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The Crystal Structure of Nickel(II)-Dehydroacetic Acid Monoimide Complex

BY SETSUO KIRYU
Faculty of Pharmaceutical Science, Kyushu University, Katakasu, Fukuoka, Japan

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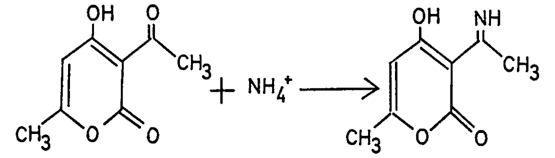
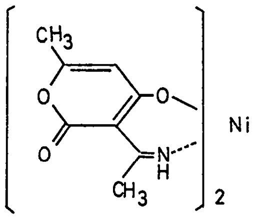
The crystal structure of nickel(II)-dehydroacetic acid monoimide complex $(C_8H_8O_3N)_2Ni$ has been determined by three-dimensional X-ray analysis. The unit-cell dimensions are: $a=18{\cdot}63$, $b=21{\cdot}64$, $c=3{\cdot}84$ Å with $Z=4$. Space group $P2_12_12_1$. The compound has a chelate structure with a square planar configuration.

Introduction

Dehydroacetic acid, which is one of the most widely used fungicides and bactericides for foods, was found to react easily with ammonium ions, primary amines and other substances containing primary amino groups, even in mild conditions such as *in vivo*.



Dehydroacetic acid   Dehydroacetic acid monoimide



Nickel(II)-DHA imide complex

These reactions were fully investigated because of their chemical and biological interest (*e.g.* Iguchi, Hisatsune, Himeno & Muraoka, 1959), and the structures of the products were studied in detail by infrared

Table 1. *Positional and thermal parameters and their standard deviations*

Standard deviations and final shifts for positional parameters are expressed in 10^{-4} Å.

	x	y	z	B	$\sigma(x)$	$\sigma(y)$	$\sigma(z)$	$\sigma(B)$
Ni	0·3640	0·1346	0·1993	*	15	16	20	*
O(1)	0·2736	0·1554	0·3373	2·15	66	67	81	0·11
O(2)	0·4556	0·1128	0·0587	2·31	67	71	89	0·13
O(3)	0·1666	0·3192	0·3581	2·84	80	76	88	0·14
O(4)	0·5538	−0·0521	−0·0293	2·41	70	75	87	0·13
O(5)	0·2571	0·3731	0·1708	3·93	85	95	102	0·17
O(6)	0·4620	−0·1067	0·1461	2·94	73	76	95	0·14
N(1)	0·3850	0·2135	0·0530	2·19	81	82	98	0·14
N(2)	0·3427	0·0565	0·3473	2·35	81	83	98	0·14
C(1)	0·2439	0·2096	0·3270	2·23	91	101	122	0·16
C(2)	0·4832	0·0583	0·0458	2·24	99	98	123	0·17
C(3)	0·1706	0·2116	0·4573	2·33	102	104	125	0·17
C(4)	0·5548	0·0551	−0·0829	2·38	100	104	127	0·17
C(5)	0·1354	0·2642	0·4649	2·38	103	98	120	0·16
C(6)	0·5891	0·0011	−0·1207	2·09	93	97	114	0·15
C(7)	0·2375	0·3218	0·2473	2·72	99	101	139	0·18
C(8)	0·4854	−0·0547	0·0971	2·44	101	103	121	0·18
C(9)	0·2770	0·2640	0·2110	2·10	89	92	120	0·18
C(10)	0·4476	0·0029	0·1563	1·95	87	92	118	0·15
C(11)	0·0592	0·2736	0·5875	2·95	113	114	142	0·20
C(12)	0·6622	−0·0084	−0·2467	2·59	102	101	138	0·17
C(13)	0·3485	0·2652	0·0738	2·01	89	94	115	0·16
C(14)	0·3786	0·0035	0·3095	1·97	86	90	120	0·15
C(15)	0·3837	0·3225	−·0600	2·30	99	100	120	0·17
C(16)	0·3431	−0·0540	0·4445	2·36	101	103	121	0·17

Shifts during final cycle	x	y	z	B
Mean	5·2	4·5	17·3	0·006
Max.	14·9	15·1	47·6	0·013

*	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
	0·00216	0·00146	0·07196	0·00009	0·00288	−0·00035
e.s.d.'s	0·00004	0·00003	0·00142	0·00006	0·00043	0·00037
Final shifts	0·00000	0·00000	0·00052	0·00000	0·00004	−0·00002

absorption and proton magnetic resonance measurements (Edwards, Page & Pianka, 1964). The resultant dehydroacetic acid monoimide (hereafter referred to as DHA imide), or 3-(1-iminoethyl)-4-hydroxy-6-methyl-2-pyrone, forms metal complexes with some metal ions such as copper(II) ion, which were expected to be chelate compounds (Iguchi & Goto, 1959). The structure determination of these complexes seems to be interesting in connexion with above investigations as well as in coordination chemistry. In view of these circumstances, an X-ray diffraction investigation of nickel(II)-DHA imide complex $(C_8H_8O_3N)_2Ni$ was undertaken.

Experimental

Nickel(II)-DHA imide complex was prepared by mixing aqueous solutions of one mole of nickel acetate and two moles of DHA imide; DHA imide was obtained as a white precipitate by keeping a solution of dehydroacetic acid mixed with an excess of ammonia for an hour at room temperature. Single crystals were prepared by slow cooling of benzene solution in a Dewar vessel and thin orange needles elongated along the *c* axis were obtained. Although well developed crystals were also obtained in solvated form from solutions of some other solvents, *e.g.* dioxane or dichloro-

