Initiation of furazanotetrazinedioxide and mixes on its basis by high-current electron beam

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Abstract. The results of study of physicochemical processes developing in the samples of furazanotetrazinedioxide (FTDO) and its mixes with dinitrodiazapentane (DNP) upon irradiation by the high-current electron beam with the energy density varied in the range of 0.05–60 J/cm² are presented. Pre-explosion processes taking place in materials under examination at below threshold modes of excitation are studied. Electron beam threshold energy densities leading to explosive decomposition of FTDO and FTDO/DNP mixes are determined. Noticeable effect of the electron beam energy density on kinetic characteristics of explosive decomposition process of FTDO is discovered. Spectra of the products of FTDO explosive decomposition are measured at explosion initiation in the atmosphere.

1. Introduction

The endothermic substance furazanotetrazinedioxide (FTDO, C₂N₆O₃) with high enthalpy of formation (4200 kJ/kg) is of great interest as a perspective high-energy material [1]. However, its practical use is complicated due to the high sensitivity to the mechanical impact which is comparable to sensitivity of lead azide. For resolving this problem it is suggested to crystallize FTDO in a mix with dinitrodiazapentane (DNP, C₃H₄N₄O₄) [2]. For the safe work with FTDO it is necessary to know not only its sensitivity to mechanical impact but its sensitivity to other energetic impacts including high-current electron beams (HCEB) action.

In the present work the results of experimental investigations of physicochemical processes developing in FTDO and its mixes with DNP (with the FTDO content of 75 weight %) under irradiation by electron beam with varied energy density are presented. Researches in this direction will allow more complete determination of explosive conversion mechanism of FTDO and mixes on its basis.

2. Experiment

FTDO samples were pressed with a pressure of 400 MPa up to density of 1.44 g/cm³ in the form of tablet with 3 mm diameter and 250–450 µm thickness. Samples of FTDO/DNP mixes of 450–800 µm thickness were prepared by crystallization of corresponding melt at the substrate temperature of 20°C. Explosive decomposition of FTDO was initiated by electron beam in atmosphere. FTDO/DNP samples were initiated in vacuum chamber at the pressure of ~10⁻³ Torr. Nanosecond electron accelerator based on pulsed high-voltage generator GIIN-600 (average electron energy ~250 keV, duration of current pulse at FWHM ~15 ns) was used as a source of excitation (initiation). The energy...
density $H$ of electron beam ejected into the atmosphere was varied in the range of $0.05–0.28 \text{ J/cm}^2$ by changing the distance between output foil of electron accelerator and sample and it was measured by radiation-chemical method. Upon irradiation of samples in a vacuum chamber the $H$ value was varied in the range of $0.05–60 \text{ J/cm}^2$ by changing the cathode-anode distance and using diaphragms.

Schemes of initiation of energy materials explosion by electron beam are given in [4]. FTDO sample under examination was fixed on a sample holder placed in atmosphere and was excited by electron beam through the anode aperture and a 30 $\mu$m thick aluminum foil that separates vacuum diode from atmosphere. Maximum energy density of HCEB ejected into the atmosphere was observed in near-foil region and reached $0.28 \text{ J/cm}^2$. Increasing distance between foil and sample led to decrease of HCEB energy density that reached $0.05 \text{ J/cm}^2$. Sample holders were prepared from different metals (aluminum, duraluminum, copper and etc.). An obstacle (20 mm $\times$ 10 mm $\times$ 2 mm aluminum plate) was placed at distance $L$ from explosive's surface. It was stopping the plasma jet formed during FTDO explosion. The purpose of using an obstacle was increasing intensity of explosive glowing and dimensional localization of plasma torch. Energy density of electron beam $H$ was 0.05, 1.0 and 60 J/cm$^2$ during experiments of FTDO/DNP mixes initiation in vacuum chamber. High $H$ values were obtained in the regime of electron beam self-focusing and were determined by the formation of erosion spots on the surfaces of metals with different sublimation temperatures. Kinetic characteristics of FTDO cathodoluminescence and glowing of the products of explosive decomposition were registered by measurement system based on grating monochromator MDR-23, PMT-84 and oscilloscope Tektronix DPO 3034 paired with PC. The resulting time resolution of measurement system was ~15 ns. Integral spectra of explosion were registered by means of a fiber-optical spectrometer AvaSpec - DUAL. Spectral range of measurements was 480–980 nm, spectral resolution ~1.5 nm. Measurements were carried out at initial temperature of 300 K.

3. Experimental results

3.1. Luminescence, destruction and explosive decomposition of FTDO samples upon excitation by electron beam in atmosphere.

