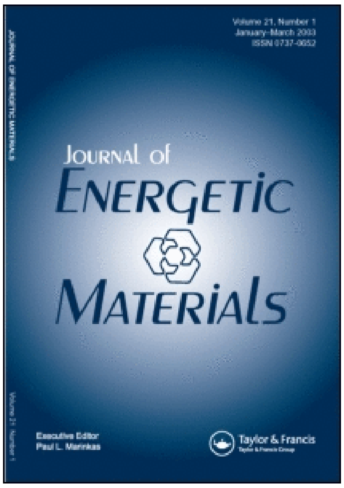

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A STUDY ON DETONATION CHARACTERISTICS OF PRESSED NTO

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Abstract. NTO is an explosive of current interest. It has been evaluated as an insensitive component to replace RDX in some explosive and propellant compositions. In our work, efforts were made to determine NTO detonation pressure and JWL equation of state of NTO detonation products. With this end in view, pressed NTO cylinders were studied experimentally by measuring the detonation velocity versus the diameter, by performing the cylinder expansion test and a water test using a SCANDIFLASH X-ray set. The results of measurements and numerical modelling of the process of copper tube expansion as well as the process of detonation of NTO charges inside a cylindrical layer of water were the basis for determining the detonation pressure and the JWL constants.

1. INTRODUCTION

3-Nitro-1,2,4-triazol-5-on (NTO) is widely accepted as an useful explosive combining comparatively high performance and insensitivity to mechanical stimulus comparable to TATB. Since the

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early 1980s when NTO was recognised as a potential explosive molecule, it has been a subject of many experimental and theoretical investigations[1+10]. NTO has been found to be far less sensitive to impact and shock than RDX and HMX whereas its explosive output is comparable to that of RDX. For these reasons NTO has appeared to be very useful for systems in which its insensitivity is more important than maximum performance. Now, despite the fact that its thermal stability (decomposition temperature of about 270 °C) is not as good as that of TATB, it is manufactured on an industrial scale being one of the most effective insensitive high explosives[8+10].

The main objective of the study was to characterise thoroughly explosive properties of NTO synthesised in our laboratory. To this end, we measured detonation velocity, carried out the water test and the cylinder test. Results of the experiments were the basis for determining such important characteristics of NTO as its detonation pressure, acceleration ability, effective exponent of isentrope and JWL equation of state for its detonation products.

2. NTO SYNTHESIS AND MAIN PROPERTIES

Synthesis of NTO was carried out in two steps including condensation of semicarbazide hydrchloride and formic acid to triazolone (TO) and its nitration to form NTO[1], Fig. 1. The final product of the synthesis was obtained by crystallisation from water. Its purity was at least 98 % what was determined by both HPLC analysis and potentiometric technique according to testing methods proposed in Ref.[11].

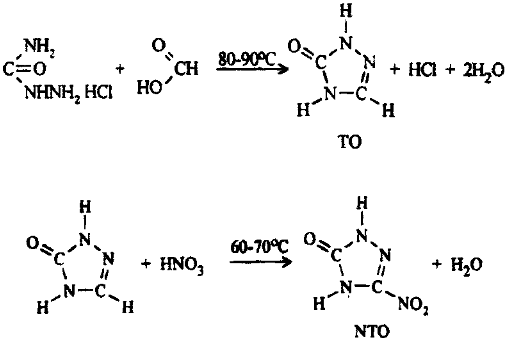

Fig.1. TO Synthesis and TO Nitration Reactions

The recrystallised NTO was ground and sieved in order to ensure the same grain size distribution in all tests. The mean particle size was about 130 μm and the crystals had cuboid shape, Figs 2 and 3.

