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Deflagration-to-Detonation Transition in Mixtures of Finely Divided Ammonium Perchlorate with Submicron Aluminum Powder

V. G. Khudaverdiev*, A. A. Sulimov, B. S. Ermolaev, and V. E. Khrapovskii
Semenov Institute of Chemical Physics, Russian Academy of Sciences, Moscow, Russia
**e-mail: vugarikkk@mail.ru*

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Abstract—Deflagration-to-detonation transition in binary mixtures of fine ammonium perchlorate (20-μm grains) with submicron ALEX-L aluminum powder (0.2-μm particles) is studied using high-speed photography and pressure recording with quartz crystal sensors. The test mixtures were loaded in thin-walled quartz tubes of inner diameter 10 mm. The charges had a porosity of ~50%. It has been shown that, even under very mild conditions (low-strength shell and a weak source of initiation), the deflagration mode of mixture combustion easily transforms into the detonation mode. The shortest length of the region of transition from deflagration to normal detonation (not more than 30 mm) was observed for a lean mixture, with an aluminum content of ~5%. The mechanism of transition to detonation involves the stage of convective combustion, resulting in the formation of a brightly luminescent crescent-shaped area behind the primary flame front, which, in turn, generates a forward (in the direction of propagation) and a backward wave. The forward wave gives rise to low-speed detonation, which later transforms into normal detonation. The pressure profile within the region of low speed detonation is measured. A comparison with similar experiments in which ALEX-L aluminum powder was replaced by ASD-4 aluminum (4 μm particles) shows that ALEX-L sensitizes the mixture, resulting in a dramatic reduction of the length of the transition region, making it possible to produce normal detonation in low-strength shells.

Keywords: deflagration-to-detonation transition, low-speed detonation, ammonium perchlorate, ALEX-L and ASD-4 aluminum powders

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INTRODUCTION

When ignited, powdered ammonium perchlorate–aluminum mixtures are known to explode. The start and development of an explosion in such mixtures was experimentally investigated in a number of works [1–3] by recording pressure–time diagrams in a constant-volume bomb. It was shown [3] that, depending on the pressure and the charge length, layer-by-layer, convective, or explosive combustion take place. The minimum length of the charge at which these modes are observed and their dependence on the mixture composition were determined. To obtain a complete picture of deflagration-to-detonation transition, elongated charges encased in a transparent shell so as to enable continuous photorecording by high-speed photography [3, 4]. Interesting data, clarifying the mechanism and characteristics of the transition of convective burning into low-speed detonation in powdery mixtures of ammonium perchlorate with ASD-4 aluminum (with a particle size of 4 μm) [3] and with polystyrene [4]. However, mixtures of ultrafine components have not yet been studied, despite a great interest in using such components in solid rocket propellants and pyrotechnic compositions to improve their performance, in particular, to increase the burning rate.

The aim of the present work is to study the characteristics of deflagration-to-detonation transition in binary mixtures of finely divided ammonium perchlorate and submicron aluminum. Particular attention is paid to the overall picture and intermediate stages of the transition of convective burning into low-speed and normal detonation.

EXPERIMENTAL

We used powdered ammonium perchlorate with a average particle size of 20 μm and ALEX-L aluminum powder with a particle size of 0.2 μm. The starting powders takes in a required proportion were thoroughly mixed by hand. Mixtures containing 3, 5, 10, 28, and 54 wt % aluminum were prepared.

The experiments were carried out in low-strength quartz glass shells with an inner diameter of 10 mm and a wall thickness of 2 mm. The nominal strength of the shells was 250 kg/cm^2. The lower end of the shell was closed with a cap. The shell was attached to a steel plate in a vertical position. The investigated mixture was poured through the upper open end of the shell in

