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Studies on Intermolecular Complex Formation. III. Crystal Structure of Thymine-*N*,*N*-Diethylmelamine Complex Monohydrate

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The crystal and molecular structure of the thymine-*N*,*N*-diethylmelamine complex monohydrate has been solved by means of the direct method. The crystals are monoclinic, with the space group of $P2_1/c$ and with $a=7.167$, $b=16.123$, $c=13.179$ Å, and $\beta=104.2°$. The final *R*-factor for 1612 reflections is 0.087. These two components are hydrogen-bonded by a NH···N and two NH···O bonds with distances of 2.881, 2.821, and 3.006 Å. The complex formation ability for thymine and uracil has also been tested for some other organic compounds with amino and nitrogen heterocycles.

Intermolecular interactions in crystalline fields have recently come into focus after Hoogsteen displayed some model complexes of DNA.[1] The pairing mechanism has been interpreted as being due to special features in the purine and pyrimidine structures, in which the atoms in these molecules are arranged in appropriate combinations of so-called "complementary hydrogen bond."[2-18] Such arrangements would have a bearing on the biological duplication mechanism. Subsequently, we have been trying to determine some role for the intermolecular complex formation between nucleotide bases and organic compounds.[19,20] It has been found that cytosine and adenine exhibit a rather strong affinity for many organic compounds, while thymine and/or uracil are less reactive and bind only with a few organic compounds that have a –C(NH$_2$)=N–C(NH$_2$)=N– grouping or a few other basic groups. In this paper we wish to describe the crystal structure of the thymine–*N*,*N*-diethylmelamine monohydrate complex and the general ability of complex formation in some organic compounds that can produce a complex with thymine and/or uracil. The biological importance of the melamine molecule and its derivatives has already been discussed in several places.[21,22] These studies may make it possible to project an illustrative intermolecular spatial system.

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TABLE 1. STRUCTURE FACTOR DATA ($\times 10$) FOR THE OBSERVED REFLECTIONS

TABLE 1. (Continued)

L	FO	FC
7	53	49
8	26	15
9	140	167
10	91	102
11	169	179
K= 3,H= 4		
-16	26	20
-15	24	19
-14	51	46
-13	85	84
-12	83	75
-10	141	125
-9	46	44
-8	155	142
-7	154	143
-6	36	36
-5	235	229
-4	310	311
-3	50	69
-2	76	71
-1	338	320
0	111	125
1	160	127
2	88	111
3	109	122
5	109	99
6	65	46
7	39	40
8	110	123
9	33	48
10	33	37
11	91	102
12	25	26
14	34	25
K= 4,H= 4		
-16	46	50
-13	24	25
-12	28	30
-11	38	31
-10	62	67
-9	43	42
-8	96	98
-7	77	78
-6	176	163
-5	40	27
-3	49	49
-2	261	251
-1	194	194
0	163	156
1	89	96
2	120	149
4	100	93
5	39	27
6	107	114
7	124	146
10	97	89
11	86	81
13	29	29
14	37	26
K= 5,H= 4		
-17	35	12
-16	43	34
-14	54	44
-13	56	55
-12	71	62
-11	43	46
-9	22	39
-8	128	140
-7	32	38
-6	125	121
-5	31	39
-4	165	143
-3	259	282
-2	206	216
-1	31	30
1	90	90
2	50	39
3	162	158
4	123	95
5	138	155
6	268	288
7	63	53
8	26	38
9	31	27
10	127	138
12	62	72
13	46	39
K= 6,H= 4		
-15	39	32
-11	51	50
-10	195	193
-9	171	184
-8	114	129
-7	94	95
-6	91	99
-5	70	78
-4	184	183
-3	191	220
-2	328	317
-1	150	119
0	170	198
1	68	63
3	35	13
4	73	71
5	76	72
6	290	308
7	199	210
8	47	42
9	25	42
10	76	88
14	55	60
K= 7,H= 4		
-15	33	38
-12	47	29
-11	140	131
-10	60	58
-9	114	108
-8	78	74
-7	171	181
-6	79	49
-5	144	141
-4	71	48
-3	214	183
-1	135	114
1	115	114
3	66	58
5	167	174
6	58	48
7	125	137
8	87	90
9	87	88
10	29	27
11	61	55
13	42	32
14	33	34

L	FO	FC
K= 8,H= 4		
-14	32	21
-13	26	17
-12	54	47
-11	85	89
-10	124	124
-9	80	89
-8	187	184
-7	350	367
-6	203	198
-4	82	85
-3	195	218
-2	178	175
-1	252	251
0	103	123
2	169	159
3	35	39
5	119	129
8	90	105
9	181	201
10	78	83
13	62	70
K= 9,H= 4		
-13	25	27
-12	46	42
-10	23	36
-8	71	46
-7	306	311
-6	435	443
-5	79	92
-4	63	43
-3	38	60
-2	196	204
-1	34	50
0	28	5
2	98	92
3	35	23
4	57	57
5	38	32
6	78	90
7	98	88
8	48	38
9	203	217
10	227	242
12	30	26
K= 10,H= 4		
-13	32	34
-9	53	46
-8	191	185
-7	29	26
-5	55	55
-4	37	29
-3	28	24
-2	145	129
-1	45	58
0	75	70
1	60	75
2	51	26
3	157	154
4	29	50
7	53	41
8	36	48
9	72	71
10	61	58
12	54	30
13	29	24
K= 11,H= 4		
-15	59	44
-14	31	28
-12	38	48
-11	52	56
-9	48	42
-8	152	160
-7	136	141
-6	69	81
-5	21	15
-4	317	325
-3	266	272
-2	176	178
-1	28	34
0	56	74
1	120	118
2	43	35
4	100	90
6	87	82
7	24	34
8	77	80
9	39	43
11	30	22
12	144	153
K= 12,H= 4		
-15	33	39
-14	27	30
-13	67	64
-10	29	35
-9	138	125
-8	32	30
-7	59	52
-4	121	122
-3	68	68
-2	113	111
-1	24	20
0	139	137
1	164	162
2	116	124
3	166	173
5	59	74
6	28	29
7	89	89
8	35	17
10	39	35
11	76	63
12	67	68
K= 13,H= 4		
-15	38	19
-13	80	73
-11	48	50
-9	44	30
-8	51	39
-7	54	54
-5	207	207
-4	31	24
-3	39	35
-1	91	82
0	50	48
2	24	30
3	121	128
4	44	33
5	35	50
8	71	64
9	82	85
11	34	39

