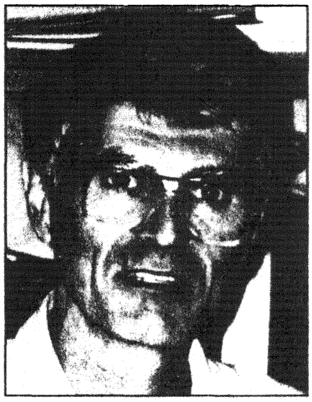

Advances and problems in DDT in solids

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Recent knowledge about deflagration-to-detonation transition (DDT) in solids has been set in outlines by the beginning of 70th (see, for instance, [1-2]). During next decades investigations were noticeable by implementation of more and more sophisticated complex experimental techniques and numerical simulation (see, for instance, [3-9]). As a result, our knowledge about DDT turned to be more argued, full and clear.

Actually, investigations into DDT cover a broad spectrum of tasks, from development and analysis of tests trying to reproduce response of specific munitions to particular stimuli, up to scientific ones aimed at exploring mechanisms and behavior of DDT. The scientific aspect is the subject of this report. Its themes are experimental methods, scenarios and mechanisms of DDT, and numerical simulation. Under consideration is state-of the art, problems, and ways to make progress.

1. EXPERIMENTAL METHODS USED FOR DDT STUDY

Well-accepted scheme of the DDT states progressive change of stages in a chain: level-by-level combustion, convective burning, low velocity detonation (or compressive burning) and full detonation. The mechanism of convective burning is connected to convective heat transfer from gaseous combustion products, filtering in pores, to pore surface. This heat transfer provides heating and ignition of explosive material. Chemical conversion of explosive material under LVD is initiated in hot spots produced in the course of compression (deformation) of material in the wave front. LVD displays all properties typical of a detonation-like process but detonation velocity's level. The low velocity is predominantly attributed to low rates of exothermic reaction and rarefaction effect of lateral expansion.

The change of the stages takes place after some conditions being reached. These conditions used to be specified by means of a set of threshold pressures.

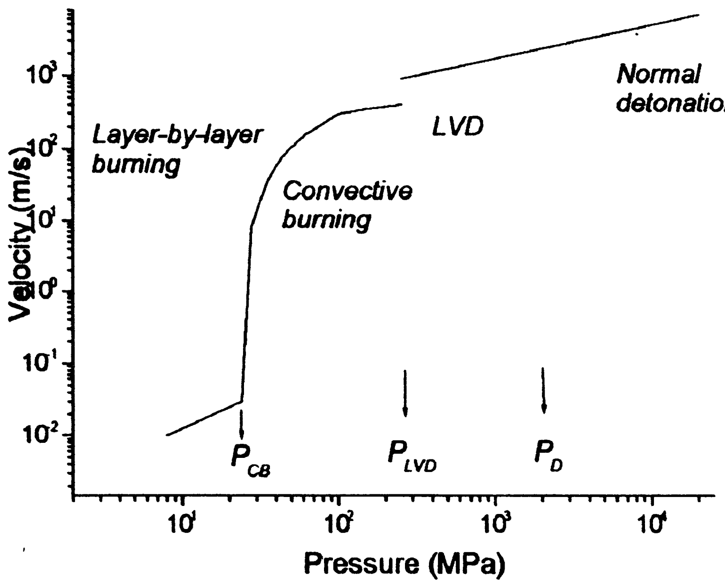


Figure 1. Schematic of DDT in coordinates of wave velocity versus wave pressure

Fig.1 illustrates a schematic of DDT for a nominal high explosive in coordinates of wave velocity versus wave pressure. Here are indicated the stages and threshold pressures : P_{CB} is critical pressure to break down layer-by-layer combustion and begin convective burning ; P_{LVD} is minimal pressure to arouse LVD ; and P_D is shock pressure to initiate full detonation. Fig. 2 and Table 1 show examples of these parameters for typical materials. The P_{CB} was determined using standard constant volume technique [1], two others were determined by means of a test of the gap test type with the strong confined charge of material tested [1,10].

