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## The Crystal and Molecular Structure of the Bis-(5-ethyl-5-isoamylbarbiturato)bis(imidazole) Complex of Nickel(II)

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The structure of the title compound has been determined from single-crystal X-ray diffraction data collected on a four-circle diffractometer. The analysis was carried out with 3741 independent reflexions and refined by full-matrix least-squares computations to a final  $R$  of 0.056. The crystals are triclinic, space group  $P\bar{1}$ , with  $a = 10.615$  (5),  $b = 10.544$  (5),  $c = 7.457$  (5) Å,  $\alpha = 111.156$  (2),  $\beta = 97.360$  (2),  $\gamma = 82.675$  (2)°,  $Z = 1$ . The nickel atom is involved in distorted octahedral coordination; the donor atoms are the deprotonated nitrogen atoms of the barbiturate anions [Ni–N: 2.078 (2) Å], the nitrogen atoms of the imidazole moieties [Ni–N: 2.055 (3) Å] and two centrosymmetrically related barbiturato oxygen atoms [Ni–O: 2.226 (2) Å]. The molecules are linked by N–H···O hydrogen bonds arising from the barbiturato and imidazole N–H groups.

### Introduction

The structure of the title compound is another in our series of X-ray determinations on compounds of the general formula  $M(II)(\text{barb})_2L_2$  where  $M$  is a transition metal, barb is the anion of a substituted barbituric acid and  $L$  is an organic base. We have reported in earlier publications the crystal structures of the bis-(5,5'-diethylbarbiturato)bispyridine complex of copper(II) (Caira, Fazakerley, Linder & Nassimbeni, 1973) the bis-(5,5'-diethylbarbiturato)bispicoline dihydrate complex of copper(II) (Fazakerley, Linder, Nassimbeni & Rodgers, 1973, 1974 *a*), the bis-[5-allyl-5-(2-bromoallyl)-

barbiturato]bispyridine dihydrate complex of copper(II) (Fazakerley, Linder, Nassimbeni & Rodgers, 1974 *b*) and the bis-(5,5'-diethylbarbiturato)bispicoline complex of zinc(II) (Nassimbeni & Rodgers, 1974). The title compound is thus the first involving nickel and an X-ray analysis was undertaken (i) to determine the molecular structure and hence the coordination type, (ii) to verify that the coordination site on the barbiturate anion is a deprotonated nitrogen atom as has been found in our earlier structures and (iii) to investigate the nature of hydrogen bonding which has been found to be a prominent feature of other structures in this series.

## Experimental

Dilute aqueous solutions of  $NiSO_4$ , sodium amylo-barbitone and imidazole were mixed at room temperature in the molar ratio 1:2:2. Small well defined light-blue crystals formed after several days.

The analysis gave:

	%C	%H	%N
Calculated:	52.11	6.51	17.37
Experimental:	51.8	6.7	17.8

These figures agreed closely enough to suggest that the complex had the composition  $NiC_{28}H_{42}N_8O_6$  corresponding to the formulation  $Ni(II)$  (5-ethyl-5-isoamyl-barbiturato)<sub>2</sub> (imidazole)<sub>2</sub>.

The method of determining the density of the crystals has been described earlier (Caira, Fazakerley, Linder & Nassimbeni, 1973). A crystal of dimensions  $0.325 \times 0.225 \times 0.100$  mm was used. The lattice constants were obtained from a least-squares analysis of the settings of 25 reflexions measured on a diffractometer with  $Mo K\alpha$  radiation ( $\lambda = 0.7107 \text{ \AA}$ ). Crystal data are given in Table 1.

Table 1. Crystal data

$a = 10.615 \text{ \AA}$	$V = 768.97 \text{ \AA}^3$
$b = 10.544$	$D_m = 1.38 (2) \text{ g cm}^{-3}$
$c = 7.457$	$D_c = 1.392 \text{ g cm}^{-3}$ for $Z = 1$
$\alpha = 111.156^\circ$	M.W. 644.7
$\beta = 97.360$	$\mu = 6.86 \text{ cm}^{-1}$ for $Mo K\alpha$ radiation
$\gamma = 82.675$	$F(000) = 342.0$
Space group = $P\bar{1}$	

## Intensity data

The intensities were collected at the National Physics Research Laboratory of the CSIR on a Philips PW

Table 3. Hydrogen atoms

Fractional atomic coordinates ( $\times 10^3$ ) and their e.s.d.'s ( $\times 10^3$ )

	<i>x</i>	<i>y</i>	<i>z</i>
H(3)	110 (3)	-501 (3)	-376 (5)
H(7,1)	344 (3)	-407 (4)	189 (5)
H(7,2)	297 (3)	-264 (4)	290 (5)
H(8,1)	82 (4)	-330 (4)	216 (6)
H(8,2)	129 (4)	-469 (4)	116 (6)
H(8,3)	165 (4)	-402 (4)	338 (6)
H(9,1)	425 (3)	-199 (3)	96 (5)
H(9,2)	456 (3)	-348 (3)	-27 (5)
H(10,1)	366 (3)	-286 (4)	-306 (5)
H(10,2)	339 (3)	-145 (4)	-181 (5)
H(11)	570 (4)	-136 (4)	-92 (5)
H(12,1)	635 (5)	-359 (5)	-187 (7)
H(12,2)	579 (5)	-370 (5)	-418 (7)
H(12,3)	703 (5)	-297 (5)	-305 (7)
H(13,1)	482 (4)	-163 (5)	-491 (7)
H(13,2)	455 (4)	-29 (5)	-321 (7)
H(13,3)	597 (4)	-92 (5)	-348 (7)
H(15)	167 (3)	226 (4)	-22 (5)
H(16)	239 (4)	216 (4)	-319 (5)
H(17)	194 (4)	12 (4)	-578 (5)
H(18)	91 (3)	-112 (3)	-430 (5)