L	FO	FC
K= 14,H= 4		
-12	70	63
-9	63	55
-8	89	78
-7	28	18
-6	48	56
-5	30	30
-4	46	57
-3	30	25
-1	66	61
0	161	166
1	163	163
2	49	56
3	65	75
4	35	48
7	47	50
8	52	54
10	46	32
K= 15,H= 4		
-12	52	53
-10	35	7
-9	47	33
-8	63	58
-7	26	16
-6	76	52
-5	63	48
-4	30	27
-2	56	68
-1	178	194
0	279	278
1	87	86
2	47	47
3	51	54
4	82	96
5	53	54
7	54	53
8	98	100
K= 16,H= 4		
-10	66	63
-9	28	26
-8	25	14
-7	55	49
-6	59	54
-4	98	96
-2	41	34
-1	109	100
0	104	94
1	49	54
6	54	44
7	28	10
8	46	51
K= 17,H= 4		
-10	47	45
-9	104	95
-8	28	19
-6	37	25
-4	41	47
-3	157	155
-2	78	80
0	29	22
1	83	75
3	67	66
6	32	33
7	51	57
K= 18,H= 4		
-10	42	28
-9	42	40
-6	26	19
-5	54	51
-4	92	99
-3	166	158
0	38	22
1	48	48
6	.38	40
K= 19,H= 4		
-7	83	78
-6	35	31
-4	75	63
-3	38	45
-2	49	45
0	86	81
1	58	48
K= 20,H= 4		
-5	42	37
-4	38	22
-2	57	48
0	58	52
1	35	27
2	60	49
3	27	18
K= 0,H= 5		
-16	176	148
-14	92	85
-12	29	30
-10	74	63
-8	336	319
-6	89	98
-4	232	220
0	478	489
4	301	315
6	44	55
8	252	259
12	48	67
K= 1,H= 5		
-14	53	44
-12	37	32
-10	70	50
-9	220	199
-8	223	226
-7	25	29
-6	77	61
-4	21	20
-2	86	91
-1	174	162
0	124	131
1	32	32
2	27	23
3	40	55
6	96	108
7	194	209
8	53	69
9	48	54
K= 2,H= 5		
-17	41	35
-16	84	69
-14	97	84
-13	194	179
-12	24	16
-11	129	123
-10	37	31
-9	89	80

L	FO	FC
-8	96	94
-7	75	68
-6	166	152
-5	130	127
-4	78	79
-3	77	85
-1	86	90
0	319	310
1	41	38
2	318	309
3	321	337
4	30	14
5	27	13
6	100	111
8	99	89
9	56	77
10	48	52
12	45	48
K= 3,H= 5		
-13	193	171
-12	119	112
-11	211	195
-10	72	60
-9	90	81
-8	32	16
-7	108	99
-6	49	53
-5	56	39
-4	161	141
-3	48	53
-2	46	54
-1	40	26
0	23	21
1	49	53
2	189	215
3	420	467
4	290	310
5	220	233
7	102	96
8	51	62
9	95	111
K= 4,H= 5		
-16	4	14
-15	37	34
-14	141	119
-13	43	40
-12	61	58
-11	24	24
-10	151	144
-9	21	32
-7	22	32
-6	31	33
-5	98	111
-3	38	61
-2	131	133
-1	51	23
0	22	25
1	136	150
2	200	205
3	141	142
5	76	91
6	105	102
7	80	95
8	71	84
9	66	61
K= 5,H= 5		
-14	86	79
-13	114	107
-11	169	163
-10	301	290
-9	125	112
-8	179	173
-7	28	18
-6	29	49
-5	30	28
-4	32	26
-3	131	144
-2	98	89
-1	91	94
2	100	116
3	132	162
4	57	51
5	225	222
6	365	398
7	59	68
8	90	95
K= 6,H= 5		
-13	76	78
-12	26	18
-11	93	87
-10	171	163
-9	184	170
-8	35	33
-7	42	34
-6	62	66
-5	57	58
-4	41	37
-2	187	118
-1	28	15
0	38	64
1	73	56
3	43	56
4	119	128
5	243	259
6	175	171
7	225	230
8	63	42
9	32	40
10	53	49
13	38	34
K= 7,H= 5		
-14	31	30
-12	43	41
-11	218	209
-8	44	58
-7	88	89
-5	75	62
-4	114	109
-3	23	22
-2	27	48
-1	30	61
1	91	75
2	93	91
3	146	160
5	71	100
6	129	124
7	75	85
K= 8,H= 5		
-17	33	13
-13	67	59
-12	58	65
-11	100	96
-10	82	94
-9	67	73

L	FO	FC
-8	68	68
-7	164	169
-5	24	36
-4	22	28
-3	66	76
-2	91	79
-1	85	79
0	28	33
2	113	108
3	75	68
5	28	37
6	56	54
7	36	47
8	52	59
9	141	146
11	26	34
K= 9,H= 5		
-15	46	47
-13	37	25
-12	78	64
-11	25	41
-10	51	43
-9	61	62
-8	23	29
-7	36	32
-6	141	152
-5	81	75
-4	79	84
-3	22	22
-2	134	132
1	185	190
2	39	43
3	76	71
4	86	89
5	100	90
6	71	79
8	55	52
10	75	76
12	36	58
K= 10,H= 5		
-16	39	31
-14	36	42
-12	85	67
-11	41	40
-9	68	69
-7	72	71
-6	56	46
-5	78	60
-4	99	86
-3	150	150
-2	61	65
-1	64	57
0	57	70
2	92	101
4	75	89
6	47	45
7	95	84
10	32	21
11	42	47
K= 11,H= 5		
-15	63	48
-12	35	35
-11	29	30
-10	25	13
-8	98	95
-6	48	58
-5	68	78
-4	23	9
-3	61	49
0	85	97
1	88	102
3	66	66
4	47	48
5	102	97
8	45	45
10	79	81
11	86	65
K= 12,H= 5		
-15	44	48
-12	47	42
-11	65	49
-9	96	101
-6	68	75
-5	88	85
-4	23	10
-3	39	35
1	74	96
2	45	53
3	23	19
4	110	116
5	103	108
7	65	64
8	55	36
10	54	45
K= 13,H= 5		
-13	44	48
-12	29	36
-7	59	53
-6	27	18
-5	56	69
-2	38	20
-1	56	58
0	64	57
1	47	45
3	35	22
5	79	80
6	45	46
8	27	27
9	53	46
K= 14,H= 5		
-13	72	73
-12	64	60
-8	66	70
-6	40	57
-5	94	98
-4	50	40
-1	26	17
0	46	42
3	145	135
4	75	86
6	42	38
7	28	23
8	129	111
K= 15,H= 5		
-6	83	68
-4	48	51
-3	66	67
-1	71	67
0	58	67
2	54	38
3	84	79
5	68	68
7	62	52