Electron beam impact on FTDO samples at energy density varied in the range of $0.05 \leq H \leq 0.28 \text{ J/cm}^2$ has been studied. A sample under examination was fixed on metal sample holder and was excited by electron beam.

**Cathodoluminescence.** It has been revealed that irradiation of samples by electron beam with the value of energy density $H \sim 0.05 \text{ J/cm}^2$ is accompanied by pulsed cathodoluminescence (CL). CL spectrum consists of a wide emission band with maximum at $\lambda = 580 \text{ nm}$. The band's full width at half maximum is ~0.36 $\text{ eV}$, decay time of luminescence $\tau \leq 10 \text{ ns}$.

**Kinetics of explosive glowing.** Explosive decomposition of samples is observed at energy density of electron beam $H \geq 0.09 \text{ J/cm}^2$. The value of threshold energy for FTDO is close to the threshold energy of initiation for heavy metal azides (HMA) and amounts ~0.1 $\text{ J/cm}^2$. Kinetics of FTDO explosive decomposition represents two pulses following one after another [4]. The first short peak corresponds to sample cathodoluminescence and can be used as a reference representing irradiation instant of sample. The second long-time peak corresponds to glowing of products of explosive decomposition. At $H^* \sim 0.09 \text{ J/cm}^2$ glowing of explosion products arises with a delay of $t_{\text{ind}} \sim 1200 \text{ ns}$ after irradiation instant. An increase of electron beam energy density within the range of $0.09 \leq H \leq 0.25 \text{ J/cm}^2$ leads to monotonous reduction of the induction period from 1200 to 30 ns and simultaneous reduction of duration of explosive luminescence pulse. Duration of induction period versus HCEB energy density relation is given on figure 1.

**Dimensional effect.** During the study a dimensional effect was discovered. This effect is that the explosion initiation probability depends on FTDO sample thick (figure 2). At $H = 0.25 \text{ J/cm}^2$ FTDO samples with thickness of 320 $\mu$m had initiation probability $P = 1$. Meanwhile the samples with thickness less than 280 $\mu$m didn't explode ($P = 0$) but they were only destructed into separate segments.
Spectra of the products of explosive decomposition. It has been found that two types of glowing are distinguished in spectra of explosion plasma of FTDO: line spectrum of atoms being a part of explosive material and a sample holder and continuous spectrum representing glowing of dense low-temperature plasma. Line and continuous spectra intensity ratio depends on a mass of explosive, sample holder material and conditions of plasma torch expansion (free expanding into vacuum and interaction with a solid obstacle). Emission spectra of products of explosive decomposition of FTDO samples fixed on aluminum and magnesium alloy (MA18) holder were measured by means of fiber-optical spectrometer AvaSpec - DUAL. They are presented on figure 3 a and b respectively.

Figure 1. Effect of density of electron beam energy on duration of induction period of FTDO

Figure 2. Dependency of the explosion initiation probability on the thickness of FTDO samples excited by HCEB ($H = 0.25 \text{ J/cm}^2$)

Figure 3. Spectra of explosive glowing of FTDO placed on aluminum (a) and magnesium alloy MA18 (b) holders at initiation by electron beam.

It can be seen that the most intensive lines in FTDO explosion spectra in visible region refer to atomic lines of alkali metals: Na I: 588.99; 589.59; Li I: 670.78; K I: 766.49; 769.89 nm. Two atomic lines Na I 588.99 & 589.99 nm are registered as one line due to insufficient spectral resolution of AvaSpec - DUAL spectrometer. A comparison of spectra obtained during explosion of FTDO samples on two different holders shows significant grow of Li I line (670.78 nm) intensity and appearance of additional Li I line (610.36 nm on figure 3, b). This evidences that the elements from holder get into plasma torch of explosion (MA18 alloy contains 10% of lithium). Analysis of experimental data shows that alkali metals enter into FTDO during the process of its producing.
3.2. Kinetics of explosive decomposition of FTDO/DNP (75/25) samples at excitation by electron beam in vacuum.