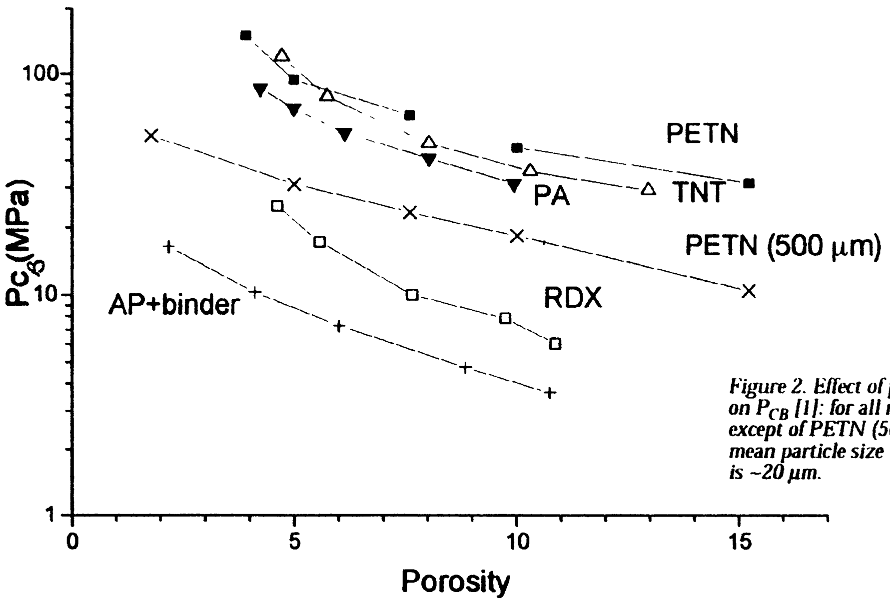


Figure 2. Effect of porosity on P_{CB} [1]: for all materials except of PETN (500 μm) mean particle size is ~20 μm.

Although these parameters are time-dependent (influence of pressure rise rate in the bomb and of profile of incoming shock in the gap test), effects of particle size and initial porosity of the charge dominate. For the most materials, except of several initiating explosives, with mass of material acceptable for laboratory and small-scale tests, DDT can be run away only with a charge in strong confinement.

Table 1. Examples of threshold pressures

Material (confinement, i.d.)	Mean particle size, mm	Porosity, %	P_{LVD}, GPa	P_D, GPa
PETN (steel, 5 mm)	0.5	20 10 2.5	0.25 0.25 0.28	0.55 0.85 1.7
Single-based grained powder (steel, 15 mm)	0.8	42 28 6	0.6 (0.23) - (0.38) 0.9	 2.3
Undamaged rocket propellant "PEKA" based on AP and HMX (steel, 15 mm)	-	-	0.8 - 1.0	2.5 - 3.0

Note : Values of P_{LVD} in brackets have been determined analyzing the change of convective burning to LVD during DDT runs [11].

Typical setup applying in the most investigations into DDT is a strong closed tube, filled with material studied and arranged with ignition unit to start burning and tools to monitor the process developing. The tubes can be of different sizes depending on the kind of high explosive and purpose of study, however in Lab tests tubes of 5 - 40 mm i.d. and 100 - 400 mm long are more often in use. The tube material is usually strong steel, sometimes with a slit covered by window for speed photography, brass or transparent plastic.

The DDT tube test is frequently arranged with ignition unit comprising electric cap and an amount of a pyrotechnic mixture. This test is used as a common test to assess propensity of material to DDT under local accidental ignition, if one can put aside particular nature of ignition stimuli, or, if being equipped with recording tools to study mechanism and behavior of DDT. Mass and kind of the igniter mixture can be different, not only to provide reliable and quick ignition of the material studied, but more often, to get different initial pressure. If this pressure exceeds P_{CB}, the process begins promptly in convective burning mode that makes measurements more reproducible. Run distance to detonation is used as a quantitative characteristic. The less run distance, the more is the propensity to DDT.

