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Crystal Structures of Condensation Products of Malononitrile

III. 2-Amino-1,1,3-tricyanopropene ("Dimeric Malononitrile")

BERNT KLEWE

Department of Chemistry, University of Oslo, Oslo 3, Norway

The crystals are monoclinic, space group $P2_1/c$, with cell dimensions $a = 12.66_3$ Å, $b = 7.54_1$ Å, $c = 7.02_9$ Å, and $\beta = 99.7_9°$. 1056 reflections were recorded as observed on an automatic four-circle diffractometer. The structure was refined by full-matrix least squares technique ($R_w = 4.1$ %, $R = 4.6$ %). Apart from the cyano group and hydrogen atoms of the $CH_2(CN)$ group, the molecule is roughly planar; the dihedral angle $(H_2N)-C-CH_2-(CN)$ is 27°. The $sp-sp^2$ C–C and sp^2-sp^2 C–C bond lengths are 1.422 Å and 1.382 Å, respectively. *Inter*-molecular hydrogen bonds between the amino group and nitrogen atoms of cyano groups form infinite double chains in the [010]-direction.

This work was undertaken as part of a program of X-ray crystallographic studies on condensation products of malononitrile. The compounds so far studied are the potassium salts of 2-cyanomethyl-1,1,3,3-tetracyanopropene [1] (or 2-dicyanomethyl-1,1,3-tricyanopropene) and 1,1,3-tricyanopropanone.[2] These papers will be referred to as $KC_9N_5H_2$ and $KC_6N_3OH_2$.

The interesting biological properties of self-condensation products of malononitrile, especially the dimer, 2-amino-1,1,3-tricyanopropene ($C_6N_4H_4$), have been the subject of several publications; *cf.* footnote 2 of Ref. 3.

Another crystal modification of $C_6N_4H_4$ is also being investigated.*

CRYSTAL DATA

$C_6N_4H_4$ was synthesized by E. G. Iversen, as described by Carboni *et al.*[4] (method **C**).

* *Note added in proof:* The crystal structure of $C_6N_4H_4$, space group $P2_1/c$, has also been determined by D. E. Zacharias (*Dissertation*, Univ. of Pittsburgh (1969); *Diss. Abstr. Int. B* **1969** 30 (3), 1077). A comparison between the investigations is presented in the study of the other crystal modification (accepted for publication in *Acta Chem. Scand.*).

The slightly yellow-coloured, needleformed crystals (crystallized from ethanol) belong to the monoclinic system, the space group being $P2_1/c$. The cell dimensions, determined on a manual four-circle diffractometer (CuK radiation), with estimated standard deviations, are: $a = 12.6629(27)$ Å, $b = 7.5409(17)$ Å, $c = 7.0285(13)$ Å, and $\beta = 99.79(1)°$. c is the needle axis. The unit cell contains four molecules ($\varrho_{calc} = 1.326$ g cm^{-3}, $\varrho_{obs} = 1.31$ g cm^{-3}).

About 1450 reflections with $2\theta < 55°$ were measured on an automatic four-circle diffractometer (Mo$K\alpha$ radiation, $\omega/2\theta$ scan) with a highly orientated graphite crystal monochromator. 1056 reflections were recorded as observed, their intensities being greater than twice the standard deviation from counter statistics, with a 2 % uncertainty in the measurements included. No absorption correction was made. A crystal of dimensions $0.21 \times 0.20 \times 0.14$ mm^3 was used for all X-ray measurements.

All programs used are included in Ref. 5.

STRUCTURE DETERMINATION

Some attempts of determining the structure using direct methods did not succeed, and eventually other methods were applied. From a sharpened Patterson synthesis, two probable orientations of the molecule (ignoring the cyano group of $-CH_2(CN)$) could be found. By moving the molecule (still excluding $-(CN)$) through the cell, and calculating the R-value for about 50 low-order reflections for each step, a set of R-minima was obtained.

Packing considerations showed that only a few of the corresponding sites were possible. The parameter set corresponding to the lowest R-value was refined by the "minimum residual method" [6] with only the 80 largest structure factors (now including the cyano group). The R-value dropped from 51 to 7 %.

The parameters arrived at were refined further by full-matrix least squares technique to a final R_w-value of 4.1 % for the 1056 observed reflections ($R = 4.6$ %). Starting positions of the hydrogen atoms were calculated from geometrical considerations. The weight analysis showed no intensity dependence, except for the smallest F-values. No secondary extinction correction was carried out. The atomic form factors were those of Cromer and Waber,[7] except for hydrogen.[8] A final difference Fourier map contained no larger density fluctuations than ± 0.22 e Å^{-3}.

Some sign sets derived by direct methods were finally examined. For one trial set, 16 out of 18 starting signs, used for determining 200 additional signs, were correct. The two wrong signs were correctly redetermined during the procedure. However, the corresponding Fourier map was not easily interpretable.

Observed and calculated structure factors are listed in Table 1, and atomic parameters in Table 2. For numbering of atoms, see Fig. 1.* The results of an analysis of the atomic vibration tensors are presented in Table 3.

The root mean square discrepancy between "observed" atomic vibration tensor components and those calculated from a rigid-body model [9] is 0.0035

* A similar numbering is used for $KC_9N_5H_2$ and $KC_6N_3OH_2$.

Table 1. Observed and calculated structure factors. (The five columns list values of h, k, l, 10 F_o, and 10 F_c.)