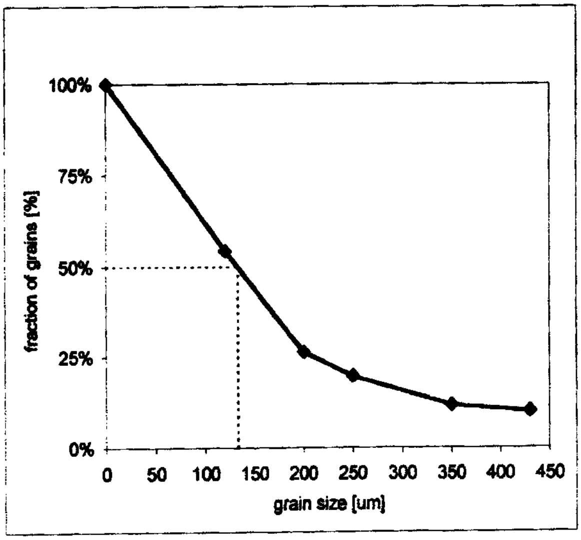

Fig.2. NTO Grain Size Distribution

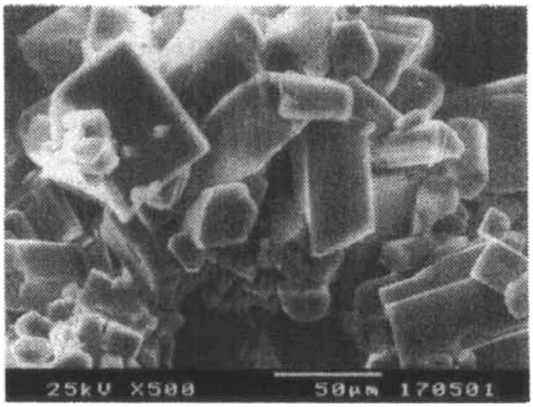

Fig.3. SEM Photograph of NTO Particles Used in Experiments

3. NTO DETONATION VELOCITY (VOD)

NTO was pressed at a density of 1.80±0.01 g/cm^3 in the form of cylindrical pellets with a diameter to length ratio of one. The pellets were 14.5 mm, 16.0 mm, 20 mm, 25.0 mm and 30.0 mm in diameter. For each diameter, a charge was made from a minimum of seven pellets. The experimental charges were initiated with a booster made of phlegmatised RDX at a density of 1.6 g/cm^3 that had the charge diameter and length of twice the diameter. In each charge, there were three measuring distances of detonation velocity. The courses were 20 mm in length, at least as they included one 20-mm, 25-mm, or 30-mm pellet or two 14.5-mm or 16-mm pellets. In order to exclude any influence of the initiation pulse on the velocity measured, the first short-cirquit sensor (consisting of two thin insulated copper wires) was fixed at a distance of triple a given charge diameter from the booster. The next sensors were located behind consecutive one or two pellets. Electrical signals from the sensors were recorded with 10-channel chronometer at accuracy of 10 ns. The distance between two sensors was measured with accuracy of 20 µm so the average error for any one datum of detonation velocity was no higher than ±40 m/s. Results of the measurements are given in Tab. 1. Each value is an average of three experimental results.

TABLE 1

Results of Detonation Velocity Measurements

Diameter [mm]	Density [g/cm^3]	Detonation Velocity VOD [m/s]
14.5	1.81	No Go
16.0	1.80	7650
20.0	1.81	7800
25.0	1.80	7820
30.0	1.80	7860

The values of VOD obtained in this work are comparable to the results for coarse NTO published in Ref.[7], where it was shown that the detonation velocity was always higher for fine NTO than for coarse one. Comparatively high critical diameter of NTO (16 mm) indicates that the time needed to destroy its molecular structure and to create molecules of detonation products is relatively long. Low rate of chemical reactions in detonation wave signifies also low shock sensitivity of NTO.

4. NTO DETONATION PRESSURE

To determine the detonation pressure of NTO, a variant of the aquarium test was applied[12]. In this method, profiles of an oblique shock wave propagating in a cylindrical layer of water during detonation of a cylindrical charge of an explosive tested is recorded with a X-ray set. The experimental profiles are then compared with results of numerical modelling of the expansion process which are in a form of relation between the position of the front of oblique shock wave in water and the exponent of isentrope (γ) of detonation products. The value of γ corresponding to the solution that overlaps the experimental profile is accepted as the exponent sought. Having determined γ, the detonation pressure is calculated according to the following equation:

$$p_{CJ} = \frac{\rho_0 D^2}{\gamma + 1} \qquad (4.1)$$

where D, p_{CJ} denote the detonation velocity and pressure, respectively, and ρ_0 is a density of the explosive tested.

The scheme of the experimental arrangement, used in this study, is shown in Fig. 4. A cylindrical NTO charge of 23.6 mm in diameter and 250 mm in length was placed inside a PCV tube with an inner diameter of 71 mm and wall thickness of 2 mm. The tube was filled with water. Short-cirquit sensors were located in the charge to measure detonation velocity and trigger the X-ray apparatus.