By changing the ignition unit, one can try to reproduce special kinds of stimuli. Spark gap reproduces hazard of static electricity [12], piston driving test is used to estimate response of material studied on impact by heavy body [4], and heating device is used to reproduce cook-off conditions [13].

For studying DDT, most effective are tools enabling us to visualize light emission and trajectory of flame front and to record pressure in several positions along the charge. They are speed streak photo, ionization pins, photodiodes, piezoelectric gauges, etc. Another tools as X-ray pulse photo to visualize density distribution, contact pins of various kinds to monitor movement of stress wave, and electro-magnetic gauges to measure particle velocity are employed occasionally.

Unfortunately, the tests available do not cover all the majority of DDT conditions and properties of energetic materials. Two problems having methodical aspects are noticed below.

1) The strong confinement providing feasibility of DDT tests simultaneously restricts extensive application of the test results. Relaying on the test results,

materials are ranged to compare their propensity to DDT or explosiveness. However these ranges are strongly connected to parameters of setup used in the test. There are no clear concepts, how the test results can be expanded to real devices, and especially, onto conditions of large masses and sizes (scaling effect).

One of unwanted effects connected to strong confinement can be explained as follows. DDT behavior is roughly ruled by interaction of two factors. The first is the intensity of chemical conversion in burning spot, which determines rate of gas and energy release, and, as a consequent, velocity of reaction propagation onto adjacent layers of explosive material and possible rate of pressure rise. The second is a discharge of the burning spot by dissemination of material due to various gas-dynamic and mechanical effects. Both factors depend on test conditions in complicated ways, and as a consequence, small change in the test conditions often results in a behavior hardly predicted. The standard tests in strong closed confinement reproduce only several possible situations, when activity of the discharge factor is significantly restricted. If confinement can be easily expanded, equilibrium between both factors may be reached under certain conditions, resulting in stabilized LVD mode. In the case of unconfined (or in soft packages) explosive material of a large mass, the discharge factor manifests itself in maximum degree, and the standard tests seem to be almost useless.

2) Under conditions of DDT Lab tests and in the course of some accidents with such materials as low porosity charges of rocket propellants and high explosives, the low-velocity detonation is often observed as a final response to different stimuli. Although most researchers recognized now the role of LVD there are no standard tests designed to assess hazard and threshold conditions of this mode.

2. SCENARIOS AND MECHANISMS OF THE DDT

In many ways behavior of DDT are closely connected to spatial wave structure of convective burning and LVD. Main elements of the structure of convective burning wave are as follows :

- (i) flame front, an interface where material ignites over the pore surface ;

- (ii), zone of filtration and heat transfer ahead of the flame front, where gas flow together with heat transfer from gas to solid takes place ;

- (iii) compaction zone also ahead of the flame front called "plug", if it almost lost permeability ;

- (iv) extended combustion zone behind of the flame front where material particles burn down by their outer surface ;

- (v) a void, adjoining to the combustion zone, which often arises at the ignited end of the charge rather in the course of movement of solid phase due to compaction than burning of particles.