h	k	l	Fo	Fc
0	0	2	868	- 874
0	0	4	98	97
0	0	6	114	113
0	0	8	89	- 90
0	1	1	310	- 318
0	1	2	10	6
0	1	3	264	261
0	1	4	236	- 232
0	1	5	82	- 82
0	1	6	42	41
0	1	7	29	34
0	2	0	34	- 31
0	2	1	54	- 49
0	2	2	242	- 237
0	2	3	178	176
0	2	4	75	- 72
0	2	5	34	- 32
0	2	6	21	28
0	2	8	24	- 19
0	3	1	169	166
0	3	2	116	117
0	3	3	85	- 88
0	3	4	66	- 68
0	3	6	74	72
0	3	7	22	- 21
0	4	0	93	- 86
0	4	1	32	- 29
0	4	2	134	135
0	4	3	17	- 21
0	4	4	26	25
0	4	5	16	10
0	4	6	59	61
0	5	2	17	21
0	5	3	34	29
0	5	5	53	- 52
0	5	6	48	- 48
0	6	0	265	274
0	6	1	120	121
0	6	2	239	- 239
0	6	3	100	- 101
0	6	5	21	14
0	6	6	57	55
0	7	1	84	- 88
0	7	2	89	87
0	7	3	155	155
0	7	4	37	- 34
0	7	5	77	- 76
0	7	6	36	28
0	8	0	91	91
0	9	1	30	- 26
0	9	3	31	20
1	0	-8	28	24
1	0	-6	162	- 164
1	0	-4	354	356
1	0	-2	320	- 325
1	0	2	1255	1299
1	0	4	485	- 493
1	0	6	140	145
1	1	-8	20	19
1	1	-7	25	29
1	1	-6	26	25
1	1	-5	28	27
1	1	-3	74	- 73
1	1	-2	440	435
1	1	-1	280	290
1	1	0	94	- 95
1	1	1	403	- 412
1	1	2	26	- 26
1	1	3	29	- 25
1	1	4	49	- 47
1	1	5	99	99
1	1	6	77	- 78
1	1	7	44	- 44
1	1	8	27	30
1	2	-5	48	- 52
1	2	-4	43	43
1	2	-3	155	161
1	2	-2	380	- 371
1	2	-1	480	- 491
1	2	0	78	76
1	2	1	222	215
1	2	2	214	- 212
1	2	3	51	48
1	2	4	121	- 122
1	2	5	25	30
1	2	6	22	24
1	3	-7	44	44
1	3	-6	52	- 55
1	3	-5	129	- 126
1	3	-4	81	- 80
1	3	-3	142	143
1	3	-2	102	104
1	3	-1	126	- 119
1	3	0	205	- 198
1	3	1	116	115
1	3	2	61	62
1	3	3	132	133
1	3	5	77	- 78
1	3	8	32	33
1	4	-8	36	- 37
1	4	-6	29	25
1	4	-5	24	26
1	4	-4	145	143
1	4	-3	41	- 41
1	4	-2	132	- 131
1	4	-1	89	89
1	4	0	311	311
1	4	1	25	- 30
1	4	2	137	- 133
1	4	3	20	- 19
1	4	4	80	80
1	4	6	60	57
1	5	-7	34	33
1	5	-5	60	- 63
1	5	-4	102	103
1	5	-3	19	- 20
1	5	-2	37	- 38
1	5	-1	55	48
1	5	0	60	- 61
1	5	1	53	- 53
1	5	2	102	103
1	5	3	39	35
1	5	4	40	- 41
1	5	5	21	- 20
1	5	7	27	- 24
1	6	-6	79	- 76
1	6	-5	61	- 56
1	6	-4	131	130
1	6	-3	30	30
1	6	-2	149	- 153
1	6	-1	99	102
1	6	0	100	- 102
1	6	1	130	- 132
1	6	2	156	161
1	6	3	73	73
1	6	4	161	- 186
1	6	5	57	- 53
1	6	6	51	52
1	7	-6	32	- 29
1	7	-3	61	- 62
1	7	-1	165	169
1	7	0	109	- 110
1	7	1	156	- 158
1	7	2	92	91
1	7	3	33	29
1	7	5	61	60
1	8	-3	28	28
1	8	-2	20	19
1	8	-1	29	- 24
1	8	0	33	30
1	8	2	61	63
1	9	-2	54	57
1	9	0	26	- 32
1	9	1	64	- 52
1	9	2	21	18
1	9	3	23	22
2	0	-8	68	69
2	0	-6	83	- 85
2	0	-4	89	- 93
2	0	-2	430	428
2	0	0	395	- 398
2	0	2	552	547
2	0	4	200	203
2	0	6	131	- 141
2	0	8	60	65
2	1	-6	23	24
2	1	-4	87	92
2	1	-3	100	100
2	1	-2	180	172
2	1	-1	32	- 28
2	1	0	311	307
2	1	1	58	- 59
2	1	2	36	- 34
2	1	3	116	- 117
2	1	4	30	- 27
2	1	5	39	42
2	1	6	35	- 39
2	2	-8	29	24
2	2	-7	54	51
2	2	-6	53	- 54
2	2	-5	22	13
2	2	-4	114	114
2	2	-3	183	- 189
2	2	-2	168	- 171
2	2	-1	327	332
2	2	0	326	- 310
2	2	1	512	- 520
2	2	2	96	91
2	2	3	230	229
2	2	4	181	- 183
2	3	-7	39	34
2	3	-6	62	- 59
2	3	-5	17	- 9
2	3	-4	91	- 92
2	3	-3	184	- 178
2	3	-1	312	302
2	3	0	58	- 59
2	3	1	174	- 163
2	3	2	54	- 52
2	3	3	104	109
2	3	5	45	43
2	3	7	55	- 56
2	3	8	23	15
2	4	-6	67	- 67
2	4	-4	142	141
2	4	-3	41	42
2	4	-2	98	100
2	4	-1	53	- 56
2	4	0	128	- 128
2	4	1	76	77
2	4	2	228	231
2	4	3	33	- 32
2	4	4	92	- 96
2	4	6	76	75
2	5	-6	32	31
2	5	-3	92	- 94
2	5	-2	160	165
2	5	-1	41	39
2	5	0	120	- 125
2	5	2	41	43
2	5	4	44	41
2	5	6	25	- 25
2	6	-6	36	- 30
2	6	-4	43	- 42
2	6	-3	49	- 50
2	6	-2	112	115
2	6	-1	89	91
2	6	0	203	- 207
2	6	2	29	33
2	6	3	64	- 62
2	6	4	18	15
2	6	5	30	29
2	6	6	80	- 72
2	7	-5	60	54
2	7	-4	57	- 54
2	7	-2	29	29
2	7	0	87	- 86
2	7	1	63	65
2	7	3	98	- 97
2	7	4	61	57
2	7	5	53	53
2	8	-3	20	- 23
2	8	-2	45	- 43
2	8	-1	75	71
2	8	0	71	67
2	8	1	63	- 64
2	8	2	32	27
2	8	4	31	22
2	9	-3	34	- 31
2	9	-2	22	25
2	9	-1	54	49
2	9	0	27	20
2	9	1	55	- 49
3	0	-6	102	107
3	0	-4	334	- 332
3	0	-2	302	288
3	0	0	48	48
3	0	2	22	21
3	0	4	273	279
3	0	6	26	- 26
3	1	-7	23	- 22
3	1	-6	24	21
3	1	-4	15	21
3	1	-3	39	39
3	1	-2	262	260
3	1	-1	223	222
3	1	0	270	257
3	1	1	223	- 218
3	1	2	64	61
3	1	3	12	- 8
3	1	4	22	- 20
3	1	5	26	- 24
3	1	6	18	- 12
3	1	7	25	25
3	2	-8	40	37
3	2	-5	127	126
3	2	-4	19	- 19
3	2	-3	93	- 92
3	2	-2	178	175
3	2	-1	200	- 207
3	2	0	416	- 407
3	2	1	183	190
3	2	2	11	- 4
3	2	3	192	- 194
3	2	4	32	- 35
3	2	5	126	125
3	2	6	66	- 68
3	2	7	23	- 15
3	3	-6	31	- 31
3	3	-5	50	52
3	3	-4	136	- 138
3	3	-3	138	- 139
3	3	-2	50	- 50
3	3	-1	19	13
3	3	0	14	- 16
3	3	1	130	125
3	3	2	77	- 81
3	3	3	50	- 47
3	3	5	58	60
3	3	6	20	- 13
3	4	-6	51	- 52
3	4	-5	21	- 15
3	4	-3	21	16
3	4	-2	198	194
3	4	-1	75	78
3	4	0	46	- 38
3	4	1	63	- 65
3	4	2	19	18
3	4	3	48	48
3	4	4	63	62
3	4	5	28	- 25
3	5	-6	48	47
3	5	-4	43	45
3	5	-3	45	- 46
3	5	-2	15	19
3	5	-1	30	- 30
3	5	0	88	85
3	5	2	64	- 67
3	5	3	16	7
3	5	4	69	70
3	6	-7	31	- 27
3	6	-6	60	55
3	6	-4	77	- 75
3	6	-3	83	- 81
3	6	-2	20	19
3	6	-1	54	54
3	6	0	28	29
3	6	1	56	55
3	6	2	131	- 132
3	6	3	52	- 47
3	6	4	51	47
3	6	5	19	- 8
3	6	6	22	- 25
3	7	-4	31	- 25
3	7	-3	54	56
3	7	-2	29	- 31
3	7	-1	24	- 21
3	7	0	18	- 11
3	7	1	21	22
3	7	2	72	- 68
3	7	4	33	35
3	7	5	26	- 29
3	8	-3	53	- 55
3	8	-1	25	21
3	8	0	24	- 21
3	8	1	48	46
3	8	2	84	75
3	8	3	63	- 59
3	9	-2	60	56
3	9	1	44	34
3	9	2	26	18
4	0	-8	42	- 41
4	0	-6	24	20
4	0	-4	108	105
4	0	-2	581	- 574
4	0	0	479	463
4	0	2	20	- 15
4	0	4	87	87
4	0	6	76	79
4	0	8	32	- 29
4	1	-8	30	28
4	1	-5	17	- 17
4	1	-4	71	73
4	1	-3	65	- 65
4	1	-2	69	63
4	1	-1	317	304
4	1	0	219	217
4	1	1	110	- 104
4	1	2	143	138
4	1	3	75	- 78
4	1	4	16	- 11
4	2	-7	38	- 35
4	2	-6	63	61
4	2	-5	51	56
4	2	-4	70	- 67
4	2	-3	115	113
4	2	-2	146	143
4	2	-1	229	- 229
4	2	0	71	- 69
4	2	1	14	- 17
4	2	2	182	- 182
4	2	4	28	24
4	2	6	48	- 45
4	2	7	47	44
4	3	-6	18	- 27
4	3	-5	23	30
4	3	-4	79	- 79
4	3	-3	23	- 28
4	3	-2	165	- 174
4	3	-1	146	- 133
4	3	1	89	85
4	3	2	46	- 46
4	3	3	44	47
4	3	4	27	- 26
4	3	5	18	- 11
4	3	7	24	18