Table 2. *Observed and calculated structure factors*

K	Fo	Fc	K	Fo	Fc	K	Fo	Fc	K	Fo	Fc	K	Fo	Fc	K	Fo	Fc	K	Fo	Fc	K	Fo	Fc
H,L=	0	0	3	10.8	9.8	20	31.4	32.0	12	31.1	29.9	14	6.5	8.6	2	11.2	11.9	16	19.0	18.7	4	28.5	22.7
2	11.3	9.8	4	36.4	29.7	21	0.0	2.8	13	0.0	4.8	15	6.3	6.7	3	20.8	23.4	17	13.5	17.9	5	15.3	10.4
4	20.9	13.0	5	3.5	1.3	22	0.0	5.7	14	13.6	13.3	16	19.0	19.6	4	0.0	3.7	18	35.5	29.5	6	42.3	39.1
6	35.0	34.1	6	60.8	52.2	23	0.0	9.2	15	0.0	0.8	17	5.7	5.9	5	12.0	12.2	19	0.0	7.2	7	42.1	39.3
8	55.0	65.7	7	59.5	49.7	24	13.5	16.0	16	28.2	27.9	18	10.6	10.1	6	11.7	13.1	20	30.5	28.8	8	29.8	25.9
10	49.2	44.0	8	58.8	63.0	25	0.0	1.3	17	17.8	18.1	19	0.0	3.9	7	23.3	21.4	21	22.5	23.5	9	39.3	34.8
12	68.4	58.1	9	62.4	53.7	26	0.0	0.7	18	6.5	8.1	20	13.7	12.2	8	6.0	6.2	22	7.6	7.1	10	56.0	48.5
14	49.7	52.7	10	4.8	7.3	H,L=	8	0	19	0.0	0.5	H,L=	16	0	9	5.7	5.3	23	23.8	25.8	11	53.5	52.4
16	76.9	72.8	11	20.3	19.1	0	66.3	84.2	20	8.4	8.9	0	9.6	8.5	10	5.3	5.9	24	0.0	1.8	12	14.0	12.4
18	50.5	47.3	12	5.3	11.1	1	35.8	22.1	21	0.0	6.5	1	19.2	16.4	11	11.0	8.3	25	11.7	12.0	13	14.6	14.9
20	13.8	12.9	13	0.0	7.6	2	67.3	56.0	22	0.0	4.4	2	6.8	7.6	H,L=	22	0	26	0.0	3.0	14	21.5	20.3
22	43.9	43.9	14	49.4	48.2	3	50.4	40.4	23	0.0	1.4	3	6.8	7.9	0	11.1	12.2	H,L=	3	1	15	43.0	39.0
24	0.0	0.5	15	20.5	19.7	4	70.1	58.7	24	12.4	12.4	4	6.8	8.6	1	0.0	0.0	0	36.7	39.3	16	11.3	13.7
26	23.5	21.5	16	25.7	29.3	5	0.0	6.0	H,L=	12	0	5	24.7	24.7	2	0.0	2.5	1	106.7	100.6	17	0.0	5.8
H,L=	1	0	17	0.0	7.9	6	41.9	34.5	0	34.1	33.3	6	6.3	9.1	3	0.0	2.8	2	53.9	54.9	18	24.6	24.6
1	66.1	61.9	18	28.0	28.6	7	14.1	13.9	1	33.2	31.5	7	0.0	3.0	4	24.0	20.6	3	60.1	45.7	19	36.4	34.4
2	91.6	82.2	19	0.0	3.6	8	52.1	45.0	2	0.0	1.8	8	0.0	6.7	5	0.0	0.0	4	62.7	55.9	20	7.9	10.7
3	29.7	34.9	20	11.8	13.0	9	37.1	32.7	3	46.0	40.5	9	0.0	7.1	6	0.0	5.2	5	18.6	13.8	21	10.8	10.6
4	49.6	41.2	21	0.0	2.5	10	37.2	31.9	4	44.2	42.1	10	0.0	0.8	7	0.0	4.3	6	13.4	6.5	22	10.1	9.6
5	68.0	58.5	22	37.7	39.2	11	10.0	9.0	5	34.7	36.4	11	0.0	3.9	8	31.1	28.0	7	46.9	37.6	23	13.7	20.7
6	40.6	45.2	23	0.0	10.0	12	24.9	25.3	6	8.5	6.8	12	15.6	17.6	H,L=	23	0	8	50.2	45.5	24	8.4	10.8
7	21.1	31.7	24	0.0	7.1	13	26.6	25.4	7	22.2	22.1	13	19.2	20.9	1	0.0	4.5	9	58.1	52.6	25	6.2	7.4
8	0.0	7.3	25	0.0	4.5	14	39.8	33.2	8	54.8	51.4	14	16.4	14.8	2	12.0	11.5	10	87.5	79.8	26	9.3	9.4
9	0.0	5.1	26	11.9	13.3	15	0.0	1.4	9	55.9	51.3	15	0.0	0.8	3	9.5	10.9	11	6.1	6.7	H,L=	7	1
10	58.8	50.6	H,L=	5	0	16	17.9	16.0	10	6.5	9.4	16	0.0	2.6	4	0.0	2.6	12	52.0	47.4	0	48.0	52.5
11	71.8	66.7	1	65.0	62.9	17	29.9	29.0	11	0.0	3.0	17	14.7	16.3	5	4.4	5.5	13	30.0	27.7	1	54.3	50.7
12	37.5	36.3	2	25.1	25.9	18	35.0	31.7	12	9.5	11.5	18	10.1	8.4	6	12.9	11.9	14	46.3	40.7	2	43.7	37.1
13	0.0	1.0	3	18.2	16.3	19	0.0	1.7	13	20.5	21.6	19	8.9	6.6	7	22.4	22.7	15	21.3	18.5	3	42.8	41.7
14	20.6	17.6	4	7.2	3.2	20	18.6	16.3	14	0.0	2.9	20	8.4	7.2	H,L=	0	1	16	15.6	12.2	4	45.9	45.7
15	58.6	59.7	5	36.9	36.0	21	0.0	6.9	15	0.0	2.2	H,L=	17	0	1	8.1	7.8	17	57.1	56.5	5	27.8	28.5
16	30.8	29.5	6	0.0	2.5	22	15.6	15.8	16	0.0	2.0	1	15.3	15.0	2	61.4	63.5	18	24.5	26.5	6	8.0	8.9
17	51.9	49.8	7	0.0	9.5	23	0.0	8.3	17	16.2	17.4	2	0.0	6.2	3	48.0	38.0	19	18.3	15.3	7	30.0	29.5
18	22.2	21.6	8	50.8	39.6	24	0.0	6.2	18	0.0	7.2	3	6.8	8.5	4	23.5	17.9	20	0.0	5.0	8	32.5	29.0
19	38.7	30.7	9	45.8	35.8	25	8.7	8.0	19	8.5	10.3	4	0.0	4.3	5	42.9	35.8	21	19.4	18.4	9	27.9	27.9
20	11.9	10.8	10	88.8	78.8	26	15.1	16.0	20	0.0	1.7	5	0.0	5.3	6	40.1	40.4	22	15.1	15.2	10	35.4	34.8
21	6.7	8.2	11	110.6	111.4	H,L=	9	0	21	0.0	3.2	6	9.6	10.9	7	34.0	27.5	23	17.4	18.0	11	20.9	23.8
22	0.0	5.6	12	31.1	29.2	1	104.9	91.7	22	0.0	8.1	7	11.7	13.7	8	56.2	42.1	24	0.0	2.3	12	46.4	45.3
23	8.8	12.3	13	0.0	6.3	2	9.3	7.7	23	0.0	4.8	8	0.0	6.8	9	31.5	24.6	25	5.7	8.8	13	13.0	14.8
24	0.0	12.4	14	0.0	8.5	3	19.6	21.4	H,L=	13	0	9	0.0	4.7	10	60.0	55.3	26	11.7	10.7	14	29.0	30.1
25	0.0	10.3	15	71.8	65.8	4	0.0	0.1	1	30.8	28.9	10	0.0	5.8	11	14.6	12.9	H,L=	4	1	15	17.8	16.9
26	0.0	2.6	16	21.6	21.6	5	37.5	35.3	2	17.1	16.0	11	17.0	18.6	12	50.6	47.3	0	11.3	0.7	16	29.2	31.3
H,L=	2	0	17	0.0	5.4	6	0.0	2.3	3	44.8	42.0	12	0.0	5.1	13	20.1	15.8	1	40.1	37.0	17	25.9	27.8
0	20.6	18.4	18	0.0	2.9	7	44.0	43.8	4	0.0	7.5	13	0.0	4.2	14	23.4	18.8	2	92.6	99.1	18	21.6	20.2
1	81.5	87.2	19	23.8	22.3	8	13.3	10.6	5	17.6	16.3	14	16.3	15.8	15	0.0	9.0	3	37.1	37.7	19	8.0	7.8
2	139.5	151.5	20	0.0	6.2	9	17.8	16.5	6	15.5	14.5	15	21.8	21.5	16	64.3	60.8	4	32.3	31.8	20	0.0	1.4
3	79.2	79.8	21	11.5	12.9	10	14.3	14.9	7	107.7	104.5	16	8.7	9.2	17	25.1	21.6	5	21.7	22.2	21	10.5	11.4
4	21.4	20.6	22	0.0	0.0	11	38.3	38.2	8	26.9	27.0	17	0.0	0.9	18	8.1	6.7	6	64.0	64.5	22	18.5	20.2
5	83.0	78.4	23	31.7	32.6	12	20.7	16.4	9	32.5	28.3	18	0.0	0.7	19	0.0	6.3	7	25.6	23.3	23	0.0	4.5
6	81.9	69.9	24	7.7	10.4	13	0.0	4.3	10	35.6	34.1	19	19.0	15.6	20	0.0	1.4	8	33.8	30.5	24	0.0	3.1
7	16.1	21.6	25	19.9	20.8	14	6.6	6.6	11	19.2	17.5	H,L=	18	0	21	7.9	9.0	9	47.3	38.5	25	10.0	9.2
