INFLUENCE OF INITIAL DENSITY ON THE REACTION ZONE FOR STEADY-STATE DETONATION OF HIGH EXPLOSIVES

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The laser interferometric system VISAR was used to investigate the detonation waves structure of pressed RDX, HMX, TNETB, and ZOX with different initial density. The experimental results are the surface velocity profiles of foils placed at the boundary between a HE sample and a water window. Critical initial densities ρ_c at which the reaction zone structure changes crucially were found: Von Neumann spike was recorded if the density was less than the critical value, otherwise monotone pressure increase in the reaction zone was observed. The ρ_c is equal to 1.72 g/cm^3, 1.84 g/cm^3, 1.56 g/cm^3, and 1.71 g/cm^3 for RDX, HMX, TNETB and ZOX respectively. The results obtained for RDX demonstrate that the ρ_c essentially depends on the sample structure and is determined not only by the HE particle size, but also by the pressing process. The results of this work can be explained by growth of the initial decomposition rate of an explosive with the increase of density if we assume that physicochemical transformations take place in a compression wave.

INTRODUCTION

According to the classical theory,[1] the detonation wave consists of a shock jump and a chemical reaction zone, in which the pressure decreases and the matter expands, i.e. Von Neumann spike is shaped. The Chapman-Jouguet point, where particle velocity relative to the shock front is equal to the local sound velocity, ends a steady-state reaction zone. Inside of this zone a subsonic flow takes place and the conditions to supply chemical energy to the shock front are realized. Such flow structure allows us to establish the selection rule for detonation wave velocity. Numerous experimental data confirm the validity of this model for heterogeneous high explosives (HE). However, it was found [2] that in RDX and HMX at high initial density the pressure increases in the reaction zone and the spike does not form. Papers [3-5] contain indirect confirmation of existence of similar detonation wave structure in high density RDX and PETN: the authors stated that they did not observe the spike.

The detonation wave without Von Neumann spike does not correspond to the classical model. Moreover, it is not clear, whether the Chapman-Jouguet state will be reached, and what the selection rule of detonation velocity is in this case. To solve these key theoretical problems it is necessary (1) to understand if the observed phenomenon is the unique property of RDX and HMX, or it reflects the property common for powerful explosives, (2) to define the initial density at which the reaction zone structure changes crucially. For these purposes the experimental investigation of the reaction zone transformation under initial density increase in pressed RDX ($C_3H_6N_6O_6$), HMX ($C_4H_8N_8O_8$), ZOX ($C_6H_8N_{10}O_{16}$, Bis (2, 2, 2 – Trinitroethyl - N - nitro) Ethylenediamine), and TNETB ($C_6H_6N_6O_{14}$, Trinitroethyl trinitrobutyrate) was conducted.

THE SCHEME OF EXPERIMENTS

The scheme of experiments is shown in Figure 1. The detonation in the samples was initiated by a shock wave with amplitude ~4 GPa, created by the HE plane generator (1). The diameter of the charges was 30 mm, the length changed from 40 up to 80 mm. Steady-state detonation wave was formed at these geometrical sizes. The wave profiles were registered by laser interferometer VISAR with 2 ns time resolution and an accuracy of the velocity determination was equal to ±10 m/s. The laser beam reflected from a 100 - 400 μm aluminum foil (2) placed between the charge and the water window (3). As the result of the
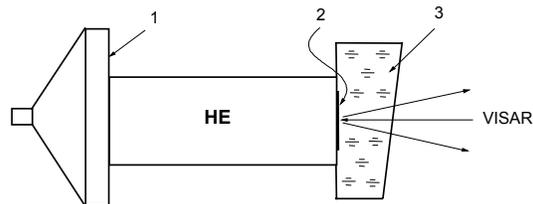

FIGURE 1. THE SCHEME OF EXPERIMENTS

experiment we have the velocity of the foil - water border, which represents all the details of the reaction zone structure in detonation wave.

EXPERIMENTS AND RESULTS

RDX and HMX

RDX samples of different initial density ρ_0 were pressed from powder with a mean particle diameter of 80 μm. To obtain ρ_0 in the range of 1.50 g/cm^3 to 1.74 g/cm^3, a small amount of acetone (less than 1 wt%) was added into pressed RDX (RDX_1). Higher density, up to 1.776 g/cm^3, was reached by increase of acetone quantity up to 10 wt% and by curing of the samples under pressure (RDX_2).