1100 computer-controlled four-circle diffractometer operating in the  $\omega$  scan mode (scan width =  $1,20^\circ$  and scan speed =  $0,04^\circ \text{ s}^{-1}$ ). The background and scan times were both 30 s. The horizontal and vertical detector apertures were respectively 2 and  $1^\circ$ .

With graphite-monochromated  $Mo K\alpha$  radiation, 4469 reflexions up to  $2\theta = 60^\circ$  were measured. A reflexion was taken above background if  $I_{rel} > 1.65\sigma(I_{rel})$ . 728 reflexions did not satisfy this criterion and were omitted as unobserved. Throughout the course of the data collection, three standard reflexions were measured after every 66 reflexions to check stability of operation and crystal decomposition. The variation in intensity of a reference reflexion was observed to be less than 3% of

Table 2. Non-hydrogen atoms

Fractional atomic coordinates and their e.s.d.'s ( $\times 10^4$ ) and anisotropic temperature factors and their e.s.d.'s ( $\times 10^4$ )

	<i>x</i>	<i>y</i>	<i>z</i>	$U_{11}$	$U_{22}$	$U_{33}$	$U_{12}$	$U_{13}$	$U_{23}$
Ni	0	0	0	193 (2)	84 (2)	164 (2)	3 (1)	62 (2)	17 (2)
N(1)	657 (2)	-2042 (2)	-1231 (3)	198 (9)	94 (8)	196 (10)	-2 (7)	51 (8)	4 (7)
C(2)	427 (2)	-3229 (2)	-2709 (3)	219 (11)	109 (10)	172 (11)	-17 (11)	65 (9)	12 (8)
N(3)	1285 (2)	-4369 (2)	-2858 (3)	256 (11)	93 (9)	260 (11)	-6 (8)	11 (9)	-26 (8)
C(4)	2368 (3)	-4415 (3)	-1681 (4)	252 (13)	138 (11)	267 (13)	-13 (9)	68 (10)	33 (9)
C(5)	2669 (2)	-3105 (2)	-3 (4)	204 (11)	14 (10)	204 (11)	2 (8)	37 (9)	45 (9)
C(6)	1667 (2)	-1938 (2)	54 (4)	206 (11)	113 (9)	201 (11)	-18 (8)	76 (9)	50 (8)
C(7)	2730 (3)	-3398 (3)	1896 (4)	398 (17)	298 (15)	276 (14)	22 (12)	23 (12)	157 (12)
C(8)	1528 (4)	-3893 (4)	2215 (6)	569 (24)	539 (23)	472 (21)	10 (18)	191 (18)	326 (19)
C(9)	3975 (3)	-2712 (3)	-230 (5)	196 (12)	268 (13)	366 (16)	-18 (10)	32 (11)	117 (12)
C(10)	3995 (3)	-2256 (4)	-1942 (5)	201 (13)	451 (18)	451 (18)	-54 (12)	44 (12)	255 (15)
C(11)	5304 (3)	-1935 (4)	-2213 (5)	234 (14)	465 (19)	443 (19)	-87 (13)	76 (13)	163 (15)
C(12)	6203 (5)	-3221 (6)	-2899 (9)	420 (24)	718 (33)	1076 (44)	160 (22)	356 (26)	329 (31)
C(13)	5198 (4)	-1137 (6)	-3559 (8)	456 (23)	851 (35)	832 (34)	-201 (23)	111 (23)	508 (30)
O(2)	-501 (2)	-3314 (2)	-3884 (3)	308 (11)	205 (10)	303 (11)	-2 (8)	-45 (9)	-27 (8)
O(4)	3057 (2)	-5462 (2)	-1947 (4)	405 (13)	167 (10)	530 (15)	111 (9)	0 (11)	15 (10)
O(6)	1771 (2)	-786 (2)	1351 (3)	279 (10)	137 (8)	211 (9)	-30 (7)	37 (7)	2 (7)
N(14)	1041 (2)	468 (2)	-1783 (3)	248 (11)	185 (10)	258 (11)	5 (8)	78 (9)	89 (9)
C(15)	1597 (3)	1648 (3)	-1423 (5)	362 (16)	220 (13)	452 (18)	-65 (12)	137 (14)	82 (13)
C(16)	2071 (3)	1625 (4)	-3014 (6)	362 (17)	387 (18)	622 (24)	-86 (14)	154 (16)	300 (17)
N(17)	1814 (3)	389 (3)	-4427 (4)	392 (16)	518 (18)	333 (15)	5 (13)	138 (12)	203 (13)
C(18)	1199 (3)	-259 (3)	-3621 (4)	310 (14)	293 (14)	239 (13)	-8 (11)	83 (11)	92 (11)

Table 4. Observed and calculated structure factors  
Reflexions marked with an asterisk are unobserved.