L	FO	FC
K= 16,H= 5		
-10	57	54
-9	33	32
-8	57	61
-5	68	58
-4	43	41
-3	55	42
-2	64	57
-1	36	25
1	42	29
4	27	26
6	78	75
7	38	36
K= 17,H= 5		
-8	27	21
-7	38	33
-4	76	77
-3	59	65
-2	133	128
0	37	31
5	37	50
6	43	42
K= 18,H= 5		
-9	37	23
-6	64	60
-5	50	54
-4	132	130
-3	120	121
-1	26	20
1	46	31
4	36	25
K= 19,H= 5		
-7	47	34
-3	79	78
-2	83	72
-1	37	29
1	50	37
2	42	26
K= 0,H= 6		
-16	223	184
-14	176	151
-12	54	45
-6	99	90
-4	93	102
-2	88	66
0	790	790
2	216	198
4	85	98
8	49	43
12	45	49
K= 1,H= 6		
-16	60	55
-14	32	8
-13	69	60
-9	33	28
-4	231	212
-3	11.	119
-2	260	269
-1	292	290
0	172	181
1	71	68
2	92	98
7	34	16
9	30	36
10	30	29
12	33	19
K= 2,H= 6		
-17	141	119
-14	105	95
-13	175	146
-10	31	26
-8	30	21
-7	54	63
-6	60	48
-5	152	134
-3	139	147
-2	92	111
-1	262	289
1	36	37
2	197	189
3	332	342
4	49	53
5	54	41
7	35	44
8	34	28
11	91	93
K= 3,H= 6		
-16	27	19
-14	60	49
-13	94	83
-12	107	95
-11	91	79
-9	71	59
-7	38	25
-5	59	79
-4	169	164
-3	296	328
-1	25	19
1	129	128
2	238	231
3	255	249
4	210	204
5	42	52
6	118	108
8	26	26
11	41	39
12	72	67
K= 4,H= 6		
-15	97	82
-14	104	93
-13	37	34
-12	65	69
-11	71	66
-9	52	39
-8	84	72
-7	33	23
-6	37	33
-5	66	84
-4	25	24
-3	55	68
-2	23	13
-1	101	99
0	150	161
1	185	195
2	45	30
3	88	89
4	66	75
5	79	80
10	39	46
11	51	60

L	FO	FC
K= 5,H= 6		
-16	54	50
-15	59	56
-14	81	69
-13	71	69
-11	59	58
-10	87	80
-9	69	64
-8	57	52
-7	50	55
-6	177	184
-5	31	20
-4	31	23
-3	48	43
-2	195	205
-1	110	109
0	66	62
1	66	70
2	115	122
3	124	128
5	97	100
7	30	34
8	78	76
9	38	38
10	50	57
K= 6,H= 6		
-15	72	58
-10	35	21
-8	71	60
-7	56	54
-5	70	89
-3	62	74
-2	57	69
-1	58	34
0	141	141
1	212	221
2	88	93
3	53	50
5	81	93
6	41	46
7	37	27
10	38	38
11	38	37
K= 7,H= 6		
-14	56	61
-12	63	62
-11	42	55
-9	30	28
-8	23	28
-7	64	57
-6	94	101
-5	83	90
-3	73	87
-2	78	79
-1	147	148
0	166	174
1	101	111
2	201	209
4	58	49
5	113	123
9	54	51
11	50	46
K= 8,H= 6		
-16	28	23
-15	65	61
-13	27	30
-11	29	24
-9	23	13
-7	83	79
-6	36	16
-5	30	33
-4	49	58
-3	114	149
-2	28	51
-1	33	32
1	136	144
2	72	71
3	71	77
4	50	56
5	42	41
6	26	28
7	40	31
9	62	58
K= 9,H= 6		
-15	31	20
-13	25	14
-10	30	23
-6	49	48
-3	72	68
-2	41	54
-1	134	127
0	82	73
1	136	142
3	79	79
5	26	19
8	34	16
9	36	43
K= 10,H= 6		
-14	54	52
-6	27	17
-4	24	12
-3	42	27
-2	57	65
-1	49	46
0	80	79
2	135	140
3	109	95
4	31	29
6	38	26
8	35	25
K= 11,H= 6		
-10	28	18
-9	30	25
-8	52	38
-7	54	42
-4	39	44
-2	24	19
-1	126	136
0	62	62
2	27	27
3	54	59
5	29	28
7	40	44
K= 12,H= 6		
-13	41	43
-10	45	50
-8	100	92
-2	68	69
0	114	113
1	33	35
4	32	28
5	48	43
7	60	40

L	FO	FC
8	56	60
K= 13,H= 6		
-13	26	6
-11	30	32
-10	69	68
-9	28	28
-6	47	40
-5	51	45
-4	35	28
-2	70	57
0	43	32
1	54	38
3	67	65
6	69	50
K= 14,H= 6		
-8	53	51
-7	32	39
-5	36	43
2	53	50
4	38	32
5	48	44
6	45	37
K= 15,H= 6		
-11	37	40
-5	40	41
-3	44	20
-2	47	41
-1	44	16
0	54	57
2	41	28
3	35	36
4	66	48
K= 16,H= 6		
-7	66	48
-6	79	67
-5	57	58
-4	46	44
-1	43	32
0	27	24
3	43	39
K= 17,H= 6		
-5	91	75
-4	37	27
0	35	21
2	45	31
K= 18,H= 6		
-6	72	52
-5	46	30
-4	29	29
-3	39	36
-1	28	22
0	56	49
K= 0,H= 7		
-16	85	80
-12	84	74
-8	112	104
-6	198	191
-4	111	119
-2	102	100
0	332	319
2	111	119
4	116	120
8	86	78
10	28	34
K= 1,H= 7		
-16	46	38
-12	33	19
-8	46	43
-7	29	23
-6	118	106
-5	199	198
-4	30	39
-3	58	64
-2	157	166
-1	178	179
0	83	85
3	60	57
4	25	34
6	45	46
8	77	83
10	37	45
K= 2,H= 7		
-12	40	31
-11	74	78
-10	71	69
-9	105	103
-8	55	43
-7	43	32
-6	98	97
-5	53	55
-2	130	130
-1	163	162
0	67	56
1	40	33
2	36	38
3	57	48
6	53	58
7	94	107
10	56	54
K= 3,H= 7		
-11	47	29
-9	77	73
-8	34	42
-7	43	43
-6	81	75
-5	61	49
-4	230	231
-3	321	347
-2	59	66
-1	65	50
2	25	13
3	27	33
4	64	63
5	51	56
7	61	66
8	53	61
10	64	66
K= 4,H= 7		
-15	39	21
-10	59	46
-8	81	73
-7	89	86
-6	87	99
-5	148	163
-4	184	203
-3	76	80
-1	37	32
1	48	49
2	47	42

