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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 = 5.212(2) Å, b = 13.923(3) Å, c = 14.206(4) Å, β = 97.898(5)°, unit-cell volume V = 1021.0(5) 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.

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

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I. INTRODUCTION

Tetrazene, first prepared in 1892 by Thiele (Thiele, 1892), is a primary explosive mainly used in the munitions industry as an energetic sensitizer particularly in percussion and stab priming compositions (Matyáš and Pachman, 2013; Hagel and Redecker, 1986). It was first studied in 1910 by Hofmann and Roth (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 (Duke, 1971). These salts, aside of few minor mentions (Patinkin *et al.*, 1955; Conduit, 1955; McNutt, 1933; Straka,and Vachovec, 1944; Bagal, 1975), were never truly studied.

We have not found this compound in the CSD database or in the PDF4+ database (Allen, 2002; ICDD, 2020). Therefore, we have decided to characterize this compound by X-ray powder diffraction technique. In our study, we present powder data for tetrazene nitrate

monohydrate, $C_2H_9N_{11}O_4$.

II. EXPERIMENTAL

A. Synthesis

The synthesis of tetrazene nitrate was inspired by the first preparation of this compound type by Hofmann and Roth (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 is filtered off and washed with ether.

B. Specimen preparation

Synthesized powder sample is very well crystallized with crystallite size suitable for X-ray powder diffraction, therefore the sample could be front-loaded into the specimen holder without grinding.

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 (λ = 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/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 1). The d-values were calculated using Cu $K\alpha_1$ radiation (λ = 1.5406 Å).

III. RESULTS AND DISCUSSION

The automatic indexing of results was obtained using Dicvol (Werner *et al.*, 1985). The experimental powder diffraction pattern is showed in Figure 2. Tetrazene nitrate monohydrate, $C_2H_9N_{11}O_4$, is monoclinic with space group $P2_1/c$ and unit-cell parameters: a = 5.212(2) Å, b = 13.923(3) Å, c = 14.206(4) Å, β = 97.898(5)°, unit-cell volume V = 1021.0(5) 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 1) were indexed and are consistent with the $P2_1/c$

space group.

The single crystal experiment was done at the temperature of 150K and the structure solution was obtained. The title compound is monoclinic with space group $P2_1/c$ and unit-cell parameters: a = 5.1417(2) Å, b = 13.9185(6) Å, c = 13.9598(6) Å, β = 97.189(2)°, unit-cell volume V = 991.18(7) Å^3 and Z = 4. The difference in unit-cell parameters from the single crystal data and the powder diffraction data is due to the temperature expansion.

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TABLE I. Indexed X-ray powder diffraction data for $C_2H_9N_{11}O_4$. Only the peaks with I_{rel} of 1 or greater are presented [a = 5.212(2) Å, b = 13.923(3) Å, c = 14.206(4) Å, β = 97.898(5)°, unit-cell volume V = 1021.0(5) Å^3, Z = 4 and space group $P2_1/c$]. All lines were indexed and are consistent with the $P2_1$ space group. The d-values were calculated using Cu $K\alpha_1$ radiation (λ = 1.5406 Å).