In each X-ray photographs, we observed a curved detonation wave front. This is a characteristic feature of explosives with a wide zone of chemical reactions that also means a low rate of the reactions. Therefore, it can be stated that the rate of chemical reaction in NTO detonation wave is fairly low if compared to other high explosives.

The density and detonation velocity of the NTO charges used in the water tests and results of the tests are given in Tab. 2. The detonation pressure was determined by comparison of measured and calculated positions of the shock wave front in a plane section located on a distance of one charge radius from the front of detonation wave. Table 2 also contains some theoretic values of the parameters calculated with the thermochemical code CHEETAH[13]. The BKWC set of coefficients of the Becker-Kistiakovski-Wilson equation of state was applied.

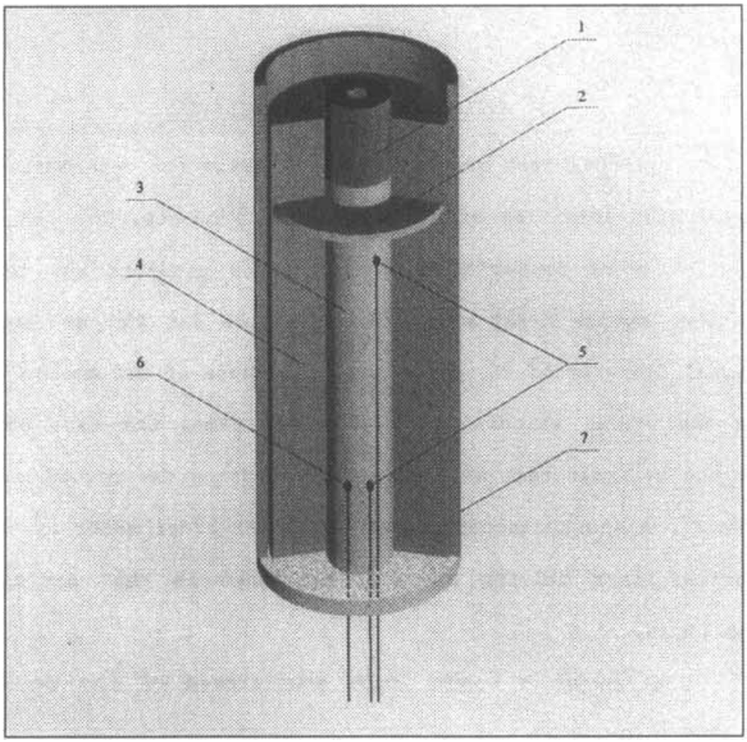

Fig.4. Water Test - Experimental Arrangement, 1 - booster, 2 - centring ring, 3 - NTO charge, 4 - water, 5 - sensors to measure VOD, 6 - sensor to trigger the X-ray set

TABLE 2

Experimental and Calculated Detonation Properties of NTO

Shot No	Density [g/cm^3]	VOD[m/s]		Exponent γ		p_{CJ} [GPa]	
		Water Test	Calc.	Water Test	Calc.	Water Test	Calc.
1	1.78	7790	8000	3.40	3.48	24.6	24.98
2	1.78	7800		3.40		24.6	

The consistence of experimental and calculated values is quite good. However, the theoretical values of VOD and p_{CJ} are still a bit higher than experimental ones. This fact implies that the conditions of detonation wave propagation in NTO charges confined with water do not enable to achieve the ideal regime of detonation.

5. NTO ACCELERATION ABILITIES

The cylinder test results were the basis for determination of acceleration abilities of NTO detonation products. The process of acceleration of copper tube by detonation products was recorded with the impulse X-ray apparatus. The tube was 250 mm long with internal diameter of 25 mm and wall thickness of 2.5 mm. To determine the radial velocity of the copper tube, the data obtained from the cylinder test were recalculated using the method proposed in Ref.[14]. A short description of the theoretical model of the cylindrical-liner driving process, published in that article, is given below.