L	FO	FC
3	48	39
4	29	22
6	40	31
8	41	42
K= 5,H= 7		
-11	38	35
-9	40	30
-8	99	91
-7	54	73
-6	189	200
-5	33	37
-4	163	172
-3	121	132
-2	79	83
-1	83	84
0	43	53
1	34	39
2	35	25
3	42	28
5	78	71
6	75	67
7	35	25
8	61	58
K= 6,H= 7		
-14	32	34
-13	31	32
-9	43	39
-7	220	215
-6	70	62
-5	23	31
-4	78	85
-3	103	101
0	37	52
2	68	71
3	45	52
9	123	128
K= 7,H= 7		
-15	27	29
-12	42	32
-11	44	40
-10	48	44
-8	29	27
-7	75	87
-6	94	87
-5	53	51
-4	28	26
-3	99	111
-2	64	67
0	84	83
1	68	73
4	45	40
5	39	45
6	35	40
8	30	35
9	52	48
K= 8,H= 7		
-15	32	28
-12	52	38
-10	23	22
-9	64	61
-6	47	40
-5	111	110
-4	132	141
-3	65	60
-2	36	33
-1	22	27
1	31	45
3	33	23
4	72	86
5	78	60
K= 9,H= 7		
-13	28	20
-11	45	30
-10	29	36
-9	66	62
-8	36	34
-6	35	47
-4	25	21
-3	72	84
-2	84	87
-1	44	43
1	69	54
2	99	88
4	38	26
5	48	51
6	47	58
7	31	38
K= 10,H= 7		
-13	28	18
-12	25	21
-9	48	48
-6	39	40
-5	39	26
-4	25	22
-3	64	81
-2	92	104
-1	86	108
4	45	49
5	24	29
7	63	61
K= 11,H= 7		
-12	32	38
-11	40	35
-10	25	35
-9	26	17
-8	44	42
-7	50	44
-3	80	78
-2	77	80
-1	26	26
0	54	38
1	36	39
2	32	11
6	35	36
K= 12,H= 7		
-10	35	23
-9	28	21
-5	93	86
-4	30	32
-3	31	28
-1	105	99
0	32	31
6	27	11
K= 13,H= 7		
-10	38	41
-9	83	73
-8	61	63
-6	33	25
-5	39	42
-4	33	33
-3	27	26

L	FO	FC
-2	38	44
-1	24	37
0	55	55
1	42	43
K= 14,H= 7		
-9	44	45
-8	68	53
-7	31	20
-6	35	23
-5	29	10
-4	75	72
-3	36	27
-2	49	42
0	27	13
3	29	30
K= 15,H= 7		
-9	30	14
-6	47	33
-3	53	50
-2	38	38
0	31	36
2	37	34
K= 16,H= 7		
-6	60	52
-5	82	74
-1	39	33
0	28	23
K= 0,H= 8		
-14	52	36
-12	96	88
-10	30	18
-8	53	47
-6	106	91
0	130	113
4	134	148
6	60	66
K= 1,H= 8		
-14	32	20
-13	31	27
-12	52	45
-11	26	15
-9	44	47
-5	65	72
-4	69	66
-3	30	24
-2	30	22
-1	57	57
0	55	48
2	35	35
3	38	45
4	56	54
7	98	104
K= 2,H= 8		
-13	41	39
-10	57	57
-9	156	145
-8	60	51
-6	96	84
-5	28	29
-4	32	29
-2	63	58
-1	28	36
0	35	33
1	41	27
2	55	58
3	25	32
5	48	48
6	43	44
7	86	80
K= 3,H= 8		
-11	53	37
-10	78	80
-8	54	67
-6	64	63
-5	44	38
-4	69	74
-2	90	84
-1	36	38
1	52	52
3	28	26
4	67	68
5	62	67
6	67	71
K= 4,H= 8		
-13	27	19
-12	40	26
-11	30	16
-9	54	58
-8	108	114
-6	69	64
-5	53	62
-4	123	116
-1	45	46
1	94	103
2	51	52
3	48	47
4	43	31
6	50	43
7	33	38
K= 5,H= 8		
-9	45	46
-7	50	47
-6	76	81
-4	31	36
-3	28	20
-1	28	15
0	34	31
1	58	56
2	41	29
4	27	18
5	39	26
K= 6,H= 8		
-7	133	142
-6	80	84
-3	36	33
-1	27	19
0	27	25
1	45	43
3	58	53
4	33	35
K= 7,H= 8		
-11	96	98
-10	42	46
-9	47	55
-8	53	62
-7	160	160
-6	34	37
-5	31	23
-2	41	27

L	FO	FC
-1	25	43
1	35	50
2	34	30
4	27	28
5	92	95
6	24	32
K= 8,H= 8		
-11	34	34
-9	40	31
-7	93	95
-3	51	55
-1	28	21
0	38	18
1	79	70
2	73	70
3	31	22
4	61	63
5	27	25
K= 9,H= 8		
-12	29	17
-11	33	36
-10	132	126
-9	83	75
-8	70	63
-4	62	53
0	39	49
1	53	48
3	28	26
5	38	39
K= 10,H= 8		
-12	71	63
-10	59	64
-9	33	32
-8	40	43
-6	58	59
-3	66	63
-1	33	30
0	31	36
2	71	74
4	77	85
K= 11,H= 8		
-11	52	50
-9	37	23
-8	34	24
-7	58	48
-1	33	27
0	32	53
1	46	45
2	34	36
K= 12,H= 8		
-6	42	54
-5	27	24
-3	58	59
0	33	23
K= 13,H= 8		
-6	49	54
-5	106	97
-1	32	29
K= 14,H= 8		
-5	28	35
-3	33	28
-2	27	35
K= 0,H= 9		
-12	37	41
-6	51	49
-2	67	56
0	48	18
2	73	75
4	59	56
K= 1,H= 9		
-13	36	35
-12	36	29
-10	53	35
-8	27	27
-7	28	16
-6	53	45
-5	55	53
-2	65	59
0	33	24
3	96	96
K= 2,H= 9		
-12	25	14
-5	55	46
-2	38	29
-1	43	39
4	48	55
K= 3,H= 9		
-7	23	22
-6	48	37
-5	28	13
-4	33	40
-3	36	29
-2	24	17
-1	47	50
0	24	24
1	35	28
2	28	39
K= 4,H= 9		
-10	42	37
-7	48	45
-6	23	9
-3	74	75
-2	52	53
-1	31	39
0	82	89
1	66	63
2	58	44
3	41	51
K= 5,H= 9		
-11	28	26
-9	58	30
-8	28	26
-6	36	34
-4	47	46
-2	65	66
-1	65	65
1	63	63
3	25	22
K= 6,H= 9		
-10	51	46
-6	30	33
-5	39	39
-3	32	36
-2	99	94

L	FO	FC
1	84	89
2	46	45
3	46	68
K= 7,H= 9		
-11	26	34
-9	41	42
-8	28	37
-7	24	27
-6	53	35
-4	47	42
-3	25	35
-2	34	33
-1	68	67
0	60	62
1	45	45
2	60	66
K= 8,H= 9		
-9	23	16
-7	28	37
-2	34	27
-1	33	22
1	90	101
K= 9,H= 9		
-7	35	43
-6	54	54
-5	26	30
-4	46	36
-3	35	42
0	39	40
K= 10,H= 9		
-7	28	24
-4	40	35
-2	25	13
K= 0,H= 10		
-8	55	28
-6	33	33
-4	79	69
-2	31	26
K= 1,H= 10		
-8	38	45
-6	31	43
-1	36	37
K= 2,H= 10		
-6	52	40
-5	105	105
K= 3,H= 10		
-3	27	27
-2	39	39
K= 4,H= 10		
-4	38	29
-3	40	49

Experimental

Equivalent moles of nucleotide bases and some nitrogen heterocycles were dissolved in hot 70% aqueous ethanol, and then allowed to stand for several days in a refrigerator. The X-ray diffraction of the materials obtained and of each component were measured up to $2\theta=50°$ (Cu Kα) with a Rigaku model D6C diffractometer.