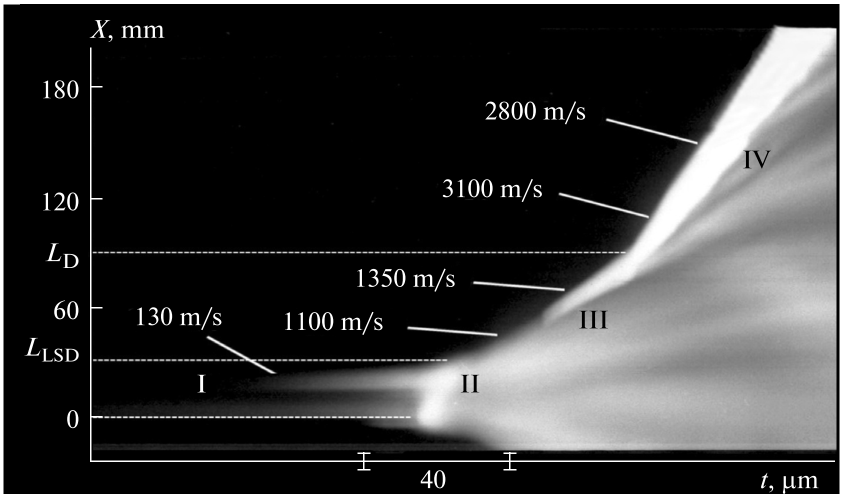

Fig. 1. Streak-camera image of deflagration-to-detonation transition in a mixture of 28% ALEX-L aluminum powder with ammonium perchlorate at $L_{LSD} = 30$ mm and $L_D = 95$ mm: (I) convective burning, (II) region of formation of the secondary wave, (III) low-speed detonation, and (IV) normal detonation.

successive portions, so that the height of each portion was equal to 2-to-3 charge diameters. After filling each portion, the shell was tapped. The charges were approximately of the same porosity, ~50%; depending on the composition of the mixture, the charge density, was 0.85–1.05 g/cm^3. In most experiments, the length of the charge was 200 mm. The mixture was ignited with a Nichrome coil. The 5-Ohm coil was housed in a textolite sleeve, protruding 3 mm deep into the charge.

The evolution of the explosion process was monitored using a ZhFR-2 streak camera. The photographic record yielded the trajectory of the luminous wave front, from the lengths of the regions of transition to low-speed detonation (L_{LSD}) and normal detonation (L_D) were determined, as well as the wave front velocity within each region, up to the onset of detonation. The lengths of the regions were measured from the location of the igniting coil. The error in determination of the wave front velocity was ~3%. These experiments have also enabled to draw certain conclusions on the mechanism of the transition process.

In addition to the experiments with photorecording of the explosion process, we conducted several experiments to obtain pressure–time diagrams. For this purpose, we used AVL-6000 piezoquartz pressure sensors with a natural frequency of ~250 kHz, which were located along the charge at different distances from the igniting coil. In these experiments, the glass tubes were replaced by Plexiglas tubes of similar dimensions. The shell wall had a 3-mm-diameter hole for accommodating the piezoelectric sensors. The sensors themselves were placed on the rear side of a steel plate, whereas the shell with the test charge was glued to the front side of the plate. The sensors were positioned 30 mm apart within the region in which, according to preliminary experiments, low-speed detonation took place. This method, which makes it possible to keep the pressure sensors intact during the destruction of the shell, was proposed and tested in [5].

EXPERIMENTAL RESULTS

Typical photorecordings showing the development of the explosion process mixtures containing 28, 10, and 5 wt % aluminum are given in Figs. 1–3, respectively. The results of photographic observations led us to the following conclusions.

The deflagration of the mixture after ignition develops very rapidly, exhibiting transition to detonation in all the experiments. Photorecording (see, e.g., Fig. 1) clearly show all the successive stages of the transition process: convective burning at velocities of ~100 m/s; the region of formation of the so-called secondary wave; low-speed detonation (1100–1400 m/s); and normal detonation (~3000 m/s).

At first glance, the mechanism of development of the explosion in the test mixtures does not differ from the transition process in powdered brisant explosives [6]. However, there is a very important difference, which consists in the fact that convective burning and transition to detonation in the test mixture occur in low-strength glass shells after a weak initiation. By contrast, powerful high explosives are capable of a similar transition process only in a strong metal shell upon an intense ignition.