A diagram of a copper tube accelerated by the detonation products is shown in Fig. 5

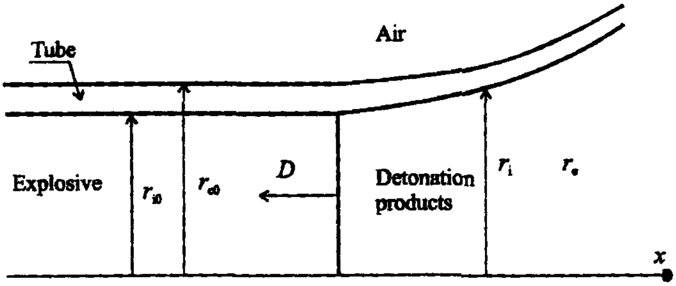

Fig.5. Diagram of Copper Tube Accelerated by Detonation Products

A plane detonation wave propagates at a velocity of D in a cylindrical charge of explosive. The gaseous detonation products expand, thus driving the tube. It is assumed that the time of detonation wave propagation is long enough to neglect the influence of the initiation of detonation. Then the motion of the detonation products and the tube material can be treated as stationary. Therefore, the axis co-ordinate and time are associated by the following relation

$$x = D \cdot t\,. \tag{5.1}$$

First, the position of the central cylinder surface is determined. Assuming a complete incompressibility of the tube material, this position can be established from the relation

$$r_m = \sqrt{r_e^2 - \frac{1}{2}\left(r_{e0}^2 - r_{i0}^2\right)}\,, \tag{5.2}$$

where r_{e0} and r_{i0} denote the initial radii of external and internal surfaces of the tube, respectively, r_e, and r_m mean the radii of external and central surfaces of the tube for given value of co-ordinate x, respectively.

By using the relation (5.1) we can replace the dependence of the tube radius on axis co-ordinate by the time function of this radius. The time dependence of central position of the tube is approximated by the following function

$$r_m = r_{m0} + \sum a_i \left\{b_i\left(t - t_0\right) - \left[1 - \exp\left(-b_i\left(t - t_0\right)\right)\right]\right\}, \tag{5.3}$$

where a_i, b_i, t_0 are parameters. In paper[14], it was proved that sufficient accuracy of approximation of the experimental results was achieved by assuming i = 2 in (5.3).

From the function (5.3), the radial velocity of the central part of the tube can be expressed by the relation

$$u_{\mathrm{m}} \equiv \frac{d\,r_{\mathrm{m}}}{d\,t} = \sum a_i\, b_i \left[1 - \exp\left(-\,b_i\,(t - t_0)\right)\right]. \qquad (5.4)$$

To determine the kinetic energy of the tube, the magnitude of velocity of the tube element must be calculated. From geometrical relations it follows that the deflection angle Θ (the angle between the line being tangential to the trajectory of the central surface of the tube and the x-axis) are specified by the relation

$$\Theta = \mathrm{arctg}\left(\frac{u_{\mathrm{m}}}{D}\right). \qquad (5.5)$$

On the other hand, the magnitude of velocity of the central part of the tube (a cylindrical liner) is expressed by the equation

$$u_{\mathrm{L}} = 2D\sin\left(\frac{\Theta}{2}\right). \qquad (5.6)$$

The acceleration ability of explosive can be described by so-called *Gurney energy,* which is defined as a sum of kinetic energies of driven tube and detonation products related to unit mass of explosive. For cylindrical envelopes, the Gurney energy is expressed by the following relation[14]

$$E_{\mathrm{G}} = \left(\mu + \frac{1}{2}\right)\frac{u_{\mathrm{L}}^2}{2}, \qquad (5.7)$$

where μ denotes the ratio of tube mass to explosive mass.

Applying this methodology to analyse X-ray records of expanding copper tubes, the dependence of the Gurney energy on the relative volume of detonation products of NTO, TNT, and TNT/RDX = 50/50 were constructed. The results of the analysis are presented in Fig. 6. Characteristics of the explosives are given in Table 3.

Charges of NTO with density 1.77 g/cm^3, placed in the copper tube, detonate at 7940 m/s. This value is almost the same as a calculated one (7960 m/s), what indicates the ideal regime of NTO detonation under these conditions.

