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The Structure of 5-Anilino-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazolo[3,4-*d*]thiazole, $C_{16}H_{12}N_4OS$

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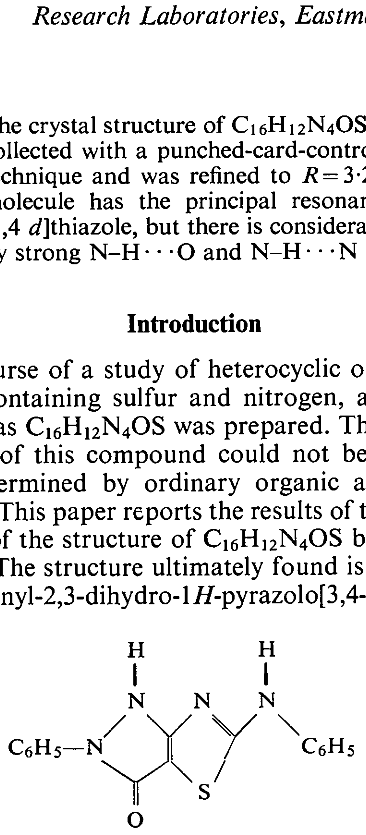
The crystal structure of $C_{16}H_{12}N_4OS$ has been determined from three-dimensional X-ray intensity data collected with a punched-card-controlled diffractometer. The structure was solved by the heavy-atom technique and was refined to $R = 3{\cdot}2\%$ by the method of least squares. Bond lengths show that the molecule has the principal resonance structure, 5-anilino-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazolo-[3,4 *d*]thiazole, but there is considerable resonance interaction. The molecules are joined to each other by strong N–H···O and N–H···N hydrogen bonds.

Introduction

In the course of a study of heterocyclic organic compounds containing sulfur and nitrogen, a compound analyzed as $C_{16}H_{12}N_4OS$ was prepared. The molecular structure of this compound could not be unambiguously determined by ordinary organic and physical methods. This paper reports the results of the complete solution of the structure of $C_{16}H_{12}N_4OS$ by X-ray diffraction. The structure ultimately found is 5-anilino-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazolo[3,4-*d*]thiazole.



Crystal data

$C_{16}H_{12}N_4OS$, M.W. 308·36, m.p. 251–256 °C (decomp.)
5-Anilino-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazolo-[3,4-*d*]thiazole.
Colorless, thick tabular crystals.
Monoclinic (Cu $K\alpha_1 = 1{\cdot}54051$ Å),
$a = 17{\cdot}107 \pm 0{\cdot}002$ Å, $b = 7{\cdot}458 \pm 0{\cdot}001$,
$c = 11{\cdot}285 \pm 0{\cdot}002$; $\beta = 106{\cdot}82 \pm 0{\cdot}01$ °.
Volume of unit cell, $1378{\cdot}1 \pm 0{\cdot}6$ Å^3.
Density, calculated ($Z = 4$), 1·486 g.cm^{-3};
measured (flotation), 1·50 g.cm^{-3}.
Linear absorption coefficient, $\mu = 21{\cdot}2$ cm^{-1} (Cu $K\alpha$).
Total number of electrons per unit cell, $F(000) = 640$.
Space group, $P2_1/c$ (C^5_{2h}).
Absent spectra: $h0l$ for l odd, $0k0$ for k odd.

Experimental

Suitable single crystals of $C_{16}H_{12}N_4OS$ were supplied by Dr K.R.Rush, of our Laboratories. Space group extinctions and preliminary lattice dimensions were obtained from precession photographs of a fairly large crystal.

A small rectangular crystal fragment $0{\cdot}10 \times 0{\cdot}10 \times 0{\cdot}18$ mm was attached to a glass fiber with Canada balsam. The crystal was then used to achieve precise alignment of a Picker four-circle automatic X-ray diffractometer with a Cu-target source and a scintillation detector. Narrow-source, open-counter ω scans (Furnas, 1957) through several reflections showed the mosaicity of the crystal to be a uniform 0·13 °. After alignment of the diffractometer, 20 reflections from the crystal were centered accurately through a very narrow vertical slit at a take-off angle of 0·5 °. Program *PICK*2 (Ibers, 1966), modified for the IBM 7044 computer, used these observations as the basis for the least-squares refinement of cell and orientation parameters. These cell parameters, given above, may be affected by systematic errors arising from the manufacture and alignment of the diffractometer and from the shape, perfection, and centering of the crystal, and thus the standard deviations are a measure of precision and not necessarily of accuracy. However, it is thought that their accuracy is sufficiently high to leave the derived interatomic distances unaffected.

Intensity data were collected with Ni-filtered Cu radiation at a take-off angle of 2·0 °. The pulse-height analyzer was set for an approximately 90% window. All 2344 unique reflections for $2\theta < 130$ ° were measured by the θ–2θ scan technique (Furnas, 1957) at a 2θ scan rate of 0·5 ° min^{-1}. The scan range varied from 1·0 ° at low θ to 1·6 ° at high θ. Stationary-crystal stationary-counter background counts of 40 sec were taken at each end of the scan. For count rates above 13000 c.p.s., attenuators were automatically inserted in the diffracted beam by the Picker system.

To check electronic and crystal stability during the period of data collection, the intensity of the 900 reflection was measured every 50th reflection. No systematic drift in this standard was observed. However, the standard deviation of 900, calculated from 56 observations, was 2·0 times what would be expected

Table 1. *Observed and calculated structure factors*

An asterisk designates 'less than'.