Explosive decomposition of desensitized FTDO/DNP (75/25) samples are observed only at the maximum energy density of HCEB ($H = 60 \text{ J/cm}^2$). At $H \geq 0.1 \text{ J/cm}^2$ only destruction of irradiated surface takes place. And the effects that are typical for explosive decomposition are not observed. Slow combustion and low-speed detonation of FTDO/DNP samples at $H = 0.1$ and 1.0 J/cm$^2$ have not been registered. Irradiation of FTDO/DNP samples by electron beam with $H \sim 1.0 \text{ J/cm}^2$ doesn't lead to their explosive decomposition. However size of produced fragments shrinks to dozens of micrometers (a powder is formed at sample location). Only a pulsed cathodoluminescence has been observed at the instance of excitation pulse. Irradiation of FTDO/DNP samples at $H^* \sim 60 \text{ J/cm}^2$ leads to detonation. The high rate of chemical reaction expansion through the sample (~9 km/s) and scabbing from the backside of metal plate carrying an energetic material sample are the facts proving this. Oscillograms of explosive glowing of FTDO/DNP (75/25) during free expansion of explosion plasma are identical to oscillograms of explosive glowing of FTDO at $H > 0.25 \text{ J/cm}^2$. That is the glowing of products of explosive decomposition of FTDO/DNP (75/25) appears with a delay of ~20–30 ns after sample irradiation instant, and duration of pulse of explosive glowing does not exceed 400 ns.

4. Discussion

Qualitative description of low-threshold initiation of FTDO samples and high-threshold initiation of FTDO/DNP (75/25) mixes on physical level can be provided in the context of thermal mechanism of process developing. This description is the following. Decomposition rate of FTDO is high and during the beam action this process supersedes gas-dynamic unloading from irradiated volume framed by beam diameter and depth of electron penetration (about 0.4 mm). So, conditions of process development are close to adiabatic, especially at high impact levels. This can explain both low energy threshold of initiation and reduction of explosion delay and duration of explosive glowing with increasing $H$. An alternative scenario occurs in case of FTDO/DNP (75/25) mixes. At relatively low excitation levels ($H \sim 1 \text{ J/cm}^2$) characteristic time of gas-dynamic unloading is less than characteristic time of heat generation from chemical reaction in heating foci. This is proved by observed processes of samples fracture and expansion of unreacted substance. It is obvious that initiation of desensitized compound is possible only at high intensities of beam energy input which accumulates necessary heat in decomposition foci. In other words it is possible at high values of $H$. It is necessary to note that the value $H = 60 \text{ J/cm}^2$ used in these experiments certainly exceeds threshold of initiation energy which will be refined in further experiments.

Analyzing explosive conversion of pressed FTDO samples it should be kept in mind that approximate estimate of substance heating gives very low values of surface layer temperature. If we take $H^*$ equal to 0.1 J/cm$^2$, take into account FTDO heat capacity that is equal to 1 J/g·K and its density of 1.5 g/cm$^3$ and take into account that electrons are being absorbed in 0.04 cm thick layer, all this will give average layer heating of 2.2 K. That is to say, here is the same situation as with the electron initiation of HMA where thresholds of explosive decomposition are lying in the range of 0.1 J/cm$^2$ [5]. In [5, 6] a model of "hot centers" formation in volume of EM during localization of energy evolved as a result of electric breakdown was suggested for description of HMA behavior. At sufficiently high rates of the reaction of chemical decomposition the process may develop out of the region of energy release of electron beam and expand on the whole mass of explosive.

Analyse of results of FTDO initiation by electron beam proves that initiation model can be based on electronic discharge model published in [5–9]. Particularly the evidence of this is dimensional effect discovered in the presented work. Namely with decreasing of FTDO sample thickness to 280 µm a part of high-energy electrons traverse the sample and are not absorbed. As a result, intensity of electric field related to negative spatial charge of electron beam injected into the sample decreases and becomes insufficient for initiation of electric breakdown.
5. Conclusion
Initiation of FTDO and crystalized FTDO/DNP (75/25) mix by high-current electron beam has been studied. It has been revealed that luminescence and samples fracture are observed at under-threshold densities of electron beam. Initiation thresholds of explosive decomposition have been measured for materials under examination. It has been established that the sensitivity of FTDO to the action of a pulse of electron beam is close to the sensitivity of HMA, and the sensitivity of FTDO/DNP mix is close to the sensitivity of PETN. In contrast to HMA, experiments with FTDO showed strong dependence of ignition delay time and duration of a pulse of explosive glowing on energy density $H$ of electron beam. With an increase of $H$ from 0.1 to 1.0 J/cm$^2$, the velocity of chemical decomposition increases by more than an order and reaches ~9 km/s. Dimensional effect proving electric-discharge mechanism of FTDO initiation by electron beam has been discovered. It has been shown that behavior of FTDO and FTDO/DNP (75/25) mixes under HCEB irradiation can be described in terms of models of electron beam initiation of HMA and PETN respectively. The results of experiments evidence possibility of using electron beam initiation method for rapid testing of varying mixes.

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