Table with multiple columns of numerical data representing structure factors. Columns are labeled with Miller indices (hkl) such as 10FO 10FC, 10FO 10FC, etc. The table contains observed values and calculated values for various reflections, with some values marked with an asterisk to indicate they are unobserved.

Table 4 (cont.)

h0k0		h0k0		h0k0		h0k0		h0k0		h0k0		h0k0		h0k0		h0k0		h0k0		
h	k	l	F	h	k	l	F	h	k	l	F	h	k	l	F	h	k	l	F	
10	99	80	3	9	98	79	3	8	97	70	3	7	96	61	3	6	95	52	3	5
8	98	71	3	7	97	62	3	6	96	53	3	5	95	44	3	4	94	35	3	4
7	97	62	3	6	96	53	3	5	95	44	3	4	94	35	3	3	93	26	3	3
6	96	53	3	5	95	44	3	4	94	35	3	3	93	26	3	2	92	17	3	2
5	95	44	3	4	94	35	3	3	93	26	3	2	92	17	3	1	91	8	3	1
4	94	35	3	3	93	26	3	2	92	17	3	1	91	8	3	0	90	-1	3	0
3	93	26	3	2	92	17	3	1	91	8	3	0	90	-1	3	-1	89	-10	3	-1
2	92	17	3	1	91	8	3	0	90	-1	3	-1	89	-10	3	-2	88	-19	3	-2
1	91	8	3	0	90	-1	3	-1	89	-10	3	-2	88	-19	3	-3	87	-28	3	-3
0	90	-1	3	-1	89	-10	3	-2	88	-19	3	-3	87	-28	3	-4	86	-37	3	-4
-1	89	-10	3	-2	88	-19	3	-3	87	-28	3	-4	86	-37	3	-5	85	-46	3	-5
-2	88	-19	3	-3	87	-28	3	-4	86	-37	3	-5	85	-46	3	-6	84	-55	3	-6
-3	87	-28	3	-4	86	-37	3	-5	85	-46	3	-6	84	-55	3	-7	83	-64	3	-7
-4	86	-37	3	-5	85	-46	3	-6	84	-55	3	-7	83	-64	3	-8	82	-73	3	-8
-5	85	-46	3	-6	84	-55	3	-7	83	-64	3	-8	82	-73	3	-9	81	-82	3	-9
-6	84	-55	3	-7	83	-64	3	-8	82	-73	3	-9	81	-82	3	-10	80	-91	3	-10
-7	83	-64	3	-8	82	-73	3	-9	81	-82	3	-10	80	-91	3	-11	79	-100	3	-11
-8	82	-73	3	-9	81	-82	3	-10	80	-91	3	-11	79	-100	3	-12	78	-109	3	-12
-9	81	-82	3	-10	80	-91	3	-11	79	-100	3	-12	78	-109	3	-13	77	-118	3	-13
-10	80	-91	3	-11	79	-100	3	-12	78	-109	3	-13	77	-118	3	-14	76	-127	3	-14
-11	79	-100	3	-12	78	-109	3	-13	77	-118	3	-14	76	-127	3	-15	75	-136	3	-15
-12	78	-109	3	-13	77	-118	3	-14	76	-127	3	-15	75	-136	3	-16	74	-145	3	-16
-13	77	-118	3	-14	76	-127	3	-15	75	-136	3	-16	74	-145	3	-17	73	-154	3	-17
-14	76	-127	3	-15	75	-136	3	-16	74	-145	3	-17	73	-154	3	-18	72	-163	3	-18
-15	75	-136	3	-16	74	-145	3	-17	73	-154	3	-18	72	-163	3	-19	71	-172	3	-19
-16	74	-145	3	-17	73	-154	3	-18	72	-163	3	-19	71	-172	3	-20	70	-181	3	-20
-17	73	-154	3	-18	72	-163	3	-19	71	-172	3	-20	70	-181	3	-21	69	-190	3	-21
-18	72	-163	3	-19	71	-172	3	-20	70	-181	3	-21	69	-190	3	-22	68	-199	3	-22
-19	71	-172	3	-20	70	-181	3	-21	69	-190	3	-22	68	-199	3	-23	67	-208	3	-23
-20	70	-181	3	-21	69	-190	3	-22	68	-199	3	-23	67	-208	3	-24	66	-217	3	-24
-21	69	-190	3	-22	68	-199	3	-23	67	-208	3	-24	66	-217	3	-25	65	-226	3	-25
-22	68	-199	3	-23	67	-208	3	-24	66	-217	3	-25	65	-226	3	-26	64	-235	3	-26
-23	67	-208	3	-24	66	-217	3	-25	65	-226	3	-26	64	-235	3	-27	63	-244	3	-27
-24	66	-217	3	-25	65	-226	3	-26	64	-235	3	-27	63	-244	3	-28	62	-253	3	-28