8	13.2	13.1	26	7.1	7.7	15	46.5	52.2	12	0.0	8.4	0	9.6	10.4	22	0.0	5.9	10	60.4	55.7	H,L=	8	1
9	105.4	91.0	27	18.6	17.7	16	21.6	21.7	13	0.0	0.2	1	0.0	0.6	23	0.0	1.3	11	8.9	9.7	0	64.0	52.8
10	28.1	16.9	H,L=	6	0	17	0.0	3.6	14	0.0	2.0	2	0.0	3.7	24	18.8	21.5	12	22.1	21.5	1	70.8	70.5
11	8.4	8.9	0	31.2	30.2	18	21.5	21.3	15	33.1	33.7	3	13.5	13.7	25	0.0	4.7	13	18.5	17.2	2	49.3	49.5
12	9.7	10.5	1	78.8	71.7	19	13.3	12.8	16	0.0	5.5	4	34.4	34.9	26	0.0	3.9	14	40.1	36.9	3	9.5	10.4
13	62.3	57.9	2	38.0	25.9	20	9.1	9.3	17	6.3	6.2	5	17.7	17.1	H,L=	1	1	15	13.2	13.0	4	23.3	25.5
14	15.2	13.3	3	11.9	16.4	21	8.6	10.1	18	0.0	1.7	6	0.0	0.3	0	93.5	111.8	16	22.3	21.8	5	29.7	29.8
15	13.5	12.5	4	6.7	6.1	22	0.0	0.8	19	26.9	26.2	7	0.0	0.4	1	148.7	187.7	17	11.4	10.2	6	35.1	32.5
16	12.6	12.6	5	54.5	55.5	23	12.6	14.9	20	0.0	5.7	8	31.8	31.9	2	9.3	10.5	18	34.7	29.8	7	10.7	11.9
17	40.3	41.5	6	26.1	28.2	24	0.0	3.2	21	0.0	6.5	9	0.0	7.0	3	33.9	29.8	19	18.3	17.4	8	42.9	44.2
18	37.8	35.4	7	0.0	6.7	25	11.9	11.3	22	0.0	2.6	10	0.0	6.5	4	63.4	56.5	20	22.8	23.5	9	9.4	13.6
19	0.0	2.6	8	32.2	24.5	H,L=	10	0	23	18.9	19.1	11	0.0	0.3	5	85.7	78.9	21	0.0	5.7	10	24.1	24.2
20	24.7	20.9	9	17.0	16.5	0	166.7	153.8	H,L=	14	0	12	14.2	15.3	6	18.6	15.2	22	10.5	10.2	11	31.4	27.7
21	13.5	15.3	10	41.0	33.4	1	83.6	83.1	0	12.6	12.8	13	0.0	5.3	7	0.0	5.1	23	0.0	5.3	12	22.4	20.6
22	0.0	0.2	11	32.5	28.6	2	0.0	0.9	1	22.9	20.6	14	9.0	8.0	8	70.1	61.7	24	24.6	27.0	13	26.7	24.7
23	12.5	14.1	12	18.8	18.4	3	7.2	7.1	2	0.0	1.6	15	0.0	3.7	9	58.4	52.1	25	11.8	11.9	14	15.8	18.7
24	10.0	7.6	13	44.7	42.9	4	41.2	38.6	3	16.9	16.1	16	0.0	5.7	10	21.1	17.8	26	0.0	5.1	15	22.8	20.8
25	20.4	19.5	14	0.0	1.5	5	58.0	55.8	4	0.0	10.4	17	0.0	4.9	11	24.0	22.4	H,L=	5	1	16	27.1	28.2
26	0.0	3.3	15	14.3	12.8	6	10.8	11.3	5	30.6	28.9	18	22.2	23.0	12	34.9	32.9	0	5.7	7.3	17	16.4	16.5
H,L=	3	0	16	20.9	20.7	7	0.0	0.4	6	0.0	3.7	H,L=	19	0	13	53.6	49.5	1	31.6	32.4	18	0.0	3.2
1	59.9	61.5	17	27.9	28.0	8	38.1	35.2	7	17.6	16.4	1	0.0	2.4	14	30.1	27.6	2	29.7	29.8	19	7.9	8.8
2	89.4	101.1	18	0.0	8.1	9	37.9	35.2	8	26.0	27.2	2	18.5	17.2	15	34.8	31.7	3	26.7	33.7	20	7.6	11.0
3	47.4	51.8	19	0.0	3.0	10	23.6	23.8	9	0.0	5.5	3	18.5	19.4	16	13.3	13.9	4	20.2	24.1	21	16.2	17.4
4	24.4	22.3	20	0.0	0.0	11	0.0	0.2	10	53.0	52.4	4	0.0	4.7	17	27.6	24.7	5	20.7	24.2	22	0.0	4.3
5	83.7	76.1	21	24.6	28.0	12	39.7	37.8	11	21.7	21.9	5	14.3	14.8	18	8.1	7.0	6	48.8	41.2	23	0.0	5.7
6	80.7	71.7	22	0.0	8.4	13	38.5	36.5	12	0.0	3.5	6	12.7	13.2	19	35.7	32.9	7	36.4	35.9	24	12.9	12.7
7	75.1	72.5	23	15.5	15.7	14	0.0	7.3	13	16.7	14.5	7	0.0	7.2	20	8.1	9.8	8	26.1	24.7	25	11.5	10.4
8	53.6	53.8	24	11.9	12.2	15	15.2	11.2	14	17.8	18.4	8	16.1	16.1	21	19.5	17.7	9	31.1	31.2	H,L=	9	1
9	89.3	69.5	25	19.1	18.6	16	30.7	27.2	15	6.5	7.4	9	0.0	3.3	22	10.8	11.8	10	16.4	14.6	0	32.7	30.8
10	8.0	9.7	26	14.2	14.6	17	18.1	17.4	16	6.3	10.5	10	9.9	9.6	23	0.0	9.7	11	6.5	4.2	1	69.4	70.7
11	14.8	12.7	H,L=	7	0	18	6.7	7.8	17	8.6	10.0	11	0.0	1.1	24	6.6	7.4	12	18.1	16.5	2	11.7	14.0
12	21.6	24.1	1	34.4	31.7	19	0.0	0.3	18	17.2	19.3	12	15.7	13.4	25	14.5	11.6	13	39.2	38.0	3	16.8	21.0
13	28.2	27.8	2	53.7	59.8	20	0.0	0.2	19	0.0	2.3	13	15.7	14.5	26	6.1	6.3	14	19.7	18.4	4	0.0	3.5
14	36.9	34.9	3	0.0	7.0	21	16.6	16.2	20	0.0	2.4	H,L=	20	0	H,L=	2	1	15	7.7	9.5	5	49.4	51.5
15	33.5	33.2	4	0.0	2.2	22	13.2	15.7	21	11.5	10.1	0	0.0	1.7	0	90.4	93.8	16	17.8	17.0	6	28.5	23.4
16	19.1	21.9	5	64.8	54.8	23	0.0	5.3	22	15.4	14.6	1	6.2	7.7	1	152.8	159.2	17	21.5	22.5	7	26.3	28.7
17	13.1	8.1	6	51.6	51.3	24	0.0	3.8	H,L=	15	0	2	12.3	13.7	2	147.5	133.3	18	16.4	16.1	8	9.5	10.1
18	0.0	4.8	7	0.0	2.1	25	8.3	7.8	1	18.7	16.9	3	15.0	17.1	3	31.3	33.8	19	11.5	11.5	9	44.9	46.7
19	24.7	23.4	8	9.8	12.0	H,L=	11	0	2	35.1	31.9	4	0.0	4.5	4	27.7	25.2	20	13.9	14.4	10	19.1	19.2
20	26.5	27.9	9	16.2	13.3	1	44.6	38.5	3	0.0	9.0	5	23.1	24.2	5	27.1	21.6	21	19.0	21.3	11	14.9	11.8
21	0.0	1.2	10	52.3	45.8	2	65.9	63.8	4	0.0	3.1	6	10.1	11.5	6	0.0	6.9	22	10.4	11.8	12	10.8	11.3
22	0.0	4.3	11	20.2	21.5	3	0.0	5.4	5	28.5	27.8	7	9.9	8.9	7	102.0	91.5	23	15.2	15.9	13	46.5	43.5
23	12.4	11.7	12	37.5	33.8	4	28.1	26.0	6	25.3	25.3	8	0.0	2.1	8	53.0	43.4	24	7.5	7.3	14	16.1	15.6
24	17.2	19.3	13	27.2	25.1	5	5.6	6.8	7	0.0	8.3	9	17.8	19.4	9	25.1	22.0	25	6.5	5.6	15	0.0	4.5
25	0.0	6.7	14	47.7	41.2	6	50.0	48.8	8	11.8	9.3	10	0.0	0.5	10	31.3	25.3	26	9.0	7.4	16	8.2	6.6
26	5.5	4.6	15	14.7	13.2	7	0.0	7.2	9	73.3	66.2	11	0.0	0.9	11	19.2	20.4	H,L=	6	1	17	29.4	29.1
H,L=	4	0	16	28.4	28.1	8	31.8	28.7	10	54.6	51.6	12	7.8	7.2	12	18.2	15.8	0	19.6	19.2	18	8.0	7.8
0	93.9	83.5	17	32.0	29.7	9	12.3	13.9	11	32.1	28.8	13	22.2	19.5	13	0.0	5.7	1	23.0	23.5	19	7.7	9.0
1	14.7	15.2	18	19.4	19.1	10	35.6	36.9	12	24.4	24.3	H,L=	21	0	14	0.0	2.8	2	47.7	49.4	20	0.0	4.9
2	17.4	17.8	19	0.0	7.8	11	0.0	3.3	13	16.3	17.0	1	8.0	8.7	15	62.3	58.9	3	42.9	39.1	21	18.5	19.7