The single crystals of thymine–*N*,*N*-diethylmelamine thus obtained, which were suitable for X-ray analysis, were used for the intensity measurements. A crystal measuring $0.5\times0.3\times0.2$ mm was chosen for recording the crystal data and the intensities. Preliminary Weissenberg photographs showed that the crystals belong to the monoclinic system, with the space group of $P2_1/c$. The lattice parameters from the diffractometer are: $a=7.167(9)$, $b=16.123(18)$, $c=13.179(15)$ Å, $\beta=104.2(1)°$, $D_{obsd}=1.39$, and $D_{calcd}=1.44$ g/cm^3. The three-dimensional intensities were collected on a Rigaku Denki four-circle automatic diffractometer with a FACOM-R computer system up to a 2θ limit of 60° using Mo Kα radiation and the $2\theta/\theta$ scan mode at a scan speed of 2° min^{-1}. A crystal was mounted along the *a*-axis, and a total of 1612 independent reflections were collected. Three standard reflections were used to monitor the crystal stability during the measurement. The data were corrected for the usual Lorentz and polarization factors, but no corrections were made for absorption.

Structure Analysis and Refinement

The reiterative application of Sayre's equation[23] was used to determine the phases of 270 reflections having $E\geqq1.50$. Three reflections (1,2,−3; 1,3,−3; 3,14,0) were chosen for specifying the origin, and three other reflections (0,0,16; 0,0,3; 5,5,6), were denoted by the symbols A, B and C respectively. The B and C symbols were uniquely determined to be −A and A respectively by subsequent calculations. Then the structure was satisfactorily solved by an interpretation of the *E*-map based on these phases. The atomic parameters were refined by the method of least-squares, using the block-diagonal least-squares program. When the *R*-factor was reduced to 0.18, the difference synthesis was calculated in order to determine the positions of the hydrogen atoms; the six atoms attached to C(11) and C(13) were vague because of disorder. The final *R*-factor reached was 0.087 for the structure factors listed in Table 1. The atomic parameters are given in Tables 2, 3 (a), and 3 (b).

Table 2. Fractional atomic coordinates in thymine—*N*,*N*-diethylmelamine complex monohyrate

Atom	*x*/*a*	*y*/*b*	*z*/*c*
N (1)	0.2413	0.9612	0.1210
C (2)	0.2674	0.8827	0.2221
N (3)	0.3082	0.7341	0.2485
C (4)	0.3255	0.6529	0.1588
N (5)	0.3004	0.7143	0.0527
C (6)	0.2607	0.8669	0.0402
N (7)	0.2361	0.9381	−0.0659
N (8)	0.2501	0.9688	0.3085
N (9)	0.3911	0.9615	0.6868
C (10)	0.4744	1.0368	0.8682
C (11)	0.3855	1.0960	0.7857
C (12)	0.4360	1.0545	0.5953
C (13)	0.3631	1.1886	0.5557
N (14)	0.0926	0.9770	0.4310
C (15)	0.1288	1.1377	0.4535
N (16)	0.1573	1.1997	0.5644
C (17)	0.1503	1.0989	0.6507
C (18)	0.1151	0.9334	0.6321
C (19)	0.0864	0.8757	0.5175
O (20)	0.0483	0.7180	0.4931
O (21)	0.1365	1.2362	0.3774
W	0.0098	0.9488	0.1899
H (7A)	0.204	0.552	0.771
H (7B)	0.274	0.602	0.887
H (8A)	0.216	0.445	0.427
H (8B)	0.246	0.620	0.376
H (16)	0.191	0.313	0.594
H (17)	0.173	0.164	0.714
H (18)	0.120	0.416	0.717
H (20A)	0.021	0.675	0.411
H (20B)	0.055	0.628	0.581

Results and Discussion

Intermolecular Interactions and Hydrogen-bond Systems. The cytosine and adenine molecules have a strong tendency to bind with acidic organic compounds, especially with molecules containing a carboxylic acid moiety. However, few organic compounds can bind with thymine or uracil under these experimental conditions. In such circumstances, we have tried to un-

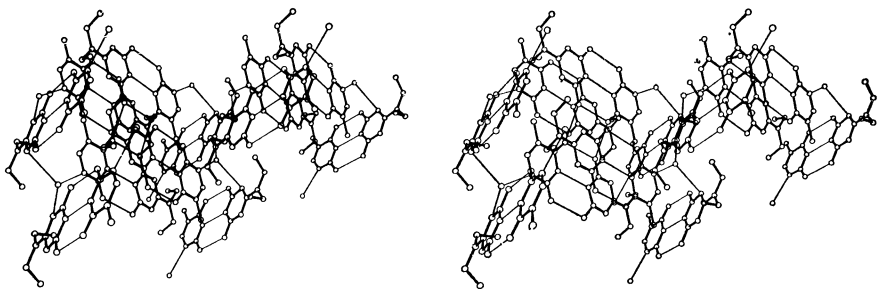


Fig. 1. Stereoscopic view of the packing and hydrogen bonding. Molecular bonds are drawn thick and hydrogen bonds thin.

23) D. Sayre, *Acta Crystallogr.*, **5**, 60 (1952).

Table 3(a). Anisotropic thermal parameters in thymine–*N*,*N*-diethylmelamine complex monohydrate

