X-ray powder diffraction data for tetrazene nitrate monohydrate, $C_2H_9N_{11}O_4$

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X-ray powder diffraction data, unit-cell parameters, and space group for tetrazene nitrate monohydrate, $C_2H_9N_{11}O_4$, are reported [$a = 5.205(1)$ Å, $b = 13.932(3)$ Å, $c = 14.196(4)$ Å, $\beta = 97.826(3)°$, unit-cell volume $V = 1019.8(4)$ Å^3, $Z = 4$, and space group $P2_1/c$]. All measured lines were indexed and are consistent with the $P2_1/c$ space group. No detectable impurities were observed. © *The Author(s), 2021. Published by Cambridge University Press on behalf of International Centre for Diffraction Data.* [doi:10.1017/S0885715621000579]

Key words: X-ray powder diffraction, tetrazene, hydrate, explosive, munitions industry

I. INTRODUCTION

Tetrazene, first prepared in 1892 by Thiele (1892), is a primary explosive mainly used in the munitions industry as an energetic sensitizer particularly in percussion and stab priming compositions (Hagel and Redecker, 1986; Matyáš and Pachman, 2013). It was first studied in 1910 by Hofmann and Roth (1910) who also prepared several anionic and cationic salts of tetrazene. This ability is attributed to the zwitterion nature of tetrazene, which was discovered after several revisions of the molecule structure in 1971 by Duke (1971). These salts, aside of few minor mentions (McNutt, 1933; Straka and Vachovec, 1944; Conduit, 1955; Patinkin *et al.*, 1955; Špičák and Šimeček, 1957; Багал, 1975), were never truly studied.

We have not found this compound in the CSD database (Groom *et al.*, 2016) or in the PDF4+ database (Gates-Rector and Blanton, 2019). Therefore, we have decided to characterize this compound by an X-ray powder diffraction (XRD) technique. In our study, we present powder data for tetrazene nitrate monohydrate ($C_2H_9N_{11}O_4$; Figure 1).

II. EXPERIMENTAL

A. Synthesis

The synthesis of tetrazene nitrate was inspired by the first preparation of this compound type by Hofmann and Roth (1910): Tetrazene (1.5 g; 7.97 mmol) was dissolved in 65% nitric acid (20 ml; 292 mmol) and diethylether (80 ml) was added dropwise. First, a layer of heavy white liquid separates in the mixture, which with further diethylether starts to coagulate to form lumps that eventually within a matter of minutes fall apart into course heavy white powder. The solid was filtered off and washed with ether.

B. Specimen preparation

Tetrazene nitrate monohydrate is a primary explosive and therefore must be handled very carefully. The synthesized powder is very well crystallized with crystallite size suitable for powder XRD, and therefore, no grinding is necessary. The sample could be front-loaded into the specimen holder and limited force was used to make the sample surface flat.

C. Diffraction data collection and reduction

The diffraction pattern for the title compound was collected at room temperature with an X'Pert[3] Powder θ–θ powder diffractometer with parafocusing Bragg–Brentano geometry using Cu$K\alpha$ radiation ($\lambda = 1.5418$ Å, Ni filter, generator setting: 40 kV, 30 mA). An ultrafast PIXCEL detector with 255 channels was employed to collect XRD data over the angular range from 5 to 80° 2θ with a step size of 0.026° 2θ and a counting time of 0.618 s per step.

The software package HighScore Plus V 4.8 (PANalytical, Almelo, Netherlands) was used to smooth the data, to fit the background, to eliminate the $K\alpha_2$ component and the top of the smoothed peaks were used to determine the peak positions and intensities of the diffraction peaks (Table I). The d-values were calculated using Cu$K\alpha_1$ radiation ($\lambda = 1.5406$ Å).

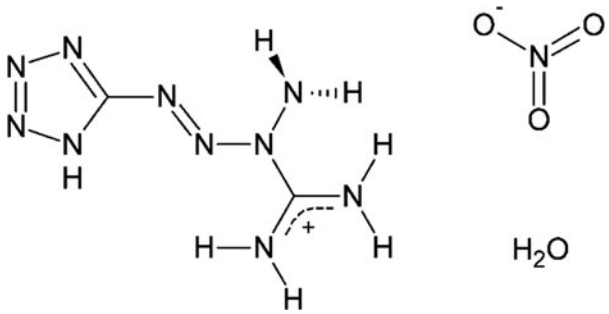

Figure 1. Structural formula of tetrazene nitrate monohydrate.

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TABLE I. Indexed X-ray powder diffraction data for $C_2H_9N_{11}O_4$.