Table 1. Continued.

h	k	l	F_o	F_c
4	4	-8	32	- 28
4	4	-5	39	40
4	4	-4	105	- 103
4	4	-3	54	- 52
4	4	-2	174	174
4	4	-1	45	41
4	4	0	51	52
4	4	1	61	66
4	4	2	41	- 38
4	4	3	43	- 46
4	4	4	41	37
4	4	5	27	23
4	5	-6	36	39
4	5	-5	22	27
4	5	-4	61	60
4	5	-3	39	- 45
4	5	-2	49	53
4	5	-1	22	- 19
4	5	0	30	33
4	5	1	15	- 13
4	5	2	15	2
4	5	4	19	12
4	5	5	20	11
4	5	6	25	25
4	6	-4	66	68
4	6	-2	108	- 116
4	6	-1	96	- 99
4	6	0	56	62
4	6	1	126	131
4	6	2	22	- 22
4	6	3	23	- 25
4	6	4	63	- 59
4	6	5	33	- 27
4	6	6	29	28
4	7	-5	36	- 42
4	7	-3	36	36
4	7	-2	61	- 53
4	7	-1	50	53
4	7	1	36	- 34
4	7	2	44	- 40
4	8	-4	24	- 22
4	8	-3	46	44
4	8	-1	78	- 78
4	8	0	28	- 30
4	8	1	73	73
4	8	2	31	29
4	8	3	20	- 21
4	8	4	33	34
4	9	-2	26	10
4	9	-1	52	- 45
4	9	0	57	53
5	0	-8	25	32
5	0	-6	103	- 105
5	0	-4	92	91
5	0	0	527	- 508
5	0	2	365	362
5	0	6	39	40
5	1	-8	24	20
5	1	-7	34	- 32
5	1	-6	22	23
5	1	-5	57	57
5	1	-3	119	- 116
5	1	-2	114	113
5	1	-1	46	45
5	1	0	151	146
5	1	1	247	243
5	1	2	181	183
5	1	3	172	- 172
5	1	4	35	36
5	1	5	24	17
5	1	6	19	- 20
5	2	-6	38	36
5	2	-5	55	- 53
5	2	-4	73	72
5	2	-3	208	211
5	2	-2	60	- 58
5	2	-1	116	- 111
5	2	0	118	115
5	2	1	124	- 124
5	2	2	96	- 95
5	2	3	28	26
5	2	4	41	- 43
5	2	5	27	- 25
5	2	7	23	26
5	3	-7	33	35
5	3	-4	63	- 66
5	3	-3	22	19
5	3	-2	134	- 140
5	3	-1	60	- 57
5	3	0	79	- 82
5	3	1	72	- 68
5	3	3	61	60
5	3	4	35	- 36
5	3	5	35	36
5	3	7	28	- 18
5	4	-8	28	- 27
5	4	-7	26	- 20
5	4	-6	48	- 47
5	4	-5	17	23
5	4	-3	55	55
5	4	-2	89	- 85
5	4	-1	112	- 113
5	4	0	190	190
5	4	1	120	124
5	4	3	21	- 19
5	4	4	22	- 21
5	5	-5	38	36
5	5	-4	65	69
5	5	-2	63	63
5	5	-1	49	- 50
5	5	0	41	41
5	5	2	18	8
5	5	3	20	- 22
5	5	5	24	20
5	6	-6	38	- 38
5	6	-5	54	- 52
5	6	-4	62	59
5	6	-2	48	53
5	6	-1	58	- 60
5	6	0	124	- 130
5	6	2	88	86
5	6	3	67	64
5	6	4	51	- 48
5	6	5	43	- 38
5	7	-5	25	16
5	7	-3	55	- 62
5	7	-1	78	82
5	7	0	65	- 57
5	7	1	20	20
5	7	3	35	- 36
5	7	4	22	- 17
5	8	-3	72	68
5	8	-1	28	- 27
5	8	0	25	- 24
5	8	3	50	41
5	9	-1	40	34
5	9	0	49	39
5	9	1	59	- 47
6	0	-6	22	18
6	0	-4	161	- 159
6	0	-2	198	187
6	0	0	284	- 282
6	0	2	86	- 87
6	0	4	167	169
6	0	6	21	- 27
6	1	-7	47	- 47
6	1	-6	44	44
6	1	-3	94	93
6	1	-2	55	61
6	1	-1	238	- 225
6	1	0	105	103
6	1	1	248	242
6	1	2	121	120
6	1	3	25	- 26
6	1	4	72	74
6	1	5	58	- 58
6	2	-8	38	36
6	2	-4	90	91
6	2	-3	22	- 23
6	2	-2	77	76
6	2	-1	213	216
6	2	0	71	- 68
6	2	1	179	- 183
6	2	2	67	66
6	2	3	27	- 23
6	2	4	47	- 44
6	2	6	26	- 24
6	3	-5	85	86
6	3	-4	56	- 60
6	3	-3	56	- 52
6	3	-2	81	- 84
6	3	-1	31	- 32
6	3	0	118	- 121
6	3	1	21	22
6	3	2	14	- 10
6	3	3	63	- 64
6	3	4	21	- 17
6	3	5	61	62
6	4	-6	86	- 86
6	4	-5	49	- 50
6	4	-4	47	- 51
6	4	-3	109	113
6	4	-2	17	13
6	4	-1	45	- 51
6	4	0	55	- 55
6	4	1	27	- 24
6	4	2	114	114
6	4	3	76	77
6	4	4	22	- 26
6	4	5	37	- 35
6	5	-5	35	38
6	5	-4	30	30
6	5	-3	35	36
6	5	-2	72	78
6	5	-1	81	- 83
6	5	0	54	54
6	5	3	21	- 15
6	6	-6	46	44
6	6	-5	27	- 18
6	6	-4	67	- 70
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6	6	-2	131	137
6	6	0	41	- 41
6	6	1	41	- 38
6	6	2	54	- 53
6	6	3	54	51
6	6	4	61	56
6	7	-4	22	- 24
6	7	-1	40	- 48
6	7	0	30	- 25
6	7	1	93	100
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6	7	3	32	- 32