Table 2 (*cont.*)

K	Fo	Fc
22	0.0	2.1
23	5.7	6.9
24	4.8	3.6
25	11.2	12.3
H,L=	10	1
0	42.8	46.2
1	30.5	33.8
2	28.1	24.6
3	53.0	52.8
4	0.0	8.7
5	28.0	30.1
6	24.3	23.7
7	40.4	42.3
8	14.2	11.1
9	24.2	22.4
10	31.8	30.7
11	27.8	25.9
12	26.1	26.7
13	0.0	4.7
14	8.1	11.2
15	14.2	17.1
16	28.3	28.8
17	16.1	15.0
18	7.8	7.7
19	0.0	5.6
20	0.0	3.3
21	0.0	2.5
22	0.0	5.4
23	6.4	8.4
24	8.9	9.2
H,L=	11	1
0	92.4	90.5
1	29.0	31.7
2	0.0	4.8
3	11.7	11.4
4	40.0	43.1
5	31.2	32.5
6	24.6	26.8
7	14.5	16.2
8	72.4	72.4
9	10.7	11.8
10	17.3	19.4
11	0.0	1.6
12	18.0	16.2
13	27.0	26.8
14	23.2	27.2
15	20.0	20.0
16	19.8	20.1
17	0.0	1.9
18	7.6	11.5
19	0.0	2.2
20	0.0	7.6
21	0.0	3.2
22	16.7	16.8
23	0.0	4.1
H,L=	12	1
0	12.2	10.2
1	33.1	35.9
2	47.2	47.1
3	22.6	23.4
4	19.1	21.6
5	22.0	21.3
6	30.8	31.7
7	40.2	39.9
8	58.3	57.5
9	20.8	18.9
10	22.6	19.1
11	19.8	19.5
12	11.5	12.5
13	8.2	8.4
14	16.3	14.6
15	19.8	21.3
16	7.9	9.6
17	0.0	5.7
18	22.0	22.4
19	0.0	6.4
20	18.1	20.6
21	7.0	5.6
22	10.5	9.9
H,L=	13	1
0	0.0	1.7
1	24.7	26.2
2	22.4	23.3
3	30.1	28.1
4	10.7	11.5
5	23.0	25.0
6	15.5	15.9
7	22.2	22.9
8	11.2	12.1
9	42.7	43.0
10	50.2	48.9
11	16.3	[illegible]
12	16.4	[illegible]
13	20.0	19.9
14	0.0	5.4
15	7.9	9.9
16	0.0	4.4
17	25.5	21.0
18	0.0	6.0
19	13.7	11.1
20	0.0	3.5
21	0.0	5.0
H,L=	14	1
0	0.0	6.7
1	0.0	7.0
2	38.2	39.1
3	11.1	13.6
4	7.9	8.5
5	11.2	13.4
6	39.3	41.8
7	14.0	15.8
8	35.5	31.7
9	31.7	29.8
10	16.4	16.4
11	21.6	20.2
12	8.1	10.4
13	0.0	6.9
14	13.6	15.9
15	10.8	13.1
16	10.3	11.7
17	6.9	7.8
18	7.9	6.7
19	12.5	11.6
20	11.1	11.4
H,L=	15	1
0	27.8	31.7
1	8.0	13.5
2	0.0	4.3
3	0.0	3.6
4	8.1	11.8
5	0.0	8.2
6	29.4	30.3
7	11.5	13.8
8	16.4	20.6
9	27.1	28.4
10	8.1	10.5
11	11.4	15.7
12	7.9	9.9
13	0.0	3.5
14	19.9	20.8
15	7.2	10.2
16	15.3	16.7
17	0.0	1.6
18	16.1	13.7
19	6.4	5.8
20	7.6	7.5
H,L=	16	1
0	0.0	2.5
1	0.0	4.1
2	16.3	18.5
3	16.3	17.6
4	11.6	11.4
5	25.9	27.5
6	8.1	9.6
7	11.5	12.3
8	8.0	10.1
9	8.0	9.5
10	37.3	38.5
11	33.2	34.9
12	0.0	4.1
13	0.0	0.6
14	0.0	4.3
15	16.5	18.6
16	8.9	8.8
17	10.0	10.0
18	8.9	6.0
19	15.0	12.9
H,L=	17	1
0	8.1	9.7
1	8.1	11.2
2	8.1	9.7
3	16.3	18.6
4	14.1	17.7
5	21.4	22.9
6	0.0	6.8
7	22.6	25.8
8	11.1	13.3
9	17.4	17.8
10	7.6	11.0
11	18.2	19.6
12	17.6	18.2
13	13.7	16.4
14	11.3	10.9
15	0.0	5.7
15	0.0	4.0
17	18.3	15.1
18	5.0	6.7
H,L=	18	1
0	8.0	12.1
1	0.0	4.5
2	15.9	15.4
	7.9	11.9
	7.9	10.0
5	7.8	9.5
6	13.4	14.2
7	10.8	8.7
8	18.4	13.3
9	7.3	10.8
10	14.3	15.0
11	0.0	3.9
12	13.1	13.7
13	8.8	9.8
14	18.8	15.9
15	0.0	2.4
16	5.7	8.4
H,L=	19	1
0	13.2	12.4
1	0.0	2.8
2	0.0	6.7
3	7.5	8.8
4	22.5	23.6
5	0.0	5.7
6	10.3	12.3
7	0.0	3.7
8	17.1	18.2
9	9.5	8.5
10	9.2	9.7
11	7.5	6.2
12	10.1	9.8
13	11.4	11.2
14	8.4	7.8
H,L=	20	1
0	0.0	2.3
1	0.0	6.8
2	7.0	5.3
3	19.8	20.4
4	11.9	13.5
5	11.8	13.5
6	9.4	10.5
7	17.1	18.5
8	10.8	12.5
9	8.5	10.2
10	8.0	7.7
11	13.0	11.2
12	4.8	7.6
H,L	21	1
0	0.0	7.7
1	15.6	17.0
2	0.0	1.6
3	0.0	2.8
4	13.7	15.3
5	12.0	12.5
6	9.2	7.5
7	13.7	11.3
8	14.6	12.1
9	17.4	15.2
10	0.0	2.6
11	5.0	7.6
H,L=	22	1
0	0.0	0.8
1	0.0	3.7
2	14.7	14.3
3	6.5	7.6
4	6.3	5.7
5	6.1	7.1
6	21.8	19.1
7	0.0	6.2
8	14.9	13.2
9	9.5	9.4
H,L=	23	1
0	0.0	7.9
1	8.7	9.7
2	4.9	6.4
3	4.7	5.9
4	9.9	9.9
H,L=	0	2
0	18.2	21.5
1	0.0	6.9
2	0.0	2.2
3	38.0	44.1
4	46.8	52.7
5	32.8	54.6
6	63.8	66.5
7	34.7	30.2
8	68.4	67.3
9	11.9	9.9
10	7.3	5.6
11	15.5	15.1
12	5.1	9.5
13	34.1	29.8
14	28.0	24.3
15	9.1	6.2
16	18.5	16.5
17	0.0	1.6
18	37.2	31.2
19	24.1	21.1
20	0.0	7.4
21	0.0	1.8
22	15.5	16.3
23	0.0	6.9
24	7.4	.6
25	14.0	12.6
H,L=	1	2
0	10.1	11.6
1	18.5	26.4
2	19.9	25.3
3	15.5	18.9
4	17.4	20.2
5	27.5	29.1
6	20.6	20.4
7	34.0	31.4
8	72.5	69.7
9	21.9	17.9
10	0.0	2.9
11	33.9	28.1
12	24.5	[illegible]
13	20.7	16.7
14	25.0	21.2
15	18.2	14.6
16	18.5	17.7
17	16.2	16.8
18	13.1	11.8
19	9.1	11.0
20	17.6	17.8
21	16.6	14.9
22	10.9	12.0
23	0.0	4.4
24	16.5	13.6
25	0.0	3.1
H,L=	2	2
0	50.4	75.0
1	36.7	50.0
2	21.6	24.9
3	24.2	29.2
4	15.7	16.9
5	23.3	25.4
6	26.6	23.2
7	17.3	16.1
8	6.5	8.3
9	55.7	50.5
10	38.0	34.4
11	7.8	8.9
12	18.4	17.5
13	34.4	30.8
14	21.8	18.4
15	9.1	12.4
16	30.8	26.3
17	28.0	26.5
18	13.1	10.9
19	20.3	18.4
20	0.0	2.8
21	0.0	7.5
22	13.2	12.5
23	16.9	14.3
24	14.5	13.6
25	9.5	10.7
H,L=	3	2
0	62.1	71.8
1	52.3	66.2
2	49.4	62.3
3	12.5	14.0
4	8.8	8.9
5	7.8	8.7
6	25.8	30.4
7	27.6	25.8
8	25.3	19.2
9	19.0	15.3
10	15.1	12.6
11	22.5	19.6
12	22.1	19.6
13	19.4	17.9
14	39.1	33.6
15	9.1	9.2
16	9.3	7.4
17	26.4	23.3
18	16.0	14.8
19	23.9	21.2
20	17.3	14.5
21	23.2	23.9
22	0.0	3.4
23	16.3	13.5
24	12.2	10.8
H,L=	4	2
0	8.2	5.7
1	23.9	31.5
2	51.5	53.5
3	75.7	86.1
4	42.7	44.4
5	0.0	7.0
6	27.9	26.0
7	27.3	28.7
8	31.3	30.1
9	16.5	14.3
10	19.0	16.2
11	27.0	26.7
12	24.0	19.9
13	19.6	17.2
14	31.3	26.1
15	9.2	9.7
16	32.3	26.9
17	22.9	21.1
18	18.4	17.2
19	26.9	25.8
20	0.0	5.2
21	8.0	10.3
22	12.8	13.2
23	11.4	9.9