TABLE 1. PARAMETERS OF EXPERIMENTAL SETUP FOR RDX.

N	ρ_0, g/cm^3	h_{Al}, μm	$\varnothing/l,mm/$ mm	Comments
1	1,51	400	30/45	spike, 70ns, RDX_1
2	1,51	200	30/45	spike, RDX_1
3	1,60	400	30/45	spike, 70ns, RDX_1
4	1,60	200	30/50	spike, RDX_1
5	1,69	200	30/40	spike, RDX_1
6	1,73	200	30/52	increase, 30 ns, RDX_1
7	1,72	200	30/40	spike, 40ns, RDX_2
8	1,776	200	30/40	spike, 30ns, RDX_2
9	1,68	200	30/45	spike, >40 ns, RDX_1
10	1,68	200	30/45	spike, >40 ns, RDX_1
11	1,74	200	30/40	increase, RDX_1
12	1,74	200	30/40	increase, RDX_1

The experimental results for RDX_1 are presented in Figures 2, 3. The initial density ρ_0, foil thickness h_{Al}, ratio of HE sample diameter $\varnothing$ to its length l, and comments concerning the reaction zone structure and characteristic time are given in Table 1. The
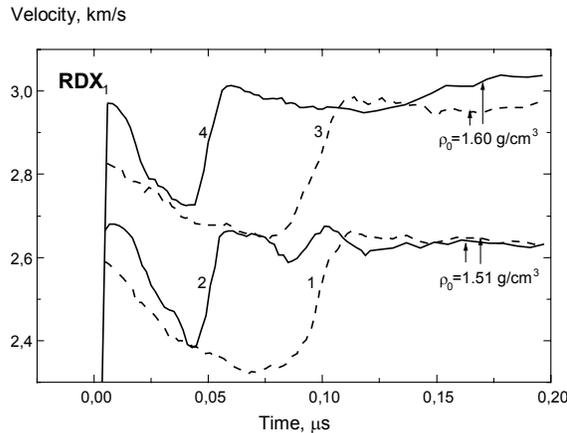
Velocity, km/s

FIGURE 2. THE VELOCITY OF AL FOIL-WATER WINDOW BOUNDARY FOR RDX_1 AT LOW INITIAL DENSITY.

Velocity, km/s

FIGURE 3. THE VELOCITY OF AL FOIL-WATER WINDOW BOUNDARY FOR RDX AT HIGH INITIAL DENSITY.

determination accuracy of the characteristic time does not exceed ±5 ns. When the initial density is less than 1.72 g/cm^3 (Figure 2) the velocity of the foil - water boundary decreases after the shock jump. Duration and amplitude of the spike are determined by Von Neumann spike in RDX. The subsequent velocity increase is caused by circulation of compression and rarefaction waves in Al foil. The attenuation of the velocity spike is recorded during its propagation through the foil. The spike amplitude decreases by 100-200 m/s as the foil thickness increases from 200 μm up to 400 μm. Therefore to find the exact value of the spike amplitude it is necessary to carry out experiments with thinner foils and to extrapolate their thickness to zero. But it was not the aim of this paper because we were interested in the qualitative change of the reaction zone structure when the initial density increases. Relatively thick foils can be used for that purpose.

For initial densities of 1.51 g/cm^3 and 1.60 g/cm^3 (Figure 2) the velocity profiles show inflection at ~30 ns while the total spike duration is equal to 70 ns. That can reflect the two-step decomposition of RDX in detonation wave. Similar chemical reaction of TNT have been observed previously.[6] The fist rapid
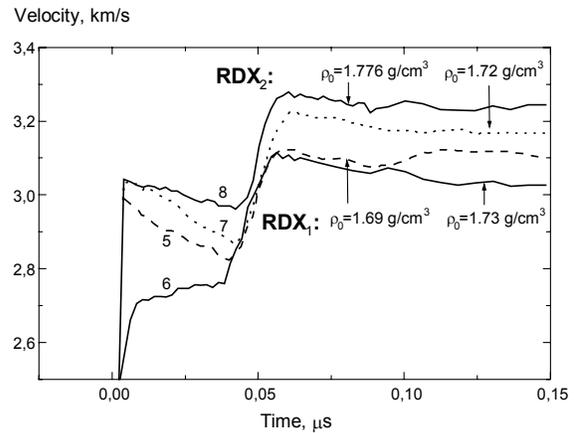
reaction (80 ns) was interpreted as the formation of gas detonation products and the second stage over an additional 200 ns was attributed to diffusion controlled solid carbon particle formation. The velocity profiles inflection in Figure 2 may be the result of similar chemical reaction of RDX.