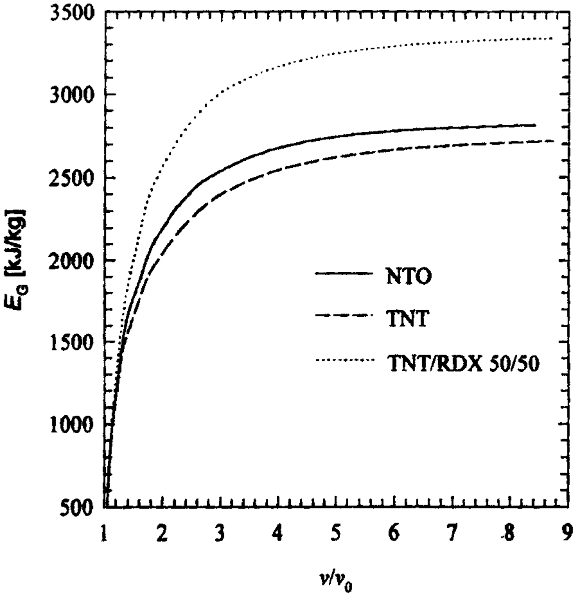

Fig.6. Dependence of the Gurney Energy on the Relative Volume of Detonation Products

From the analysis of curves shown in Fig. 6, it follows that the changes in the Gurney energy of NTO during the tube expansion are similar to those of other high explosives. NTO acceleration abilities are only slightly higher than that of TNT. Likely reasons for such a poor acceleration ability of NTO are discussed at the end of Section 8.

6. NTO DETONATION ENERGY

The results of cylinder test can also be used to estimate the detonation energy. In Ref.[14] it was shown that there was a correlation between the velocity of driven tube at the infinitive volume of the detonation products and the detonation energy of an explosive.

The relation can be written down as follows

$$\frac{E_0}{E_0^s}=\frac{\left(\mu+\frac{1}{2}\right)\rho_0}{\left(\mu^s+\frac{1}{2}\right)\rho_0^s}\left(\frac{u_L}{u_L^s}\right)^2 \qquad (6.1)$$

where E_0 and E_0^s are the detonation energy of a given explosive and a standard explosive, respectively u_L and u_L^s denote the tube velocity determined at the infinitive volume of detonation products of the explosives, μ and μ_s denote the ratio of tube mass to explosive mass, ρ_0 and ρ_0^s are densities of the explosives.

In order to estimate the velocities u_L and u_L^s, the velocity of copper tube was determined from the results of cylinder test and the dependency of velocity square on reciprocal volume of detonation products was constructed. After that, the dependence was extrapolated to null and the velocities corresponding to the infinitive volume were found.

Using phlegmatized RDX as a standard explosive for which the detonation energy (E_0^s = 5263.5 kJ/kg) was taken from Ref.[15], the detonation energies of the explosives tested (E_0) was calculated from (6.1). The values obtained are given in Tab. 3

TABLE 3

Detonation Energy Estimated on the Basis of Cylinder Test

Explosive	Density ρ_0 [g/cm^3]	VOD [m/s]	Detonation Energy E_0 [MJ/m^3]
NTO	1.77	7940	7100
TNT	1.59	6910	6830
TNT/RDX 50/50	1.64	7610	8100

From Table 3, it follows that the detonation energy of NTO is a bit higher than that of TNT but considerably lower than for TNT/RDX = 50/50. This seems to be one of the reasons that justify the poor acceleration ability of NTO.

7. NTO EFFECTIVE EXPONENT OF ISENTROPE

So-called *effective exponent of isentrope* is often used to calculate the detonation parameters of explosives or some characteristics of the detonation products. The value of the effective exponent is not determined from the parameters in the CJ point but on the basis of the real isentrope of the detonation products[16]. The effective exponent of isentrope can be estimated from results of the cylinder test. In Ref.[17] the effective exponent is determined by comparison of the experimental profile of the copper tube with that obtained from numerical modelling of the expansion process. The detonation products, driving the tube, are described by the constant-γ equation of state. The algorithm used to determine the effective isentropic exponent is as follows.

The problem of driving the cylindrical liner is solved numerically for n values of the exponent γ_i $(i = 1, n)$. For each γ_i a discrete dependence of the outer tube radius on the axial co-ordinate is derived. This dependence is interpolated by spline functions and the values of $r_{ej}(\gamma_i)$ at chosen points x_j (j = 1, m) are calculated. The effective exponent γ_{ef} is determined by minimising the function

$$f(\gamma)=\sum_{j=1}^{m}\left[r_{ej}-r_{ej}(\gamma)\right]^{2}, \tag{7.1}$$

where r_{ej} is the experimental dependence obtained from the cylinder test.

This method was applied in the present work to estimate the effective exponent of isentrope for NTO detonation products. The experimental and calculated profiles of the copper tubes driven by the detonation products of NTO and TNT (for comparison) are presented in Fig. 7. Density and detonation velocity of the explosives are listed in Table 3.