24	0.0	2.2
H,L=	5	2
0	46.6	54.2
1	5.4	13.0
2	42.8	44.2
3	45.3	52.8
4	29.3	25.8
5	35.5	35.4
6	29.5	30.4
7	38.0	42.0
8	27.2	25.4
9	27.5	21.9
10	21.1	18.8
11	24.9	22.2
12	8.6	7.7
13	17.8	14.5
14	25.8	23.6
15	9.3	9.7
16	24.7	22.8
17	13.1	11.8
18	[illegible]	40.2
19	38.6	38.5
20	11.9	13.9
21	7.8	8.8
22	0.0	5.1
23	0.0	1.6
24	5.1	6.6
H,L=	6	2
0	8.4	9.4
1	31.6	37.8
2	38.8	45.6
3	51.9	58.2
4	22.3	20.9
5	23.2	22.1
6	31.2	34.0
7	24.1	26.1
8	25.2	22.6
9	29.6	29.5
10	8.2	7.9
11	0.0	9.3
12	8.8	7.9
13	27.1	24.0
14	9.2	10.7
15	13.2	16.7
16	9.3	11.8
17	20.7	17.9
18	9.0	8.3
19	0.0	5.3
20	8.2	9.4
21	7.6	6.0
22	9.8	8.2
23	12.7	10.5
24	5.7	5.8
H,L=	7	2
0	15.9	16.5
1	43.2	42.3
2	38.8	39.2
3	9.5	8.1
4	9.7	8.6
5	25.7	25.5
6	31.3	31.8
7	22.9	19.3
8	0.0	4.3
9	27.2	25.2
10	33.9	30.3
11	0.0	6.7
12	23.7	19.7
13	18.3	14.4
14	22.8	18.7
15	13.2	15.1
16	9.3	10.5
17	9.1	7.3
18	21.8	18.9
19	0.0	4.6
20	21.2	18.1
21	10.4	10.7
22	8.0	6.1
23	6.8	5.7
H,L=	8	2
0	46.6	48.7
1	15.7	12.8
2	10.0	11.0
3	12.5	7.9
4	34.6	37.0
5	13.1	9.3
6	34.8	30.3
7	21.2	19.5
8	29.8	27.5
9	17.0	17.5
10	19.5	20.5
11	40.1	35.4
12	30.3	30.7
13	18.5	17.1
14	16.2	15.5
15	9.3	10.9
16	20.6	18.6
17	9.0	10.9
18	0.0	8.1
19	8.2	7.5
20	0.0	2.0
21	8.6	7.3
22	13.1	9.9
23	8.6	6.5
H,L=	9	2
0	28.1	30.8
1	15.1	14.8
2	0.0	7.1
3	27.8	25.7
4	45.7	45.0
5	19.6	19.8
6	32.7	34.0
7	33.5	30.3
8	27.7	29.2
9	21.5	17.1
10	20.0	14.4
11	30.3	25.7
12	30.7	30.4
13	22.9	19.6
14	9.3	7.4
15	16.0	13.9
16	9.1	9.5
17	8.8	7.6
18	11.9	12.8
19	15.9	15.2
20	7.3	7.3
21	11.4	9.0
22	6.9	4.4
H,L=	10	2
0	17.9	17.1
1	16.0	11.6
2	14.0	10.8
3	16.3	11.8
4	24.8	23.6
5	25.2	23.8
6	32.0	28.9
7	53.7	50.8
8	15.3	13.6
9	49.5	44.7
10	0.0	8.6
11	27.8	26.3
12	0.0	4.9
13	9.3	8.8
14	24.5	21.9
15	9.1	6.6
16	12.5	11.4
17	17.1	16.4
18	16.2	13.6
19	13.1	13.7
20	11.9	10.6
21	10.5	10.9
22	10.4	9.6
H,L=	11	2
0	14.6	9.2
1	11.9	12.7
2	26.9	25.2
3	8.5	8.8
4	0.0	2.9
5	8.7	7.8
6	26.6	22.9
7	33.7	31.0
8	44.7	43.4
9	26.1	24.0
10	0.0	1.6
11	26.4	21.3
12	20.9	18.5
13	18.5	17.0
14	15.8	14.3
15	0.0	3.6
16	19.2	17.3
17	16.4	16.6
18	7.7	6.6
19	8.6	5.9
20	13.4	11.1
21	11.3	8.5
H,L=	12	2
0	0.0	1.9
1	36.4	34.7
2	8.8	8.4
3	17.8	16.7
4	12.7	13.6
5	12.8	12.7
6	15.8	12.7
7	13.0	12.8
8	22.7	18.1
9	40.7	34.4
10	16.2	14.7
11	13.2	11.6
12	0.0	5.2
13	18.2	15.8
14	21.8	21.0
15	12.1	11.5
16	11.6	9.4
17	7.7	8.8
18	10.1	10.6
19	7.9	4.6
20	6.8	3.6
H,L=	13	2
0	30.3	28.8
1	35.4	39.9
2	15.8	18.5
3	15.9	14.3
4	9.2	9.0
5	0.0	1.2
6	0.0	2.3
7	16.1	15.4
8	9.3	5.8
9	0.0	1.7
10	0.0	3.1
11	29.1	25.6
12	18.1	15.3
13	8.8	8.0
14	8.5	9.0
15	23.2	21.1
16	0.0	6.0
17	10.2	10.9
18	8.0	7.4
19	7.0	3.7
H,L=	14	2
0	16.1	16.2
1	0.0	4.0
2	24.6	26.6
3	0.0	4.2
4	26.4	21.7
5	16.2	14.1
6	9.3	7.2
7	16.1	15.3
8	16.1	13.4
9	0.0	8.0
10	0.0	6.3
11	15.5	13.6
12	17.4	16.5
13	0.0	2.6
14	11.4	9.8
15	20.3	19.0
16	0.0	5.7
17	17.3	15.2
18	14.1	9.8
19	8.0	6.1
H,L=	15	2
0	0.0	2.0
1	9.3	11.4
2	18.7	17.8
3	22.8	25.4
4	9.3	8.9
5	18.6	17.4
6	26.1	25.6
7	9.1	10.2
8	9.0	8.2
9	23.7	19.7
10	23.2	19.6
11	0.0	5.7
12	8.2	6.9
13	7.9	6.3
14	10.6	10.2
15	7.0	5.0
16	17.5	14.0
17	15.5	11.8
18	9.7	7.5
H,L=	16	2
0	0.0	2.2
1	18.4	19.5
2	13.0	13.8
3	9.1	8.9
4	15.8	12.1
5	18.1	17.1
6	26.9	22.4
7	0.0	6.7
8	17.4	17.7
9	8.5	9.3
10	0.0	2.2
11	0.0	7.6
12	0.0	7.1
13	15.4	12.9
14	8.2	7.9
15	7.5	7.4
16	9.4	8.3
17	5.2	5.9
H,L=	17	2
0	12.6	12.1
1	12.5	10.6
2	0.0	2.7
3	17.6	15.1
4	17.5	16.8
5	0.0	3.6
6	8.5	9.6
7	14.5	12.3
8	8.1	11.6
9	23.8	22.8
10	23.0	23.2
11	14.6	14.8
12	16.9	12.0
13	0.0	3.0
14	10.0	6.3
15	8.7	6.4
H,L=	18	2
0	16.8	17.8
1	8.3	10.3
2	0.0	5.2
3	8.2	6.7
4	21.7	19.0
5	16.1	15.4
6	0.0	5.6
7	13.3	14.3
8	0.0	7.4
9	12.4	12.9
10	0.0	4.9
11	28.4	23.2
12	25.2	22.1
H,L=	19	2
0	15.4	16.1
1	7.6	6.8
2	17.8	10.0
3	7.5	6.9
4	14.9	16.2
5	0.0	4.5
6	21.3	22.1
7	6.8	8.6
8	19.7	15.4
9	13.2	8.4
10	12.3	9.2
11	15.7	12.2
12	0.0	6.9
13	3.3	10.9
H,L=	20	2
0	14.4	7.5
1	14.4	8.8
2	8.2	6.7
3	0.0	3.0
4	0.0	1.7
5	17.2	9.4
6	10.5	6.3
7	17.2	13.8
8	14.7	9.1
9	17.8	12.0
H,L=	21	2
0	11.8	0.6
1	6.8	8.6
2	6.7	3.7
3	11.3	6.4
4	6.3	8.3
5	0.0	7.2
6	16.6	2.1
H,L=	0	3
1	0.0	0.7
2	7.2	6.5
3	0.0	0.8
4	10.2	16.2
5	13.2	13.5
6	16.1	15.7
7	20.6	23.4
8	13.5	18.3
9	29.0	35.5
10	12.9	10.2
11	0.0	2.9
12	16.7	12.0
13	17.0	18.1
14	0.0	6.6
15	0.0	4.2
16	0.0	10.4
17	18.5	23.3
18	0.0	7.5
19	0.0	8.7
20	7.3	10.4
21	15.4	18.4
22	0.0	11.1
H,L=	1	3
0	0.0	2.4
1	0.0	9.4
2	0.0	1.8
3	12.9	16.7
4	10.4	13.4
5	16.3	19.0
6	26.3	27.6
7	22.2	27.2
8	11.8	8.1
9	19.6	20.5
10	9.1	6.7
11	0.0	6.0
12	0.0	8.9
13	0.0	7.6
14	0.0	8.3
15	9.7	10.1
16	16.5	14.3
17	9.2	7.4
18	15.1	15.3
19	9.1	10.9
20	15.5	13.9
21	0.0	11.7
22	9.4	11.1
H,L=	2	3
0	7.7	13.5
1	0.0	6.8
2	0.0	2.7
3	11.5	12.9
4	25.4	35.9
5	6.9	4.7
6	10.5	8.7
7	25.2	36.3
8	14.6	12.3
9	17.7	19.7
10	16.0	16.6
11	13.4	15.8
12	19.4	16.3

