al oxidation in detonation products.

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