6	8	-1	51	46
6	8	0	30	- 24
6	8	1	42	- 43
6	8	3	33	29
7	0	-8	77	- 73
7	0	-6	78	77
7	0	-4	113	- 115
7	0	-2	66	- 65
7	0	0	88	83
7	0	2	245	- 249
7	0	4	81	82
7	0	6	48	44
7	1	-7	20	21
7	1	-5	113	- 113
7	1	-4	40	42
7	1	-3	137	137
7	1	-2	50	- 52
7	1	-1	104	- 103
7	1	0	117	124
7	1	2	66	67
7	1	3	117	119
7	1	4	57	53
7	1	5	77	- 78
7	1	6	24	26
7	2	-8	24	18
7	2	-7	35	- 39
7	2	-6	60	60
7	2	-4	37	- 38
7	2	-2	192	193
7	2	-1	48	45
7	2	0	13	- 14
7	2	1	47	49
7	2	2	21	- 28
7	2	3	98	- 101
7	2	4	38	41
7	2	6	37	- 31
7	2	7	22	- 1
7	3	-7	71	- 72
7	3	-6	19	21
7	3	-5	52	54
7	3	-3	95	98
7	3	-2	109	- 111
7	3	-1	176	- 174
7	3	0	45	- 45
7	3	1	75	77
7	3	2	56	- 58
7	3	3	24	- 25
7	3	5	29	- 25
7	4	-7	37	36
7	4	-5	35	- 35
7	4	-4	167	- 172
7	4	-3	23	- 21
7	4	-2	37	38
7	4	-1	129	133
7	4	0	15	- 17
7	4	1	107	- 110
7	4	3	52	56
7	4	4	57	59
7	4	6	36	- 35
7	5	-7	24	- 18
7	5	-5	20	14
7	5	-3	62	66
7	5	-2	55	53
7	5	-1	38	- 40
7	5	0	70	72
7	5	1	32	- 31
7	5	3	21	13
7	6	-6	69	68
7	6	-3	50	- 42
7	6	-2	25	- 30
7	6	0	127	131
7	6	1	27	- 32
7	6	2	95	- 93
7	6	4	23	23
7	6	5	22	16
7	7	-5	61	- 62
7	7	-3	45	45
7	7	-2	30	- 26
7	7	-1	33	- 37
7	7	0	32	32
7	7	2	49	- 49
7	7	3	47	52
7	8	-3	26	- 24
7	8	-1	47	45
7	8	2	31	- 32
8	0	-8	44	- 39
8	0	-6	68	- 64
8	0	-4	114	113
8	0	-2	226	- 228
8	0	0	51	47
8	0	2	53	- 56
8	0	4	84	- 83
8	0	6	69	69
8	1	-7	26	29
8	1	-6	21	26
8	1	-5	17	- 22
8	1	-3	98	- 99
8	1	-2	14	15
8	1	-1	146	148
8	1	0	23	- 23
8	1	1	112	- 113
8	1	2	85	87
8	1	3	82	83
8	1	4	41	43
8	1	6	24	16
8	2	-8	33	- 34
8	2	-7	45	- 46
8	2	-6	101	97
8	2	-5	32	- 31
8	2	-3	30	33
8	2	0	205	206
8	2	1	58	60
8	2	2	90	- 90
8	2	3	40	- 41
8	2	4	20	20
8	2	5	36	- 36
8	3	-7	42	39
8	3	-6	47	45
8	3	-5	146	- 145
8	3	-3	224	230
8	3	-2	61	- 61
8	3	-1	83	- 86
8	3	0	68	- 70
8	3	1	108	- 109
8	3	2	27	- 33
8	3	3	108	109
8	3	5	68	- 65
8	4	-7	21	11
8	4	-6	60	58
8	4	-5	49	46
8	4	-4	101	- 102
8	4	-3	89	- 92
8	4	-2	158	- 161
8	4	-1	80	81
8	4	0	134	136
8	4	2	81	- 82
8	4	3	47	- 42
8	4	4	19	24
8	4	5	46	43
8	5	-6	58	- 54
8	5	-5	31	- 34
8	5	-4	44	41
8	5	-3	76	77
8	5	0	85	85
8	5	1	43	- 50
8	5	2	30	28
8	6	-5	23	17
8	6	-4	90	91
8	6	-3	35	- 33
8	6	-2	48	- 48
8	6	-1	24	- 14
8	6	0	47	46
8	6	1	32	- 38
8	6	2	33	36
8	6	4	49	- 47
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8	7	-3	50	- 54
8	7	-2	47	- 42
8	7	-1	38	43
8	7	1	23	- 28
8	7	3	42	32
8	8	1	28	27
9	0	-8	41	- 35
9	0	-6	31	- 37
9	0	-4	37	- 35
9	0	-2	14	21
9	0	0	91	- 91
9	0	2	22	- 25
9	0	4	49	- 55
9	1	-8	25	- 18
9	1	-7	40	38
9	1	-6	22	- 20
9	1	-4	69	70
9	1	-3	24	- 23
9	1	-2	69	- 69
9	1	-1	49	- 49
9	1	0	24	19
9	1	1	52	51
9	1	4	40	40
9	1	5	29	23
9	1	6	32	22
9	2	-7	56	51
9	2	-6	23	- 18
9	2	-5	115	- 118
9	2	-4	164	167
9	2	-3	21	21
9	2	-2	120	- 122
9	2	-1	66	69
9	2	0	145	145
9	2	1	23	- 27
9	2	2	60	62
9	2	3	45	42
9	2	4	62	- 61
9	2	5	40	- 42
9	2	6	34	33
9	3	-7	44	41
9	3	-4	96	96
9	3	-3	126	- 120
9	3	-2	68	- 69
9	3	-1	286	291
9	3	0	42	- 41
9	3	1	212	- 218
9	3	2	21	- 26
9	3	5	44	42
9	4	-7	42	- 37
9	4	-6	69	- 67
9	4	-5	50	53
9	4	-4	101	98
9	4	-2	230	- 236
9	4	-1	67	- 66
9	4	1	92	95
9	4	2	87	88
9	4	3	49	- 45
9	4	4	76	- 70