The convective combustion region produces a rather weak and unstable luminescence, which manifests itself in the form of a few nearly horizontal streaks separated by dark intervals. Streaks end in brightly glowing kernels, which merge into a crescent-shaped region that marks the beginning of the formation of the secondary luminescent wave. The front of this wave, propagating through fresh mixture, gives rise to

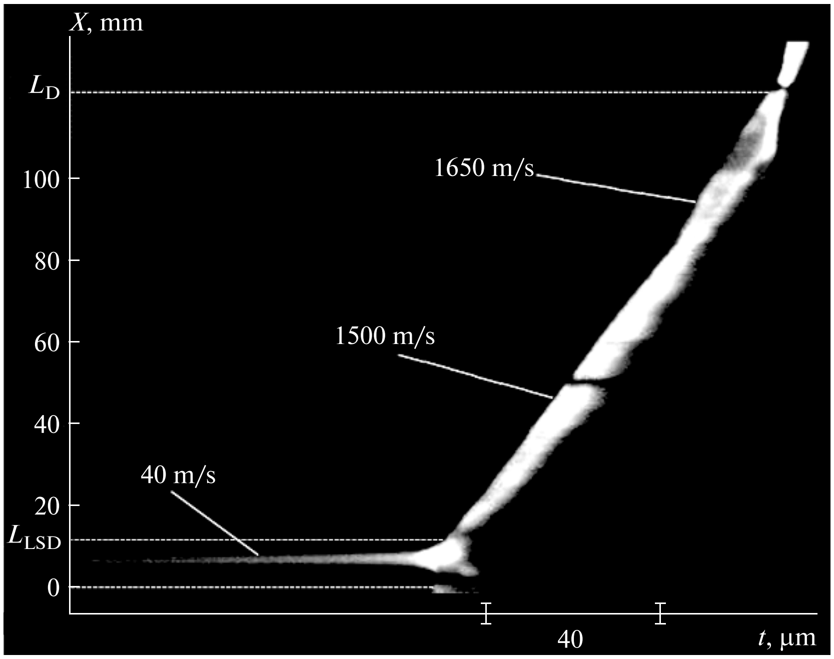

Fig. 2. Streak-camera image of deflagration-to-detonation transition in a mixture of 10% ALEX-L aluminum powder with ammonium perchlorate at $L_{LSD} = 12$ mm and $L_D = 120$ mm.

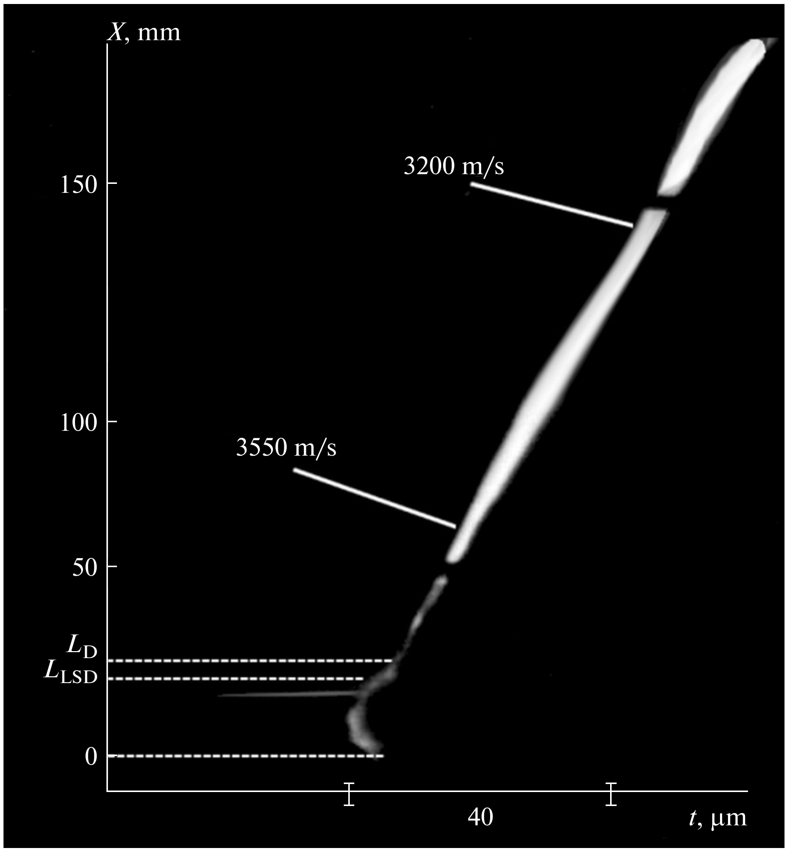

Fig. 3. Streak-camera image of deflagration-to-detonation transition in a mixture of 5% ALEX-L aluminum powder with ammonium perchlorate at $L_{LSD} = 25$ mm and $L_D = 30$ mm.

