DIFFRACTION STUDIES OF TATB

***Ten K.A.,*[*1] *Pruuel E.R.,*[1] *Lukyanchikov L.A.,*[1] *Tolochko B.P.,*[2] *Shmakov A.N.,*[3] *Zhulanov V.V.,*[4] *Shehtman L.I.,*[4] *Aminov Yu. A.,*[5] *Muzyrya A.K.,*[5] *Kostisyn O.V.,*[5] *Smirnov E.B.*[5]

[1]*LIH SB RAS, Novosibirsk, Russia,* [2]*ISSCM SB RAS, Novosibirsk, Russia,* [3]*BIC SB RAS, Novosibirsk, Russia,* [4]*BINP SB RAS, Novosibirsk, Russia,* [5]*RFNC–VNIITF, Snezhinsk, Russia*

[*]*ten@hydro.nsc.ru*

This paper presents the results of diffraction studies of TATB-based explosives with the use of synchrotron radiation from the VEPP-3 accelerator complex; more specifically, measurement of diffraction reflections of TATB under heating up to 240°C and compression in diamond anvils to pressures of up to 6.5 GPa.

Keywords: synchrotron radiation diffraction reflections, TATB.

Introduction. Detonation characteristics of explosives are in strong dependence on internal parameters of initial compounds. Conditions of initiation, as well as crest values in the cumulative process, depend on the presence of voids (pores) and their size distribution. Monitoring of such parameters as irregularity size distribution and variation of lattice parameters as a function of temperature are a topical task. Diffraction techniques allow one to find out internal parameters of TATB explosives in a nondestructive way. The object of the study were new explosives, promising from the industrial point of view, which were manufactured on the basis of 1,3,5-triamino-2,4,6-trinitrobenzene (TATB). TATB has high effective temperature resistance and ability to decompose (without explosion) under further heating [1].

Lawrence Livermore National Laboratory pioneered the use of synchrotron radiation to study the internal structure of explosives [2]. Works [3, 4] present data on the internal structure (irregularities) of U.S. TATB-based compounds. Those data were obtained on the Advanced Photon Source accelerator at Argonne National Laboratory. In our country, the first application of SR to this area was performed by BINP on VEPP-3 [4]. Irregularities of up to 3-5 μm in size were detected by the method of X-ray microtomography [5].

Below are presented the results on the internal structure of TATB samples as studied by two diffraction techniques:

- Determination of the TATB crystal structure under heating up to 240°C via the measurement of the deviation of diffraction peaks through large angles (up to 90°C) with the detector OD-3M (3000 registration channels) [6].
- Determination of the TATB crystal structure under isothermal compression up to 6.5 GPa in diamond anvils.

Study of the crystal structure of samples of TATB-based explosives under heating up to 240°C. The structure of TATB crystals was measured at the "Precision diffractometry II" station on VEPP-3 (Fig. 1). A thermostat of the station *(Anton Paar XRK900)* ensured a set rate of heating and temperature control with an accuracy of 1%. The maximum operating temperature of the water-cooled thermostat was 700°C. The diffractograms were recorded for compressed TATB samples (15 mm in diameter and 2 mm high) with the density ρ= 1.819 g/cm^3.