$2\theta_{obs}$ (°)	d_{obs} (Å)	I_{obs}	h	k	l	$2\theta_{cal}$ (°)	d_{calc} (Å)	$\Delta 2\theta$
8.911	9.9156	1	0	1	−1	8.927	9.8977	0.016
12.565	7.0391	3	0	0	2	12.578	7.0318	0.013
14.089	6.2809	2	0	1	2	14.097	6.2776	0.008
17.166	5.1615	19	1	0	0	17.183	5.1563	0.017
18.314	4.8405	23	1	1	0	18.332	4.8358	0.018
18.623	4.7607	1	1	1	−1	18.611	4.7639	−0.013
19.902	4.4576	7	1	0	−2	19.902	4.4577	−0.001
20.082	4.4180	11	0	3	1	20.120	4.4098	0.038
21.410	4.1470	15	1	2	0	21.423	4.1445	0.013
21.690	4.0941	4	1	2	−1	21.664	4.0989	−0.026
22.874	3.8848	7	0	2	3	22.847	3.8892	−0.027
23.606	3.7658	11	1	1	2	23.603	3.7663	−0.003
24.647	3.6092	3	1	1	−3	24.685	3.6037	0.038
25.295	3.5181	28	0	0	4	25.311	3.5159	0.016
25.811	3.4489	9	1	3	0	25.797	3.4508	−0.014
26.065	3.4159	13	1	2	2	26.104	3.4109	0.039
27.044	3.2945	11	0	3	−3	27.004	3.2992	−0.040
27.683	3.2198	6	1	3	−2	27.717	3.2159	0.034
28.647	3.1137	100	1	0	−4	28.695	3.1086	0.048
29.367	3.0389	13	1	1	−4	29.416	3.0340	0.049
30.731	2.9071	10	1	3	−3	30.716	2.9085	−0.015
30.926	2.8892	14	1	4	0	30.958	2.8863	0.032
31.852	2.8072	1	0	3	4	31.900	2.8032	0.048
32.619	2.7430	3	1	4	−2	32.599	2.7446	−0.019
33.363	2.6835	4	1	1	4	33.340	2.6853	−0.024
34.486	2.5986	3	1	4	2	34.449	2.6014	−0.038
34.795	2.5763	3	2	0	0	34.768	2.5782	−0.027
35.248	2.5442	5	1	4	−3	35.223	2.5459	−0.025
36.252	2.4760	7	0	4	−4	36.276	2.4744	0.023
36.649	2.4501	3	1	5	0	36.629	2.4514	−0.020
36.845	2.4375	3	2	1	1	36.828	2.4386	−0.017
37.318	2.4077	3	0	3	5	37.347	2.4059	0.029
38.143	2.3575	3	1	3	4	38.139	2.3577	−0.004
38.754	2.3217	7	0	6	0	38.748	2.3220	−0.006
39.003	2.3074	4	1	1	5	39.016	2.3067	0.013
39.399	2.2852	2	1	3	−5	39.410	2.2846	0.011
39.756	2.2655	2	2	3	−1	39.704	2.2683	−0.051
39.956	2.2546	3	2	3	0	39.965	2.2541	0.009
40.517	2.2246	1	2	3	−2	40.512	2.2249	−0.005
40.936	2.2028	2	2	2	2	40.959	2.2017	0.022
41.306	2.1839	1	0	5	−4	41.310	2.1838	0.003
42.695	2.1161	6	1	5	3	42.680	2.1168	−0.015
43.440	2.0815	4	0	6	−3	43.456	2.0808	0.016
44.276	2.0441	2	2	2	3	44.253	2.0451	−0.024
44.717	2.0250	1	1	3	−6	44.677	2.0267	−0.039
45.004	2.0127	1	1	1	6	45.048	2.0108	0.044
45.941	1.9738	1	1	6	−3	46.002	1.9714	0.061
46.475	1.9524	2	1	5	4	46.473	1.9525	−0.002
46.872	1.9368	2	2	1	4	46.867	1.9370	−0.005
47.564	1.9102	1	1	5	−5	47.561	1.9103	−0.003
48.105	1.8900	2	1	6	3	48.095	1.8903	−0.011
48.893	1.8613	2	1	3	6	48.883	1.8617	−0.010
50.174	1.8168	1	1	7	−2	50.156	1.8174	−0.018
51.011	1.7889	1	0	6	−5	50.957	1.7907	−0.054
51.249	1.7812	2	2	5	2	51.186	1.7832	−0.063
51.441	1.7750	3	1	7	2	51.474	1.7739	0.033
52.616	1.7380	4	1	0	−8	52.626	1.7377	0.010
53.946	1.6983	3	1	7	3	53.955	1.6980	0.009
54.424	1.6845	1	3	2	−1	54.465	1.6834	0.041
55.977	1.6414	1	2	6	2	55.976	1.6414	−0.001
56.589	1.6251	1	3	3	−1	56.589	1.6251	0.000
57.341	1.6055	1	1	5	−7	57.336	1.6057	−0.005
57.626	1.5983	1	1	0	8	57.608	1.5987	−0.018
58.309	1.5812	3	2	7	−1	58.345	1.5803	0.036
59.402	1.5547	3	3	1	−5	59.401	1.5547	−0.001
59.841	1.5443	2	2	1	−8	59.824	1.5447	−0.017
61.018	1.5173	2	0	7	−6	61.025	1.5171	0.007
61.381	1.5092	1	3	2	3	61.332	1.5103	−0.049
61.818	1.4996	1	1	6	−7	61.806	1.4999	−0.012
62.728	1.4800	1	3	4	−4	62.745	1.4796	0.017
63.248	1.4691	2	1	8	4	63.240	1.4692	−0.009
64.037	1.4529	1	1	4	8	64.031	1.4530	−0.006
64.398	1.4456	1	3	5	−3	64.434	1.4449	0.035
64.577	1.4420	1	2	8	0	64.521	1.4431	−0.056
65.473	1.4244	1	2	8	1	65.464	1.4246	−0.009
65.950	1.4153	1	3	3	−6	65.953	1.4152	0.003
67.032	1.3950	1	2	7	4	67.031	1.3951	−0.001
78.375	1.2191	1	4	2	2	78.361	1.2193	−0.014