H	L	10FO	10FC
***** K= 0 *****			
1	0	592	557
2	0	133	138
3	0	630	616
4	0	586	599
5	0	533	529
6	0	850	851
7	0	152	154
8	0	133	131
9	0	864	803
10	0	107	106
11	0	122	131
12	0	175	177
13	0	123	123
14	0	22*	23
15	0	21*	18
16	0	199	199
17	0	109	111
18	0	44	41
19	0	66	69
0	2	990	1129
1	2	136	142
-1	2	1355	1418
2	2	157	162
-2	2	53	51
3	2	356	361
-3	2	57	64
4	2	755	775
-4	2	230	249
5	2	286	294
-5	2	378	392
6	2	316	322
-6	2	261	263
7	2	56	53
-7	2	155	169
8	2	182	183
-8	2	641	591
9	2	21	28
-9	2	222	227
10	2	218	228
-10	2	364	368
11	2	152	154
-11	2	510	498
12	2	228	231
-12	2	130	131
13	2	248	250
-13	2	156	152
14	2	87	85
-14	2	313	304
15	2	26	30
-15	2	173	170
16	2	52	59
-16	2	21*	4
17	2	90	86
-17	2	274	288
18	2	35	54
-18	2	93	91
-19	2	178	176
0	4	500	511
1	4	687	678
-1	4	900	960
2	4	109	107
-2	4	672	682
3	4	407	408
-3	4	126	124
4	4	253	248
-4	4	852	851
5	4	93	88
-5	4	23	6
6	4	618	594
-6	4	59	57
7	4	454	434
-7	4	602	599
8	4	298	307
-8	4	563	554
9	4	142	146
-9	4	59	67
10	4	571	559
-10	4	175	170
11	4	220	221
-11	4	75	75
12	4	173	183
-12	4	193	200
13	4	21*	11
-13	4	542	547
14	4	48	52
-14	4	27	19
15	4	209	202
-15	4	35	40
16	4	175	170
-16	4	148	150
-17	4	83	77
-18	4	115	111
-19	4	227	216
-20	4	114	110
0	6	479	474
1	6	66	72
-1	6	823	842
2	6	349	330
-2	6	539	545
3	6	261	243
4	6	353	353
-4	6	128	140
5	6	451	458
-5	6	188	176
6	6	291	283
-6	6	388	386
7	6	211	213
-7	6	147	145
8	6	202	205
-8	6	18*	11
9	6	404	401
-9	6	71	72
10	6	129	132
-10	6	29	28
11	6	168	170
-11	6	36	43
12	6	20*	8
-12	6	393	394
13	6	121	120
-13	6	87	95
14	6	189	185
-14	6	131	123
-15	6	132	143
-16	6	28	35
-17	6	149	144
-18	6	206	214
-19	6	51	54
0	8	21*	7
1	8	231	224
-1	8	24	18
2	8	414	421
-2	8	148	142
3	8	509	508
-3	8	315	309
4	8	97	94
-4	8	160	170
5	8	172	167
-5	8	92	98
6	8	97	92
-6	8	440	440
7	8	37	36
-7	8	235	234
8	8	115	116
-8	8	43	43
9	8	113	107
-9	8	92	98
10	8	140	140
-10	8	73	76
11	8	60	62
-11	8	270	273
-12	8	318	326
-13	8	352	352
-14	8	145	139
-15	8	331	329
-16	8	60	46
-17	8	50	43
-18	8	152	154
0	10	73	68
1	10	316	317
-1	10	95	105
2	10	293	292
-2	10	254	241
3	10	33	27
-3	10	129	116
4	10	255	257
-4	10	230	231
5	10	140	130
-5	10	62	68
6	10	291	286
-6	10	115	124
7	10	198	192
-7	10	136	137
8	10	173	170
-8	10	254	269
-9	10	129	131
-10	10	114	120
-11	10	56	61
-12	10	42	41
-13	10	157	160
-14	10	75	76
-15	10	98	93
-16	10	196	188
-17	10	88	87
0	12	20*	17
1	12	124	123
-1	12	101	97
2	12	139	131
-2	12	83	88
-3	12	261	246
-4	12	20*	9
-5	12	314	322
-7	12	142	137
-8	12	22*	24
-9	12	133	134
-10	12	84	89
-11	12	22*	12
-12	12	104	110
-13	12	76	69
***** K= 1 *****			
1	0	748	757
2	0	139	142
3	0	208	211
4	0	489	498
5	0	322	334
6	0	150	141
7	0	271	262
8	0	384	372
9	0	237	240
10	0	219	215
11	0	293	289
12	0	166	172
13	0	108	109
14	0	214	224
15	0	203	205
16	0	63	70
17	0	35	39
18	0	96	97
19	0	104	108
0	1	110	109
1	1	96	101
-1	1	632	621
2	1	105	107
-2	1	116	123
3	1	265	277
-3	1	438	442
4	1	190	197
-4	1	182	190
5	1	104	106
-5	1	302	320
6	1	332	315
-6	1	354	355
7	1	35	34
-7	1	16*	5
8	1	45	30
-8	1	235	227
9	1	267	263
-9	1	19	17
10	1	217	218
-10	1	235	228
11	1	208	208
-11	1	359	358
12	1	264	268
-12	1	321	329
13	1	220	231
-13	1	57	57
14	1	370	372
-14	1	91	95
15	1	296	299
-15	1	135	147
16	1	32	32
-16	1	108	115
17	1	101	107
-17	1	21*	3
18	1	97	95
-18	1	26	26
-19	1	85	90
0	2	510	536
1	2	360	377
-1	2	473	507
2	2	535	537
-2	2	196	194
3	2	163	171
-3	2	303	302
4	2	251	238
-4	2	16*	13
5	2	121	128
-5	2	69	58
6	2	177	170
-6	2	363	380
7	2	169	149
-7	2	485	477
8	2	263	271
-8	2	129	127
9	2	142	133
-9	2	230	215
10	2	134	138
-10	2	116	97
11	2	20*	10
-11	2	21	19
12	2	151	149
-12	2	257	268
13	2	252	259
-13	2	142	147
14	2	269	271
-14	2	100	100
15	2	59	54
-15	2	41	27
16	2	54	59
-16	2	192	201
17	2	86	88
-17	2	54	51
18	2	58	58
-18	2	20*	8
-19	2	43	44
0	3	224	227
1	3	57	60
-1	3	407	424
2	3	117	114
-2	3	393	393
3	3	199	194
-3	3	266	267
4	3	503	496
-4	3	161	160
5	3	99	95
-5	3	116	122
6	3	244	223
-6	3	32	29
7	3	77	67
-7	3	610	600
8	3	62	35
-8	3	704	662
9	3	177	137
-9	3	341	330
10	3	124	128
-10	3	108	93
11	3	89	99
-11	3	32	30
12	3	28	33
-12	3	81	83
13	3	170	169
-13	3	122	124
14	3	162	168
-14	3	261	260
15	3	127	133
-15	3	174	169
16	3	66	68
-16	3	296	299
17	3	25	7
-17	3	202	192
-18	3	47	50
-19	3	20*	5
0	4	122	113
1	4	208	199
-1	4	152	158
2	4	448	437
-2	4	39	25
3	4	56	58
-3	4	469	472
4	4	365	362
-4	4	64	69
5	4	102	100
-5	4	179	168
6	4	77	83
-6	4	372	365
7	4	28	24
-7	4	164	171
8	4	25	24
-8	4	583	561
9	4	174	175
-9	4	452	434
10	4	227	233
-10	4	47	42
11	4	264	269
-11	4	68	65
12	4	27	33
-12	4	369	367
13	4	60	57
-13	4	103	104
14	4	60	59
-14	4	212	209
15	4	77	76
-15	4	146	154
16	4	22*	6
-16	4	64	65
-17	4	241	239
-18	4	84	76
-19	4	42	42
0	5	156	161
1	5	242	242
-1	5	222	219
2	5	135	146
-2	5	245	253
3	5	170	172
-3	5	56	66
4	5	416	415
-4	5	299	310
5	5	265	263
-5	5	66	54
6	5	24	16
-6	5	129	133
7	5	20*	7
-7	5	104	106
8	5	20*	19
-8	5	244	229
9	5	21*	5
-9	5	355	351
10	5	450	462
-10	5	38	38
11	5	133	138
-11	5	115	119
12	5	26	25
-12	5	127	124
13	5	45	53
-13	5	282	284
14	5	20*	5
-14	5	223	224
15	5	26	21
-15	5	108	120
-16	5	162	170
-17	5	114	117
-18	5	99	94
-19	5	126	123
0	6	98	102
1	6	508	496
-1	6	167	160
2	6	111	102
-2	6	204	213
3	6	74	72
-3	6	120	120
4	6	275	272
-4	6	287	285
5	6	153	153
-5	6	85	89
6	6	57	55
-6	6	142	142
7	6	52	52
-7	6	123	125
8	6	22*	26
-8	6	228	224
9	6	94	100
-9	6	25	12
10	6	405	409
-10	6	164	169
11	6	71	65
-11	6	122	127
12	6	26	26
-12	6	21*	0
13	6	21*	31
-13	6	383	379
14	6	24*	16
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-15	6	71	65
-16	6	89	91
-17	6	73	71
-18	6	57	48
-19	6	141	133
0	7	149	154
1	7	212	208
-1	7	77	84
2	7	153	150
-2	7	102	99
3	7	20*	18
-3	7	105	101
4	7	130	140
-4	7	59	68
5	7	218	229
-5	7	55	53
6	7	54	53
-6	7	135	135
7	7	44	45
-7	7	289	276
8	7	92	91
-8	7	347	349
9	7	54	57
-9	7	424	422
10	7	192	197
-10	7	148	147
11	7	36	35
-11	7	205	208
12	7	23	26
-12	7	154	158
13	7	91	93
-13	7	319	317
-14	7	22	14
-15	7	147	141
-16	7	70	63
-17	7	79	80
-18	7	40	37
-19	7	39	32
0	8	214	217
1	8	150	154
-1	8	78	80
2	8	48	49
-2	8	218	227
3	8	98	97
-3	8	25	19
4	8	112	107
-4	8	20*	4
5	8	111	117
-5	8	51	49
6	8	29	17
-6	8	91	86
7	8	130	131
-7	8	298	294
8	8	27	24
-8	8	271	275
9	8	84	83
-9	8	410	405
10	8	51	48
-10	8	189	191
11	8	20*	5
-11	8	53	53
-12	8	114	112
-13	8	128	130
-14	8	22*	22
-15	8	64	57
-16	8	197	203
-17	8	47	42
-18	8	77	79
0	9	127	136
1	9	121	123
-1	9	178	182
2	9	226	221
-2	9	64	66
3	9	196	195
-3	9	335	350
4	9	21*	8
-4	9	22*	22
5	9	84	86
-5	9	29	26
6	9	30	36
-6	9	22	28
7	9	38	36
-7	9	160	152
8	9	20*	1
-8	9	348	349
9	9	41	44
-9	9	561	558
10	9	36	29
-10	9	295	300
-11	9	31	35
-12	9	73	66
-13	9	98	103
-14	9	21*	5
-15	9	26	29
-16	9	139	137
-17	9	87	93
0	10	144	150
1	10	21*	3
-1	10	28	21
2	10	214	212
-2	10	124	122
3	10	164	164
-3	10	391	397
4	10	20*	7
-4	10	51	62
5	10	32	21
-5	10	22*	12
6	10	117	126
-6	10	24	10
7	10	59	56
-7	10	194	199
8	10	24*	11
-8	10	88	84
-9	10	319	325
-10	10	190	185
-11	10	21*	14
-12	10	44	46
-13	10	96	94
-14	10	22	21
-15	10	64	61
-16	10	22*	28
0	11	21	15
1	11	67	63
-1	11	21*	12
2	11	116	119
-2	11	123	123
3	11	145	145
-3	11	127	128
4	11	45	40
-4	11	137	142
5	11	45	38
-5	11	59	63
-6	11	76	81
-7	11	89	93
-8	11	49	44
-9	11	88	87
-10	11	135	135
-11	11	35	40
-12	11	71	70
-13	11	127	123
-14	11	124	119
-15	11	89	93
0	12	118	115
1	12	99	99
-1	12	46	37
2	12	25*	24
-2	12	128	130
-3	12	55	59
-4	12	87	86
-5	12	39	43
-6	12	133	134
-7	12	63	62
-8	12	137	136
-9	12	134	128
-10	12	23*	30
-11	12	22*	22
-12	12	78	76
-13	12	53	52
-3	13	101	110
-4	13	24*	4
-5	13	34	36
-6	13	125	128
-7	13	121	116
-8	13	132	133
***** K= 2 *****			
0	0	273	291
1	0	802	833
2	0	449	462
3	0	203	210
4	0	402	407
5	0	39	42
6	0	40	38
7	0	319	308
8	0	53	51
9	0	64	71
10	0	37	43
11	0	134	131
12	0	20*	9
13	0	44	39
14	0	125	129
15	0	20*	8
16	0	20*	15
17	0	27	27
18	0	20*	6
0	1	2453	2616
1	1	31	31
-1	1	1306	1311
2	1	370	377
-2	1	31	31
3	1	43	48
-3	1	381	376
4	1	257	261
-4	1	536	553
5	1	360	369
-5	1	39	44
6	1	509	513
-6	1	307	312
7	1	134	131
-7	1	178	193
8	1	304	308
-8	1	176	168
9	1	309	298
-9	1	481	476
10	1	101	103
-10	1	468	463
11	1	179	177
-11	1	78	74
12	1	145	149
-12	1	71	69
13	1	168	169
-13	1	70	64
14	1	85	85
-14	1	187	178
15	1	53	46
-15	1	42	48
16	1	120	126
-16	1	133	131
17	1	20*	2
-17	1	179	187
18	1	30	36
-18	1	78	78
-19	1	68	65
0	2	1034	1064
1	2	99	101
-1	2	641	647
2	2	163	163
-2	2	187	183
3	2	361	362
-3	2	280	289
4	2	38	37
-4	2	325	328
5	2	145	148
-5	2	119	121
6	2	51	50
-6	2	73	75
7	2	59	58
-7	2	247	245
8	2	106	103
-8	2	145	143
9	2	186	177
-9	2	38	37
10	2	110	109
-10	2	129	118
11	2	98	102
-11	2	60	56
12	2	192	191
-12	2	27	22
13	2	107	114
-13	2	274	277
14	2	105	111
-14	2	75	73
15	2	33	43
-15	2	24	31
16	2	20*	16
-16	2	21*	15
17	2	59	53
-17	2	51	46
-18	2	33	34
-19	2	24*	29
0	3	141	153
1	3	348	349
-1	3	18	13
2	3	266	267
-2	3	159	159
3	3	401	406
-3	3	449	453
4	3	415	412
-4	3	444	442
5	3	27	31
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6	3	42	42
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13	0	167	172
14	0	219	222
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Table 1 (*cont.*)