-25	65	-226	3	-26	64	-235	3	-27	63	-244	3	-28	62	-253	3	-29	61	-262	3	-29
-26	64	-235	3	-27	63	-244	3	-28	62	-253	3	-29	61	-262	3	-30	60	-271	3	-30
-27	63	-244	3	-28	62	-253	3	-29	61	-262	3	-30	60	-271	3	-31	59	-280	3	-31
-28	62	-253	3	-29	61	-262	3	-30	60	-271	3	-31	59	-280	3	-32	58	-289	3	-32
-29	61	-262	3	-30	60	-271	3	-31	59	-280	3	-32	58	-289	3	-33	57	-298	3	-33
-30	60	-271	3	-31	59	-280	3	-32	58	-289	3	-33	57	-298	3	-34	56	-307	3	-34
-31	59	-280	3	-32	58	-289	3	-33	57	-298	3	-34	56	-307	3	-35	55	-316	3	-35
-32	58	-289	3	-33	57	-298	3	-34	56	-307	3	-35	55	-316	3	-36	54	-325	3	-36
-33	57	-298	3	-34	56	-307	3	-35	55	-316	3	-36	54	-325	3	-37	53	-334	3	-37
-34	56	-307	3	-35	55	-316	3	-36	54	-325	3	-37	53	-334	3	-38	52	-343	3	-38
-35	55	-316	3	-36	54	-325	3	-37	53	-334	3	-38	52	-343	3	-39	51	-352	3	-39
-36	54	-325	3	-37	53	-334	3	-38	52	-343	3	-39	51	-352	3	-40	50	-361	3	-40
-37	53	-334	3	-38	52	-343	3	-39	51	-352	3	-40	50	-361	3	-41	49	-370	3	-41
-38	52	-343	3	-39	51	-352	3	-40	50	-361	3	-41	49	-370	3	-42	48	-379	3	-42
-39	51	-352	3	-40	50	-361	3	-41	49	-370	3	-42	48	-379	3	-43	47	-388	3	-43
-40	50	-361	3	-41	49	-370	3	-42	48	-379	3	-43	47	-388	3	-44	46	-397	3	-44
-41	49	-370	3	-42	48	-379	3	-43	47	-388	3	-44	46	-397	3	-45	45	-406	3	-45
-42	48	-379	3	-43	47	-388	3	-44	46	-397	3	-45	45	-406	3	-46	44	-415	3	-46
-43	47	-388	3	-44	46	-397	3	-45	45	-406	3	-46	44	-415	3	-47	43	-424	3	-47
-44	46	-397	3	-45	45	-406	3	-46	44	-415	3	-47	43	-424	3	-48	42	-433	3	-48
-45	45	-406	3	-46	44	-415	3	-47	43	-424	3	-48	42	-433	3	-49	41	-442	3	-49
-46	44	-415	3	-47	43	-424	3	-48	42	-433	3	-49	41	-442	3	-50	40	-451	3	-50
-47	43	-424	3	-48	42	-433	3	-49	41	-442	3	-50	40	-451	3	-51	39	-460	3	-51
-48	42	-433	3	-49	41	-442	3	-50	40	-451	3	-51	39	-460	3	-52	38	-469	3	-52
-49	41	-442	3	-50	40	-451	3	-51	39	-460	3	-52	38	-469	3	-53	37	-478	3	-53
-50	40	-451	3	-51	39	-460	3	-52	38	-469	3	-53	37	-478	3	-54	36	-487	3	-54
-51	39	-460	3	-52	38	-469	3	-53	37	-478	3	-54	36	-487	3	-55	35	-496	3	-55
-52	38	-469	3	-53	37	-478	3	-54	36	-487	3	-55	35	-496	3	-56	34	-505	3	-56
-53	37	-478	3	-54	36	-487	3	-55	35	-496	3	-56	34	-505	3	-57	33	-514	3	-57
-54	36	-487	3	-55	35	-496	3	-56	34	-505	3	-57	33	-514	3	-58	32	-523	3	-58
-55	35	-496	3	-56	34	-505	3	-57	33	-514	3	-58	32	-523	3	-59	31	-532	3	-59
-56	34	-505	3	-57	33	-514	3	-58	32	-523	3	-59	31	-532	3	-60	30	-541	3	-60
-57	33	-514	3	-58	32	-523	3	-59	31	-532	3	-60	30	-541	3	-61	29	-550	3	-61
-58	32	-523	3	-59	31	-532	3	-60	30	-541	3	-61	29	-550	3	-62	28	-559	3	-62
-59	31	-532	3	-60	30	-541	3	-61	29	-550	3	-62	28	-559	3	-63	27	-568	3	-63
-60	30	-541	3																	