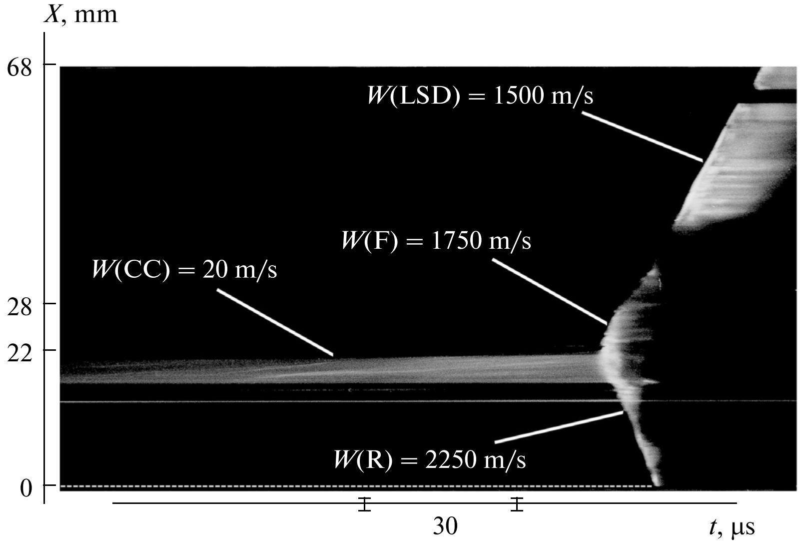

Fig. 4. Magnified streak-camera image of the region of transition of deflagration to low-speed detonation in a mixture of 10% ALEX-L aluminum. The brightly luminescent crescent-shaped region generates two waves: a forward one (F), which transforms a low-speed detonation wave and a reverse one (R), which travels towards the igniting coil.

detonation. Note that a similar pattern is observed in the combustion of finely grained high explosives [7].

The point in the crescent-shaped region that corresponds to the shortest time can be considered as the origin of the secondary wave. In most experiments, this point is located behind the convective combustion front at some distance from it (5–20 mm). This situation is clearly visible in Fig. 3. However, in some experiments, typically aimed at obtaining a high-magnification image (Fig. 4), the point of origin of the secondary wave is located nearly at the convective combustion front. Careful analysis of this and other photographic recording allows us to assume that one or two streaks, almost invisible because of a low intensity of luminescence, are indeed located above the streak seen in the image displayed in Fig. 4. This means that the point of origin of the secondary wave is located behind the front.

For mixtures with 5 to 28% aluminum, the transition from combustion to low-speed detonation occurs at a short distance from the igniting coil. The length of the transition region L_{LSD} is typically 5–30 mm. The length of the region corresponding to low-speed detonation, i.e., the region between L_{LSD} and L_{D}, for the 5% aluminum experiments (Fig. 3) and some 10% aluminum experiments is very small, not more than 5 mm. In these experiments, L_{D}, the total length of the region of transition to normal detonation, does not exceed 30 mm. The normal detonation velocity is 2800–3200 m/s. It should be noted that, in some experiments, the velocity of detonation within the initial region exceeded its steady-state value, i.e., deflagration-to-detonation transition occurred through the stage of overdriven detonation (Figs. 1, 3), which was also observed [8] for mixtures based on potassium perchlorate.

The other experiments with the 10% aluminum mixture (Fig. 2) and with stoichiometric (28% aluminum) mixture (Fig. 1), the length of the region of transition to normal detonation L_{D} was significantly longer, 90–120 mm. In these experiments, the region of low-speed detonation turned out to be more extended; it featured the growth of the wave velocity from 1100 to 1700 m/s.