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8	0	167	170
9	0	216	215
10	0	252	253
11	0	165	162
12	0	28	28
13	0	213	217
14	0	58	55
15	0	95	93
16	0	93	94
17	0	67	74
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-13	6	106	102
-14	6	159	158
-15	6	20*	6
-16	6	33	29
-17	6	166	157
0	7	57	59
1	7	25	22
-1	7	120	124
2	7	230	233
-2	7	84	77
3	7	68	74
-3	7	71	65
4	7	62	62
-4	7	123	122
5	7	25	28
-5	7	41	43
6	7	44	39
-6	7	67	65
7	7	47	50
-7	7	83	87
8	7	33	34
-8	7	90	85
9	7	20*	9
-9	7	150	155
10	7	88	92
-10	7	49	42
-11	7	25	19
-12	7	51	50
-13	7	91	95
-14	7	101	101
-15	7	88	88
-16	7	21*	7
0	8	35	31
1	8	114	112
-1	8	154	159
2	8	251	255
-2	8	32	39
3	8	181	175
-3	8	226	226
4	8	37	39
-4	8	111	111
5	8	21*	13
-5	8	112	115
6	8	40	33
-6	8	168	164
7	8	20*	13
-7	8	143	143
8	8	72	68
-8	8	65	68
9	8	35	26
-9	8	39	36
-10	8	123	128
-11	8	128	120
-12	8	263	262
-13	8	75	74
-14	8	20*	10
-15	8	36	35
-16	8	82	79
0	9	87	84
1	9	74	72
-1	9	21*	9
2	9	74	73
-2	9	157	158
3	9	36	35
-3	9	123	116
4	9	22	19
-4	9	21*	12
5	9	83	83
-5	9	70	67
6	9	175	174
-6	9	64	56
7	9	50	46
-7	9	127	125
-8	9	39	41
-9	9	111	111
-10	9	29	26
-11	9	102	106
-12	9	20*	15
-13	9	20*	1
-14	9	46	48
-15	9	59	58
0	10	52	50
1	10	205	205
-1	10	72	71
2	10	111	110
-2	10	51	48
3	10	35	36
-3	10	71	73
4	10	151	149
-4	10	56	57
-5	10	24	4
-6	10	41	42
-7	10	170	171
-8	10	114	117
-9	10	122	125
-10	10	46	46
-11	10	21*	19
-12	10	21*	13
-13	10	70	72
0	11	79	83
1	11	83	83
-1	11	51	47
-2	11	21*	21
-3	11	63	66
-4	11	20*	4
-5	11	137	135
-6	11	49	49
-7	11	64	69
-8	11	51	49
-9	11	67	63
-10	11	25*	17
-11	11	24*	1