Table 4 (cont.)

A large grid of numerical data with multiple columns and rows, containing various numbers and labels such as 'M=100', 'M=1000', etc.

its mean value. Lorentz-polarization corrections were applied. However, absorption corrections were ignored as the variation in  $\mu R$  was between 0.03 and 0.14 with corresponding  $A^*$  values of 1.00 and 1.18 for the  $\theta$  range 0 to  $30^\circ$ . This was regarded as insignificant.

### Structure determination and refinement

In space group  $P\bar{1}$ , the Ni atom must lie on a centre of symmetry and the other atoms in general positions. All 22 non-hydrogen atoms were located in a Fourier synthesis phased on Ni at 0,0,0. Before refinement  $R = \sum(|F_o| - |F_c|) / \sum|F_o|$  was 0.371. Initially, atoms 16 and 17 in the imidazole ring were arbitrarily assigned as nitrogen and carbon respectively. After 4 cycles the isotropic refinement (*CRYLSQ*: X-RAY System, 1972) terminated at an  $R$  of 0.112. At this stage the bond lengths and angles within the imidazole ring were calculated (*BONDLA*: X-RAY System, 1972). Consideration of these, together with the temperature factors, indicated that atoms 16 and 17 had in fact been incorrectly assigned. They were accordingly interchanged and anisotropic temperature factors of the form:  $T = \exp[-2\pi^2(U_{11}h^2a^{*2} + U_{22}k^2b^{*2} + U_{33}l^2c^{*2} + 2U_{12}hka^*b^* + 2U_{13}hla^*c^* + 2U_{23}klb^*c^*)]$  were introduced into the refinement. After 2 cycles,  $R$  dropped to 0.070. All 21 hydrogen atoms were located in a difference synthesis and were assigned the isotropic temperature factors of the atoms to which they were bonded. After the final cycle of refinement the average e.s.d. in the positional and anisotropic temperature factors of the non-hydrogen atoms was more than 50 times the average parameter shift. The average e.s.d. in the hydrogen atomic positions was about five times the average parameter shift. The final  $R$  was 0.056. As a final check of the correctness of the structure, a difference synthesis with the structure factors calculated in the last cycle of refinement was computed. This map was practically featureless.

Scattering factors were obtained from Cromer & Mann (1968). The nickel was treated as  $Ni^0$  and the anomalous dispersion correction ( $\Delta f' = 0.4$  for Mo  $K\alpha$  radiation) was applied. Extinction was not regarded as being significant and no correction was made. Each reflexion was assigned unit weight.

The final positional and thermal parameters for the non-hydrogen atoms are given in Table 2 and the positional parameters of the hydrogen atoms in Table 3. The observed and calculated structure factors are listed in Table 4.

All computations were performed on a Univac 1106 computer.

### Description of the structure and discussion

The structure of the molecule is shown in Fig. 1 [*PLUTO*: Motherwell, W. S. D. (Cambridge). To be published]. The intramolecular bond lengths and angles and their associated e.s.d.'s are given in Tables 5 and 6

Table 5. Intramolecular bond lengths and their e.s.d.'s ( $\text{\AA}$ )

Ni—N(1)	2.078 (2)	N(3)—H(3)	0.78 (3)
N(1)—C(2)	1.362 (3)	C(7)—H(71)	0.97 (4)
C(2)—N(3)	1.392 (3)	C(7)—H(72)	0.92 (3)
N(3)—C(4)	1.361 (4)	C(8)—H(81)	0.92 (4)
C(4)—C(5)	1.529 (4)	C(8)—H(82)	0.96 (4)
C(5)—C(6)	1.513 (4)	C(8)—H(83)	0.91 (5)
C(6)—N(1)	1.331 (3)	C(9)—H(91)	0.98 (3)
C(2)—O(2)	1.221 (3)	C(9)—H(92)	0.95 (3)
C(4)—O(4)	1.210 (4)	C(10)—H(101)	0.91 (3)
C(6)—O(6)	1.258 (3)	C(10)—H(102)	0.98 (4)
C(5)—C(7)	1.546 (5)	C(11)—H(11)	1.00 (3)
C(7)—C(8)	1.515 (6)	C(12)—H(121)	0.97 (6)
C(5)—C(9)	1.543 (4)	C(12)—H(122)	0.98 (4)
C(9)—C(10)	1.522 (6)	C(12)—H(123)	0.98 (5)
C(10)—C(11)	1.526 (5)	C(13)—H(131)	1.01 (4)
C(11)—C(12)	1.518 (6)	C(13)—H(132)	1.03 (4)
C(11)—C(13)	1.511 (8)	C(13)—H(133)	0.87 (5)
Ni—N(14)	2.055 (3)	C(15)—H(15)	0.89 (3)
N(14)—C(15)	1.369 (4)	C(16)—H(16)	0.75 (5)
C(15)—C(16)	1.338 (6)	N(17)—H(17)	0.97 (4)
C(16)—N(17)	1.382 (5)	C(18)—H(18)	0.93 (3)
N(17)—C(18)	1.328 (6)		
C(18)—N(14)	1.327 (4)		
Ni—O(6)	2.226 (2)		

Table 6. Intramolecular bond angles and their e.s.d.'s ( $^\circ$ )