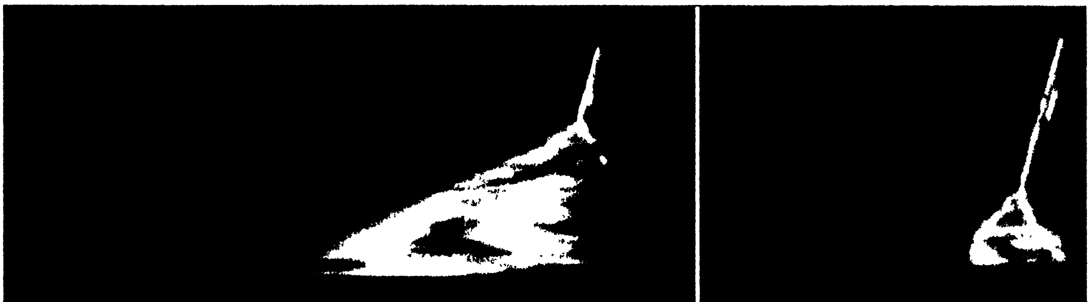
Large dimensions of the combustion zone composed by burning particles are inherent also to LVD stage. Before transition to full detonation only a small part of material has time to burn down.

Two following scenarios of DDT were extensively discussed (see [1, 2, 5, 6], etc.) :

1) DDT with successive change through both intermediate stages to full detonation. The stimuli causing change of the stages arise at/near the flame front of the wave and often include formation and evolution of the so-called plug. Some investigators studying high porosity charges of fine-grained high explosives observed detonation appeared at a distance ahead of the flame front, due to pore collapse at the leading edge of the plug.

2) DDT with sudden formation of a secondary wave of violent reaction and high pressure at a distance behind the flame front of convective burning. Here the full detonation builds up after this secondary wave overtakes the leading front.

These two scenarios are illustrated by examples of streak photos shown in Figure 3. Although a few studies gave evidences that the secondary wave really is a suddenly arising wave of reaction and pressure, the mechanism of formation of the secondary wave as well as domain of conditions favorable of second type of DDT remains debatable. As is seen at Table 2, typical objects appear to be loose-packed and slightly pressed charges of explosives of fine particles or with wax additive. Trying to define our point of view to this problem [14,15], we supposed that the difference in the scenarios must result in different trends. We have considered effects of the particle size on the run distance to detonation and characteristic time t^* of pressure rise. The t^* was determined as a mean value in the interval of several kbars by treatment of a signal of pressure gauge located near the igniter in coordinates of $ln(P)$ versus t/t^*. The general trends which can be surely connected to the first scenario of DDT are that both parameters reduce if particle size decreases, and t^* with a factor of 1,5 - 2,5 equals to a time scale $t_0 = d_{po}/(4BRT_b\rho_{ko})$. Here d_{po} is pore diameter, B constant in pressure law of combustion rate $U_p = BP$, T_b adiabatic temperature of combustion, R gas constant, ρ_{ko} TMD of material. The t_0 determines the rate of pressure rise under so called ideal conditions of pyrostatics. Quite different trends can be associated to the scenario with the secondary wave. The first trend is a reverse dependence of run distance to detonation on particle size. This reverse dependence first were described in [16] for HMX, RDX and PETN in a range of particle size below ~ 100 mm. Now a few additional evidences are available. Another trend is that the time t^* estimated for explosives listed in Table 2 in several tens times exceeds t_0.



A) B)

Figure 3. Streak photo records of DDT in PETN [1]. Steel tube of 5 mm i.d. with a split. Length of the charge exposed through the slit is 95 mm. A) First scenario of DDT (particle size is 500 μm, charge density 87% TMD); B) Scenario of DDT with a secondary wave (particle size is 20 μm, charge density 80% TMD)

Summarizing, we arrive at a conclusion, that the factors, which more likely cause the secondary wave, are the followings :

(i) Kinetic restrictions hampering ignition and normal combustion inside pores. Favorable conditions are pores of small sizes or covered by wax, when P_{CB} is high and porous bed is easily compacted.

(ii) Occurrence of a rather extended void in combination with a rather short part of the charge covered by convective burning. This void forms at ignited end of the charge due to movement and compaction of the porous bed downstream and is occupied by combustion products of the igniter and high explosive.