$2\theta_{obs}$ (deg)	d_{obs} (Å)	I_{obs}	h	k	l	$2\theta_{cal}$ (deg)	d_{calc} (Å)	$\Delta 2\theta$
8.911	9.9156	1	0	1	-1	8.928	9.8970	0.017
12.565	7.0391	3	0	0	2	12.571	7.0358	0.006
14.089	6.2809	2	0	1	-2	14.092	6.2795	0.003
17.166	5.1615	19	1	0	0	17.164	5.1621	-0.002
18.314	4.8405	23	1	1	0	18.315	4.8401	0.001
19.902	4.4576	7	1	0	-2	19.869	4.4649	-0.033
20.082	4.4180	11	0	3	1	20.131	4.4074	0.049
20.851	4.2569	1	1	1	-2	20.877	4.2516	0.026
21.410	4.1470	15	1	2	0	21.412	4.1465	0.003
21.690	4.0941	4	1	2	-1	21.647	4.1020	-0.043
22.874	3.8848	7	0	2	-3	22.843	3.8899	-0.031
23.606	3.7658	11	1	1	2	23.596	3.7675	-0.010
24.647	3.6092	3	1	1	-3	24.650	3.6087	0.004
25.295	3.5181	28	0	0	4	25.297	3.5179	0.002
25.811	3.4489	9	1	3	0	25.794	3.4512	-0.017
26.065	3.4159	13	1	2	2	26.100	3.4114	0.035
27.044	3.2945	11	1	2	-3	27.063	3.2921	0.020
27.683	3.2198	6	1	3	-2	27.703	3.2176	0.020
28.647	3.1137	100	1	0	-4	28.654	3.1129	0.007
29.367	3.0389	13	1	1	-4	29.377	3.0379	0.010
30.731	2.9071	10	1	3	-3	30.695	2.9104	-0.036
30.926	2.8892	14	1	4	0	30.962	2.8859	0.036
31.852	2.8072	1	0	3	4	31.896	2.8035	0.044
32.619	2.7430	3	1	4	-2	32.593	2.7451	-0.026
33.363	2.6835	4	1	1	4	33.333	2.6859	-0.031
34.486	2.5986	3	1	4	2	34.456	2.6008	-0.030
34.795	2.5763	3	2	0	0	34.728	2.5810	-0.067
35.248	2.5442	5	1	4	-3	35.210	2.5468	-0.038
36.252	2.4760	7	0	4	4	36.278	2.4743	0.026
36.845	2.4375	3	2	2	-1	36.836	2.4381	-0.009
37.318	2.4077	3	0	3	5	37.338	2.4064	0.020
38.143	2.3575	3	1	3	4	38.139	2.3577	-0.004
38.754	2.3217	7	2	0	2	38.746	2.3222	-0.009
39.003	2.3074	4	1	1	5	39.008	2.3072	0.004
39.399	2.2852	2	1	3	-5	39.378	2.2864	-0.021
39.956	2.2546	3	1	0	-6	39.946	2.2551	-0.010
40.517	2.2246	1	1	1	-6	40.490	2.2261	-0.028

40.936	2.2028	2	2	2	2	40.936	2.2028	0.000
41.306	2.1839	1	0	5	4	41.318	2.1833	0.012
42.695	2.1161	6	1	5	3	42.693	2.1162	-0.003
43.440	2.0815	4	0	6	-3	43.476	2.0798	0.036
44.276	2.0441	2	2	2	3	44.234	2.0460	-0.042
44.717	2.0250	1	2	1	-5	44.700	2.0257	-0.017
44.976	2.0139	1	2	3	-4	45.026	2.0118	0.050
45.941	1.9738	1	1	6	-3	46.005	1.9712	0.064
46.475	1.9524	2	1	5	4	46.484	1.9520	0.009
46.872	1.9368	2	0	6	4	46.866	1.9370	-0.006
47.564	1.9102	1	1	5	-5	47.544	1.9110	-0.020
48.105	1.8900	2	1	6	3	48.113	1.8897	0.008
48.893	1.8613	2	1	3	6	48.878	1.8619	-0.015
50.174	1.8168	1	1	7	-2	50.172	1.8168	-0.002
51.255	1.7810	2	2	5	2	51.180	1.7834	-0.075
51.441	1.7750	3	1	7	2	51.499	1.7731	0.058
52.616	1.7380	4	1	6	-5	52.578	1.7392	-0.039
53.946	1.6983	3	1	7	3	53.980	1.6973	0.034
54.424	1.6845	1	3	2	-1	54.396	1.6853	-0.028
55.977	1.6414	1	2	6	2	55.977	1.6414	0.000
56.589	1.6251	1	3	1	-4	56.561	1.6258	-0.028
57.341	1.6055	1	1	5	-7	57.305	1.6065	-0.036
57.626	1.5983	1	2	3	-7	57.592	1.5992	-0.034
58.309	1.5812	3	2	7	-1	58.341	1.5804	0.032
59.402	1.5547	3	3	4	-1	59.422	1.5542	0.020
59.841	1.5443	2	3	3	-4	59.863	1.5438	0.022
61.018	1.5173	2	0	7	6	61.036	1.5169	0.018
62.728	1.4800	1	3	1	-6	62.734	1.4799	0.006
63.248	1.4691	2	0	9	-3	63.246	1.4691	-0.003
64.398	1.4456	1	3	5	-3	64.367	1.4462	-0.031
65.473	1.4244	1	2	8	1	65.476	1.4244	0.003
65.950	1.4153	1	3	4	3	65.990	1.4145	0.041
67.032	1.3950	1	2	7	4	67.041	1.3949	0.009
78.375	1.2191	1	4	4	-1	78.378	1.2191	0.003

Figure Captions

Figure 1. Structural formula of tetrazene nitrate monohydrate.

Figure 2. X-ray powder diffraction pattern of title compound using Cu *Kα* radiation (λ = 1.5418 Å).