For the mixtures with 3 and 54% aluminum, the process terminated at the stage of convective burning because of the destruction of the tube. The velocity of the combustion wave did not exceed 300 m/s. Note the enormous difference caused by a small increase in the content of aluminum: for 3% aluminum mixtures, the transition process is terminated at the stage of convective burning, whereas for 5% aluminum mixtures, the transition from deflagration to normal detonation takes the shortest length (L_{D} = 25–30 mm) in this series of experiments. Thus, the propensity of lean mixtures of finely divided ammonium perchlorate with submicron aluminum to deflagration-to-detonation transition is similar to that of initiating explosives, such lead styphnate, for which $L_{\mathrm{D}} \sim 10$ mm for tests in a steel shell [9].

The photo shown in Fig. 4 makes it possible to examine in detail the process of onset of low-speed detonation. The brightly luminescent crescent-shaped area, located at a distance of 22 mm from the igniting coil, generates two waves. The forward wave travels 6–7 mm at a velocity of ~1700 m/s, and before generating the low-speed detonation wave, it dramatically slows down as its luminescence decreases sharply. The backward wave travels to the igniting coil at a velocity of

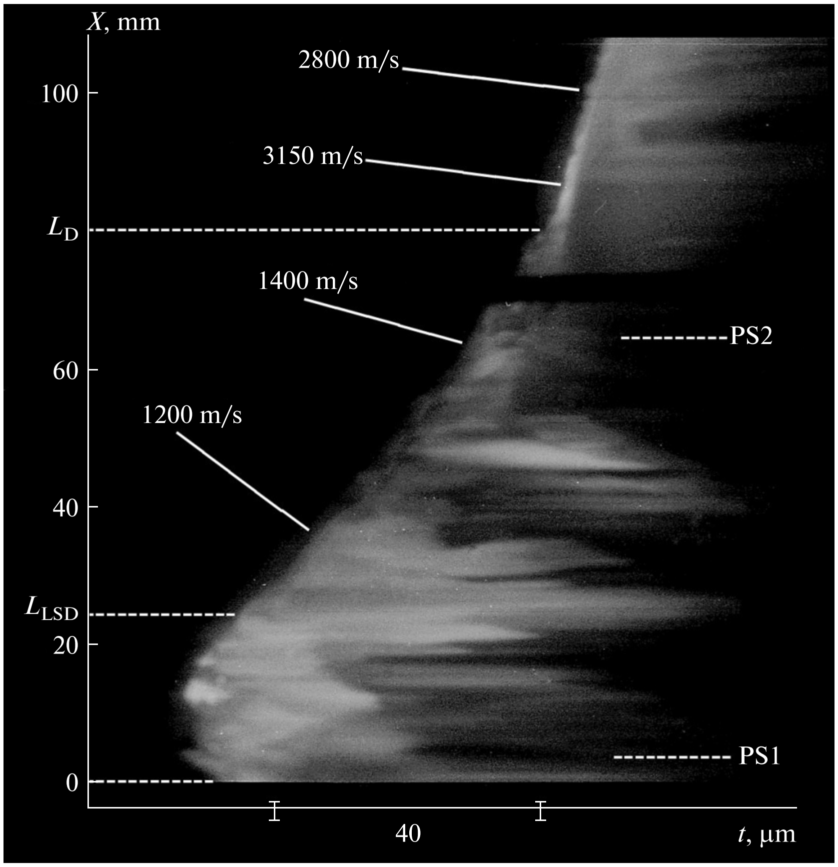

Fig. 5. Streak-camera image of deflagration-to-detonation transition for a mixture of 28% ALEX-L aluminum powder with ammonium perchlorate obtained in an experiment with pressure recording.

~2200 m/s. The length of the transition region is L_{LSD} is ~32 mm.

Figures 5 and 6 show the results of piezometric measurements and photorecording of the transient process in the stoichiometric mixture containing 28% ALEX-L aluminum.

The photorecording shows the onset of low-speed detonation with a velocity of 1200–1400 m/s and its transformation to normal detonation within a transition region of length $L_D = 80$ mm. Because of a weak luminescence, the streaks corresponding to the convective combustion region are not visible in the photo. The positions of the pressure sensors are indicated by dashed lines. One sensor is located behind the point of formation of the crescent-shaped area, whereas the second, at the end the low-speed detonation region. The pressure recordings in crescent-shaped area and in low-speed detonation region have a triangular profile with a rise time of 2–3 μs and an amplitude of 560 and 670 MPa, respectively. The values the signal rise time and pressure amplitude are approximate due to an insufficiently high frequency resolution of the data acquisition system.