Reasons of the kinetic restrictions arising in the course of combustion inside small pores are obvious and considered elsewhere [13,14,18]. However in order

Table 2. List of documented examples of DDT with a secondary wave

Energetic material (confinement, i.d.)	Particle size, μm	Charge density, TMD	Technique of evidence	Ref.
PETN (steel, 5 mm)	20	80%	Streak photo	[1]
HMX (plastic, 25.4 mm)	15	44.2%	Streak photo	[3]
RDX/wax 95/5 (steel, 16 mm)	150 - 200	73%	Ionization pins + shock pins	[5]
Tetryl (steel, 16.3 mm)	20	76.6%	Ionization pins + strain gauges	[17]
Tetryl/wax 97/3 (steel, 16.3 mm)	470	66.5	Ionization pins + strain gauges	[17]
Picric acid (steel+plastic, 5 mm)	70	40%	Streak photo + pressure gauges	[6]
Fluid type single-based powder (steel+plastic, 5 mm)	35	53%	Streak photo + pressure gauges	[6]
AP/polystyrene 90/10 (steel+plastic, 5 mm)	25/10	36%	Streak photo + pressure gauges	[6]

to confirm these ideas further investigations, especially including numerical modeling are necessary. Our attempt of simulation implemented for fluid type single-based powder [11] reproduces formation of the secondary wave only in a distant degree. For getting quantitative agreement the model needs further improvements taking into account kinetic restrictions.

Contribution of convective burning into the run distance to detonation falls if initial porosity of the charge decreases. Finally, in high-density charges (porosity of 5 % and less) no part of the streak photo records can be apparently identified with convective burning. Besides, confinement must be stronger otherwise even for sensitive high explosives the process may not reach full detonation stabilizing at some low velocity detonation. The investigations conducted in [1,19] and found out influence of confinement properties on LVD velocity in pressed charges of high explosives turn out to be useful in several aspects. It was shown that LVD is steady, and the radial extension is the key factor of stabilization mechanism. Besides, by comparing experimental data with theoretical modeling, intensity of chemical conversion in LVD wave was estimated in the term of specific burning surface.

New strong arguments for stability of this LVD mode were obtained for composite propellants and PBX in [10,20]. Authors of [20] investigated LVD in a low porosity HMX-based composition varying properties of confinement. The process initiated by heating propagates along charge of 1 m long at steady velocity near 2 - 2.5 km/s and pressure near 2 GPa. It was shown that observed cases of transition to full detonation are provoked by heterogeneity of the charge (air gap artificially inserted into the charge).

Condition of the compaction zone affects ways of change of convective burning to LVD. If grained material is easily compacted (a lot of crystalline high explosive), this zone readily comes into the plug hardly permeable of gas and convective burning. The corresponding behavior is clearly demonstrated by the piston driving test [4] with the detonation-like mode originating at the leading edge of the plug at a distance ahead of the burning front. The similar behavior at thermal initiation of the process is not expressed evidently. Really there are streak photo records, which can be interpreted in the same way [1,21]. However, experiments implemented for the same materials by other researchers [2,7] have not revealed a break in the front trajectory and make to suppose that the break, if it was observed, may be a result of very low light emission of the front, when it propagates through the compacted bed.

Beds of single-based grained powders are heavier compacted. For this material the compaction zone appears not to lose permeability, and change of convective burning to LVD manifests itself with no break of the front trajectory and only in sharp increase of the front velocity [22]. However the LVD without reference to any compaction is also possible.

The compaction zone of convective burning evidently forms due to plastic waves propagate along porous bed at velocity exceeding the velocity of the flame front. This situation is ordinary, however reverse situation is also possible. At least, two corresponding examples are known and both in charges of loose-packed density. The first is a single-based coarse-grained powder [23,24] and the second is the initiation by a spark discharge of PETN and RDX [12]. Streak photo records [23] with tracks of burning grains shown in Figure 4, as well as electromagnetic measurements of particle velocity coupled with signals of the photodiode and pressure record at the same position [24] produce direct evidences, that in these cases the LVD develops with no formation of the compaction zone ahead of the flame front.