***** K= 5 *****

H	L	10FO	10FC
1	0	151	149
2	0	191	191
3	0	37	26
4	0	227	224
5	0	79	73
6	0	91	91
7	0	29	3
8	0	21*	2
9	0	61	58
10	0	112	112
11	0	37	36
12	0	123	127
13	0	117	117
14	0	20*	6
15	0	74	72
0	1	121	118
1	1	100	94
-1	1	69	71
2	1	313	311
-2	1	51	53
3	1	124	124
-3	1	165	167
4	1	29	25
-4	1	173	176
5	1	130	134
-5	1	45	48
6	1	122	125
-6	1	138	132
7	1	22*	19
-7	1	176	175
8	1	110	109
-8	1	142	146
9	1	61	61
-9	1	56	58
10	1	123	124
-10	1	55	57
11	1	184	184
-11	1	170	174
12	1	66	68
-12	1	180	182
13	1	144	140
-13	1	54	50
14	1	145	147
-14	1	42	42
15	1	131	132
-15	1	21*	10
-16	1	49	51
0	2	228	222
1	2	304	300
-1	2	61	59
2	2	200	199
-2	2	30	27
3	2	197	194
-3	2	155	152
4	2	69	66
-4	2	70	63
5	2	164	161
-5	2	32	30
6	2	145	147
-6	2	100	101
7	2	70	66
-7	2	97	95
8	2	87	80
-8	2	64	66
9	2	138	142
-9	2	22*	6
10	2	127	128
-10	2	106	107
11	2	47	44
-11	2	42	36
12	2	20*	18
-12	2	151	148
13	2	50	54
-13	2	283	282
14	2	21*	20
-14	2	47	45
-15	2	53	45
-16	2	26	17
0	3	20	11
1	3	33	25
-1	3	34	35
2	3	39	37
-2	3	35	30
3	3	42	40
-3	3	61	52
4	3	113	114
-4	3	143	144
5	3	74	78
-5	3	50	36
6	3	21*	15
-6	3	96	100
7	3	49	46
-7	3	152	144
8	3	119	120
-8	3	248	243
9	3	41	42
-9	3	247	248
10	3	69	65
-10	3	91	92
11	3	55	56
-11	3	81	83
12	3	49	52
-12	3	58	59
13	3	111	107
-13	3	23	33
-14	3	108	106
-15	3	210	204
-16	3	114	113
0	4	172	169
1	4	44	45
-1	4	121	117
2	4	53	53
-2	4	38	29
3	4	30	26
-3	4	158	157
4	4	105	106
-4	4	22	15
5	4	60	58
-5	4	47	49
6	4	146	147
-6	4	161	164
7	4	21*	17
-7	4	42	35
8	4	196	203
-8	4	22*	6
9	4	91	90
-9	4	137	141
10	4	42	38
-10	4	32	23
11	4	93	92
-11	4	45	44
12	4	52	52
-12	4	128	130
-13	4	21*	14
-14	4	37	35
-15	4	64	66
-16	4	57	50
0	5	155	153
1	5	41	40
-1	5	22*	20
2	5	90	89
-2	5	185	185
3	5	106	111
-3	5	95	90
4	5	187	185
-4	5	69	66
5	5	55	62
-5	5	184	183
6	5	86	84
-6	5	136	141
7	5	83	84
-7	5	78	77
8	5	21*	9
-8	5	120	124
9	5	99	106
-9	5	74	73
10	5	125	126
-10	5	83	85
11	5	130	131
-11	5	179	177
-12	5	59	61
-13	5	19*	11
-14	5	29	25
-15	5	66	68
-16	5	134	134
0	6	280	280
1	6	153	152
-1	6	209	214
2	6	142	142
-2	6	182	189
3	6	20*	19
-3	6	243	244
4	6	56	53
-4	6	39	43
5	6	40	47
-5	6	83	85
6	6	35	35
-6	6	96	100
7	6	39	45
-7	6	165	166
8	6	32	38
-8	6	78	78
9	6	107	107
-9	6	86	87
10	6	73	72
-10	6	153	153
-11	6	40	44
-12	6	68	69
-13	6	21*	23
-14	6	66	65
-15	6	21*	2
0	7	172	169
1	7	183	181
-1	7	63	63
2	7	22*	2
-2	7	22*	0
3	7	79	72
-3	7	22*	7
4	7	60	64
-4	7	21*	10
5	7	126	126
-5	7	119	121
6	7	20*	3
-6	7	97	96
7	7	34	34
-7	7	142	143
8	7	31	33
-8	7	158	154
9	7	23*	12
-9	7	188	187
-10	7	88	86
-11	7	70	65
-12	7	153	156
-13	7	92	93
-14	7	21*	17
-15	7	35	39
0	8	160	161
1	8	164	163
-1	8	21*	7
2	8	21*	19
-2	8	45	39
3	8	23	32
-3	8	72	70
4	8	120	115
-4	8	103	104
5	8	54	45
-5	8	21*	16
6	8	122	120
-6	8	55	56
7	8	100	100
-7	8	24	24
-8	8	21*	6
-9	8	67	74
-10	8	97	100
-11	8	51	52
-12	8	25	30
-13	8	22	29
-14	8	24*	11
0	9	102	106
1	9	19*	4
-1	9	144	142
2	9	86	90
-2	9	31	22
3	9	73	69
-3	9	61	63
4	9	59	51
-4	9	118	116
5	9	42	41
-5	9	54	53
-6	9	63	63
-7	9	40	41
-8	9	107	111
-9	9	265	262
-10	9	235	233
-11	9	23	25
-12	9	68	63
0	10	58	60
1	10	56	54
-1	10	54	50
-2	10	20*	6
-3	10	77	79
-4	10	73	70
-5	10	40	41
-6	10	81	81
-7	10	137	138
-8	10	113	114
-9	10	22*	25
-10	10	91	95

***** K= 6 *****

H	L	10FO	10FC
0	0	219	221
1	0	180	186
2	0	232	231
3	0	120	119
4	0	60	61
5	0	21*	23
6	0	28	26
7	0	65	65
8	0	68	67
9	0	21*	7
10	0	21*	19
11	0	110	110
12	0	86	83
13	0	89	88
14	0	24*	12
0	1	151	151
1	1	41	44
-1	1	166	164
2	1	208	209
-2	1	55	51
3	1	57	59
-3	1	122	116
4	1	21*	9
-4	1	64	63
5	1	22*	11
-5	1	27	25
6	1	152	146
-6	1	21*	6
7	1	50	53
-7	1	32	31
8	1	63	61
-8	1	126	130
9	1	21*	15
-9	1	172	173
10	1	53	58
-10	1	223	222
11	1	120	118
-11	1	36	32
12	1	61	67
-12	1	20*	4
13	1	112	115
-13	1	144	146
-14	1	24*	8
0	2	120	129
1	2	249	250
-1	2	27	16
2	2	79	80
-2	2	36	36
3	2	183	187
-3	2	107	108
4	2	21*	7
-4	2	28	18
5	2	22*	19
-5	2	63	63
6	2	21*	16
-6	2	66	67
7	2	25	31
-7	2	59	58
8	2	22	30
-8	2	23	2
9	2	60	58
-9	2	55	49
10	2	99	99
-10	2	21*	13
11	2	29	28
-11	2	43	49
12	2	41	44
-12	2	20*	6
-13	2	96	94
-14	2	155	157
0	3	116	110
1	3	112	112
-1	3	168	173
2	3	207	210
-2	3	67	68
3	3	64	62
-3	3	142	143
4	3	115	114
-4	3	193	191
5	3	22	18
-5	3	41	41
6	3	21*	7
-6	3	44	40
7	3	26	17
-7	3	21*	4
8	3	29	24
-8	3	84	81
9	3	112	114
-9	3	85	85
10	3	85	85
-10	3	206	204
11	3	50	47
-11	3	74	76
12	3	23*	16
-12	3	20*	4
-13	3	42	42
-14	3	48	46
0	4	47	49
1	4	66	64
-1	4	239	239
2	4	145	141
-2	4	33	27
3	4	48	50
-3	4	59	59
4	4	48	38
-4	4	90	92
5	4	97	96
-5	4	64	63
6	4	75	72
-6	4	21*	12
7	4	112	107
-7	4	22*	15
8	4	179	180
-8	4	21*	4
9	4	27	29
-9	4	21*	3
10	4	21*	20
-10	4	34	38
11	4	23*	12
-11	4	39	36
-12	4	20*	5
-13	4	41	40
-14	4	112	112
0	5	133	137
1	5	21*	4
-1	5	91	85
2	5	134	129
-2	5	121	121
3	5	69	67
-3	5	66	69
4	5	60	57
-4	5	71	64
5	5	78	73
-5	5	21*	20
6	5	81	83
-6	5	44	39
7	5	43	42
-7	5	124	126
8	5	75	78
-8	5	56	53
9	5	41	41
-9	5	110	112
-10	5	61	54
-11	5	79	77
-12	5	33	28
-13	5	133	127
-14	5	85	81
0	6	35	38
1	6	86	86
-1	6	162	160
2	6	55	57
-2	6	96	99
3	6	55	59
-3	6	53	53
4	6	20*	14
-4	6	72	81
5	6	65	63
-5	6	21*	4
6	6	63	59
-6	6	37	30
7	6	156	158
-7	6	53	55
8	6	25*	16
-8	6	53	52
-9	6	119	119
-10	6	51	53
-11	6	20*	6
-12	6	35	39
-13	6	40	46
0	7	45	49
1	7	21*	8
-1	7	91	95
2	7	86	86
-2	7	59	55
3	7	65	61
-3	7	175	171
4	7	75	77
-4	7	25	9
5	7	45	44
-5	7	60	61
6	7	99	99
-6	7	39	29
-7	7	21*	16
-8	7	49	46
-9	7	112	113
-10	7	95	96
-11	7	164	162
-12	7	115	111
0	8	21*	7
1	8	92	90
-1	8	41	43
2	8	90	93
-2	8	103	109
3	8	21*	13
-3	8	87	85
4	8	24*	13
-4	8	34	40
-5	8	107	103
-6	8	20*	17
-7	8	20*	14
-8	8	59	58
-9	8	85	87
-10	8	22*	5
-11	8	106	108
0	9	48	46
1	9	139	136
-1	9	23*	24
-2	9	21*	17
-3	9	31	34
-4	9	20*	5
-5	9	112	111
-6	9	71	67
-7	9	114	112
-8	9	73	76
-9	9	31	35