The thermal parameters are given in the form

$$T = \exp[-(B_{11}h^2+B_{22}k^2+B_{33}l^2+2B_{12}hk+2B_{13}hl+2B_{23}kl)].$$

Atom	B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
N (1)	0.00356	0.01109	0.00485	0.00081	0.00162	0.00021
C (2)	0.00326	0.01237	0.00448	−0.00025	0.00158	0.00043
N (3)	0.00335	0.01149	0.00435	0.00062	0.00155	0.00012
C (4)	0.00318	0.01402	0.00542	0.00077	0.00163	0.00013
N (5)	0.00353	0.01401	0.00439	0.00107	0.00133	−0.00108
C (6)	0.00277	0.01152	0.00450	0.00071	0.00097	0.00034
N (7)	0.00487	0.01332	0.00473	0.00112	0.00231	−0.00020
N (8)	0.00422	0.01475	0.00501	0.00021	0.00209	0.00069
N (9)	0.00649	0.02709	0.00912	−0.00394	0.00313	−0.00202
C (10)	0.00670	0.02456	0.01369	0.00013	0.00180	0.00063
C (11)	0.00672	0.02299	0.00962	−0.00329	0.00235	−0.00127
C (12)	0.01205	0.02420	0.01401	−0.00234	0.00253	−0.00085
C (13)	0.00601	0.04780	0.01340	−0.00072	0.00244	−0.00559
N (14)	0.00249	0.00944	0.00383	−0.00026	0.00119	−0.00029
C (15)	0.00224	0.00996	0.00345	0.00043	0.00101	−0.00010
N (16)	0.00336	0.01152	0.00377	−0.00002	0.00148	−0.00041
C (17)	0.00283	0.01233	0.00347	0.00044	0.00086	−0.00025
C (18)	0.00270	0.01055	0.00384	−0.00035	0.00137	0.00058
C (19)	0.00252	0.01150	0.00360	−0.00005	0.00109	−0.00000
O (20)	0.00423	0.01532	0.00652	0.00088	0.00159	−0.00058
O (21)	0.00338	0.01293	0.00436	−0.00077	0.00133	0.00058
W	0.00362	0.01530	0.00420	−0.00008	0.00076	−0.00032

Table 3(b). Isotropic thermal parameters of some hydrogen in thymine–*N*,*N*-diethylmelamine complex monohydrate

Atom	B_{iso}	Atom	B_{iso}	Atom	B_{iso}
H (7A)	5.64	H (8B)	3.99	H (18)	9.06
H (7B)	4.57	H (16)	2.36	H (20A)	4.76
H (8A)	4.20	H (17)	1.12	H (20B)	5.30

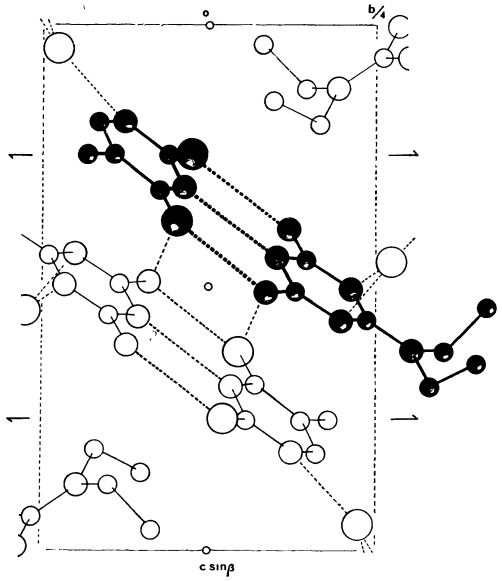


Fig. 2. Molecular packing and hydrogen bonding in thymine–*N*,*N*-diethylmelamine complex monohydrate.

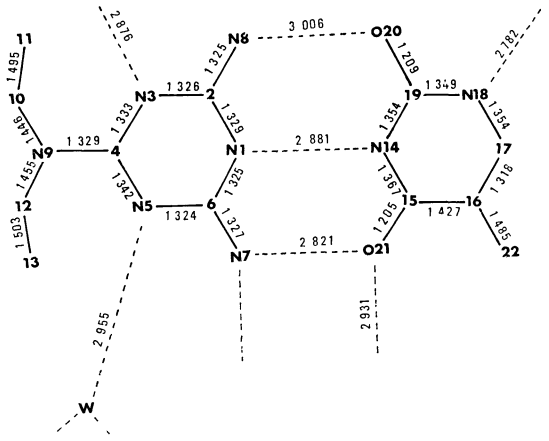
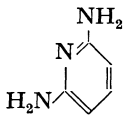
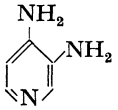
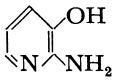
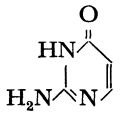
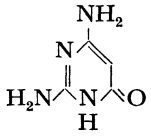
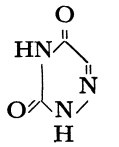
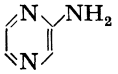
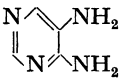
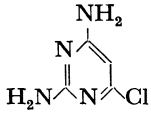
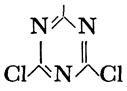
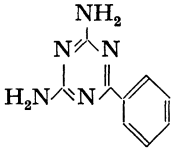
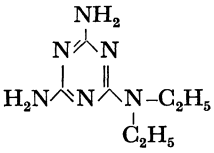
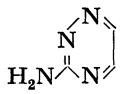
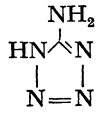
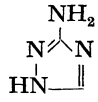
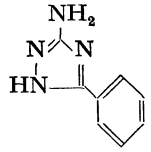
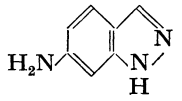
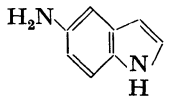
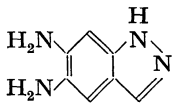
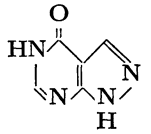
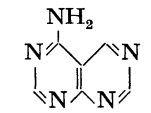
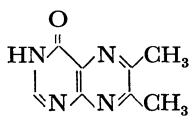
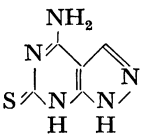


Fig. 3(a). Interatomic distances in thymine–*N*,*N*-diethylmelamine complex monohydrate.

TABLE 4. INTERMOLECULAR COMPLEX FORMATION BETWEEN SOME NITROGEN HETEROCYCLES AND NUCLEOTIDE BASES

	A	C	T	U
 2,6-diamino-pyridine	−	−	−	−
 3,4-diamino-pyridine	−	−	−	−
 2-amino-3-hydroxy pyridine	−	−	−	−
 isocytosine	+	?	−	−
 2,4-diamino-6-hydroxypyrimidine	−	+	+	+
 6-azauracil	+	−	−	−
 aminopyrazine	−	−	−	−
 4,5-diamino-pyrimidine	−	−	−	−
 4-chloro-2,6-diaminopyrimidine	−	−	+	+
 cyanuric chloride	+	+	−	−
 2,4-diamino-6-phenyl-5-triazine	+	+	−	+
 2,4,6-triamino *N*,*N*-diethyl-1,3,5-triazine	+	+	+	+

