

## URGENT COMMUNICATION

# Laser Initiation of Crystallized Mixtures of Furazanotetrazine Dioxide and Dinitrodiazapentane

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**Experiments were performed to determine the threshold laser pulse energy density resulting in explosive transformation of the mixtures studied. The experiments showed the possibility of using the laser initiation technique for rapid testing of mixtures of variable composition and structure.**

**Key words:** laser initiation, crystallized mixtures, furazanotetrazine dioxide, dinitrodiazapentane.

## INTRODUCTION

The endothermic substance 5,6-(3',4'-furazano)-1,2,3,4-tetrazine-1,3-dioxide (C<sub>2</sub>N<sub>6</sub>O<sub>3</sub>, FTDO), which has a high enthalpy of formation, is of great interest as a promising energetic material [1]. However, its practical use is hampered by its high impact sensitivity comparable to that of lead azide [2]. To overcome this obstacle, it is proposed to crystallize FTDO in a mixture with the linear nitramine 2,4-dinitro-2,4-diazapentane (DNP) [3]. Such mixtures are characterized by the presence of two eutectics FTDO/DNP (18/82 and 65/35) and a molecular compound (49/51). It has been found that the mixtures burn steadily at atmospheric pressure but an increase in the pressure to several tens of atmospheres makes the combustion unsteady [4]. The higher the content of FTDO in the mixture, the lower the threshold pressure of steady-state combustion.

An analysis of data on the transition from steady-state to unsteady combustion for the examined crystallized mixtures suggests that this phenomenon occurs

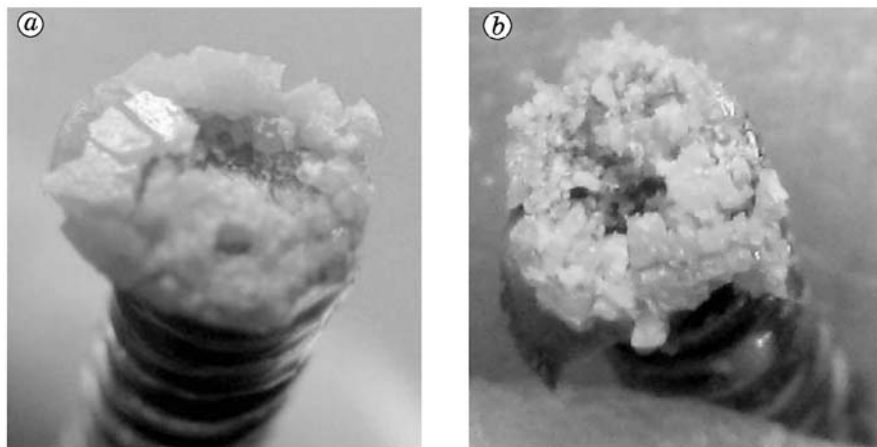
when the rate of temperature rise in the condensed-phase reaction zone exceeds the critical level. Indeed, at a burning rate of 20 mm/sec, the calculated maximum temperature gradient in the condensed phase is  $\approx 100$  K/ $\mu$ m. In this case, the temperature rise rate reaches a value of  $\approx 10^6$  K/sec. This is responsible for high thermal stresses, which can lead to mechanical destruction of the crystals and combustion transition to a mode close to explosive transformation. Predominant initiation of explosive transformation during combustion can occur in local regions with an increased content of FTDO.

In the present work, an attempt is made to estimate the sensitivity of crystallized FTDO/DNP mixtures to the action of a thermal pulse which simulates combustion wave conditions. The thermal pulse was produced by the focused radiation of a CO<sub>2</sub> laser. The laser was adjusted to the 10P32 line (932.96 cm<sup>-1</sup>), which coincides with the maximum of one of the absorption bands in the IR spectrum of the FTDO/DNP mixture. The pulse had a shape typical of CO<sub>2</sub> lasers: an initial peak 0.2  $\mu$ sec long and a nitrogen tail about 1.5  $\mu$ sec long. The energy of the initial peak was about 30% of the total laser pulse energy. The total pulse energy was varied in the interval of 0.1–0.5 J.