The velocity spike duration changes insignificantly, but its amplitude notably drops as the density increases. When ρ_0 exceeds ρ_c=1.72 g/cm^3 the situation changes crucially: instead of velocity decrease after the shock jump the monotonic increase is observed (curve 6 for ρ_0 =1.73 g/cm^3 in Figure 3). It is necessary to note, that the particle velocity of explosion products increases with the increase of initial density except for critical density, where the abnormal velocity behavior is registered. For ρ_0= 1.73 g/cm^3 the particle velocity is the same as for 1.69 g/cm^3, whereas it has to increase by more than 50 m/s. It is possible to explain this by transition to underdriven detonation when Von Neumann spike disappears.

Figure 3 shows the experimental data for RDX_2. Here the velocity spike is registered for all initial densities. This result differs essentially from the result that one would
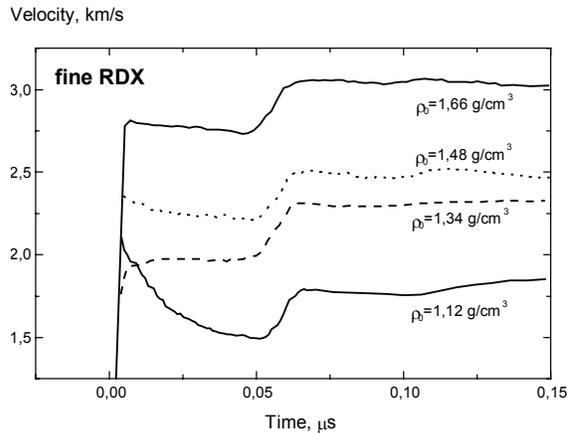
Velocity, km/s

FIGURE 4. THE VELOCITY PROFILES FOR FINE RDX. h_{Al}=200 μm.

expect on the basis of the RDX_1 data extrapolation to higher densities. The spike amplitude drops approximately twice when the ρ_0 increases from 1.72 up to 1.776 g/cm^3, and, by analogy with RDX_1, it is possible to expect that for RDX_2 $\rho_c \approx 1.78$ g/cm^3. This conclusion agrees with the data.[2] It can also be seen, that the particle velocity at 1.72 g/cm^3 density (curve 7) is approximately 100 m/s higher than the velocity at 1.73 g/cm^3 density for RDX_1, when the spike fades. That fact confirms that underdriven detonation is possible when the density exceeds ρ_c.

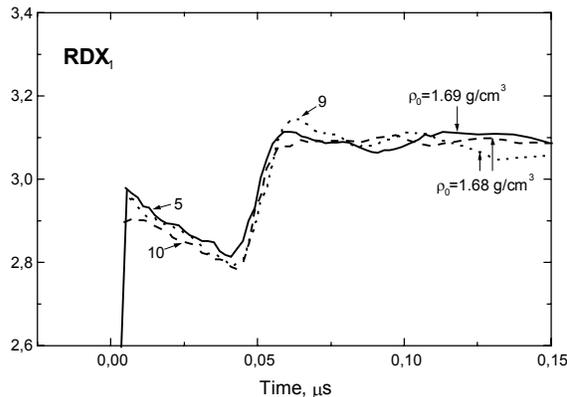The obtained results demonstrate that the

ρ_c depends essentially on the sample structure and is determined not only by the RDX particle size, but also by the pressing process. It is known,[7] that at pressing many of the explosive particles are cracked and sheared. Pressing with a small quantity of acetone (RDX_1) creates a lot of the potential centers of reaction, and the decomposition rate and explosive part reacting in a shock front increase. Therefore the detonation wave without Von Neumann spike is formed at $\rho_0 >$ 1.72 g/cm^3. Pressing with a large quantity of acetone (RDX_2) gives the same density at smaller damage of RDX particles, which decreases the decomposition rate and the spike is recorded.