N(1)—Ni—N(1 <sup>4</sup> )	180.0 (1)	C(4)—C(5)—C(7)	108.0 (3)
N(1)—Ni—N(14)	89.3 (1)	C(6)—C(5)—C(7)	109.2 (2)
N(1)—Ni—N(14 <sup>1</sup> )	90.7 (1)	C(5)—C(7)—C(8)	114.8 (3)
N(14)—Ni—N(14 <sup>1</sup> )	180.0 (1)	C(4)—C(5)—C(9)	108.5 (2)
Ni—N(1)—C(2)	144.1 (2)	C(6)—C(5)—C(9)	109.3 (2)
N(1)—C(2)—N(3)	118.1 (2)	C(7)—C(5)—C(9)	109.8 (2)
C(2)—N(3)—C(4)	126.2 (2)	C(5)—C(9)—C(10)	114.6 (2)
N(3)—C(4)—C(5)	117.6 (2)	C(9)—C(10)—C(11)	114.3 (3)
C(4)—C(5)—C(6)	112.0 (2)	C(10)—C(11)—C(12)	111.5 (3)
C(5)—C(6)—N(1)	123.6 (2)	C(10)—C(11)—C(13)	111.2 (4)
C(6)—N(1)—C(2)	122.4 (2)	C(12)—C(11)—C(13)	111.3 (4)
C(6)—N(1)—Ni	93.5 (1)	Ni—N(14)—C(15)	128.0 (2)
N(1)—C(2)—O(2)	122.0 (2)	N(14)—C(15)—C(16)	110.3 (3)
N(3)—C(2)—O(2)	119.9 (2)	C(15)—C(16)—N(17)	106.3 (4)
N(3)—C(4)—O(4)	120.9 (2)	C(16)—N(17)—C(18)	106.6 (3)
C(5)—C(4)—O(4)	121.5 (3)	N(17)—C(18)—N(14)	112.0 (3)
C(5)—C(6)—O(6)	120.1 (2)	C(18)—N(14)—Ni	126.7 (2)
N(1)—C(6)—O(6)	116.3 (2)	C(18)—N(14)—C(15)	104.9 (3)

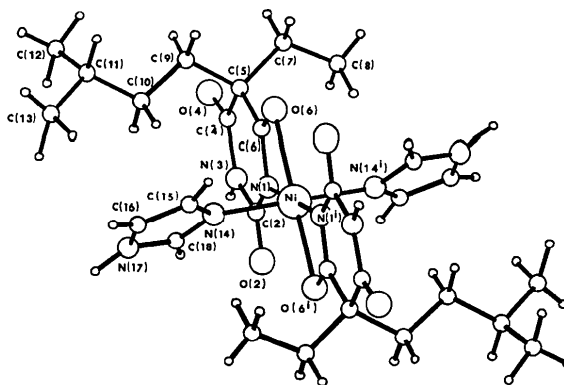


Fig. 1. The molecular structure of the complex. The superscripted atoms are centrosymmetrically related to the corresponding atoms without superscripts.

respectively (*BONDLA: X-RAY System*, 1972). Table 7 lists computed least-squares planes with their equations and the distances of various atoms from these planes.

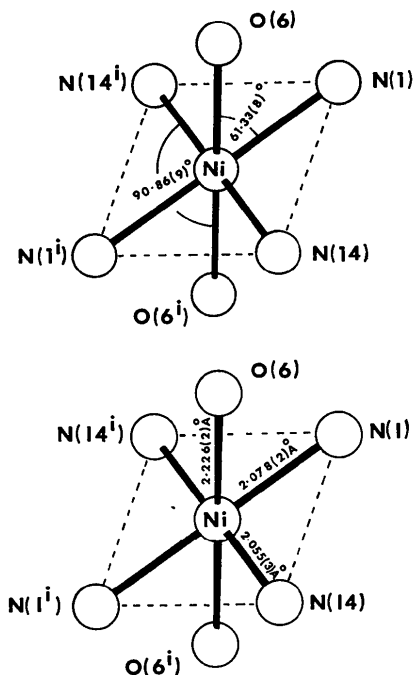


Fig. 2. The nickel coordination sphere.

#### The nickel coordination sphere

The nickel atom is involved in distorted octahedral coordination and is situated in the centre of a plane defined by the deprotonated donor nitrogen atoms N(1) and N(1<sup>i</sup>) of the barbiturate anions and the donor nitrogens N(14) and N(14<sup>i</sup>) of the imidazole moieties (plane I). The octahedron is completed by two centrosymmetric barbiturato oxygen atoms, O(6) and O(6<sup>i</sup>) which lie above and below this plane.

The Ni–N bond lengths are 2.078 (2) and 2.055 (3) Å for the barbiturate and imidazole ligands respectively. These may be compared with the Ni–N distances of 2.107 (3) and 2.082 (3) Å reported for the structure diaquobis-(2,2'-biimidazole)nickel(II) dinitrate (Mighell, Reimann & Mauer, 1969).

The Ni–O bond length is 2.226 (2) Å. In the structure reported by Mighell *et al.* the water oxygens occupy the octahedral positions at a distance of 2.103 (3) Å while in the structure diaquobisglycinatonickel(II) (Freeman, Gus & Sinclair, 1968) the Ni–O bond distance is 2.10 (1) Å.

The bond angles N(1)–Ni–N(14) and N(1)–Ni–N(14<sup>i</sup>) with values 89.3 (1) and 90.7 (1)° respectively indicate regular geometry. However, the distorted octahedral configuration arises from the positions of O(6) and O(6<sup>i</sup>) which lie off the normal to plane I (*i.e.* off the 'Z axis'). This may be seen from the angles N(1)–Ni–O(6) and N(14)–Ni–O(6) which are 61.33 (8) and 89.14 (9)° respectively while the 'off the Z axis angle' is 28.73°.

