Preexplosion Phenomena in Heavy Metal Azides


INTRODUCTION

The explosive decomposition of heavy metal azides (HMA) has attracted the attention of researchers for about half a century [1]. This unflagging interest is motivated by the fact that HMA are simple, and, hence, adequately studied representatives of initiating explosives and are used as a model object for this class of systems [2-4].

By the beginning of the 1980s, a great amount of experimental data had been accumulated and the basic concepts of the mechanism of explosive decomposition of HMA had been formulated [2] and became the basis of most of the subsequent studies (see, for example, [5, 6]). However, an analysis of experimental data and relevant physical models shows the lack of a clear understanding of the mechanism of explosive decomposition of HMA.

Leaving aside work of purely applied direction, it is possible to distinguish two lines of experimental studies. The first is concerned with studying slow processes of HMA decomposition on exposure to heat, light, ionizing radiations, and electric and magnetic fields [6, 7]. Obviously, using data of such studies to develop an adequate model of the explosive decomposition is rather problematic [3, 4].

The second line involves direct experimental investigation of explosive decomposition. An explosion is initiated by a pulsed action (shock, light pulse, accelerator pulse, etc.). Experiments are performed so as to record the explosion [2] or, at best, the time period (induction period) between the initiating pulse and the moment of explosion, determined by a light flash [8] or fracture of the sample (high-speed video recording) [2]. It is clear that in such experiments, researchers can record only integral parameters of the process and cannot obtain the most important information — variations in the sample's characteristics (or phenomena accompanying these variations) during explosive decomposition.

Because of the lack of adequate experimental material, the existing concepts of the mechanism of explosive decomposition of HMA are of a speculative nature [2, 5]. According to these concepts, the energetics of the explosion is ensured by the exothermic reaction

\[ 2N_3 \longrightarrow N_6 \longrightarrow 3N_2 + Q. \] (1)

According to quantum-chemical calculations (for isolated radicals \( N_3^* \)), we have \( Q \approx 9-12 \text{ eV} \) [2]. In solids, this reaction is possible when two holes are located at neighboring sites of the lattice. The most probable location is assumed to be a cation vacancy [5]. For development of an explosion, it is necessary that even part of the energy \( Q \) be expended in reproduction and multiplication of holes. Various pathways of multiplication are considered. Most researchers believe that the energy is released in the form of phonons and local heating of the lattice.
ensures thermal generation of several electron-hole pairs [2, 8]. A so-called thermal explosion occurs in this case [9]. The minority of authors [5] hold to the concept of a chain explosion [9]. In this case, hole multiplication is assumed to proceed by the mechanism of photomultiplication [5] or generation of a hot hole directly as a result of reaction (1) with subsequent multiplication by collision ionization [10]. It should be emphasized that even the basic idea of the main role of reaction (1) in the explosive decomposition of HMA have not been supported experimentally, let alone the chain or thermal character of the explosion.

In view of the foregoing, progress in the understanding of the mechanism of HMA explosive decomposition can be achieved only by developing new experimental approaches to recording preexplosion variations in the characteristics (physical properties) of samples in real time. We developed such an approach based on the flash techniques used in radiation physics [11], which were adapted to the specific features of examination of exploded samples. The present paper considers results of implementation of this approach and an explosive decomposition model developed on the basis of these results.

1. OBJECTS AND TECHNIQUE

The objects of investigation were the azides of silver (AgN₃), thallium (TlN₃), and lead [Pb(N₃)₂]. The azide powder were synthesized by the method of double-jet crystallization [12]. The concentrations of the main impurities (Fe, Si, Ca, Mg, A1, and Na) were determined by polarography and chelatometry and did not exceed 10¹⁶ - 10¹⁷ cm⁻³.

The examined samples of thallium and lead azides were pressed pellets having a diameter of 10 mm and a thickness of 300–400 µm for thallium azide and a diameter of 2.5 mm and a thickness of 30-40 µm for lead azide. Silver azide samples were filamentary crystals with typical dimensions of 0.1 x 0.05 x 10 mm and macrocrystals with typical dimensions of 0.5 x 3 x 3 mm. The crystals were grown from solution by the procedure of [12]. The concentration of cation vacancies in the crystals did not exceed 10¹⁶ cm⁻³ [12].

The complex of apparatus (Fig. 1) comprises experimental facilities employed in flash radiolysis and photolysis [11]. The sources of excitation (initiation) were a GIN-600 high-current electron accelerator (effective electron energy 300 keV, current density 1000 A/cm², and pulse duration 3 nsec) and a YAG Nd₃⁺-laser (λ = 1064 nm, pulse duration 30 nsec, and pulse energy 0.5–30 mJ).

Laser initiation was used to study the kinetics of signals, and the luminescence spectrum was examined using initiation by a pulsed electron accelerator (the identity of the spectral glow distribution for both types of initiation was especially tested).

The recording instrumentation incorporates several paths synchronized by reference pulses that arise when an initiating pulse acts on a detector (action of dispersed laser light on the photodetector, direct action of a laser pulse on the acoustic detector, etc.). The accuracy of time referencing for signals from different paths is ±3 nsec. A detailed description of the apparatus complex is given in [3, 4, 13, 14].

2. EXPLOSIVE CONDUCTIVITY

A series of experiments on the conductivity of HMA during explosive decomposition (explosive conductivity) was performed as follows. A filamentary crystal of silver azide with typical cross-sectional dimensions of 100 x 150 µm was fixed in a 2-mm-wide air (or vacuum) interelectrode gap. Explosion was initiated by a laser pulse. The initiation was homogeneous because the beam covered the entire length of the interelectrode gap and the energy of the initiating photons (λ = 1064 nm) was in the range of transparency of the crystal (optical width of the forbidden band of silver azide ≈3.5 eV [15]).

A typical shape of the explosive conductivity pulse is shown in Fig. 2a. The increase in conductivity in the first peak occurs when the crystal has not yet failed (preexplosion conductivity), the decay of the first peak is attributed to the discontinuity of the sample due to propagation of the mechanical stresses resulting from the decomposition, and the subsequent increase in conductivity is related to the conductivity of the explosion products (plasma).

To verify this assumption, we performed the following series of experiments. The lateral face of the sample was fixed on the acoustic detector window to synchronize measurements of the conductivity signal and the acoustic signal. The beginning of sample deformation, followed by mechanical failure (explosion), was determined from the leading edge of the acoustic detector.

The conductivity of the sample ahead of the leading edge of the acoustic signal (Fig. 2b) corresponds to the yet undeformed sample, i.e., can be identified as preexplosion conductivity. A detailed examination of the detected preexplosion conductivity revealed a number of regularities [3, 4, 13, 16].