It follows from these results that the reaction zone structure has to depend on both the initial density and the initial particle size of HE powder. To check this assumption the RDX samples were pressed from powder with a mean particle diameter of 5 μm. Figure 4 shows the experimental results for fine RDX. Maximum initial density did not exceed 1.66 g/cm^3, but from this figure we notice that the amplitude and duration of fine RDX are smaller than those of coarse RDX at the same density. It is conceivable that the critical density of fine RDX is below 1.72 g/cm^3. The most unusual result is the disappearance of
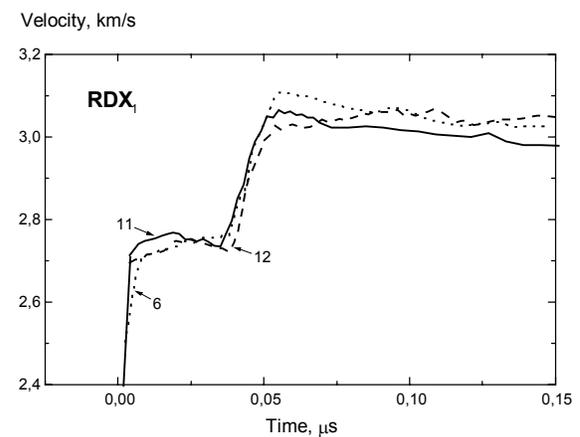
Velocity, km/s

FIGURE 5. EXPERIMENTAL DATA FOR RDX_1 AT $\rho_0 < \rho_c$.

Velocity, km/s

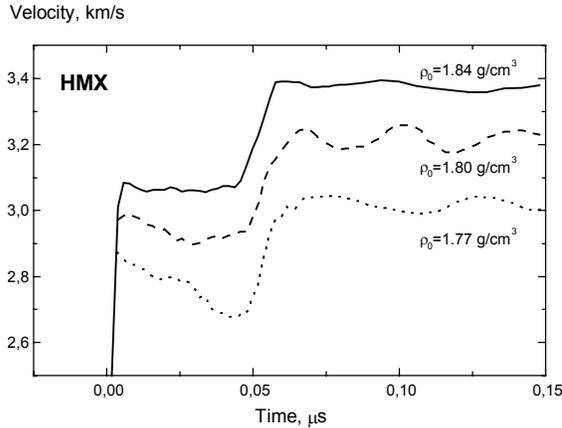**FIGURE 6. EXPERIMENTAL DATA FOR RDX_1 AT $\rho_0 \geq \rho_c$.**

Velocity, km/s

FIGURE 7. THE VELOCITY PROFILES FOR HMX. h_{Al}=200 μm.

Von Neumann spike in fine RDX at $\rho_0 \approx 1.3$ g/cm^3, whereas it is recorded at densities both below and above this density.

Reproducibility of experimental results is an important aspect for heterogeneous medium. In pressed HE the typical pore sizes change from one to few tens of microns, so one would be expect a marked difference between experimental data. But it follows from our experiments that it is not so. Figures 5, 6 show the results for initial density lower and higher than the critical density. A good reproducibility is the result of experimental setup. In the first place, the shock wave passes through the foil, the thickness of which significantly exceeds the pore sizes, and heterogeneity becomes negligible. In the second place, the foil velocity is recorded by laser interferometer which picks a signal off a spot with a diameter ~100 μm and averages the velocity too. Consequently, the replication of velocity profiles from shot to shot is good enough.

The similar experiments were conducted for HMX. Samples were pressed from powder with a mean particle diameter of 150 μm. The quantity of acetone added in the HMX at pressing did not exceed 0.5 wt%. Figure 7 shows the experimental results. The velocity

spike with the duration of ~30 ns is recorded at the ρ_0=1.77 g/cm^3. When the density is equal to ρ_c=1.84 g/cm^3 the spike disappears, and the velocity remains constant after the shock jump. This result agrees well with the data given in paper,[2] where the pressure increase in the reaction zone was found at the initial density of 1.87 g/cm^3.

TNETB and ZOX

TNETB [8] is an explosive with low negative oxygen balance (-4.15 %) and density of a single crystal 1.839 g/cm^3. The detonation heat and sensitivity of TNETB are at the level of RDX. The experimental dependence of the detonation velocity D on the initial density can be approximated by linear relationship: D=3.87+3.92(ρ_0-0.6), km/s, where the density is measured in g/cm^3. The TNETB was pressed without any solvents.

TABLE 2. PARAMETERS OF EXPERIMENTAL SETUP FOR TNETB.