Table 2 (*cont.*)

K	Fo	Fc
13	9.8	10.3
14	0.0	0.9
15	13.8	11.9
16	0.0	9.1
17	12.9	12.4
18	15.0	16.5
19	13.9	15.4
20	0.0	5.7
21	0.0	5.0
22	8.7	8.7
H,L=	3	3
0	19.8	24.1
1	0.0	4.2
2	13.2	19.7
3	12.6	13.2
4	21.5	33.2
5	20.6	23.4
6	13.4	18.7
7	14.2	12.9
8	14.9	16.7
9	9.0	7.6
10	9.3	9.6
11	23.5	26.4
12	13.8	13.5
13	9.8	6.8
14	0.0	1.5
15	9.7	9.5
16	16.4	15.1
17	0.0	5.3
18	0.0	5.0
19	13.7	16.2
20	0.0	9.1
H,L=	4	3
0	0.0	4.1
1	24.9	32.3
2	18.7	23.4
3	9.8	12.0
4	7.3	8.9
5	21.7	22.8
6	14.0	16.8
7	14.7	14.1
8	12.5	10.3
9	20.5	19.3
10	16.4	16.0
11	0.0	1.6
12	13.8	11.1
13	19.7	16.7
14	0.0	2.5
15	13.6	13.8
16	0.0	5.8
17	8.9	8.3
18	18.8	19.7
19	13.4	13.4
20	14.5	15.2
H,L=	5	3
0	15.9	23.4
1	14.3	16.0
2	38.7	54.2
3	32.8	38.5
4	17.5	19.4
5	19.9	23.8
6	20.7	27.2
7	8.7	10.9
8	0.0	6.6
9	9.3	11.3
10	16.6	19.5
11	9.7	9.0
12	0.0	6.5
13	13.9	15.6
14	16.9	13.7
15	0.0	6.9
16	18.4	17.8
17	12.4	9.1
18	0.0	5.1
19	9.1	8.4
20	8.0	6.7
H,L=	6	3
0	0.0	6.1
1	27.1	27.1
2	15.8	19.1
3	11.4	14.1
4	35.3	40.9
5	25.7	29.5
6	12.5	12.0
7	18.2	18.3
8	0.0	7.6
9	0.0	6.3
10	19.4	20.1
11	17.0	19.0
12	17.1	14.6
13	0.0	6.4
14	0.0	8.4
15	21.0	23.5
16	9.0	9.1
17	8.5	7.6
18	23.8	21.9
19	0.0	6.5
20	10.5	10.1
H,L=	7	3
0	0.0	0.3
1	16.8	18.7
2	14.6	20.3
3	24.3	30.6
4	29.1	38.5
5	22.0	22.7
6	18.3	18.1
7	16.2	16.9
8	16.5	18.1
9	0.0	3.0
10	9.8	9.7
11	29.6	31.5
12	0.0	4.7
13	0.0	6.1
14	13.4	11.2
15	0.0	7.2
16	0.0	11.1
17	8.2	9.8
18	16.1	13.7
19	16.4	16.4
20	9.4	12.3
H,L=	8	3
0	15.3	21.3
1	8.9	11.8
2	8.9	11.3
3	12.8	12.3
4	15.9	18.2
5	26.4	29.9
6	25.0	27.6
7	21.5	24.5
8	9.7	9.4
9	27.8	30.3
10	22.0	22.2
11	0.0	2.3
12	16.9	13.2
13	16.6	17.1
14	0.0	5.7
15	0.0	7.0
16	0.0	5.6
17	13.6	15.3
18	0.0	1.5
19	10.6	9.7
H,L=	9	3
0	9.3	10.6
1	0.0	7.3
2	0.0	0.8
3	16.3	14.3
4	0.0	8.1
5	19.2	17.3
6	25.6	20.9
7	13.8	16.2
8	31.1	31.9
9	13.9	14.2
10	9.8	7.5
11	0.0	4.4
12	9.6	12.4
13	18.7	17.8
14	0.0	3.6
15	0.0	1.8
16	14.0	14.4
17	7.4	7.0
18	8.0	5.5
H,L=	10	3
0	0.0	4.2
1	0.0	6.7
2	0.0	6.7
3	0.0	6.1
4	16.8	13.1
5	13.8	12.5
6	0.0	8.8
7	26.1	22.0
8	13.9	15.6
9	17.0	18.2
10	0.0	5.2
11	0.0	2.7
12	18.7	18.8
13	0.0	11.3
14	0.0	6.5
15	11.5	11.8
16	0.0	4.0
17	14.4	10.4
18	9.9	9.5
H,L=	11	3
0	0.0	7.1
1	0.0	7.0
2	0.0	4.6
3	17.0	15.8
4	9.8	8.9
5	0.0	5.1
6	0.0	2.9
7	13.9	6.6
8	9.7	4.9
9	9.6	11.5
10	21.2	21.1
11	16.1	8.3
12	12.7	9.3
13	8.6	8.1
14	8.1	10.5
15	10.7	10.4
16	8.4	6.9
17	7.3	6.3
H,L=	12	3
0	19.7	21.2
1	13.9	10.8
2	0.0	4.1
3	13.9	12.1
4	0.0	1.7
5	9.8	6.9
6	9.7	11.9
7	0.0	9.1
8	0.0	11.0
9	0.0	8.8
10	0.0	2.9
11	0.0	10.2
12	12.1	12.6
13	14.0	11.3
14	10.7	12.4
15	0.0	4.8
16	14.7	13.2
H,L=	13	3
0	9.7	7.2
1	13.8	10.3
2	16.8	14.5
3	0.0	7.2
4	0.0	5.6
5	9.6	8.8
6	16.4	16.3
7	0.0	2.4
8	15.9	11.1
9	12.7	12.2
10	15.1	14.9
11	0.0	4.4
12	11.2	9.3
13	7.4	7.8
14	14.4	11.5
15	10.2	10.2
H,L=	14	3
0	13.4	10.9
1	16.4	17.0
2	18.9	18.4
3	0.0	3.0
4	9.3	8.2
5	13.0	9.5
6	0.0	4.7
7	0.0	10.3
8	0.0	2.1
9	0.0	7.0
10	14.0	11.8
11	10.8	10.8
12	0.0	4.4
13	0.0	5.2
14	12.0	10.9
15	8.7	8.0
H,L=	15	3
0	15.7	16.4
1	0.0	9.7
2	0.0	7.1
3	17.9	19.7
4	8.8	11.3
5	0.0	2.8
6	8.5	7.8
7	11.7	9.9
8	11.3	8.8
9	0.0	1.4
10	12.6	11.3
11	13.5	13.9
H,L=	16	3
0	0.0	3.9
1	0.0	8.3
2	0.0	5.3
3	16.5	18.6
4	15.3	15.7
5	7.9	8.2
6	11.0	11.1
7	0.0	5.0
8	12.4	7.0
9	11.6	7.4
H,L=	17	3
0	0.0	3.3
1	13.1	13.3
2	13.0	12.5
3	9.0	8.5
4	0.0	4.1
5	12.1	9.0
6	16.5	13.2
7	0.0	5.2
8	10.2	8.9