***** K= 7 *****

H	L	10FO	10FC
1	0	29	27
2	0	162	163
3	0	128	127
4	0	28	74
5	0	36	35
6	0	87	88
7	0	72	75
8	0	19*	0
9	0	63	63
10	0	21*	11
11	0	147	148
0	1	89	85
1	1	111	110
-1	1	20*	6
2	1	119	122
-2	1	37	32
3	1	43	43
-3	1	33	28
4	1	21*	6
-4	1	91	91
5	1	57	60
-5	1	21*	23
6	1	31	37
-6	1	64	67
7	1	29	37
-7	1	21*	13
8	1	26	28
-8	1	21*	18
9	1	20	5
-9	1	20	9
10	1	80	77
-10	1	21*	17
11	1	23*	8
-11	1	22*	26
0	2	21*	4
1	2	96	94
-1	2	21*	3
2	2	219	216
-2	2	21*	6
3	2	21*	10
-3	2	38	37
4	2	23	7
-4	2	35	41
5	2	28	30
-5	2	42	34
6	2	22	18
-6	2	77	79
7	2	25	27
-7	2	91	93
8	2	147	141
-8	2	130	132
9	2	21*	9
-9	2	90	91
10	2	52	58
-10	2	20*	7
-11	2	33	42
-12	2	81	85
0	3	196	194
1	3	81	85
-1	3	89	86
2	3	75	77
-2	3	32	34
3	3	32	35
-3	3	70	68
4	3	47	50
-4	3	21*	4
5	3	89	89
-5	3	30	23
6	3	87	84
-6	3	20*	13
7	3	20	11
-7	3	22	16
8	3	87	87
-8	3	43	38
9	3	88	91
-9	3	43	42
-10	3	21*	16
-11	3	21*	8
-12	3	92	92
0	4	119	119
1	4	77	70
-1	4	93	91
2	4	22	26
-2	4	33	26
3	4	21	25
-3	4	104	100
4	4	85	82
-4	4	59	61
5	4	61	61
-5	4	89	94
6	4	52	50
-6	4	20*	4
7	4	96	93
-7	4	36	42
8	4	72	70
-8	4	73	75
-9	4	86	85
-10	4	130	129
-11	4	23*	16
0	5	21*	23
1	5	31	27
-1	5	118	118
2	5	52	48
-2	5	20*	7
3	5	21	19
-3	5	81	84
4	5	21*	21
-4	5	31	29
5	5	41	42
-5	5	21*	14
6	5	24*	17
-6	5	108	107
-7	5	54	48
-8	5	56	51
-9	5	46	48
-10	5	35	40
-11	5	24*	17
0	6	117	113
1	6	120	120
-1	6	62	59
2	6	65	64
-2	6	64	66
3	6	21*	4
-3	6	31	28
4	6	26	25
-4	6	36	37
5	6	44	47
-5	6	111	111
-6	6	100	103
-7	6	56	55
-8	6	21*	9
-9	6	28	33
-10	6	24*	14
0	7	90	91
1	7	127	126
-1	7	44	50
2	7	44	45
-2	7	50	54
-3	7	41	47
-4	7	25	18
-5	7	20*	4
-6	7	25	24
-7	7	47	52
-8	7	24*	14

***** K= 8 *****

H	L	10FO	10FC
0	0	39	31
1	0	20*	5
2	0	20*	20
3	0	36	34
4	0	20*	5
5	0	20*	6
6	0	21*	10
7	0	45	36
0	1	20*	27
1	1	134	137
-1	1	75	75
2	1	20*	3
-2	1	49	50
3	1	56	59
-3	1	26	21
4	1	33	27
-4	1	20*	12
5	1	20*	2
-5	1	20*	2
6	1	22*	17
-6	1	35	33
7	1	28	25
-7	1	22*	6
-8	1	49	43
0	2	39	40
1	2	20*	2
-1	2	102	99
2	2	139	145
-2	2	21*	17
3	2	31	30
-3	2	106	106
4	2	22*	24
-4	2	54	51
5	2	25	22
-5	2	20*	4
6	2	56	54
-6	2	20*	15
-7	2	23*	11
-8	2	24*	3
0	3	36	39
1	3	43	48
-1	3	43	45
2	3	111	115
-2	3	20*	1
3	3	68	69
-3	3	39	39
4	3	24*	22
-4	3	39	40
5	3	43	48
-5	3	46	46
-6	3	31	35
-7	3	25	21
0	4	49	51
1	4	21*	15
-1	4	21*	11
2	4	93	92
-2	4	31	33
3	4	31	26
-3	4	38	34
-4	4	64	67
-5	4	22*	14
-6	4	51	49
-7	4	35	34
0	5	24*	15
-1	5	81	79
-2	5	25*	24
-3	5	24*	13
-4	5	24*	16
-5	5	24*	17

on the basis of counting statistics alone. This indicates that some unknown factor is affecting the intensities.

The reduction of the intensity data to structure amplitudes was accomplished with the program *DACOR*, written by the author. The background was approximated by a straight line between the two measured background points. The intensities, corrected for background, were next corrected for Lorentz and polarization effects but not for absorption. Standard deviations $\sigma(I)$ were based on counting statistics and were corrected to $\sigma(F)$. Of the 2344 measured reflections, 313 were less than 2 $\sigma(I)$ and were considered unobserved. The intensities of these unobserved reflections were then set equal to 2 $\sigma(I)$. For the 2031 observed reflections, the statistical R value $R_s = \Sigma\,\sigma(F)/\Sigma\,F$ was 0·016.

Determination and refinement of the structure

The Fourier and least-squares programs used in this analysis were local modifications of programs obtained from Professor J. Trotter (1965). The least-squares program was block-diagonal in coordinates and diagonal in temperature factors. It minimized $\Sigma\,w(|F_o|-|F_c|)^2$. All least-squares cycles were based on observed reflections only. The atomic scattering factors used for all atoms were those tabulated by Ibers (1962). All computations were performed on the IBM 7044 and 360 computers.

The position of the sulfur atom was determined from an unsharpened three-dimensional Patterson map. A three-dimensional electron density map, phased on the sulfur atom, yielded the positions of the other 21 non-hydrogen atoms. Initially, these 21 atoms were assigned carbon scattering factors. Three cycles of least-squares with individual isotropic thermal parameters and unit weighting reduced $R_1 = \Sigma\,||F_o|-|F_c||/\Sigma\,|F_o|$ to 0·16 for observed reflections. Inspection of the output temperature factors of cycle 4 enabled the oxygen and the four nitrogen atoms to be identified. This identification was in complete accord with calculated bond lengths and with chemical evidence. Three additional cycles of least-squares reduced R_1 to 0·13. A difference Fourier then indicated considerable anisotropic thermal motion. Several cycles of anisotropic least-squares refinement with Hughes (1941) weighting yielded $R_1 = 0{\cdot}062$. A second difference Fourier clearly showed the 12 hydrogen atoms. The hydrogens were assigned isotropic thermal parameters and refinement was resumed, all parameters being allowed to vary. After several cycles, R_1 converged to 0·032.

In the course of the refinement, between 17 and 26 reflections were considered to be severely enough affected by extinction to be excluded from the refinement. A plot of $(F_c/F_o)^2$ *versus* $\left(\dfrac{1+\cos^4 2\theta}{(1+\cos^2 2\theta)\sin 2\theta}\right) F_o^2 = fF_o^2$ yielded the extinction parameter $g = 2{\cdot}5\times10^{-5}$ (Zachariasen, 1963, 1965). The observed data were then corrected for extinction by the formula $F^2_{\text{corr}} = F_o^2(1+gfF_o^2)$. The least-squares analysis was resumed with weights now defined as:

$$w=[\sigma(F_o)+0{\cdot}01F_{\text{corr}}+0{\cdot}1(F_{\text{corr}}-F_o)]^{-2}\,.$$

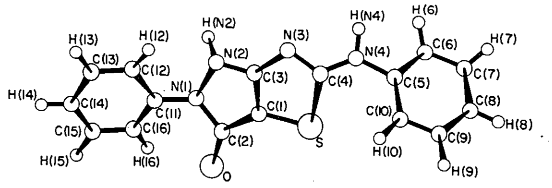

Fig. 1. Configuration and labeling of the molecule.