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**Fig. 1.** Photographs of the surfaces of the molecular compound (a) and eutectic mixture (b) after exposure to radiation pulses of subthreshold intensity.

In this work, we studied samples of crystallized FTDO/DNP mixtures of compositions 49/51 (molecular compound) and 65/35 (energetic eutectic). The samples were prepared by heating of the mixed initial powders to melting with subsequent crystallization of tiny droplets (weighing 8–12 mg) of the mixture on a cover glass or fluoroplastic film. The droplets were crystallized at various substrate temperatures: the lower limit was liquid nitrogen temperature, and the upper limit was at 50–55°C (near the melting point of the corresponding mixture).

The structure of samples of the molecular compound and eutectic mixture crystallized at substrate temperatures  $T_s = 20$  and  $-18^\circ\text{C}$  were studied by x-ray microtomography on the synchrotron radiation source of the Institute of Nuclear Physics, Siberian Division, Russian Academy of Sciences. Reconstructed images of thin sections of samples of the eutectic mixtures were obtained. Their analysis allowed us to find the void fraction (the ratio of the volume of the pores to the volume of the sample) and the pore size distribution. It was found that the void fraction of the molecular compound decreases with increasing cooling rate of the samples, with the pore distribution curve shifting toward small sizes: for  $T_s = 20$  and  $-18^\circ\text{C}$  the void fraction was 1.3 and 0.8%, respectively, and the modal (number averaged) pore size  $D_{10}$  was 16 and 13  $\mu\text{m}$ , respectively. For the eutectic mixture, the void fraction in both cases was 0.9%, and the modal sizes  $D_{10}$  were 16 and 14  $\mu\text{m}$  for  $T_s = 20$  and  $-18^\circ\text{C}$ , respectively.

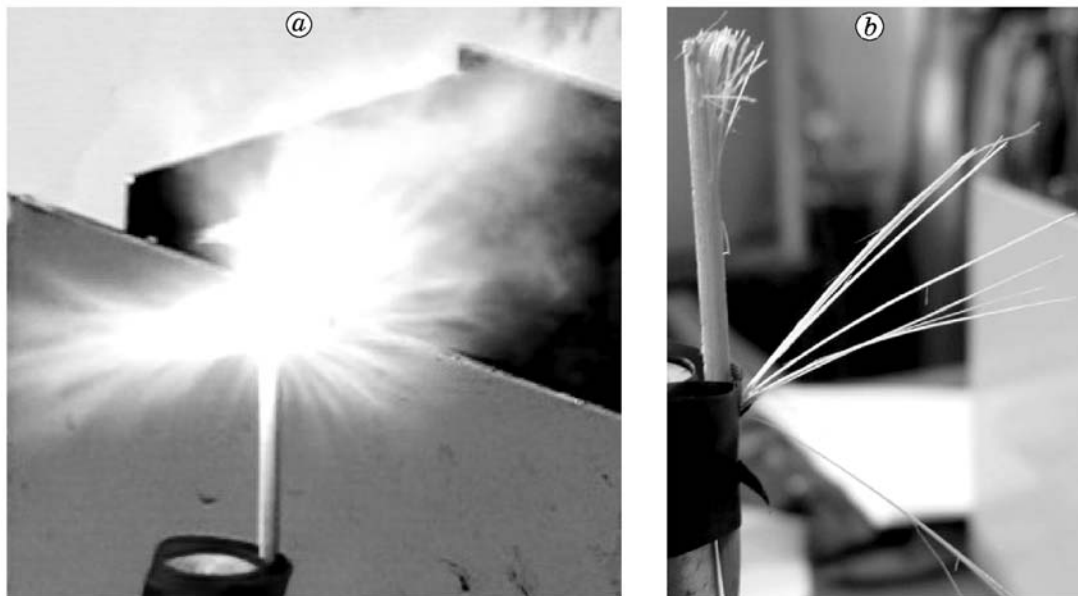
Samples of the examined crystallized mixtures were irradiated by single pulses of a  $\text{CO}_2$  laser. Lenticular samples of 3–5 mm diameter with the flat base facing the laser output window were placed in the region of the

laser beam waist focused by a long-focus ( $f = 250$  mm) NaCl lens. The total radiation energy pulse was measured by an IMO-2M device.