ethane, they were very labile and not suitable for X-ray analysis.

Analysis. Calc. for $C_{16}H_{16}O_6N_2Ni$: C 49·2, H 4·13, N 7·17, Ni 15·0%. Found: C 49·2, H 4·31, N 7·18, Ni 15·5%.

The lattice constants of the orthorhombic unit cell were determined from oscillation and zero-level Weissenberg photographs about three crystallographic axes with Cu $K\alpha$ ($\lambda = 1{\cdot}5418$ Å) radiation:

$a = 18{\cdot}63 \pm 0{\cdot}02$, $b = 21{\cdot}64 \pm 0{\cdot}02$ and $c = 3{\cdot}84 \pm 0{\cdot}01$ Å .

The cell contains four molecules; the observed density is 1·68 g.cm^{-3}, calculated 1·65 g.cm^{-3}. Systematic absences of $h00$ for h odd, $0k0$ for k odd, and $00l$ for l odd indicate the space group to be $P2_12_12_1$. Linear absorption coefficient for Cu $K\alpha$ radiation: $\mu = 22{\cdot}8$ cm^{-1}. Total number of electrons per unit cell: $F(000) = 808$.

The dimensions of the specimen used for collecting the intensity data were 0·2 × 0·1 × 0·3 mm. Equi-inclination Weissenberg photographs, $hk0$–$hk3$, $h0l$–$h2l$, and $0kl$, were taken on a Nonius integrating Weissenberg camera with Cu $K\alpha$ radiation, using the multiple film technique. Intensities were estimated visually with a calibrated scale. The number of the examined unique reflexions was 1798, which contains 401 reflexions of zero intensity. The range of measured intensities was 1 to 5400. Corrections were applied for Lorentz and polarization factors, but not for absorption and extinction.

Structure determination

Since the short c axis suggested that the molecules were planar and packed parallel to (001), the analysis was started with the c axis projection, using $hk0$ reflexion data. The structure was solved by the usual heavy atom method. Two-dimensional Patterson synthesis showed that the nickel atom was located at the position with $x = 0{\cdot}114$, $y = 0{\cdot}134$. The x, y, coordinates and isotropic temperature factors of all atoms were determined through three cycles of Fourier and four cycles of $F_o - F_c$ synthesis ($R = 0{\cdot}15$). Three-dimensional Patterson synthesis was then carried out. It was, however, difficult to estimate the precise z coordinate of the nickel atom from the sections $P(\frac{1}{2}, v, w)$, $P(u, \frac{1}{2}, w)$, and $P(u, v, \frac{1}{2})$ because of the short c axis and proximity of the nickel atom position to $z = \frac{1}{4}$. Therefore, the value $z = \frac{1}{4}$ was assumed for the first three-dimensional Fourier synthesis, in which the phases used were those determined by the nickel atom only.

To estimate the z coordinates of all light atoms, the Fourier map and the Patterson function were carefully

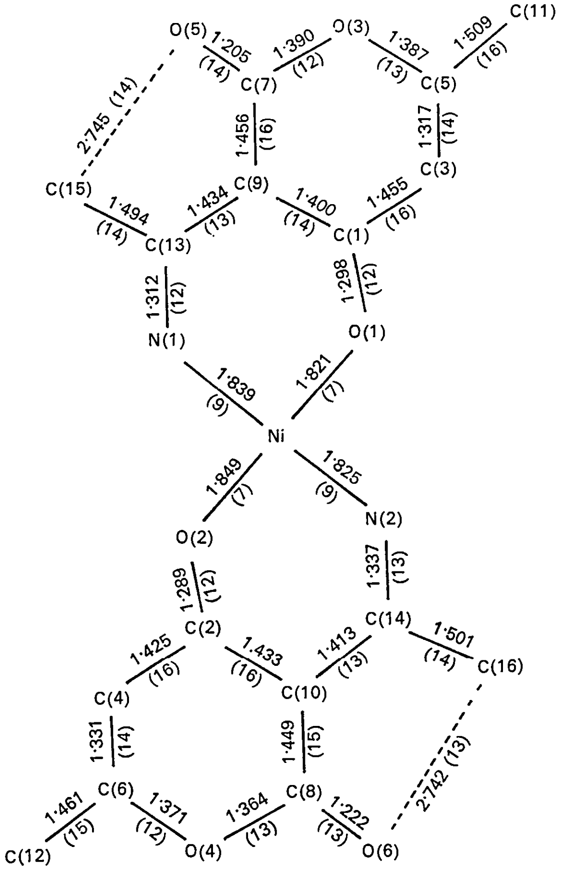


Fig. 1. Bond lengths, and their standard deviations in parentheses (× 1000).