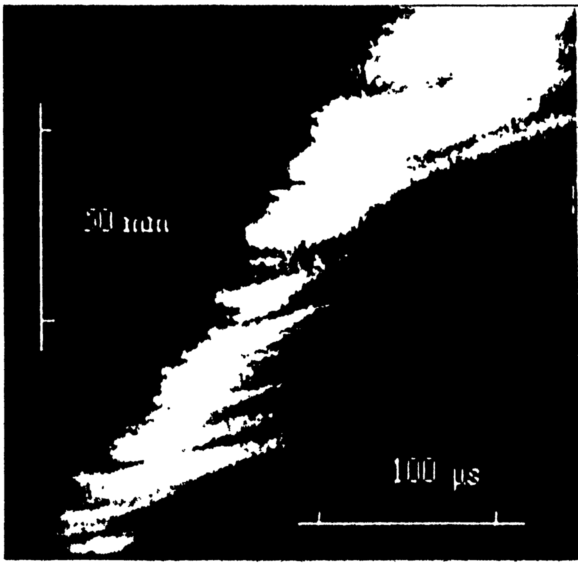


Figure 4. An example of streak photo record of LVD at the front velocity ~ 0 8 km/s with tracks of burning grains [23]. Single-based powder of 3.3 mm effective grain diameter in a multi-section DDT steel tube 15 mm i.d and 700 mm long with a slit for photography at a distance 280 – 420 mm from igniter. Shape of the tracks with an initial horizontal leg of ~ 30 μs long gives evidence that grains ignited with no contribution of a compaction process.

The experimental data and their interpretation by means of numerical simulation [11] demonstrate, that in these cases the LVD has got a different structure, and energetic material ignites with no pore collapse (at low particle velocity and porosity close to initial ones) due to convective heat transfer from strongly compressed and preheated gases filling pores. This mechanism has been referred to as the "gas compressive" LVD.

This mechanism is totally in accord with two-phase nature of the porous material and can be deduced considering conservation equation of energy of gases filtered in pores ahead of the flame front [25]. This equation, in addition to the convective term, contains also terms resulting in heating of gas due to work of gas compression and work of friction at the pore surface. Just these latter terms provide ignition in the front of "gas compressive" LVD.

Every time when destructive accidental explosions (such as explosion of 3 railway cars transporting RDX and HMX at Arzamas station, Russia, 04.06.88, and explosion of AN at AZF, Toulouse, France, 21.09.01) have happened, scenarios of DDT taking place during accidents and involving large masses of powdered explosive materials attract attention once again. Let us consider a powdered explosive material stored in a pile, open container or soft packages. What are possible ways of DDT starting from a burning spot initiated by fire or other accidental stimuli ? One can distinguish two key situations : (i) the burning spot is located on an opened surface of the material and (ii) the burning spot buried under the bed of the material. In the former situation, there seems to be no possibility for combustion to penetrate inside the pile, and explosive material could burn down easy. However, experience of accidental explosions makes us to try for possible DDT scenario.

The scenario supposed relays on evidences that a short time before a detonation of energetic material involved into the fire occurs, a violent intensification of flame in plume takes place. So, the scenario comprises a following chain of events. Combustion of explosive material due to buoyancy of hot gases produces thermal plume. As area of combustion spot increases, the size of the plume and velocity of upward flow of gases also increase. Vortex flows are capable to blow up from the surface particles of explosive material and involve them into the plume. Burning of these particles results in an avalanche-like increase of intensity of the fire and capability of the upward flow to strip up and involve into the flow new and new particles thus forming burning aerosol. These events culminate in a formation of a strong blast wave in this aerosol, whose interaction with the bed of the explosive material results in detonation build up or DDT in the whole pile.