Only the peaks with I_{rel} of 1 or greater are presented [$a = 5.205(1)$ Å, $b = 13.932(3)$ Å, $c = 14.196(4)$ Å, $\beta = 97.826(3)°$, unit-cell volume $V = 1019.8(4)$ Å^3, $Z = 4$, and space group $P2_1/c$]. All lines were indexed and are consistent with the $P2_1/c$ space group. The d-values were calculated using Cu$K\alpha_1$ radiation ($\lambda = 1.5406$ Å).

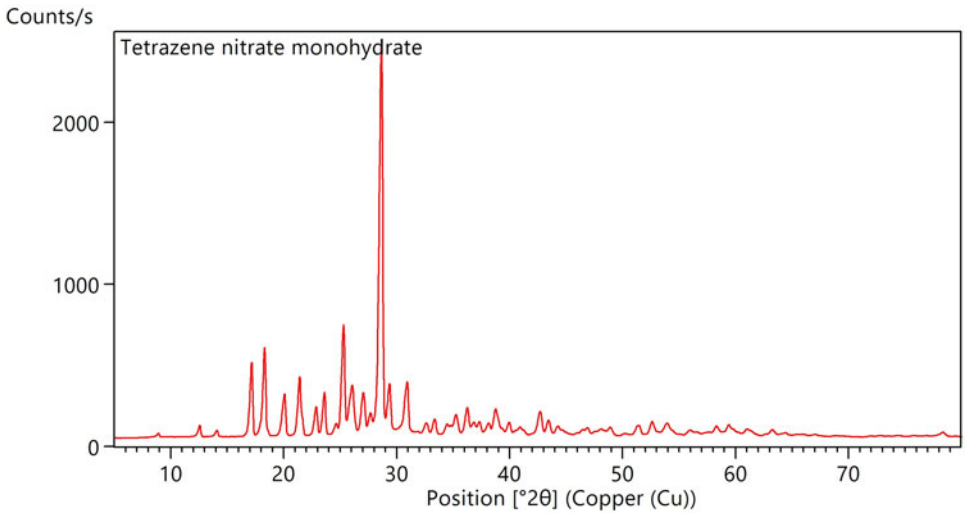

Figure 2. X-ray powder diffraction pattern of tetrazene nitrate monohydrate using Cu$K\alpha$ radiation ($\lambda = 1.5418$ Å).

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III. RESULTS AND DISCUSSION

The automatic indexing of results was obtained using Dicvol (Boultif and Louër, 2004). The experimental powder diffraction pattern is shown in Figure 2. Tetrazene nitrate monohydrate, $C_2H_9N_{11}O_4$, is monoclinic with the space group $P2_1/c$ and unit-cell parameters: $a = 5.205(1)$ Å, $b = 13.932(3)$ Å, $c = 14.196(4)$ Å, $\beta = 97.826(3)°$, unit-cell volume $V = 1019.8(4)$ Å^3, and $Z = 4$. The figures of merits are $F_{20} = 16.3(0.0205, 60)$ and $M_{20} = 8.8$ (de Wolff, 1968; Smith and Snyder, 1979). All measured lines (Table I) were indexed and are consistent with the $P2_1/c$ space group.

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