It is worth noting that, for experiments performed in Plexiglas shells, inferior to quartz shells in the degree of smoothness of the inner surface, the regions of transition to low-speed and normal detonation were shorter. The effect of roughness of the shell on the combustion process was studied in [10].

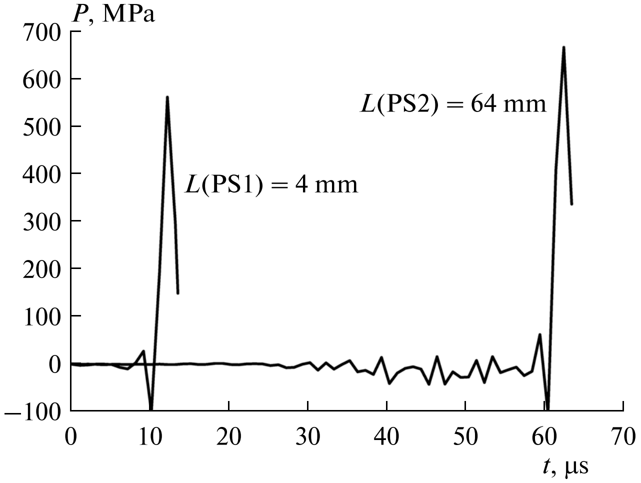

Fig. 6. Pressure time history recorded at two points along the charge length for a mixture of 28% ALEX-L aluminum powder with ammonium perchlorate: pressure sensors PS1 and PS2 are located 4 and 64 mm, respectively, from the igniting coil.

Thus, in almost all the experiments carried out in quartz tubes with mixtures containing from 5 to 28% ALEX-L aluminum, the explosion process results in normal detonation propagating at velocity of 2800–3200 m/s. Such a high activity of mixtures has been observed neither for finely powdered mixtures of ammonium perchlorate with polystyrene [4] nor for similar mixtures with coarser aluminum, such as ASD-4 (with a particle size of ~4 μm) [3]. For both mixtures in thick-walled Plexiglas shells or in Plexiglas tubes reinforced with slotted steel collars, only the transition to low-speed detonation occurred. In this case, the lengths of the transition region L_{LSD} for the most active compounds (with 5 and 10% aluminum) exceeded 150 mm. Experiments in quartz tubes with mixtures containing this type aluminum demonstrated the disruption of the transition process because of the destruction of the tubes at the stage of convective combustion.

CONCLUSIONS

Mixtures of fine ammonium perchlorate with submicron aluminum in low-strength glass shells were demonstrated to exhibit a high propensity to an explosion with transition to detonation upon ignition with an incandescent wire coil. The transition process occurred in mixtures with an aluminum content within 5–28% and included all typical stages, such as convective combustion, low-speed detonation, and normal detonation. Mixtures with an aluminum content of 3 and 54% exhibited a disruption of the transition process at the stage of convective combustion because of the destruction of the shell.

Low-speed detonation, with a velocity from 1100 to 1700 m/s, is initiated by the secondary wave generated by a crescent-shaped brightly luminescent area formed behind the convective combustion front. The pressure profile in the crescent-shaped area and low-speed detonation wave has a triangular form with an amplitude of ~600 MPa and a rise time of a few microseconds. The length of the region of transition to low-speed detonation L_{LSD} does not exceed 30 mm. The onset of low-speed detonation can be very fast. For a 5% aluminum mixture, the length of the region of transition from low-speed detonation to normal detonation is only 5–7 mm. As a result, in experiments with this mixture, normal detonation sets in at a distance of 20–30 mm from the igniting coil.

The high activity of the mixtures is largely associated with the aluminum particles having a submicron size, which significantly intensifies the combustion and speeds up deflagration-to-detonation transition detonation as compared to mixtures with micron-sized aluminum powder. According to [11], the sensitizing effect of submicron aluminum particles also manifests itself in the case of aluminum–ammonium nitrate mixtures, for which deflagration-to-detonation transition is difficult to produce.

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