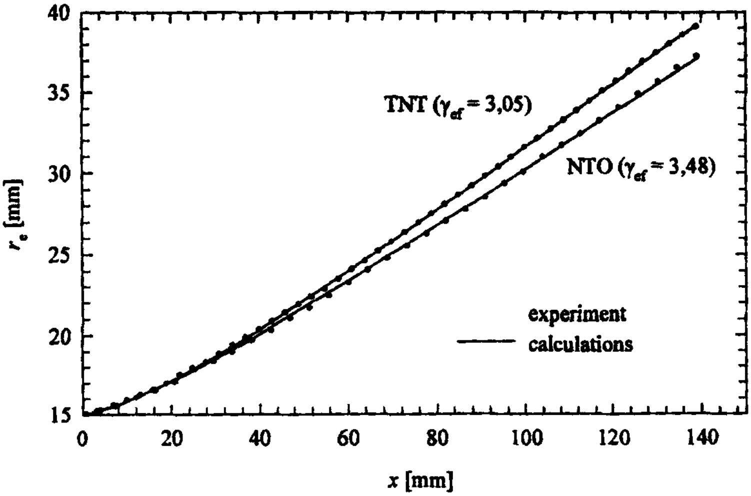

Fig.7. Experimental and Calculated Profiles of Tubes Driven by Detonation Products of NTO and TNT

A satisfying conformity of experimental and theoretical profiles were achieved when the effective exponents (γ) were 3.48 and 3.05 for NTO and TNT, respectively. Reliable value obtained for TNT detonation products confirms correctness of the method applied.

8. NTO JWL ISENTROPE

Jones, Wilkins and Lee proposed the equation of the isentrope for the detonation products of explosives in the following form[18,19]

$$p = Ae^{-R_1 V} + Be^{-R_2 V} + CV^{(-1-\omega)} \qquad (8.1)$$

where $V=v/v_0$. The following equation of state (JWL EOS) corresponds to this isentrope

$$p = A\left(1-\frac{\omega}{R_1 V}\right)e^{-R_1 V} + B\left(1-\frac{\omega}{R_2 V}\right)e^{-R_2 V} + \frac{\omega\ E}{V} \qquad (8.2)$$

where A, B, C, R_1, R_2 and ω are constants for given explosive. The basic method of determination of these coefficients is cylinder test. Besides it, some connections between coefficients following from the conservation laws written for the CJ point are used in this method[20]. As a result, parameters A, B, and C are expressed as functions of R_1, R_2, ω and ρ_0, D, E_0 and p_{CJ}. Density of explosive ρ_0 as well values of detonation velocity D, detonation energy E_0 and pressure p_{CJ} at the CJ point are established experimentally. Thus, only the constants R_1, R_2 and ω remain to be determined.

They are calculated by the method in which the experimental dependence of radial displacement of the outer tube wall on the axial co-ordinate is compared with that obtained from a numerical simulation[20]. The set of JWL constants is chosen for which the experimental and simulated displacements are sufficiently close to each other. The R_1, R_2 and ω are obtained from comparison of the experimental and calculated radial position of the tube wall at chosen m values of the axial co-ordinate x_j. So, the values of these parameters are determined by minimising the function

$$f(R_1,R_2,\omega)=\sum_{j=1}^{m}\left[r_{ej}-r_{ej}(R_1,R_2,\omega)\right]^2 , \tag{8.3}$$

where r_{ej} i $r_{ej}(R_1, R_2, \omega)$ are the experimental and calculated positions of external surface of the tube, respectively.

Using the model, proposed in Ref.[20], as well as the values of detonation velocity, detonation pressure and detonation energy of NTO, obtained with methods described in Sections 2, 4 and 6, the constants of the JWL equation of state of NTO were calculated. Results of the calculations are given in Tab. 4.

TABLE 4

JWL Isentrope of NTO

Explosive/Characteristics	JWL EOS Constants
NTO	A = 1025.489 GPa,
ρ_0 = 1.77 g/cm^3,	B = 8.4815229 GPa,
VOD = 7940 m/s,	C = 0.7041268 GPa,
P_{CJ} = 25.4 GPa,.	R_1 = 5.03, R_2 = 1.20,
	ω = 0.25, E_0 = 7.1 GPa.