	A	C	T	U
 3-amino-1,2,4-triazine	−	−	−	−
 5-amino-tetrazole	−	−	−	−
 3-amino-1,2,4-triazole	−	−	−	−
 3-amino-5-phenyl-1,2,4-triazole	−	−	−	−
 6-aminoindazole	−	−	−	−
 5-aminoindole	−	−	−	−
 5,6-diamino-indazole	−	−	−	−
 4-hydroxy pyrazolo-[3,4d]pyrimidine	+	−	−	−
 4-aminopyrimido [4,5d]pyrimidine	−	−	−	−
 2-amino-6,7-dimethyl-4-hydroxypteridine	−	−	−	−
 4-amino-6-mercapto pyrazolo[3,4d]-pyrimidine	−	−	−	−

cover some specific atomic arrangement or any molecule that can associate with thymine or uracil by means of hydrogen bonding. Table 4 shows the materials tested in this experiment. Some molecules with a number of exocyclic amino groups in combination with endocyclic –N= parts, especially in an arrangement which corresponds to a $NH_2\ N\ NH_2$ group, were found to

be able to associate with these molecules. Therefore, combining this finding with the results for cytosine and adenine, it can be said that at least a partially molecular basicity or acidity plays an important role in binding the two components, presumably a so-called acid-and-base interaction. From this point of view, adenine, cytosine, thymine, and uracil can be classified into two groups; cytosine and adenine act as basic nucleotide bases which can bind with acidic components, such as carboxylic acids, while the other two are acidic nucleotide bases which can bind with basic components, such as melamine. However, the binding force is sometimes seriously affected by some other molecular

TABLE 5. ATOMIC DISTANCES OF THREE HYDROGEN BONDS

Components	Distances (Å)			Type	Ref.
9-Ethylguanine–1-methyl-5-bromocytosine	2.91	2.95	2.86	B	2)
Deoxyguanosine–5-bromodeoxycytidine	2.83	2.92	2.78	B	4)
1-Methyl-5-iodouracil–9-ethyl-2,6-diaminopurine	2.92	2.91	2.87	A	5)
1-Methylthymine–9-ethyl-2,6-diaminopurine	2.99	2.99	2.87	A	14)
1-Methyl-5-fluorouracil–9-ethyl-2,6-diaminopurine	2.89	2.88	2.81	A	18)
Isocytosine-isocytosine	2.904	2.908	2.861	B	37)
Cytosineacetic acid–cytosineacetic acid	2.790	2.823	2.790	B	36)
Cytosine–cytosine (trimer complex with resorcylic acid)	2.889	2.815	2.778	B	35)
Thymine–*N*,*N*-diethylmelamine	3.006	2.881	2.821	A	This work

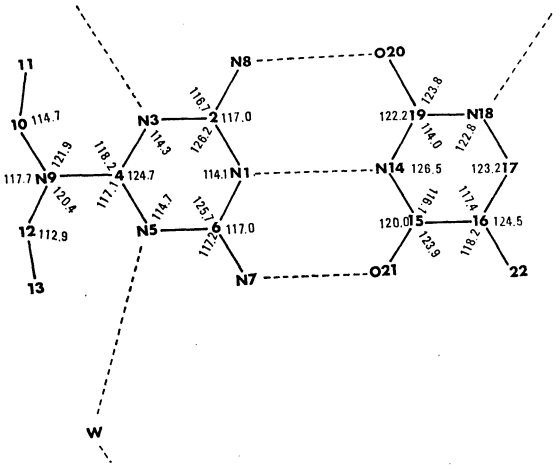
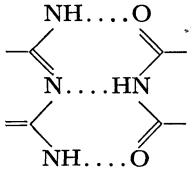


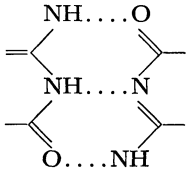
Fig. 3(b). Interbond angles in thymine–*N*,*N*-diethylmelamine complex monohydrate.

feature; thus, not all similar molecular structures can always bind with each other. The local atomic basicity or acidity may be considered to be appropriate for association at each specific site, but steric hindrance may be the determining factor in the formation of molecular complex crystals. Detailed molecular geometry can be evaluated only by X-ray analysis.

Description of the Crystal Structure. The crystal and molecular structure is shown in a stereographic plot in Fig. 1, while a projectional view along the *a*-axis is shown in Fig. 2. The thymine and melamine molecules are associated with each other mainly by means of two NH···O hydrogen bonds (2.821 and 3.006 Å) and one NH···N bond (2.881 Å). This hydrogen-bond system is defined as consisting of the specific nitrogen and oxygen atoms in both molecules, which form the tips of the capital letter "W"; two such molecules bind with each other, with both tops of the letter, thus forming three hydrogen bonds.



I (A)



I (B)

There are several analogous three-hydrogen bond ystems, I(A) and I(B), which are summarized in

TABLE 6. INTERMOLECULAR ATOMIC DISTANCES, LESS THAN 3.7 Å BETWEEN THE MOLECULES

x, *y*, *z*			N (8) ·····O (21)	3.297
N (1) ·····N (14)	2.881		N (9) ·····C (16)	3.541
N (7) ·····O (21)	2.821		N (9) ·····C (17)	3.427
N (8) ·····O (20)	3.006		C (12)·····C (17)	3.584
N (18)·····W (23)	2.782		2−*x*, 1−*y*, 1−*z*	
1+*x*, *y*, *z*			N (7) ·····O (21)	2.931
N (7) ·····N (8)	3.380		−*x*, 1/2+*y*, 1/2−*z*	
O (22)·····O (20)	3.413		W (23)·····C (2)	3.507
C (21)·····N (8)	3.672		W (23)·····N (3)	2.876
C (21)·····O (20)	3.446		W (23)·····N (8)	3.186
1−*x*, −*y*, 1−*z*			W (23)·····C (10)	3.424
C (11)·····C (13)	3.696		W (23)·····C (11)	3.396
1−*x*, 1−*y*, 1−*z*			*x*, 1/2−*y*, 1/2+*z*	
N (1) ·····N (14)	3.394		C (10)·····C (19)	3.711
N (1) ·····C (15)	3.621		C (10)·····O (20)	3.539
N (1) ·····C (21)	3.633		C (11)·····C (19)	3.542
C (2) ·····N (14)	3.636		C (11)·····O (20)	3.506
C (2) ·····C (15)	3.351		1−*x*, 1/2+*y*, 1/2−*z*	
N (3) ·····C (15)	3.295		C (17)·····N (3)	3.658
N (3) ·····C (16)	3.477		C (17)·····C (4)	3.633
N (3) ·····C (21)	3.380		C (21)·····N (3)	3.632
C (4) ·····N (14)	3.668		C (21)·····N (8)	3.634
C (4) ·····C (15)	3.460		W (23)·····N (5)	2.955
C (4) ·····C (16)	3.395		W (23)·····C (6)	3.282
C (4) ·····C (17)	3.526		W (23)·····N (7)	3.272
N (5) ·····N (14)	3.424		W (23)·····C (13)	3.661
N (5) ·····N (18)	3.491		1+*x*, 1/2−*y*, 1/2+*z*	
N (5) ·····C (19)	3.332		C (13)·····N (8)	3.707
C (6) ·····N (14)	3.202		C (13)·····O (20)	3.437
C (6) ·····O (20)	3.626		*x*, 1/2−*y*, −1/2+*z*	
N (7) ·····N (8)	3.624		C (19)·····C (11)	3.542
N (7) ·····N (14)	3.630		O (20)·····C (10)	3.539
N (7) ·····C (19)	3.543		O (20)·····C (11)	3.506
N (7) ·····C (20)	3.318		−1+*x*, 1/2−*y*, −1/2+*z*	
			O (20)·····C (13)	3.437