Table 1. Continued.

h	k	l	F_o	F_c
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9	5	-4	53	- 54
9	5	-2	61	61
9	5	-1	86	84
9	5	1	53	- 56
9	5	2	71	64
9	6	-5	51	53
9	6	-4	31	23
9	6	-2	56	54
9	6	-1	38	- 34
9	6	2	43	41
9	6	3	33	- 31
9	7	-2	28	27
9	7	-1	49	- 44
9	7	0	50	- 44
9	7	1	28	24
9	7	2	34	32
9	8	-1	25	23
10	0	-6	109	- 105
10	0	-4	60	60
10	0	-2	92	- 93
10	0	4	69	- 63
10	1	-7	32	30
10	1	-6	23	- 19
10	1	-5	35	32
10	1	-4	34	- 35
10	1	-3	67	- 71
10	1	-2	101	108
10	1	-1	40	43
10	1	0	117	- 119
10	1	1	54	- 53
10	1	2	24	33
10	1	3	28	28
10	1	4	22	19
10	2	-7	26	- 23
10	2	-6	43	- 46
10	2	-5	72	71
10	2	-4	83	83
10	2	-3	177	- 182
10	2	-2	72	75
10	2	-1	92	93
10	2	0	66	- 68
10	2	1	54	55
10	2	2	126	129
10	2	3	40	- 40
10	2	4	28	- 21
10	3	-7	60	- 62
10	3	-5	77	80
10	3	-4	20	16
10	3	-3	80	- 83
10	3	-2	106	105
10	3	0	126	- 124
10	3	1	139	137
10	3	3	154	- 150
10	3	4	21	- 14
10	3	5	52	48
10	4	-5	54	- 55
10	4	-4	47	- 45
10	4	-3	86	89
10	4	-2	25	25
10	4	-1	44	- 45
10	4	0	182	- 181
10	4	2	58	60
10	4	3	43	41
10	4	5	27	- 26
10	5	-4	22	- 20
10	5	-2	47	- 48
10	5	-1	40	39
10	5	0	56	56
10	5	3	36	- 34
10	5	4	28	19
10	6	-4	37	33
10	6	-3	50	52
10	6	-1	40	- 48
10	6	0	25	28
10	6	1	25	- 25
10	6	2	26	17
10	7	-3	49	- 40
10	7	-2	39	35
10	7	-1	31	21
10	7	1	47	- 45
11	0	-4	109	- 107
11	0	-2	92	101
11	0	0	102	- 104
11	1	-5	53	56
11	1	-4	23	- 21
11	1	-2	31	- 35
11	1	-1	46	- 47
11	1	0	69	74
11	1	1	28	35
11	1	2	84	- 87
11	1	3	40	- 35
11	1	4	32	31
11	1	5	28	30
11	2	-5	46	- 44
11	2	-4	24	- 21
11	2	-3	64	66
11	2	-2	111	115
11	2	-1	147	- 148
11	2	0	21	- 23
11	2	1	101	105
11	2	2	21	24
11	2	4	45	41
11	2	5	27	- 20
11	3	-6	26	32
11	3	-5	39	- 35
11	3	-3	48	54
11	3	-2	47	47
11	3	-1	53	- 57
11	3	0	26	27
11	3	1	46	48
11	3	2	75	- 76
11	3	4	23	18
11	4	-3	60	- 59
11	4	-2	40	- 38
11	4	-1	91	90
11	4	0	51	- 50
11	4	1	48	- 51
11	4	2	49	- 51
11	4	3	20	14
11	4	4	36	35
11	5	-5	29	23
11	5	-4	45	- 42
11	5	-3	27	- 30
11	5	-2	27	22
11	5	-1	49	48
11	5	0	66	- 61
11	5	2	60	58
11	6	-2	55	45
11	6	-1	23	28
11	6	1	25	- 31
11	6	2	31	24
11	7	-1	24	- 23
12	0	-6	27	- 22
12	0	-4	30	- 34
12	0	-2	40	- 33
12	0	0	26	31
12	0	2	28	- 41
12	1	-6	55	- 55
12	1	-5	33	31
12	1	-4	24	24
12	1	0	27	- 25
12	1	3	20	7
12	1	4	36	- 35
12	2	-6	29	28
12	2	-4	30	- 29
12	2	-3	55	- 55
12	2	-2	51	48
12	2	-1	32	30
12	2	0	41	39
12	2	1	55	- 54
12	2	3	63	64
12	2	4	28	27
12	3	-6	21	16
12	3	-5	58	- 58
12	3	-4	23	26
12	3	-2	39	38
12	3	-1	24	25
12	3	0	29	30
12	3	1	19	- 23
12	3	3	31	28
12	4	-5	29	32
12	4	-4	21	- 13
12	4	-1	36	- 34
12	4	0	61	- 57
12	4	1	61	57
12	4	2	31	- 31
12	4	3	35	- 34
12	5	-4	81	- 79
12	5	-2	25	- 14
12	5	1	44	49
12	5	2	48	- 38
12	6	-3	58	48
12	6	-1	34	- 19
12	6	0	26	24
13	0	-4	90	- 82
13	0	-2	24	18
13	0	0	24	- 12
13	1	-6	37	- 35
13	1	-4	40	- 42
13	1	-3	35	37
13	1	-2	21	30
13	1	-1	34	- 30
13	2	-5	21	16
13	2	-4	25	17
13	2	-1	42	- 41
13	2	0	39	37
13	3	-4	42	37
13	3	-3	97	- 92
13	3	-2	24	14
13	3	-1	57	61
13	3	0	53	47
13	3	1	20	2
13	3	2	21	- 5
13	4	-5	29	- 21
13	4	-4	59	49
13	4	-3	45	42
13	4	-1	34	- 34
13	4	0	28	- 27
13	5	-2	89	- 87
13	5	-1	24	10
13	5	0	27	25
14	0	-2	70	- 66
14	0	0	26	28
14	1	-4	62	- 56
14	1	-3	25	21
14	1	3	21	- 12
14	2	-5	55	51
14	2	-2	37	30
14	2	-1	30	- 33
14	3	-3	21	- 16
14	3	-2	36	41
14	3	-1	87	- 81
14	3	1	89	85
14	4	-2	67	62
14	4	0	44	- 32
14	4	1	26	- 27
15	0	-4	33	- 32
15	1	-3	50	43
15	1	-2	60	- 59
15	1	0	27	32
15	1	1	27	14
15	2	-4	28	- 27
15	2	-3	64	59
15	2	-1	33	- 36
15	2	0	42	33
15	2	1	21	- 25
15	3	-3	73	62
15	3	-1	70	- 66
15	3	0	36	37
16	0	-2	70	- 60
16	0	0	28	25
16	1	-1	22	17