Table 2. *Final parameters of non-hydrogen atoms*

$$\text{T.F.} = \exp\left[-\tfrac{1}{4}(h^2a^{*2}B_{11}+k^2b^{*2}B_{22}+l^2c^{*2}B_{33}+2hka^*b^*B_{12}+2hla^*c^*B_{13}+2klb^*c^*B_{23})\right].$$

	$10^5 x$	$10^5 y$	$10^5 z$	B_{11} (Å^2)	B_{22} (Å^2)	B_{33} (Å^2)	B_{12} (Å^2)	B_{13} (Å^2)	B_{23} (Å^2)
S	6842 (2)	38324 (7)	35202 (4)	2·26 (1)	4·65 (2)	1·98 (1)	0·23 (1)	0·81 (1)	0·27 (1)
O	27968 (7)	30099 (20)	51801 (10)	2·86 (4)	6·32 (7)	2·02 (4)	0·35 (5)	0·81 (3)	0·70 (5)
N(1)	29054 (8)	33955 (21)	31868 (11)	2·23 (4)	4·41 (7)	1·80 (5)	0·25 (5)	0·56 (4)	0·26 (5)
N(2)	23952 (8)	39419 (21)	20158 (11)	2·33 (4)	4·53 (7)	1·75 (5)	0·26 (5)	0·47 (4)	0·18 (5)
N(3)	8972 (8)	42868 (22)	13234 (12)	2·48 (4)	4·40 (7)	2·13 (5)	0·24 (5)	0·71 (4)	0·16 (5)
N(4)	−4676 (8)	42978 (23)	12763 (12)	2·34 (4)	5·02 (8)	2·32 (5)	0·22 (5)	0·71 (4)	0·31 (5)
C(1)	16619 (9)	37448 (25)	33767 (14)	2·41 (5)	3·99 (8)	2·28 (6)	0·03 (6)	0·97 (4)	0·15 (6)
C(2)	24741 (9)	33252 (24)	40688 (14)	2·53 (5)	3·71 (8)	2·08 (6)	0·03 (6)	0·80 (4)	0·20 (6)
C(3)	16332 (9)	40210 (24)	21756 (14)	2·42 (5)	3·54 (8)	2·17 (6)	0·03 (6)	0·73 (4)	0·01 (5)
C(4)	3296 (9)	41788 (24)	19088 (14)	2·56 (5)	3·35 (8)	2·23 (6)	0·12 (6)	0·68 (5)	0·06 (5)
C(5)	−11664 (9)	41303 (24)	16944 (15)	2·47 (5)	3·30 (8)	2·92 (6)	0·29 (6)	0·87 (5)	0·28 (6)
C(6)	−18810 (11)	36388 (29)	8105 (17)	2·92 (6)	5·41 (10)	3·62 (7)	−0·37 (7)	0·80 (6)	−0·62 (8)
C(7)	−25986 (12)	35044 (35)	11247 (21)	2·98 (8)	7·40 (14)	6·00 (10)	−1·03 (9)	1·25 (7)	−1·10 (10)
C(8)	−26161 (11)	38450 (34)	23216 (19)	3·45 (6)	6·44 (12)	5·84 (9)	−0·03 (8)	2·53 (6)	0·48 (9)
C(9)	−19111 (11)	43476 (31)	31881 (17)	4·09 (8)	5·26 (10)	3·95 (7)	0·93 (8)	2·09 (5)	0·79 (8)
C(10)	−11806 (10)	45043 (29)	28900 (15)	3·12 (6)	4·84 (9)	3·01 (6)	0·39 (7)	1·15 (5)	0·10 (7)
C(11)	37626 (9)	34357 (24)	34065 (14)	2·36 (5)	3·36 (8)	2·24 (6)	0·10 (6)	0·75 (4)	−0·30 (5)
C(12)	41071 (10)	44348 (27)	26428 (15)	2·92 (6)	4·11 (9)	2·82 (6)	0·01 (6)	0·82 (5)	0·41 (6)
C(13)	49450 (10)	45128 (29)	28964 (16)	3·07 (6)	4·55 (9)	3·71 (7)	−0·22 (7)	1·31 (5)	0·36 (7)
C(14)	54471 (10)	36161 (28)	39056 (17)	2·53 (6)	4·45 (9)	3·84 (7)	−0·01 (7)	0·83 (5)	−0·25 (7)
C(15)	51014 (10)	26004 (28)	46456 (17)	2·79 (6)	4·48 (9)	3·13 (7)	0·69 (7)	0·61 (5)	0·19 (7)
C(16)	42616 (10)	24862 (26)	43974 (15)	2·87 (6)	3·87 (8)	2·67 (6)	0·33 (6)	0·92 (5)	0·26 (6)

The final discrepancy factors for all 2031 observed reflections were $R_1=0{\cdot}032$ and $R_2=[\Sigma\ w(F_{\text{corr}}-F_c)^2/\Sigma\ wF^2_{\text{corr}}]^{1/2}=0{\cdot}039$. The standard deviation of an observation of unit weight was 1·66. Of the 313 reflections too weak to be observed, only 21 had $F_c>F_{\text{min}}$ and none had $F_c>1{\cdot}7F_{\text{min}}$. A final difference Fourier contained values between $-0{\cdot}22$ and $+0{\cdot}16$ e.Å^{-3} and thus verified the structure.

The final values of the observed and calculated structure factors are compared in Table 1. The final positional and thermal parameters and their standard deviations are given for non-hydrogen atoms in Table 2 and for hydrogen atoms in Table 3. The average standard deviations of the positional parameters expressed in Å are 0·0004 for S, 0·0013 for O, 0·0014 for N, 0·0019 for C, and 0·022 for H. These standard deviations were derived from the semi-diagonalized least-squares process and are to be considered optimistic by a factor of two or three.

Description and discussion of the structure

The general shape and configuration of the molecule can be seen in Fig. 1. Bond lengths and angles, uncorrected for thermal motion, are given in Tables 4 and 5 and Fig. 2. The standard deviations given in Tables 4

Table 3. *Final parameters of hydrogen atoms*

	10^4x	10^4y	10^4z	B
H(N2)	2483 (12)	3251 (28)	1330 (18)	3·3 (5) Å^2
H(N4)	−562 (13)	4564 (33)	467 (20)	4·3 (6)
H(6)	−1875 (13)	3365 (31)	−42 (19)	4·0 (6)
H(7)	−3111 (14)	3208 (32)	431 (21)	4·8 (6)
H(8)	−3134 (14)	3793 (35)	2534 (21)	5·3 (7)
H(9)	−1933 (13)	4629 (31)	4039 (18)	4·0 (6)
H(10)	−693 (12)	4906 (31)	3504 (18)	3·5 (5)
H(12)	3740 (12)	5101 (30)	1928 (18)	3·4 (5)
H(13)	5179 (13)	5217 (36)	2336 (20)	4·9 (6)
H(14)	6068 (13)	3738 (29)	4096 (19)	3·3 (5)
H(15)	5448 (12)	1975 (30)	5384 (19)	3·4 (5)
H(16)	4023 (12)	1771 (30)	4935 (18)	3·7 (6)

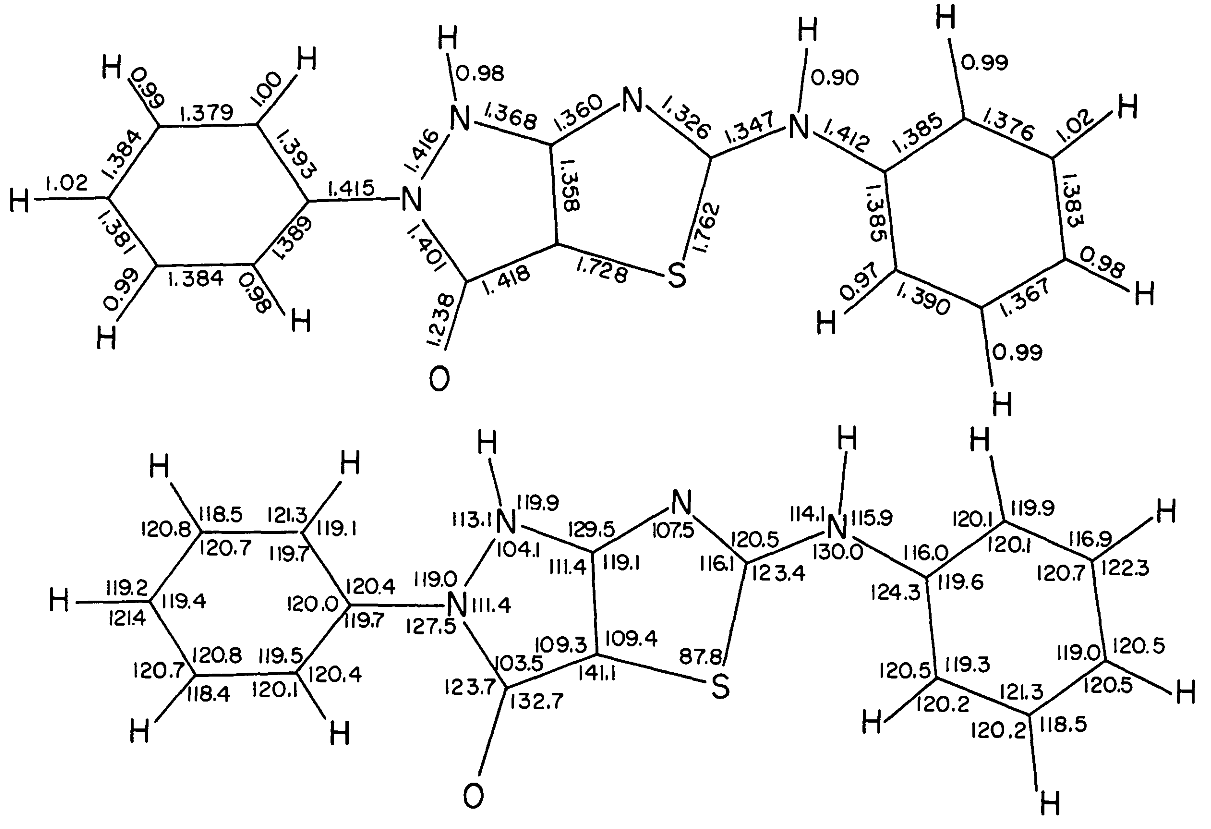

Fig. 2. Bond lengths and angles.