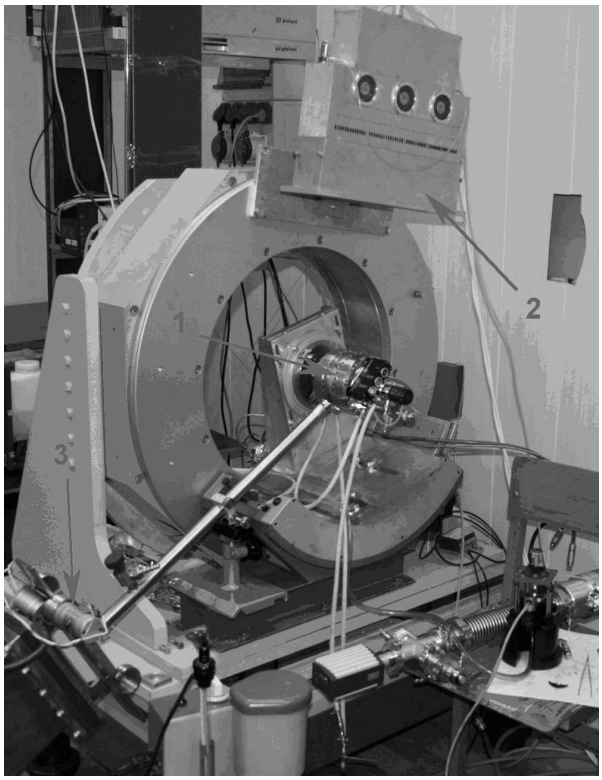

Figure 1. General view of the "Precision diffractometry II" station. 1 — the thermostat with the sample, 2 — the detector OD-3M, 3 — the shutter.

The angle measurement interval of the detector OD-3M is $\sim 30°$ (3000 channels), so the measurements were conducted in two steps: 1) in the range of 30°– 60°, and 2) in the range of 25°– 55°. For the purpose of precise reference of diffraction peaks, the samples were calibrated in the range of 10°– 90°. The main maximum corresponds to the angle $2\theta = 28.301$ degrees (the precision is ± 0.015 degrees), and the second one to $2\theta = 58.502$ degrees.

In both cases, we measured the angle of deviation of diffraction peaks with the temperature of sample varying from T_0 = 27°C (room temperature) to 240°C. The heating was carried out at a rate of 1°C/min for 20 minutes (i.e. 20°C) with subsequent holding for 20 minutes. Then again, heating up to 20°C and so on. After holding for 20 minutes, the sample was left to cool for 30 minutes. During the cooling, the diffractograms were recorded every 5 minutes.

In Fig. 2 one can see the dynamics of main peak location vs. temperature, and Fig. 3 shows the initial and final positions of the peaks. The uniform heating of the sample leads to a linear shift of the peak position. When the sample temperature was increased by

213°C, the peak shifted by 1.45 degrees.

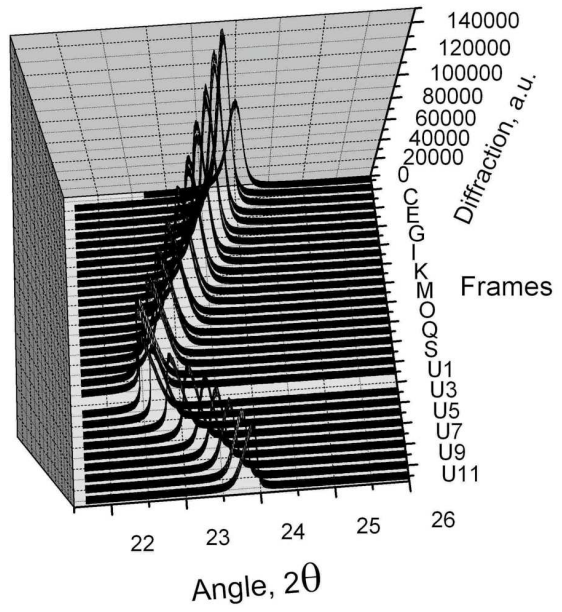

Figure 2. Variation of the position of the main maximum (peak) under heating up to 240°C and subsequent cooling.

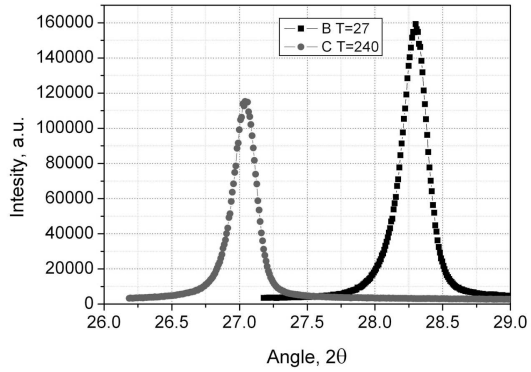

Figure 3. Position of the peaks (the angle range $2\theta = 26°$–$29°$) at room temperature (T = 27°C) and under heating up to T = 240°C.

The distance between the TATB layers is found from the formula $2d\sin(\theta) = \lambda$, where $\lambda = 1.731\mathring{A}$ is the wavelength of the monochromator. $d = 3.5404\mathring{A}$ corresponds to the first peak.