Å² for the whole molecule, and 0.0033 Å² if the cyano group C6 – N6 of the $-CH_2(CN)$ group is omitted. The reduced r.m.s. translational amplitudes corresponding to the discrepancy of 0.0035 Å² are 0.199, 0.171, and 0.163 Å, and the r.m.s. librational amplitudes are 6.8, 3.1, and 1.2°. The axis of greatest libration is nearly parallel to the direction of C1 – C2.

"Riding" motion corrections [10] show that C1, C3, and N2 "ride" on C2. Contrary to what might be expected, C6 does not "ride" on C3.

Bond distances, bond angles, and dihedral angles of the molecule, short *intra*-molecular contacts, hydrogen bond lengths and angles, and other short contacts are given in Table 4. Librational corrections in bonds correspond to the rigid-body calculation of the whole molecule, except for C – N bonds, which are corrected for "riding" motion. The standard deviations in bond lengths and bond angles have been calculated from the correlation matrix of the last least squares refinement cycle, ignoring the E.S.D.'s of the cell parameters. σ(C – C) and σ(C – N) are 0.002 Å, σ(C – H) and σ(N – H) 0.02 Å, σ(C – C – C) and σ(C – C – NH_2) 0.15°, σ(C – C – N) 0.22°, σ(C – C – H) and σ(C – N – H) 1.0°, and σ(H – C – H) and σ(H – N – H) 1.5°. Bond distances (uncorrected) and angles are also given in Fig. 1.

Table 2. Fractional atomic coordinates and thermal parameters with estimated standard deviations ($\times 10^5$). The temperature factor is given by $\exp-(B_{11}h^2+B_{22}k^2+B_{33}l^2+B_{12}hk+B_{13}hl+B_{23}kl)$. Isotropic temperature factors (Å^2) are given for the hydrogen atoms. For numbering of atoms, see Fig. 1.

Atom	x	y	z	B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
C1	31397	15017	30835	453	886	1885	−105	503	111
	12	19	23	12	28	44	30	34	59
C2	25338	−153	31400	406	951	1504	42	365	24
	12	19	21	11	28	38	28	31	55
C3	13557	1938	31536	397	1007	2374	57	510	140
	13	22	31	12	31	51	31	38	69
C4	42401	14181	29005	587	1238	2913	−396	768	328
	16	22	28	14	36	56	36	44	71
C5	26706	32132	30845	579	1076	2101	−381	420	135
	13	22	25	13	32	48	34	38	64
C6	8640	−12841	40165	372	1295	1969	−25	461	−396
	12	21	25	11	34	44	31	36	61
N2	29243	−16263	31180	433	882	3232	71	791	16
	13	19	23	11	27	51	29	36	59
N4	51203	13557	27335	631	2616	5383	−506	1511	315
	14	22	31	14	46	74	39	52	90
N5	22810	45919	30785	900	1066	3484	−31	618	69
	13	19	24	14	28	55	34	42	64
N6	4662	−24206	46784	626	1716	2492	−430	759	54
	11	20	22	12	33	44	32	35	64
H1	12300	12818	38327	4.04					
	134	209	263	.41					
H2	9939	3484	18166	4.85					
	141	243	299	.47					
H3	36491	−17977	30421	4.03					
	162	205	243	.42					
H4	25069	−26006	31387	3.83					
	133	235	251	.41					

Table 3. The root mean square amplitudes of vibrations $\overline{(u^2)}^{1/2}$ (Å) and B-values (Å^2) along the principal axes given by the components of a unit vector in fractional coordinates ($\times 10^3$).

Atom	$\overline{(u^2)}^{1/2}$	B	e_x	e_y	e_z
	.216	3.69	33	5	140
C1	.186	2.72	−70	43	32
	.156	1.93	23	125	−16
	.193	2.93	37	8	137
C2	.176	2.44	69	30	−44
	.165	2.15	−18	129	2
	.241	4.57	18	9	144
C3	.175	2.41	67	66	−11
	.168	2.24	40	−115	5
	.268	5.67	25	10	143
C4	.225	4.01	−63	76	10
	.166	2.19	43	108	−21
	.226	4.05	−3	24	139
C5	.224	3.95	72	−55	37
	.162	2.06	34	118	−11

Table 3. Continued.