and 5 were calculated from the coordinate standard deviations obtained from the least-squares process. More realistic estimates of the standard deviations were obtained from consideration of the presumably equivalent bond lengths and angles of the two phenyl groups. These standard deviations are 0·007 Å for C–C, 0·017 Å for C–H, 0·7° for C–C–C, and 1·3° for C–C–H.

Table 4. *Intramolecular bond distances*

S—C(1)	1·728 Å	C(11)–C(12)	1·393 Å
S—C(4)	1·762	C(12)–C(13)	1·379
C(1)—C(2)	1·418	C(13)–C(14)	1·384
C(1)—C(3)	1·358	C(14)–C(15)	1·381
C(2)—O	1·238	C(15)–C(16)	1·384
C(2)—N(1)	1·401	C(16)–C(11)	1·389
N(1)—C(11)	1·415	N(2)—H(N2)	0·976
N(1)—N(2)	1·416	N(4)—H(N4)	0·902
N(2)—C(3)	1·368	C(6)—H(6)	0·987
C(3)—N(3)	1·360	C(7)—H(7)	1·017
N(3)—C(4)	1·326	C(8)—H(8)	0·984
C(4)—N(4)	1·347	C(9)—H(9)	0·993
N(4)—C(5)	1·412	C(10)–H(10)	0·965
C(5)—C(6)	1·385	C(12)–H(12)	1·000
C(6)—C(7)	1·376	C(13)–H(13)	0·991
C(7)—C(8)	1·383	C(14)–H(14)	1·024
C(8)—C(9)	1·367	C(15)–H(15)	0·989
C(9)—C(10)	1·390	C(16)–H(16)	0·982
C(10)–C(5)	1·385		

Estimated standard deviations:
Bonds without hydrogen 0·003 Å,
Bonds with hydrogen 0·024 Å.

Inspection of the molecular configuration and the bond lengths reveals that C(1)–C(3) and N(3)–C(4) are double bonds. Therefore, the molecule is 5-anilino-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazolo[3,4-*d*]thiazole. However, all bond lengths [except S–C(4)] not involving hydrogen are considerably shorter than normal single-bond distances and it is evident that these bonds have considerable aromatic π character. For example, consideration of the covalent radii for sulfur (Pauling, 1960) and trigonal carbon (Bastiansen & Traetteberg, 1962) leads one to expect the sulfur–carbon single bond to be 1·78 Å and the double bond to be 1·61 Å. Thus the S–C(4) bond (1·762 Å) is not much different from a single bond, but the S–C(1) bond (1·728 Å) has considerable double-bond character and plays an important role in any resonance scheme. Likewise, C(1)–C(3) (1·358 Å) is close to the accepted C=C distance of 1·34 Å, and C(1)–C(2) (1·418 Å) is much shorter than the C–C distance of 1·48 Å characteristic for trigonal carbons (Lide, 1962; Stoicheff, 1962; Bastiansen & Traetteberg, 1962; Sutton, 1965). Carbon–nitrogen bonds are expected to be 1·47 Å for C–N and 1·27 Å for C=N (Sutton, 1965). The N(3)–C(4) bond (1·326 Å) is somewhat longer than a normal double bond, whereas the six other C–N bonds, ranging from 1·347 to 1·415 Å, are considerably shorter than a single bond. The N(1)–N(2) bond (1·416 Å) is somwhat shorter than the 1·45 Å for the N–N single bond in hydrazine (Sutton, 1965) and the C–O bond (1·238) is slightly longer

Table 5. *Intramolecular bond angles*

C(4)-S—C(1)	87·8°	C(16)–C(11)–C(12)	120·0°
S—C(1)—C(3)	109·4	C(11)–C(12)–C(13)	119·7
C(1)-C(3)—N(3)	119·1	C(12)–C(13)–C(14)	120·7
C(3)-N(3)—C(4)	107·5	C(13)–C(14)–C(15)	119·4
N(3)-C(4)—S	116·1	C(14)–C(15)–C(16)	120·8
C(3)-C(1)—C(2)	109·3	C(15)–C(16)–C(11)	119·5
C(1)-C(2)—N(1)	103·5	N(1)—N(2)—H(N2)	113·1
C(2)-N(1)—N(2)	111·4	C(3)—N(2)—H(N2)	119·9
N(1)-N(2)—C(3)	104·1	C(4)—N(4)—H(N4)	114·1
N(2)-C(3)—C(1)	111·4	C(5)—N(4)—H(N4)	115·9
S—C(1)—C(2)	141·1	C(5)—C(6)—H(6)	120·1
N(2)-C(3)—N(3)	129·5	C(7)—C(6)—H(6)	119·9
C(1)-C(2)—O	132·7	C(6)—C(7)—H(7)	116·9
N(1)-C(2)—O	123·7	C(8)—C(7)—H(7)	122·3
N(3)-C(4)—N(4)	120·5	C(7)—C(8)—H(8)	120·5
S—C(4)—N(4)	123·4	C(9)—C(8)—H(8)	120·5
C(4)-N(4)—C(5)	130·0	C(8)—C(9)—H(9)	118·5
N(4)-C(5)—C(6)	116·0	C(10)–C(9)—H(9)	120·2
N(4)-C(5)—C(10)	124·3	C(9)—C(10)–H(10)	120·2
C(10)-C(5)—C(6)	119·6	C(5)—C(10)–H(10)	120·5
C(5)-C(6)—C(7)	120·1	C(11)–C(12)–H(12)	119·1
C(6)-C(7)—C(8)	120·7	C(13)–C(12)–H(12)	121·3
C(7)-C(8)—C(9)	119·0	C(12)–C(13)–H(13)	118·5
C(8)-C(9)—C(10)	121·3	C(14)–C(13)–H(13)	120·8
C(9)-C(10)-C(5)	119·3	C(13)–C(14)–H(14)	119·2
C(2)-N(1)—C(11)	127·5	C(15)–C(14)–H(14)	121·4
N(2)-N(1)—C(11)	119·0	C(14)–C(15)–H(15)	120·7
N(1)-C(11)-C(12)	120·4	C(16)–C(15)–H(15)	118·4
N(1)-C(11)-C(16)	119·7	C(15)–C(16)–H(16)	120·1
		C(11)–C(16)–H(16)	120·4

Estimated standard deviations: Angles without hydrogen 0·2°
Angles with hydrogen 1·3°.

than the 1·22 Å found for C=O in ketones and aldehydes (Sutton, 1965). With these bond-length considerations in mind, it is not surprising to find that atoms C(1), C(2), C(3), C(4), N(1), N(2), N(3), S, and O all lie within 0·07 Å of a plane (Schomaker, Waser, Marsh & Bergman, 1959; Blow, 1960) whose equation is

$$0{\cdot}0753X+0{\cdot}9792Y+0{\cdot}1887Z=3{\cdot}497\ ,$$

referred to the a,b,c^* axes. These observations are all consistent with the existence of lone pairs of electrons on the N, S, and O atoms and it is reasonable to expect that the transfer of charge from these lone-pair orbitals into a conjugated system involving double bonds greatly increases the stability of the molecule. Many structures closely related to the present one have been determined with quite good agreement in bond features. Among some recent ones are antabuse (Karle, Estlin & Britts, 1967), uric acid (Ringertz, 1966), isocytosine (Sharma & McConnell, 1965), alloxan (Singh, 1965), purine (Watson, Sweet & Marsh, 1965), $C_{17}H_{15}Cl_3N_2O$ (Karle & Karle, 1967) and $(C_9H_9N_3S)_2$ (Allmann, 1967).