At T = 240°C the angle $2\theta = 27.0417$°C, which corresponds to the distance between the layers $d_2 = 3.70189\mathring{A}$. Under heating, the 2θ angle variation is 1.25832°C. In our settings, this angle variation corresponds to 147 scale divisions of the detector OD-3M, i.e. 1.45°C per detector scale division. Thus, the sensitivity of this technique (at temperature variation) is 1.45°C/division.

The increase in the distance between the layers (offset) under heating is $\Delta d = 0.1615\mathring{A}$.

The relative offset is $\frac{\Delta d}{d} = 0.046$. The angle θ is determined with accuracy $\Delta\theta = 0.03$ degrees (900 over 3000 channels of the detector). The wavelength $\lambda = 1.731\mathring{A}$ is determined with a relative accuracy of 10^{-4}.

The accuracy of determination of d depends on the accuracy of evaluation of λ and θ and equals $\frac{\Delta d}{d} = \sqrt{(\frac{\Delta\lambda}{\lambda})^2 + (\frac{\Delta\theta}{\theta})^2} = \sqrt{10^{-8} + 0.01^2} = 0.01$. That is, the absolute accuracy of determination of interplanar distances is $\Delta d = 3.54\mathring{A} * 0.01 = 0.035\mathring{A}$.

The lattice parameter of TATB crystal equals twice the distance between the layers and is $C = 7.081\mathring{A}$.

Measurement of the TATB crystal structure under compression in diamond anvils. The measurements were conducted at the "Diffractometry at high pressure" station, which is located on channel 4 of the VEPP-3 storage ring. The measurement design is shown in Fig. 4. A sample (TATB powder) was placed inside a metal gasket, which was crimped between two diamond anvils.

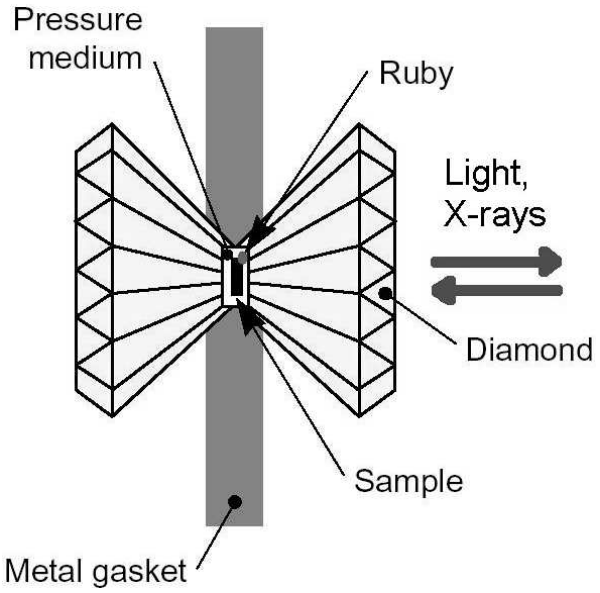

Figure 4. Design of the measurement of diffraction reflections under compression in diamond anvils.

For the compression to be uniform, the TATB powder was placed in a liquid (10 parts of methanol per 1 part of ethanol). The pressure was calibrated from the shift of the lines of a ruby crystal placed beside the sample inside a metal cage.

The initial density of the TATB powder was 1.1 g/cm^3; the measurements were performed at T_0 = 27°C; the time of exposure was 1 to 4 hours. Two series of experiments were conducted: in the first, the pressure was increased up to 3.15 GPa and in the second, up to 6.5 GPa (Fig. 5).

The main peak, which corresponds to the interplanar distance, falls on the angle $2\theta = 4.64$ degrees and shifts to 4.99 degrees with the pressure increased up to 6.5 GPa (Fig. 6). The changes in the unit cell are shown in Table 1. In this pressure range, our data are in good agreement with data on U.S. TATB compounds [7].