	.224	3.96	19	−50	133
C6	.188	2.79	16	122	52
	.167	2.20	−76	13	18
	.281	6.23	20	1	144
N2	.178	2.49	75	35	−12
	.158	1.97	−20	128	2
	.364	10.45	23	6	143
N4	.283	6.31	27	−125	1
	.194	2.96	72	44	−18
	.291	6.69	9	2	144
N5	.266	5.60	80	−3	9
	.175	2.42	2	133	−2
	.253	5.07	50	−41	118
N6	.237	4.45	−27	96	78
	.191	2.87	57	82	−32

The deviations of atoms from some least squares planes are listed in Table 5. Fig. 2 is a schematical drawing of the structure, viewed along the *c*-axis, showing the hydrogen bonding system.

DISCUSSION

Apart from the cyano group and hydrogen atoms of the $CH_2(CN)$ group, the conformation of the molecule can roughly be described as planar (plane **1** of Table 5), the largest deviation being 0.063 Å. The hydrogen atoms of the amino group do not deviate significantly from this plane. C1 – C2 makes an angle of 3.7° with the planar $C(CN)_2$ group (plane **2**). A similar bending of the $C(CN)_2$ group is also observed for $KC_9N_5H_2$ and $KC_6N_3OH_2$. The angle between the planes **3** (through C2, C3, and N2) and **2** is 6.2°. The differences in deviations of C4,N4 and C5,N5 from plane **3** (those of C4,N4 being about three times the corresponding values for C5,N5) show that the $C(CN)_2$ group also is twisted about C1 – C2. C2 is 0.016 Å above the plane through C1, C3, and N2.

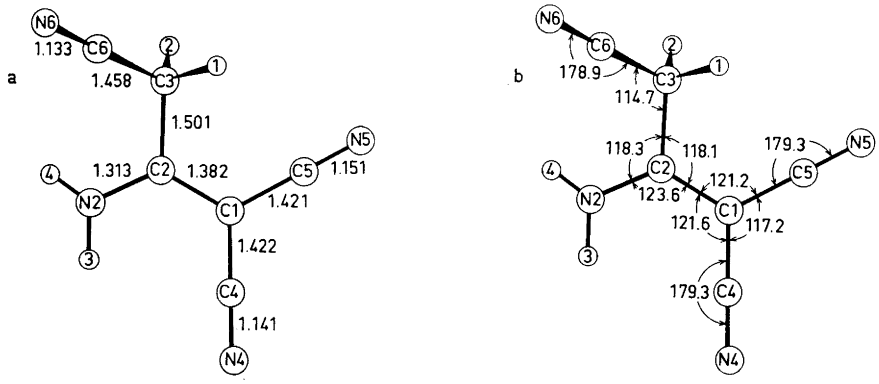

Fig. 1. Schematical drawings of the molecule, showing bond distances (a) and bond angles (b). Small circles indicate hydrogen atoms.

Table 4. Bond distances, bond angles, dihedral angles, short *intra*-molecular contacts, hydrogen bond lengths and corresponding angles, and other short contacts (equivalent position numbers in parentheses as defined below). Estimated standard deviations in bond lengths between heavy atoms are 0.002 Å, in C–C–C and C–C–NH_2 angles 0.15°, and in C–C–N 0.22°. Distances in parentheses are corrected for anisotropic thermal motion (see text).

Bond distances (Å)		
C1–C2	1.382	(1.384)
C2–C3	1.501	(1.511)
C1–C4	1.422	(1.429)
C1–C5	1.421	(1.429)
C3–C6	1.458	(1.461)
C2–N2	1.313	(1.319)
C4–N4	1.141	(1.187)
C5–N5	1.151	(1.180)
C6–N6	1.133	(1.158)
C3–H1	.98	
C3–H2	.98	
N2–H3	.94	
N2–H4	.91	

Intra-molecular contacts (Å)	
C4···N2	2.86
C6···N2	2.80
C4···H3	2.54
C5···H1	2.46
C6···H4	2.48

Hydrogen bond lengths (Å)	
N2···N4(1)	3.05
N2···N5(2)	2.96
H3···N4(1)	2.23
H4···N5(2)	2.14

Other contacts (Å)	
C6···N5(3)	3.36
C6···N6(4)	3.16
C6···C6(5)	3.39
N6···C3(6)	3.27
N6···C3(7)	3.31
N6···H1(5)	2.69
N6···H2(6)	2.69
N6···H2(7)	2.59

1:$(1-x,-\frac{1}{2}+y,\frac{1}{2}-z)$
2:$(x,-1+y,z)$
3:$(x,\frac{1}{2}-y,\frac{1}{2}+z)$
4:$(x,-\frac{1}{2}-y,-\frac{1}{2}+z)$
5:$(-x,-y,1-z)$
6:$(x,-\frac{1}{2}-y,\frac{1}{2}+z)$
7:$(-x,-\frac{1}{2}+y,\frac{1}{2}-z)$

Bond angles (°)	
C4–C1–C2	121.6
C5–C1–C2	121.2
C4–C1–C5	117.2
C1–C2–C3	118.1
C1–C2–N2	123.6
C3–C2–N2	118.3
C2–C3–C6	114.7
C1–C4–N4	179.3
C1–C5–N5	179.3
C3–C6–N6	178.9
C2–C3–H1	110
C2–C3–H2	108
C6–C3–H1	109
C6–C3–H2	109
H1–C3–H2	106
C2–N2–H3	120
C2–N2–H4	122
H3–N2–H4	118

Dihedral angles (°)	
C4–C1–C2–C3	174.5
C5–C1–C2–C3	−1.5
C4–C1–C2–N2	−3.1
C5–C1–C2–N2	−179.1
C1–C2–C3–C6	155.0
N2–C2–C3–C6	−27.2

Hydrogen bond angles (°)	
N2–H3···N4	147
N2–H4···N5	152
H3···N4–C4	137
H4···N5–C5	147

The cyano group bonded to C3 is rotated by 27° about the C2–C3 bond out of plane **1**; the CN group being in *anti* position with respect to the $C(CN)_2$ group.

Table 5. Deviations of atoms from some least squares planes (Å). Plane No. **1** is through all heavy atoms except the cyano group of $-CH_2(CN)$, plane No. **2** is through the $C(CN)_2$ group, and plane No. **3** through the rest of the heavy atoms, defining plane No. **1**. Deviations of atoms not defining the planes in parentheses.