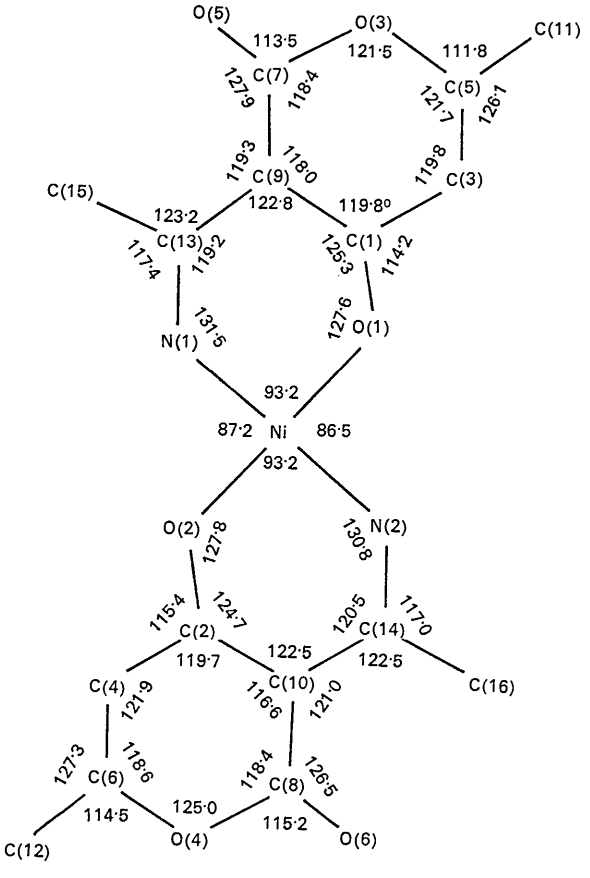


Fig. 2. Bond angles. Mean value of standard deviations is 0·9°.

compared with the (001) projection of the structure. Using the estimated values of z coordinates and the other parameters determined by the (001) projection, the second Fourier synthesis was calculated. At this stage, the R value was not satisfactorily low because of the inaccurate initial value of the z parameter of the nickel atom. Therefore, all the positional and isotropic thermal parameters were refined through further four cycles of $F_o - F_c$ synthesis ($R = 0\cdot19$).

Further refinement was carried out by a block diagonal least-squares method with anisotropic temperature factors for the nickel atom only. The weighting scheme was:

$$\sqrt{w}=0 \text{ for } F_o<1\cdot8,\quad \sqrt{w}=1/F_o \text{ for } F_o\geq 1\cdot8\,.$$

The final R value was 0·166 (0·103 for observed reflexions). The final positional and thermal parameters with their standard deviations are listed in Table 1. Maximum shifts of positional and thermal parameters during the final cycle were about a third of the standard deviations.

The atomic scattering factors used were those of Thomas & Umeda (1957) for nickel and those of Berghuis, Haanappel, Potters, Loopstra, McGillavry & Veenendaal (1955) for carbon, nitrogen and oxygen.

Most of the calculations were carried out on OKITAC 5090H with programs written by I. Ueda (Kyushu University) and least-squares calculation on HITAC 5020 with a program by T. Ashida (Osaka University). At the initial stage of the analysis, the IBM 7074 computer was also used.

Description of the structure

The interatomic distances and the bond angles calculated on the basis of the parameters obtained are shown in Figs. 1 and 2.

It is shown that the complex has a chelate structure with a square planar configuration. Four ligand atoms are exactly coplanar with the nickel atom, none of these atoms deviating more than 0·005 Å from the mean plane. The Ni–O distances 1·82 and 1·85 Å and the Ni–N distances 1·83 and 1·84 Å agree well with the following distances reported for some diamagnetic nickel(II) complexes: Ni–O 1·84 and Ni–N 1·84 Å in bis(salicylaldiminato)nickel(II) (Stewart & Lingafelter, 1959) and Ni–O 1·80 and Ni–N 1·90 Å in bis-(*N*-methylsalicylaldiminato)nickel(II) (Frasson, Panattoni & Sacconi, 1959). However, they are much shorter than the Ni–O distance 2·02 Å found in diaquobis(salicylaldehydato)nickel(II), in which the nickel atom has an octahedral coordination configuration (Stewart, Lingafelter & Breazeale, 1961). There are no other ligand atoms bonded to the nickel atom in the direction perpendicular to the plane of the square, so that the elec-

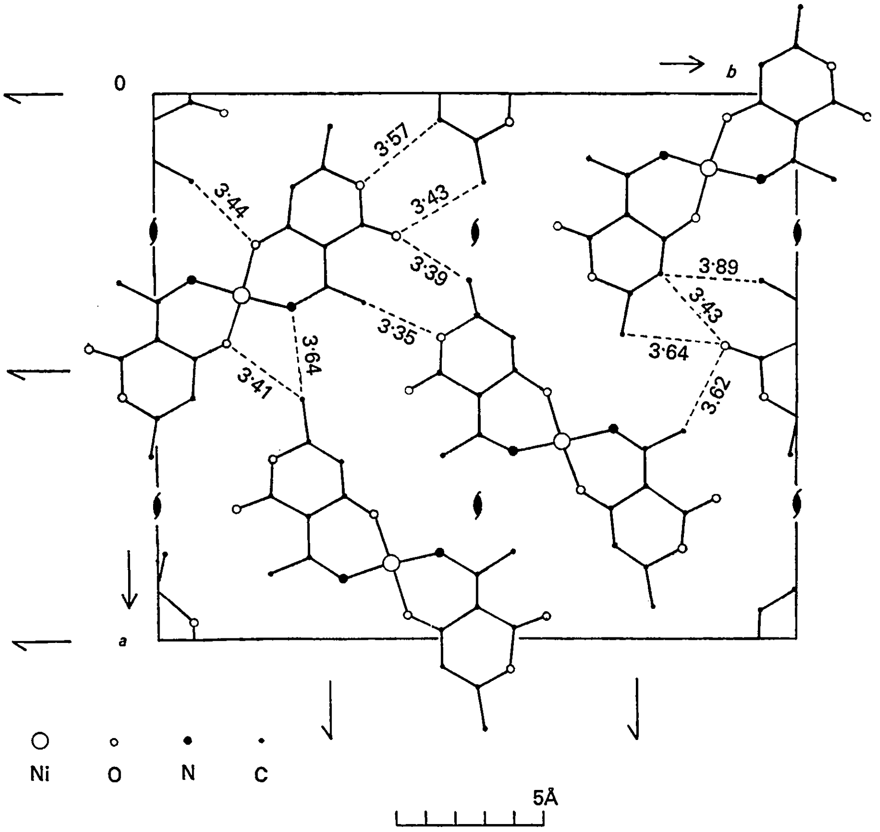


Fig. 3. Arrangement of molecules on the projection along the c axis.

tronic structure of the nickel atom is considered to be of the typical *dsp*^{2} type.

The C–O bond lengths 1·29 and 1·30 Å and the C–N distances 1·31 and 1·34 Å in the chelate rings are also in good agreement with those of the corresponding bonds in the above compounds and rather close to the values of 1·23 and 1·28 Å respectively for isolated double bonds. Most of the bond lengths of six-membered rings of the DHA imide residues are close to those of benzenoid rings except the C(3)–C(5) and C(4)–C(6) distances, 1·32 and 1·33 Å, which show almost an isolated double-bond character. The lengths of C(9)–C(13) and C(10)–C(14) bonds, which are members of both the chelate rings and CH_3CNH-groups, 1·43 and 1·41 Å, are also close to those of benzenoid C–C bonds.

The deviations of some atoms from the mean plane of the molecule were found to be appreciable. The least-squares plane of the six-membered ring of a DHA imide residue is given by the following equation:

$$0{\cdot}1000X+0{\cdot}0440Y+0{\cdot}2764Z=1\,,$$

where X, Y and Z are coordinates expressed in Å. The deviations of the atoms from this plane are C(1) 0·003, C(3) 0·012, C(5) −0·008, O(3) −0·020, C(7) 0·039, C(9) −0·029, C(11) −0·020, C(13) −0·067, C(15) −0·140, O(5) 0·052, N(1) −0·143, O(1) 0·053 Å.

A similar examination was carried out for another plane through the atoms C(1), C(3), C(5), C(11) and O(3), because it is expected from the isolated double-bond character of the C(3)–C(5) bond that these five atoms will be exactly coplanar. The equation is

$$0{\cdot}0997X+0{\cdot}0468Y+0{\cdot}2722Z=1\,.$$

The deviations are C(1) −0·006, C(3) 0·010, C(5) −0·001, C(11) −0·005, O(3) 0·003, O(1) 0·009, C(7) 0·060, O(5) 0·084, C(9) −0·027, C(13) −0·067, C(15) −0·380, N(1) −0·093 Å.

The above analysis shows that out-of-plane distortions occur in the DHA imide residue. It is noticeable that the CH_3CNH group departs from either plane above mentioned and that O(5) deviates to the opposite side. Similar distortions are also found in the other residue.

All the contact distances between the molecules appear to be reasonable (Fig. 3). The shortest Ni–Ni distance, 3·84 Å, is a little longer than the lengths of the intermolecular metal–metal bonds found in some nickel(II) chelate compounds such as dimethylglyoxime (Banks & Barnum, 1958).

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