N	ρ_0, g/cm^3	h_{Al}, μm	$\varnothing/l$, mm/mm	Comments
1	1.48	200	30/42	spike, 50 ns
2	1.48	400	30/42	spike, 50 ns
3	1.51	200	30/42	spike, 40 ns
4	1.56	200	30/42	plateau
5	1.61	200	30/40	increase, 30 ns
6	1.61	200	30/80	increase, 30 ns
7	1.71	100	30/38	increase, >30 ns
8	1.71	200	30/38	increase, 30 ns

The experimental results are presented in Figures 8, 9 and in the Table 2. At 1.48 g/cm^3 initial density (curve 1) the spike with ~50 ns duration is observed. The increasing of ρ_0 up to 1.51 g/cm^3 (curve 3) leads to decrease of spike amplitude, at ρ_0=1.56 g/cm^3 (curve 4) the velocity spike disappears and behind the shock jump the parameters are constant. At further initial density increase, the velocity
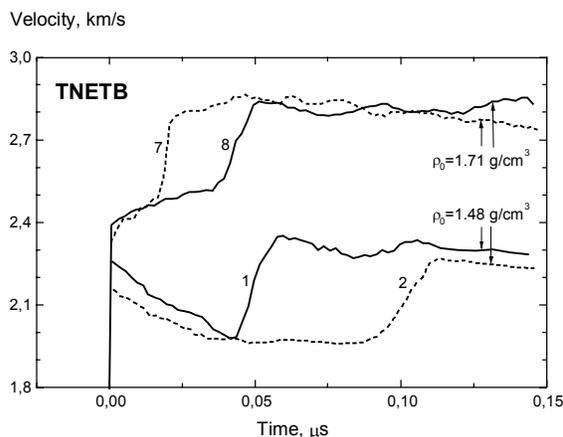
Velocity, km/s

FIGURE 8. THE VELOCITY OF AL FOIL-WATER WINDOW BOUNDARY FOR TNETB.

Velocity, km/s

FIGURE 9. THE VELOCITY OF AL FOIL-WATER WINDOW BOUNDARY FOR TNETB NEARLY ρ_c.

increases by ~100 m/s in ~30 ns time behind the jump. Good reproducibility of the experimental results is observed for the double length charge (curves 5, 6 in Figure 9). It means that steady-state detonation is realized in all the experiments. As well as for RDX, the abnormal change of particle velocity at critical density ρ_c=1.56 g/cm^3 (Figure 9) was found: for ρ_c the velocity is approximately 50 m/s lower than for 1.51 g/cm^3. It can be explained by transition to underdriven detonation at the moment of disappearance of Von Neumann
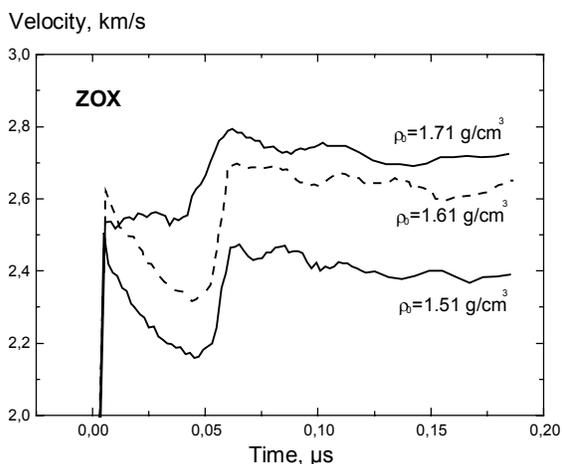
Velocity, km/s

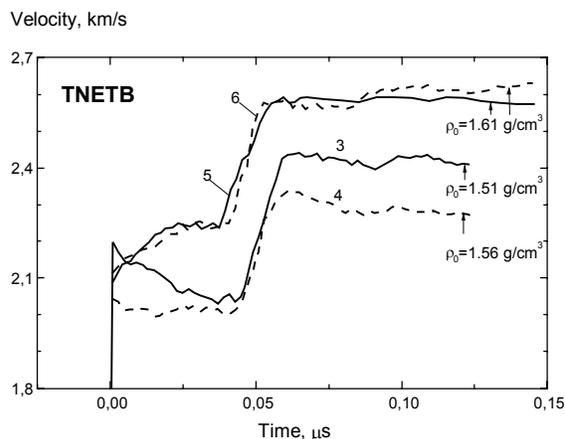**FIGURE 10. THE VELOCITY OF AL FOIL-WATER WINDOW BOUNDARY FOR ZOX. h_{Al}=200 μm.**

spike.