Atom	1	2	3
C1	0.042	−0.003	(−0.047)
C2	0.020	(−0.093)	0.000
C3	−0.063	(−0.221)	0.000
C4	0.005	0.005	(−0.166)
C5	0.020	0.002	(−0.063)
N2	0.022	(−0.118)	0.000
N4	−0.040	−0.002	(−0.275)
N5	−0.003	−0.001	(−0.080)
H3	(0.027)		(−0.046)
H4	(−0.005)		(0.019)

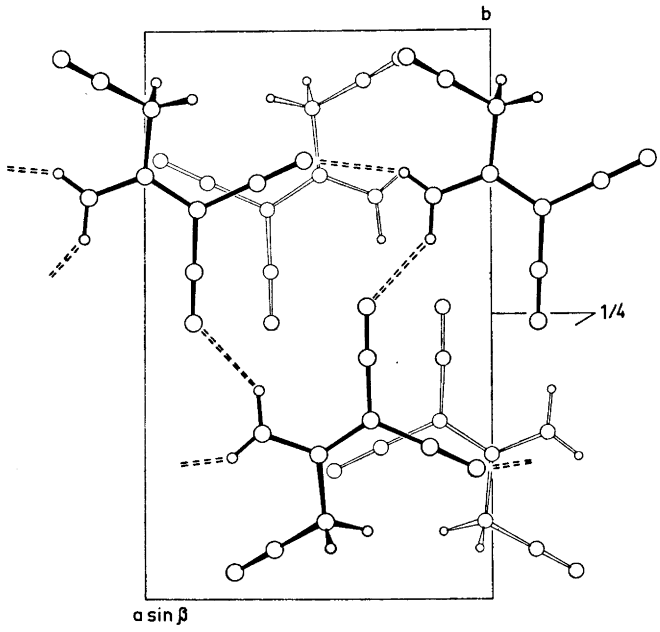

Fig. 2. Schematical drawing of the structure viewed along the *c* axis, showing the packing and the hydrogen bond system.

Opening of the angles C1 – C2 – N2, C2 – C1 – C4, and C2 – C1 – C5 from a predicted value of 120° may partly be caused by the hydrogen bonding system (Fig. 2). All other bond angles are normal.

The analysis of the thermal motions has been described earlier. The largest vibrational amplitudes of all atoms (Table 3) are roughly normal to plane **1**. In contradiction to the results of rigid-body calculations on $KC_9N_5H_2$ and

$KC_6N_3OH_2$, the root mean square discrepancy between "observed" atomic vibration tensor components and those calculated from a rigid-body model is not significantly lowered if the cyano group (C6 – N6) of the cyanomethyl group is omitted (0.0035 Å^2 and 0.0033 Å^2). For $KC_9N_5H_2$ and $KC_6N_3OH_2$, the decreases are 0.0014 Å^2 and 0.0012 Å^2, respectively. The C6 – N6 groups of those two compounds are coordinated to cations whereas van der Waals contacts only are present in $C_6N_4H_4$.

"Riding" corrections of the C – N triple bonds are quite large (0.046 Å for C4 – N4) and may be based upon unwarranted assumptions. The centroid of charge density of terminal atoms generally does not coincide with the nuclear position. The corrected values for the C – N bonds of the $C(CN)_2$ group are greater (1.187 Å and 1.180 Å) than the one of the third C – N triple bond (1.158 Å). This corresponds to a decrease of bond order, due to resonance stabilization. The two former nitrogen atoms are engaged in hydrogen bonding. A similar lengthening of C – N bonds coordinated to K^+ seems to be present in $KC_9N_5H_2$. However, semiempirical LCAO – MO calculations for CN^- and a series of XCN molecules [11] indicate that addition at nitrogen results in increase (relative to CN^-) in the CN σ and total overlap energies. Also, in a number of cases observed, the corresponding stretching frequencies increase upon complex formation.[12–14]

Uncorrected values of other bonds will be discussed. Molecular orbital calculation of π-electron densities of 2-amino-1,1-dicyanoethene has been carried out by Lofthus, using a method[15] based on extended HMO theory. The bond order values are as follows: $sp-sp^2$ C – C (0.48), sp^2-sp^2 C – C (0.58), sp^2-sp^2 C – N (0.69), and $sp-sp$ C – N (0.82). These values are assumed to be applicable for the present molecule (A). Similar bond order values for $KC_9N_5H_2$ (B) and the anion of hexacyanobutylene (C) have been reported.[1] Observed bond lengths of Table 4 and Fig. 1 are in satisfactory agreement with the bond order calculations of (A). Bond lengths and bond orders in parentheses for (A), (B), and (C) are as follows: $sp-sp^2$ C – C; 1.422 Å (0.48), 1.425 Å (0.46), 1.421 Å (0.49), and for sp^2-sp^2 C – C; 1.382 Å (0.58), 1.391 Å (0.62), 1.424 Å (0.52).

The C2 – N2 bond length of 1.313 Å ("riding" corrected value 1.328 Å) corresponds closely to the cyclopropanecarboxamide [16] value of 1.317 Å (uncorrected). Normal values of C2 – C3, C3 – C6 and the bonds to hydrogen atoms are also found.

The packing and hydrogen system are shown in Fig. 2. Hydrogen bond lengths and angles, and other short contacts are listed in Table 4. The hydrogen bonds are *inter*-molecular, and form infinite double chains in the [010]-direction. Pairs of single chains, formed by N2 – H4···N5 bonds between translational equivalent molecules, are linked by N2 – H3···N4 bonds between molecules related by the two-fold screw axis (through $x=\frac{1}{2}$, and $z=\frac{1}{4}$). Plane **1** of Table 5 makes an angle of only 0.6° with the *b* axis; the single chains are thus nearly planar. The N2 – H3···N4 bonds link parallel planes about 0.4 Å apart. The deviation of C4,N4 from plane **3** may be due to this bond.

The plane through the single chains make an angle of 76.5° with *c*. This corresponds to a molecule "thickness' of 3.42 Å, and indeed all distances between atoms of the planar parts of glide plane related molecules (in the [001]-

direction) are found to be larger than 3.4 Å. All short *inter*-molecular contacts are between C6 or N6 and other atoms. The shortest are C···C 3.39 Å, C···N 3.16 Å, and N···H 2.59 Å.

Hydrogen bonding with $-NH_2$ as donor, and cyano nitrogen as acceptor, has been found in dicyandiamide,[17] cyanamide,[18] and diaminomaleonitrile.[19] The presence of a bifurcated hydrogen bond in cyanamide is rather unlikely (the N···N distance of 3.22 Å is probably a van der Waals contact). The cyano nitrogen atoms of those compounds are all but one engaged in two hydrogen bonds with N···N distances in the region of 3.04 to 3.21 (average value 3.13 Å). (A value of 3.18 Å is reported for the C–H···N bond of hydrogen cyanide.[21]) In the present study, the N···N distances are found to be 2.96 Å and 3.05 Å. The angles H3–N2···N4 and H4–N2···N5 of 24° and 20°, respectively, are quite typical.[20]

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