The two phenyl groups are quite normal. The average bond lengths are 1·383 Å for C–C and 0·993 Å for C–H, and the average C–C–C and C–C–H bond angles are both 120·0°. The planes of the phenyls are tilted relative to the approximate plane of S, N(1)···N(3), C(1)···C(4) by 27·8° for the C(5)···C(10) phenyl and by −28·7° for the C(11)···C(16) phenyl. These tilts coupled with distorted angular distributions at N(1), N(4), and C(5) serve to relieve the strain caused by several close intramolecular contacts. The short contacts are C(16)···O (2·914 Å), H(16)···O (2·380 Å), S···C(10) (3·102 Å) and S···H(10) (2·483 Å), all of which are considerably shorter than the sums of the van der Waals radii. The short contacts possibly suggest weak intramolecular hydrogen bonds, but more likely represent a compromise among the requirements of molecular packing, atomic size, and resonance structures.

Fig. 3 shows the molecular packing and hydrogen-bond network in the unit cell, and Table 6 gives the relevant distances and angles. Table 7 lists other intermolecular approaches (excluding hydrogen) less than 3·5 Å. Molecules related by the *c* glide plane are connected by strong N–H···O bonds to form infinite ribbons. In addition, alternate pairs of N–H···N bonds, in the form of eight-membered rings about centers of symmetry, join each ribbon with the ribbon above and the ribbon below to form sheets of hydrogen-bonded molecules.

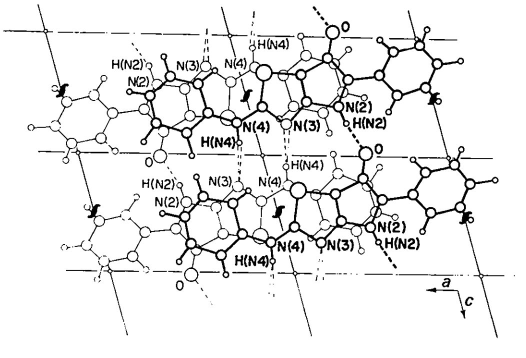

Fig. 3. A view down the *b* axis showing contents of a unit cell. Broken lines represent hydrogen bonds.

Table 6. *Hydrogen bond distances and angles*

N(2)$_{\mathrm{I}}\cdots$O$_{\mathrm{II}}$	2·776 Å
H(N2)$_{\mathrm{I}}\cdots$O$_{\mathrm{II}}$	1·806
N(4)$_{\mathrm{I}}\cdots$N(3)$_{\mathrm{III}}$	3·002
H(N4)$_{\mathrm{I}}\cdots$N(3)$_{\mathrm{III}}$	2·115
N(2)$_{\mathrm{I}}$–H(N2)$_{\mathrm{I}}\cdots$O$_{\mathrm{II}}$	171·9°
H(N2)$_{\mathrm{I}}\cdots$O$_{\mathrm{II}}$–C(2)$_{\mathrm{II}}$	133·6
N(4)$_{\mathrm{I}}$–H(N4)$_{\mathrm{I}}\cdots$N(3)$_{\mathrm{III}}$	167·3
H(N4)$_{\mathrm{I}}\cdots$N(3)$_{\mathrm{III}}$–C(4)$_{\mathrm{III}}$	117·7

I, II and III refer to the equivalent positions x,y,z; $x,\ \frac{1}{2}-y,\ -\frac{1}{2}+z$, and $-x,\ 1-y,\ -z$, respectively.

Table 7. *Short intermolecular contacts*

C(9)$_{\mathrm{I}}\cdots$O$_{\mathrm{II}}$	3·344 Å
O$_{\mathrm{I}}\cdots$C(9)$_{\mathrm{II}}$	3·344
N(4)$_{\mathrm{I}}\cdots$S$_{\mathrm{III}}$	3·417
C(14)$_{\mathrm{I}}\cdots$C(16)$_{\mathrm{IV}}$	3·437
C(16)$_{\mathrm{I}}\cdots$C(14)$_{\mathrm{IV}}$	3·437
C(10)$_{\mathrm{I}}\cdots$C(3)$_{\mathrm{III}}$	3·452
C(8)$_{\mathrm{I}}\cdots$N(1)$_{\mathrm{III}}$	3·453
C(10)$_{\mathrm{I}}\cdots$C(1)$_{\mathrm{III}}$	3·470

I, II, III and IV refer to the equivalent positions $x,\ y,\ z$; $-x,\ 1-y,\ 1-z$; $-x,\ \frac{1}{2}+y,\ \frac{1}{2}-z$; and $1-x,\ 1-y,\ 1-z$, respectively.

In the N–H···O bond, the N···O separation is 2·776 Å, considerably shorter than the normal N–H···O separation of 2·93 Å, given by Wallwork (1962) and somewhat shorter than the normal N^+–H···O^- distance of 2·83 Å. This indicates that an important resonance form of the mulecule involves species with $N(2)^+$ and O^-. In the N–H···N bonds, the N···N separation of 3·002 Å is shorter than the normal 3·07 Å.

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Polytypes of the Two-Packet Chlorites

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The systematic derivation of all the possible chlorite polytypes with a two-packet unit cell is carried out. The derivation is completed for trioctahedral, dioctahedral and di-trioctahedral chlorites. The diffraction features which should identify the different chlorite polytypes are calculated.

The chlorites are layer-lattice silicates of variable composition. They consist of regularly alternating talc- and brucite-type layers (Pauling, 1930). The talc-type layer is three-storied, consisting of an octahedral net occupying the centre and two tetrahedral nets adjoining the octahedral net from above and below. The brucite-type layer is one-storied, consisting of one octahedral network. Each pair of three-storied and one-storied layers forms a chlorite packet. It would be accepted that a chlorite packet comprises one three-storied layer and two halves of two different one-storied layers, one covering it and one underlying it.

Chlorites are divided into trioctahedral, where all the octahedra are occupied by cations, and dioctahedral, in which only $\frac{2}{3}$ of all the octahedral positions are occupied. The ideal structural formula for trioctahedral chlorites is

$$(\mathrm{Mg}_{6-x-y}\mathrm{Fe}^{2+}_{y}\mathrm{Al}_{x})\,(\mathrm{Si}_{4-x}\mathrm{Al}_{x})\mathrm{O}_{10}(\mathrm{OH})_{8}\,,$$

and for dioctahedral chlorites,

$$(\mathrm{Al}_{4+x/3})\,(\mathrm{Si}_{4-x}\mathrm{Al}_{x})\mathrm{O}_{10}(\mathrm{OH})_{8}\,.$$

The structure of the chlorites is made up from chlorite packets superimposed one upon another in a varying manner. Scientific publications usually describe one-packet and semi-random chlorites (McMurchy, 1934; Zvyagin, 1964; Brown & Bailey, 1962; 1963; Shirozu & Bailey, 1965).

However, McMurchy (1934), and later Garrido (1949) and Brindley, Oughton & Robinson (1950) pointed out the occurrence of chlorites with a different periodicity of packets along the *c* axis, and discussed some polytypes possible for chlorites with more than one packet per repeat period. Chlorites with a two-packet monoclinic unit cell have been described by Drits & Lazarenko (1967) and Wlasow & Drits (1967). This paper gives a systematic determination of all the possible chlorite polytypes with a repeat period of two chlorite packets and their diffraction features are calculated in order to determine the structure of the new two-packet chlorites which we have investigated (Wlasow & Drits, 1967; Drits & Lazarenko, 1967). In deriving these polytypes the analytical method of Zvyagin (1964) was used, providing a simpler solution of this problem.

Table 1. *Components of the displacements σ and τ along the a and b axes which are possible in structures of layer silicates*

Symbols of the displacements	σ_1 τ_4	σ_2 τ_5	σ_3 τ_6	σ_4 τ_1	σ_5 τ_2	σ_6 τ_3	τ_0	τ_+	τ_-
Components of the displacements	$\frac{1}{3}, \frac{1}{3}$	$-\frac{1}{3}, \frac{1}{3}$	$\frac{1}{3}, 0$	$-\frac{1}{3}, -\frac{1}{3}$	$\frac{1}{3}, -\frac{1}{3}$	$-\frac{1}{3}, 0$	0, 0	$0, \frac{1}{3}$	$0, -\frac{1}{3}$

Displacement components are expressed in multiples of *a* and *b*.