Table 5. The molecular complexes of similar hydrogen-bonding arrangements have been reported by Sobell *et al.*[14] and Rich *et al.*[18] These facts suggest that an atomic arrangement such as that in I(A) is one of the fundamental combination schemes for binding among molecules. The paired-molecular complex formed in this manner is connected by the remaining

Table 7. Interatomic bond distances and angles in thymine–*N*,*N*-diethylmelamine monohydrate complex

N (1)–C (2)	1.329 Å	C (2)–N (1)–C (6)	114.1°
N (1)–C (6)	1.325	N (3)–C (2)–N (8)	116.7
C (2)–N (3)	1.326	N (1)–C (2)–N (3)	126.2
C (2)–N (8)	1.325	N (1)–C (2)–N (8)	117.0
N (3)–C (4)	1.333	C (2)–N (3)–C (4)	114.3
C (4)–N (5)	1.342	N (3)–C (4)–N (5)	124.7
C (4)–N (9)	1.329	N (3)–C (4)–N (9)	118.2
N (5)–C (6)	1.324	N (5)–C (4)–N (9)	117.1
C (6)–N (7)	1.327	C (4)–N (5)–C (6)	114.7
N (9)–C (10)	1.446	N (1)–C (6)–N (5)	125.7
N (9)–C (12)	1.455	N (5)–C (6)–N (7)	117.2
C (10)–C (11)	1.495	N (1)–C (6)–N (7)	117.0
C (12)–C (13)	1.503	C (4)–N (9)–N (10)	121.9
N (14)–C (15)	1.367	C (4)–N (9)–C (12)	120.4
N (14)–C (19)	1.355	C (10)–N (9)–C (12)	117.7
C (15)–C (16)	1.427	N (9)–C (10)–C (11)	114.7
C (16)–C (17)	1.318	N (9)–C (12)–C (13)	112.9
C (17)–N (18)	1.354	C (15)–N (14)–C (19)	126.5
N (18)–C (19)	1.349	N (14)–C (15)–C (16)	116.1
C (19)–O (20)	1.209	C (16)–C (15)–C (21)	123.9
C (16)–O (22)	1.485	N (14)–C (15)–C (21)	120.0
C (15)–C (21)	1.205	C (15)–C (16)–C (17)	117.4
		C (17)–C (16)–C (21)	124.5
		C (15)–C (16)–C (21)	118.2
		C (16)–C (17)–N (18)	123.2
		C (17)–N (18)–C (19)	122.8
		N (14)–C (19)–N (18)	114.0
		N (18)–C (19)–O (20)	123.8
		N (14)–C (19)–O (20)	122.2

NH···O hydrogen bond at a distance of 2.931 Å, and such units are stacked in such a way that they are parallel to the alternate (0, 1, 1, and 0,1,–1) crystallographic planes. One water molecule is associated by means of hydrogen bonds between these triazine-ring systems in the direction of the *a*-axis. The component molecules are almost coplanar, and the dihedral angle of both least-squares planes is 4.9°. Parallel sheets of the component molecules are separated from each other at an average spacing of 3.35 Å, since the maximum deviation between the atoms in some charge-transfer complexes is 3.26 Å.[24] Therefore, for the complex formation of this series, in some way the molecular stacking or van der Waals forces are second-order in importance. Sakurai *et al.*[25,26] reported some charge-transfer complexes between the molecules of thymine and quinone, which would imply that stacking forces between these hygroscopic moieties play an important role in the complex formation.

In the three-hydrogen-bond system the distances of the two bondings on the outside differ by about 0.2 Å even though *N*,*N*-diethylmelamine is a fairly symmetric molecule. It is hard to explain why hydrogen bonding between the atoms N(8) and O(20) is weak in such symmetrical three-hydrogen bonds. The other intermolecular atomic distances are shown in Table 6.

Table 8. Deviations of the atoms from the least-squares planes (Å)

N,*N*-Diethylmelamine		Thymine	
N (1)	−0.004	N (14)	−0.0141
C (2)	0.004	C (15)	−0.002
N (3)	−0.016	C (16)	−0.004
C (4)	0.000	C (17)	−0.012
N (5)	0.033	N (18)	−0.014
C (6)	−0.004	C (19)	0.003
N (8)	−0.009	O (20)	0.010
N (9)	−0.008	C (21)	0.024

Description of the Molecular Structure. The intramolecular atomic bond lengths and angles are shown in Figs. 3(a) and 3(b) and in Table 7. As was discussed earlier, the existence of a hydrogen atom attached to nitrogen may be indicated by its bond angle, in the manner of Singh;[27] the usual –NH– angle is 125°, while for –N= it is 116°. In this complex both components agree well with this hypothesis, since the mean C–N–C bond angle of melamine is 114.4° and that of thymine is 124.7°. The difference synthesis at the *R*-factor, 0.14, indicated that there are no protons around the triazine nitrogen hetero-atoms, while there are two protons attached to N(14) and N(18) in the thymine molecule. The bond lengths of the endocyclic part of thymine differ significantly from those found in the literature; *i.e.* the C=O distances of 1.209 and 1.205 Å are slightly shorter than those found in thymine monohydrate[28] (1.234 and 1.231), methylthymine[29] (1.237 and 1.214 Å) and uracil (1.230 and 1.241 Å). The mean bond length of all the N–C distances in *N*,*N*-diethylmelamine is 1.330 Å, and that in thymine is 1.356 Å. These values are slightly shorter than the expected values between 1.37 and 1.38 Å observed for uracil,[30] thymine monohydrate,[28] barbiturate derivatives,[31,32] but they are compatible with those in melamine[33] and purine,[34] with values from 1.34 to 1.35 Å. The deviations of atoms from each molecular plane are listed in Table 8.

24) A. W. Hanson, *ibid.*, **19**, 610 (1965).
25) T. Sakurai and M. Okunuki, *ibid.*, **B27**, 1445 (1971).
26) T. Sakurai and H. Tagawa, *ibid.*, **B27**, 1453 (1971).
27) C. Singh, *ibid.*, **19**, 861 (1965).
28) R. Gerdil, *ibid.*, **14**, 333 (1961).
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30) G. S. Parry, *ibid.*, **7**, 313 (1954).
31) W. Bolton, *ibid.*. **16**, 166 (1963).
32) D. Mootz and G. A. Jeffery, *ibid.*, **19** 717 (1965).
33) E. W. Hughes, *J. Amer. Chem. Soc.*, **63**, 1737 (1941).
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35) to be published.
36) R. E. Marsh, R. Biersted and E. L. Eichhorn, *Acta Crystallogr.*, **15**, 310 (1962).
37) B. D. Sharma and J. F. McConnel, *ibid.*, **19**, 797 (1965).