Table 7. Least-squares planes

The equations of the planes are expressed in orthogonalized space as  $PI + QJ + RK = S$ .

Plane I	Square planar coordination around the nickel atom								
Atoms defining the plane	Ni	N(1)	N(14)	N(1 <sup>i</sup> )	N(14 <sup>i</sup> )				
Atoms not included in the plane						O(2)	O(6)		
Distance from the plane, Å	0.0	0.0	0.0	0.0	0.0	–2.016	1.952		
Equation	0.71545I + 0.13755J + 0.68499K = 0.00								
Plane II	Through the 5 atoms of the imidazole ring								
Atoms defining the plane	N(14)	C(15)	C(16)	N(17)	C(18)				
Distance from the plane, Å	–0.001	0.001	–0.002	0.001	0.000				
Equation	0.78785I – 0.42039J + 0.45007K = 0.19227								
Plane III	Through the 6 pyrimidine ring atoms								
Atoms defining the plane	N(1)	C(2)	N(3)	C(4)	C(5)	C(6)			
Atoms not included in the plane							O(2)	O(4)	O(6)
Distance from the plane, Å	0.003	0.006	–0.009	0.003	0.005	–0.008	0.019	–0.001	–0.011
Equation	0.64340I + 0.25957J – 0.72018K = –0.05739								
Plane IV	Through the 9 atoms of the trioxypyrimidine ring								
Atoms defining the plane	N(1)	C(2)	N(3)	C(4)	C(5)	C(6)	O(2)	O(4)	O(6)
Distance from the plane, Å	0.001	0.000	–0.014	0.003	0.010	–0.004	0.008	0.000	–0.004
Equation	0.64601I + 0.26122J – 0.71724K = –0.05807								
Plane V	Through the atoms of the hydrocarbon chain attached at C(5)								
Atoms defining the plane	C(5)	C(7)	C(8)	C(9)	C(10)	C(11)			
Atoms not included in the plane							C(12)	C(13)	
Distance from the plane, Å	–0.020	0.055	–0.030	–0.027	0.049	–0.026	–1.147	0.426	
Equation	–0.21224I + 0.95481J + 0.20809K = –3.16104								
Intersection angles (°)	I & II	35.50	II & III	85.78					
	I & III	89.85	III & IV	0.25					
	I & IV	89.61	III & V	87.79					

Fig. 2 is an illustration of the nickel coordination sphere. Table 8 lists all relevant bond lengths and angles.

Table 8. Bond lengths ( $\text{\AA}$ ) and angles ( $^\circ$ ) and their *e.s.d.*'s in the nickel coordination sphere

Ni—N(1)	2.078 (2)	N(1)—Ni—O(6)	61.33 (8)
Ni—N(14)	2.055 (3)	N(14)—Ni—O(6)	90.86 (9)
Ni—O(6)	2.226 (2)	N(1)—Ni—O(6 <sup>1</sup> )	118.67 (8)
N(1)—O(6)	2.199 (3)	N(14)—Ni—O(6 <sup>1</sup> )	89.14 (9)
N(14)—O(6)	3.052 (4)	O(6)—N(1)—N(14)	71.92 (9)
N(14 <sup>1</sup> )—O(6)	3.006 (4)	O(6)—N(1)—N(14 <sup>1</sup> )	69.94 (9)
N(1)—N(14)	2.906 (4)	O(6)—N(14)—N(1)	43.23 (7)
N(1)—N(14 <sup>1</sup> )	2.939 (4)	O(6)—N(14)—N(1 <sup>1</sup> )	76.33 (9)

#### The imidazole ring

The bond lengths within the ring agree within experimental error with those reported by Wang & Craven (1971) for the bis-(5,5'-diethylbarbiturato)bisimidazole complex of Zn(II) and with those reported by Lundberg (1966) for the complex diimidazolezinc(II) dichloride. The ring is planar, the greatest deviation from the least-squares plane through the five atoms (plane II) being that of C(16) at a distance of 0.002  $\text{\AA}$ . This plane and plane I intersect at an angle of 35.50°.

#### The barbiturate ligand structure

The bond lengths for the barbiturate ligand show good agreement with those reported for two polymorphs of the parent 5-ethyl-5-isoamylbarbituric acid (Craven & Vizzini, 1969). In this latter compound, the internal ring angle C(2)—N(1)—C(6) was found to have a mean value of 126.6 (3)° while in the present complex it is 122.4 (2)°. Thus the result of deprotonating N(1) in forming the complex is a closing of the angle despite the fact that N(1) is bonded to nickel.