Although nobody has investigated this scenario in detail, one can find useful information elsewhere [26]. The authors studied plumes generating after burning of a few pyrotechnic compositions. The samples of 1, 5 and 25 kg in mass were placed on a support as a circle layer of identical thickness. Most of the samples burnt easy with no explosion. However, for 3 compositions the increase of the mass (and, hence, diameter of the circle) results in detonation-like explosion instead of quiet burning. That is an object for numerical modeling. Theoretical analysis based on aerodynamics of hot gases ascending in gravity space gives reasonable estimation for the critical sizes of the burning spot capable to strip up particles from the surface and involved them into the plume.

In regard to the second situation (the buried burning spot) the burning following gases can penetrate into the pores of the explosive material bed surrounding the burning spot. However pressure rise in the spot can easily disseminate the bed and prevent development of explosion. In order to estimate hazard of explosion in this situation and get adequate information on the discharge effect, we apply a test, referred to as critical height bed test [27]. The steel cylinder vessel with open upper end is fitted with igniter (heating wire) and pressure gauge both placed at the bottom of the vessel. The test procedure includes several runs starting with a small amount of material studied which being ignited does not bring explosion. In this case the gauge records a small pressure increase, and the material turns out to be almost totally ejected from the vessel, burning down outside or remaining not burnt on the base. In the next run, which is carried out with increased mass (and height of the bed) of the material, the behavior repeats, but the maximum on the pressure diagram slightly grows. By increasing the height of the bed, run after run, it finally exceeds some critical value, when the behavior drastically changes demonstrating violent explosion, more often with a detonation.

Increase of the bed height makes more and more difficult discharge of the burning spot, therefore critical height of the bed provides threshold conditions of equilibrium, at which the discharge factor is still capable to make up for the factor of burning intensity and to prevent unlimited pressure rise. As shown in Table 3, if diameter of the vessel increases, the critical height of the bed grows and then levels out, manifesting attenuation of effect of the vessel wall. The maximum pressure at the pressure – time diagram recorded at the critical height of the bed practically does not depend on the vessel diameter and approximately equals to the PCB of the energetic material tested. The results of the test are useful for design explosion-safety devices in reactors and containers filled with explosive materials, as well as for estimation of explosion-dangerous mass of materials stored in piles and bags and involved into a fire. For material tested the leveled values of the critical height of the bed significantly exceed run distance to detonation. This is in accord with experience that under favorable conditions a large mass of high explosive can burn up easy with no explosion.

Table 3. Critical height of the bed (in mm) for a few materials. Effect of the charge diameter

Energetic material (mean particle size, μm)	Charge diameter, mm				
	16	40	125	250	500
Fine HMX (120)		150	330	400	450
Coarse HMX (400)		150	340	480	550
Fine RDX (50)	20		80	270	
Coarse RDX (500)	95	170		600	
Ball powder (440)	200	380	500	600	700

3. NUMERICAL MODELING AND PREDICTIVE ABILITY OF THE MODELS

The DDT is predominantly empirical science. Theory and numerical modeling are used for interpreting and better understanding of experiments. There are several models and computer codes developed by different teams (see, for instance, [9,28-33], etc.). These models are based on a common approach comprising the following key items :

- (i) application of mechanics of multiphase reacting media with volume-averaged variables ;

- (ii) the hypothesis that heating of energetic material to a point providing start of exothermic reactions at pore surface is implemented through two mechanisms: convective heat transfer from hot gases filtering into pores, and energy dissipation during pore collapse ;

- (iii) effects connected to finite rate of chemical reactions in the flame are ignored at the stages of convective burning and LVD.

The last item is easily reduced to assumption that intensity of chemical reaction in a unit volume equals the product of specific surface of pores (or particles) by rate of level-by-level combustion of the material. In anywise, models include mesoscale processes, mainly in order to define a moment when burning of material at pore surface begins.