After determining the JWL isentrope, the expansion work of detonation products was calculated from equation:

$$w(v) = -e_c + \int_{v_{CJ}}^{v} p_i dv' \qquad (8.4)$$

where $e_c = (p_{CJ} - p_0)(v_0 - v_{CJ})/2$ denotes the energy of explosive compressed at the front of detonation wave, p_i is the pressure on the isentrope.

Dependence of the expansion work on the relative volume of detonation products is presented in Fig. 8.

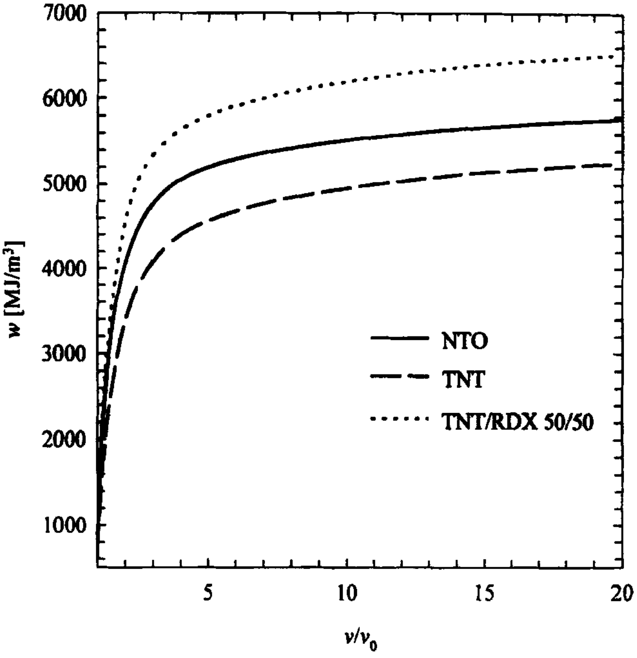

Fig.8. Expansion Work as a Function of the Relative Volume of Detonation Products

NTO expansion work is higher than that of TNT by about 10 %, and it reaches 5800 MJ/m^3 at the relative volume of detonation products of 20. It is a surprising outcome especially if we take into consideration the fact that density and detonation parameters (detonation velocity and pressure) of NTO are considerably higher than those of TNT. However capability to perform work and metal acceleration ability depend also on values of the detonation energy and the isentrope exponent (γ and γ_{ef}). The detonation energy of NTO (Table 3) is only a bit higher than TNT detonation energy whereas the effective exponent of isentrope of NTO ($\gamma_{ef} = 3.48$) is substantially higher than that of TNT ($\gamma_{ef} = 3.05$). High value of the exponent means that alongside an increase in the volume of detonation products their pressure decreases sharply and, consequently, their influence on surroundings is less effective.

The values of detonation energy and isentrope exponent are affected mainly by the composition of detonation products. From an analysis of detonation product composition in the CJ point, calculated with CHEETAH, it follows that nitrogen constitutes almost 50 % of gaseous NTO detonation products. In the case of TNT and TNT/RDX mixture detonation products contain only 25 % and 33 % of nitrogen, respectively. Therefore we are of opinion that the composition of gaseous detonation products, especially the presence of high amount of nitrogen, is the reason for comparatively poor NTO acceleration ability.

SUMMARY

1. NTO detonation velocity is quite high. It attains 7860 m/s in unconfined charges at a density of 1.8 g/cm^3. With confinement in a form of copper tube, used in the cylinder test, the velocity reaches 7940 m/s at density of 1.77 g/cm^3. Critical diameter

of detonation of NTO with mean particle size of 130 μm is 16 mm in charges pressed at 1.8 g/cm^3.

2. The pressure of detonation of NTO pressed at 1,78 g/cm^3 is 25.4 GPa. This value is comparable to that of Composition B.
3. The velocity of copper tube, driven by NTO detonation products, increases rapidly in the initial stage of expansion and, then, the increase is slow. The final acceleration ability of NTO is comparable to that of TNT:
4. Detonation energy of NTO at density 1.77 g/cm^3 is a bit higher than that of TNT and reaches 7100 MJ/m^3.
5. The effective exponent of isentrope is 3.48. The set of constants of JWL EOS is as follows: A = 1025.489 GPa, B = 8.4815229 GPa, C = 0.7041268 GPa, R_1 = 5.03, R_2 = 1.20, ω = 0.25, E_0 = 7.1 GPa. The expansion work of NTO, calculated with using the JWL EOS, is higher by about 10 % than that of TNT.

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