The dynamics of the maximum peak ($2\theta = 6.7°$) turns out to be interesting. This peak appears because

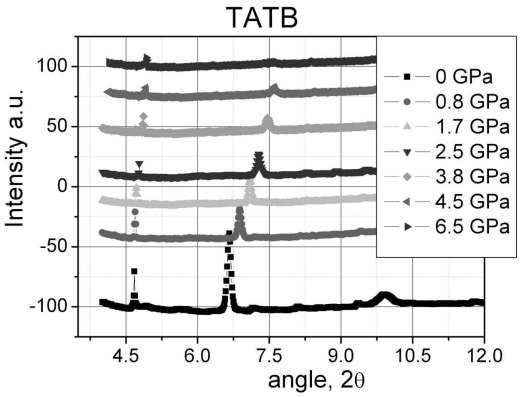

Figure 5. Diffractograms of TATB under isothermal compression to a pressure of 6.5 GPa (2θ, the angle of radiation scattering, is laid along the x axis and the radiation intensity is plotted along the y axis.

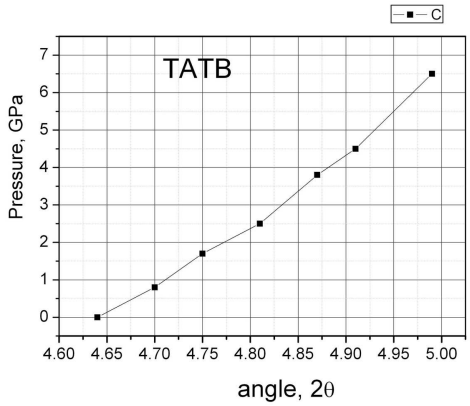

Figure 6. Change in the position of the main peak of TATB under isothermal compression to a pressure of 6.5 GPa (2θ, the angle of radiation scattering, is laid along the x axis).

Table 1. Change in unit cell volumes under isothermal compression of TATB

Preasere, GPa	V/V_0
0	1
0.8	0.97457
1.7	0.94399
2.5	0.91468
3.8	0.8649
4.5	0.84393
6.5	0.80399

of the ordered arrangement of TATB powder (the powder is composed of ordered groups of crystals). When the pressure is increased, the powder is distributed uniformly, and this peak disappears. The same peak was observed in [7] and studied in more detail in [8].

Conclusion. Measurement of diffraction reflections makes it possible to determine their position with an accuracy of 10^{-2} degrees and thus determine the interplanar distances with an accuracy of $0.03\mathring{A}$. This technique is sensitive to changes in the temperature of sample. An offset of peaks is recorded when the temperature varies by 1.45°C. It makes sense to measure the interplanar distances in TATB-based compounds at different initial densities and different rates of heating (including periodic ones). The influence of the time of high-temperature holding on lattice parameters is also of interest.

Under isothermal compression up to pressures of 6.5 GPa, the crystal structure of TATB changes almost linearly. The unit cell volumes in a crystal lattice vary within 0.804. During compression of TATB powder in a liquid, ordered groups of TATB crystals experience rearrangement.

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1. Garmasheva N. V., Filin V. P., Chemagina I. V. *et al.* // Proceedings of the International Conference VII Zababakhin scientific readings. VNIITF, Snezhinsk, 2003, P. 2.
2. Willey T. M., van Buuren T., Lee J. R. *et al.* // Propellants, Explosives, Pyrotechnics, V. 31, No. 6, 2006, P. 466.
3. Willey T. M., Overturf G. // 40th International Annual Conference of ICT Karlsruhe, Germany. June 26, 2009.
4. Kuper K. E., Ten K. A., Pruuel E. R. // Physics of Extreme states of Matter—2009 / Ed. by Fortov V. E. *et al.* Chernogolovka: IPCP RAS, 2009. P. 130.
5. Kuper K. E., Ten K. A., Pruuel E. R. *et al* // Proceedings of the International Conference X Zababakhin scientific readings. VNIITF, Snezhinsk, 2010, P. 101 [in Russian].
6. Aulchenko V. M., Zhulanov V. V. *et al* // Nuclear Instruments and Methods in Physics research A, 2009, V. 603, P. 76.
7. Stevens L. L., Velisavljevic N., Hooks D. E., Dattelbaum D. M. // Propellants, Explosives, Pyrotechnics, V. 33, No. 4, 2008, P. 286.
8. Tumanov N. A. Combined use of single-crystal and powder X-ray diffraction for challenging studies of new phases, including the metastable ones, of molecular crystals. Thesis work for the degree of Candidate of Chemical Science. Novosibirsk State University, 2010 [in Russian].