There are a few works demonstrated agreement between numerical simulation and experimental data in DDT behavior as well as in characteristics of separate stages. See, for instance : (i) DDT in high explosive CP with V-shape curve of run distance to detonation versus porosity [8] ; (ii) DDT in a double-based propellant demonstrating agreement on pressure rise and trajectory of fronts [32] ; (iii) LVD in PETN demonstrating effect of confinement [19] ; (iv) convective burning, transition to LVD and LVD behavior in single-based grained propellants [11], etc.

One can put questions : what stands for the observed agreement, and to what extent models are suitable for making quantitative predictions ? Our comment is as follows. First of all, the assumption of intensity of chemical conversion being predominantly controlled by layer-by-layer burning over pore surface hits the right nail. In contrary, it would be impossible to exactly reproduce pressure rise rate, which makes dominant effect on DDT behavior. The second, models include set of input parameters, specifying the different properties of material in broad range of pressure and temperature. Some of them are usually unknown, or express the corresponding properties in a qualified mode, and virtually used as fitting parameters. They are, for instance, effective viscosity of solid phase and ignition point of material, etc. In case of materials of low porosity or crystalline high explosives with easily crashed particles, specific pore surface (effective particle size) plays the same role, being varied to get the best consent with experiment. Finally, the agreement is observed for energetic materials demonstrating first scenario of DDT, and no good agreement for DDT with a secondary wave.

So, one can conclude that different improvements of the model are required depending on energetic material considered. The following objects can be selected.

- Single and double-based grained propellants. They seem to conform to the recent models in the best way.

- Powdered high explosives. Models need in improvement to take into account kinetic limitations in the case of fine size particles. Besides, effect of particle fragmentation on their burning surface for coarse-particle materials should be finally studied and correctly incorporated into the models.

- PBX and rocket propellants of low porosity. Data on specific burning surface are required for the surface preset by initial pores as well as for the surface arising in the course of DDT due to deformations. One can easy anticipate that the comparison of simulation and experiment is a real way to determine this value.

- Heterogeneous mixtures (aluminized mixture of AP, etc.), AN-based industrial explosives. All these objects now are beyond a concern of modeling. New versions of DDT models should be developed taking into account heterogeneous reactions. Headache here is description of real complicated structure of pores and development of a model to simulate the mesoscale processes.

The improvement of models may be successful only in close cooperation with implementation of experiments to get empirical information required, and check and confirm simulations. Special attention should be paid to getting agreement in conditions of the stage change.

4. CONCLUSIVE REMARKS

Knowledge about processes composing DDT in solids is rather full and clear, and methods of study are informative, if applied in complex. Unfortunately, the tests available do not cover all the majority of DDT conditions and properties of energetic materials. Results of distinct tests ranging explosive materials in respect to their propensity to DDT can not be directly expanded to real conditions.

Recent tendency to lay hopes on development of predictive computer codes comes to DDT study. Numerical simulation already turned out to be very helpful in explanation of experiments and shedding light at mechanisms of DDT. However the predictive ability of models is still limited by few factors. First is deficit of extensive empirical information specifying different aspects of behavior of energetic material during all stages of DDT. Besides white spots remain in understanding and description of important processes at meso-scale level which accompany thermal and mechanical loading (fragmentation of particles, formation of hotspots, separation of explosive material from wall of confinement, etc.). The complex approach, when numerical modeling coordinates with experimental study aimed to elucidate meso-scale processes and properties of separate stages of DDT will provide progress.

Hitherto investigations into DDT were mainly stimulated by hazard of accident explosions under manufacture and handling of explosive materials and rocket propellants, especially with involving their large masses as well as by risk of destruction of munitions and devices filled by these materials. However, as knowledge and skill to control the DDT were accumulated, attractive prospects to make in use of properties of convective burning and LVD in applications are recognizing. As an example, we can notice a new concept of propellant charge of elevated density composed of grains covered with thin polymer film for enhancing performance of gun systems [34]. Other productive directions are connected to development of charges, which burn down in modes of controlled convective burning or LVD and designate for impulse rocket motors, as well as gas or heat generators.

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