ZOX [9] is a powerful explosive completely balanced on oxygen, with the density of a single crystal 1.87 g/cm^3. The dependence of the detonation velocity D on the initial density can be approximated by the following relationship: $D=4.10+3.92(\rho_0-0.6)$, km/s. Figure 10 shows the experimental results for ZOX. The disappearance of Von Neumann spike is watched at ρ_c=1.71 g/cm^3: after the jump the velocity remains practically constant. Whereas at smaller initial density the spike is registered. At ρ_0=1.51 g/cm^3 its amplitude equals ~300 m/s, and duration is ~50 nc. The increase of initial density to 1.61 g/cm^3 reduces amplitude and duration of the velocity spike approximately by 20 %.

DISCUSSION

The results of this work confirm the possibility of detonation wave propagation without Von Neumann spike in a powerful HE. The reaction zone structure changes qualitatively at a critical initial density. It can be explained by growth of the initial decomposition rate of an explosive with the increase of density, if we assume that
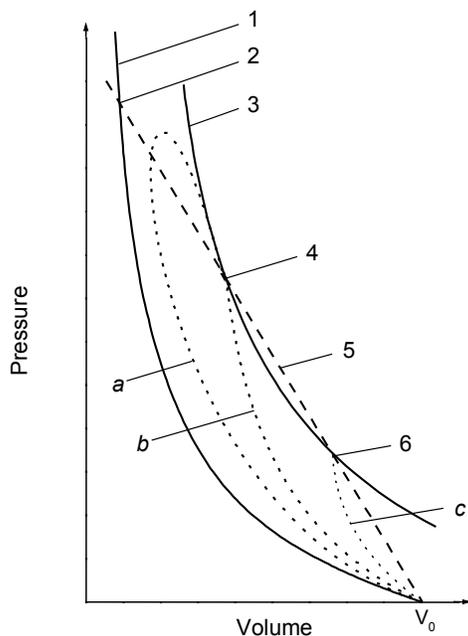
Pressure

Volume V_0

FIGURE 11. PHASE TRAJECTORY IN THE PLANE PRESSURE – VOLUME.

physicochemical transformations take place in a compression wave. The influence of RDX charge structure on the critical density agrees well with this assumption. Besides when the pressure increases in the reaction zone, the final state of detonation products can be on the weak part of a detonation Huhoniot.

Steady-state detonation wave without Von Neumann spike does not correspond to the classical model, which ignores the chemical reaction in a shock zone. Nevertheless, a strong coupling between the shock and reaction zones was found.[10 - 13] In the book [12] the main attention is given to the fact, that after the shock jump the non-equilibrium HE Hugoniot is realized. Further there is a relaxation to an equilibrium HE Hugoniot and chemical reaction.

Mathematical model of detonation phenomenon with the reaction in shock compression is analyzed in the book.[13] It is proposed that the shock front thickness is produced by viscosity. Figure 11 shows the results of these investigations in the plane pressure – volume. According to the classical theory,[1] shock jump compresses the material from the initial state to the point 2 on the Hugoniot of HE (1) and initiates the chemical reaction. Then the reaction products expand along the Rayleigh line (5) from the point 2 to the point 4 on the Hugoniot for the reaction products (3). If we consider the HE decomposition inside of the shock front, the matter state changes steadily along the trajectory *a*, which does not coincide neither with the HE Hugoniot nor with the Rayleigh line. At low decomposition rate the steady-state detonation corresponds to the classical model. The Von Neumann spike amplitude decreases only and the part of explosive reacts in the shock front. However when the decomposition rate increases the structure of detonation wave changes significantly: the final state 4 on the detonation Hugoniot is reached as a result of monotonic pressure increase (trajectory *b*). At very fast decomposition rate the final state can belong to the weak part of the reaction product Hugoniot (point 6). Thus underdriven detonation can be realized. Curve *c* in figure 11 shows the phase trajectory in this case. Attention must be paid to the next result: the selection rule of the detonation velocity gives a classical Chapman-Jouguet detonation when the final state belongs to the strong part of the reaction product Hugoniot (point 4), and it does not matter whether the Von Neumann spike exists or not. But if the underdriven detonation is realized the detonation velocity will be determined by the kinetics of chemical reaction and shock wave front, and not thermodynamics and gas dynamics only, as it takes place in the classical theory of detonation.

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