The energy density distribution in the beam waist region was measured by scanning the beam section with a pinhole plate. The distribution profile was found to be nearly Gaussian one with a width  $a = 1.4$  mm at the level of  $\exp(-1)$ . Accordingly, the maximum energy density  $E_0$  at the center of the waist was calculated by the formula  $E_0 = 4W/(\pi a^2)$ , where  $W$  is the total laser pulse energy.

Part of the laser radiation incident on the focusing lens was reflected by a NaCl plate onto a MG-33 pyroelectric detector. The signal from the detector was used to record the laser pulse energy in fire experiments. The detector was calibrated using an IMO-2M measuring instrument directly before and after a series of experiments. The laser pulse energy was varied by changing the laser electric discharge energy and the laser mixture pressure and using attenuating optical KRS-5 glass filters.

In the experiments on laser initiation of FTDO/DNP mixtures, the following results were obtained. For pulse energy densities varied in the range of 10–35  $\text{J}/\text{cm}^2$ , samples of the molecular compound show only a short-term flash with subsequent quenching. The irradiated surface has a circular region with partial ablation and melting traces of the initial material. The frames (camera speed of 30 frames/sec) show a flame in the pyrolysis product jet expanding from the surface and subsequent quenching. On exposure to the maximum energy radiation pulse, the sample undergoes fracture with the formation of a central crater and large fragments (Fig. 1a). Threshold



**Fig. 2.** Photographs of explosion of the eutectic mixture (a) and the split wood rod (b).

values of the specific energy density for the initiation of explosive decomposition were measured in experiments with the 65/35 FTDO/DNP eutectic mixture. As in the case of the molecular compound, a short-term flash and subsequent quenching were recorded on exposure to low energy. This was accompanied by a specific destruction of the irradiated surface with the formation of a central crater and separate weakly connected fragments with characteristic sizes of 0.1–0.5 mm (see Fig. 1b). The attainment of the threshold radiation energy level was characterized by a pop, scattering of the combustion products on a circular sphere, and layering of the wood rod used to hold the sample (Fig. 2). It was found that the threshold radiation energy corresponding to explosive transformation depended on the structural properties of the sample. In particular, the 65/35 FTDO/DNP eutectic mixture crystallized at  $T_s = -196^\circ\text{C}$  exploded at an energy density  $E \geq 18 \text{ J/cm}^2$ , and the mixture crystallized at  $T_s = 20^\circ\text{C}$  exploded at  $E \geq 21 \text{ J/cm}^2$ .

The difference in the threshold values of the energy density for initiation of modifications of the eutectic mixtures crystallized at two different substrate temperatures cannot be explained unambiguously. On the one hand, the low substrate temperature (high cooling rate of the droplet) promotes the formation of smaller crystals. On the other hand, the crystals formed in this case can have a less regular shape than those formed at moderate crystallization rate at room substrate temperature. The pore sizes, as follows from the measurements

at  $T_s = 20$  and  $-18^\circ\text{C}$ , decrease with decreasing substrate temperature (data for liquid nitrogen temperature are absent). It is required to perform more detailed studies, in particular, of the characteristics of the crystal structure under various cooling conditions, using advanced methods of high-speed visualization of the laser initiation process.

Thus, it is shown that the results of laser initiation experiments are a specific characteristic of thermal transformation of irradiated energetic materials. The qualitative similarity of explosive transformation characteristics under laser initiation and during combustion at increased pressure means that the laser initiation technique can be used for rapid testing of mixtures of variable composition and structure. In addition, advantages of laser initiation are the possibilities of handling explosive samples of limited mass and providing reliable safe test conditions.

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