The pyrimidine ring is planar (plane III), the largest deviation being that of atom N(3) at a distance of -0.009  $\text{\AA}$ . When the three exocyclic oxygen atoms are included in the calculation, it is seen that the entire trioxypyrimidine ring is planar (plane IV) the largest deviation being that of N(3), again, at a distance of -0.014  $\text{\AA}$ . The coplanarity of the exocyclic oxygen atoms and the pyrimidine ring atoms is further illustrated by the small angle of intersection of planes III and IV (0.25°). Planes I and III intersect at 89.85°, *i.e.* the plane Ni, N(1), N(14) and the pyrimidine ring plane are perpendicular. Similarly the planes through the two different moieties of the molecule, *i.e.* through the imidazole ring (plane II) and the pyrimidine ring

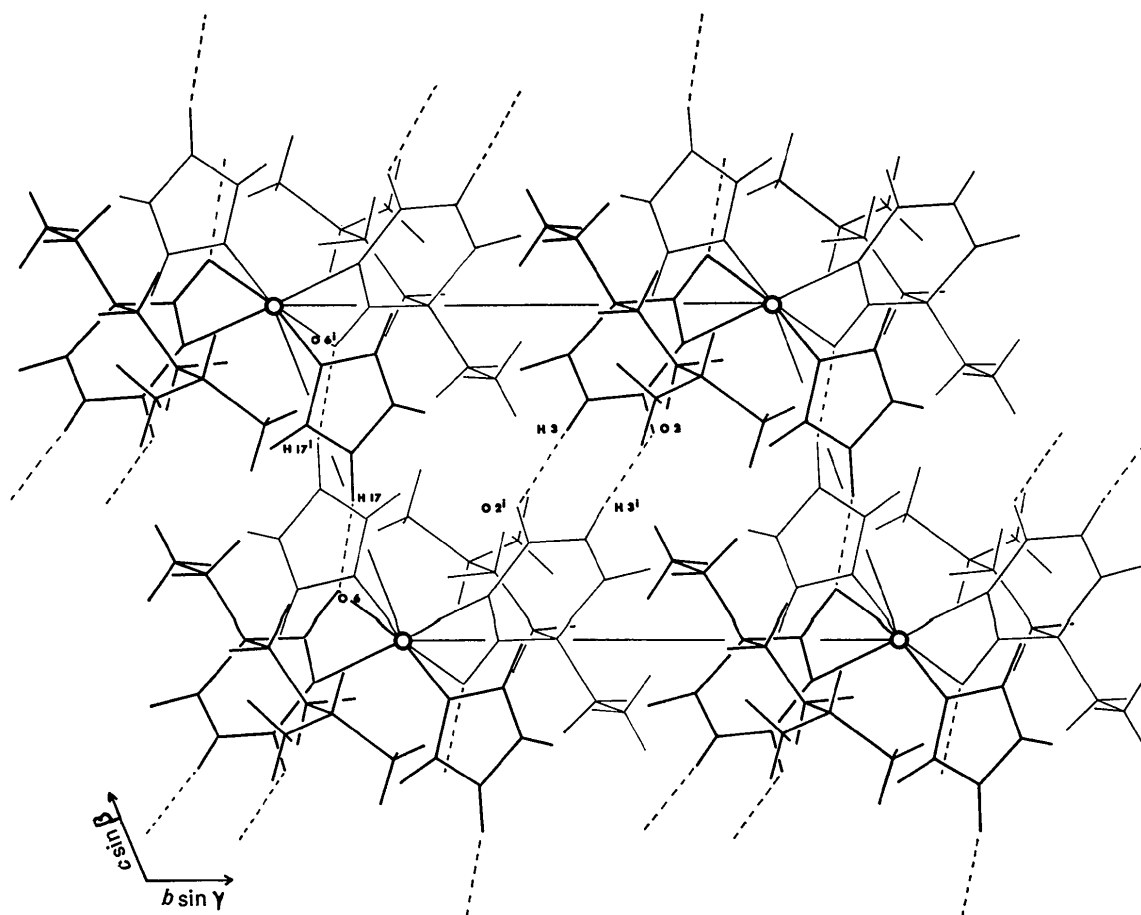


Fig. 3. The [100] projection of the structure. The intermolecular hydrogen bonding is represented by dotted lines.



(plane III), are approximately normal to each other ( $85.78^\circ$ ). The hydrocarbon chain from C(8) through C(5) to C(11) forms a plane (plane V) which is almost normal ( $87.79^\circ$ ) to the pyrimidine ring plane (plane III). This is consistent with that observed in the parent acid.

#### Hydrogen bonding

The intermolecular hydrogen bonding is shown as dotted lines in Figs. 3 and 4, the [100] and [010] projections of the structure respectively while the geometry of the interactions is illustrated in Fig. 5. All relevant bond lengths and angles are listed in Table 9.

Each discrete molecule of the complex is linked to four others by  $N-H \cdots O$  hydrogen bonds arising from the interaction of the imidazole NH and barbiturate NH groups with barbiturate oxygen atoms.

Table 9. Bond lengths ( $\text{\AA}$ ) and angles ( $^\circ$ ) and their *e.s.d.*'s involved in the hydrogen bonding

H(3)—O(2 <sup>1</sup> )	2.037 (3)
N(3)—O(2 <sup>1</sup> )	2.878 (4)
N(3)—H(3)—O(2 <sup>1</sup> )	176.7 (2)
H(17)—O(6)	1.961 (2)
N(17)—O(6)	2.935 (4)
N(17)—H(17)—O(6)	161.6 (2)

H(17) of the molecule at  $x, y, z$  is hydrogen bonded to O(6) of the molecule at  $x, y, z-1$ . As a result of centrosymmetry, H(17<sup>1</sup>) of the molecule at  $x, y, z$  is hydrogen bonded to O(6<sup>1</sup>) of the molecule at  $x, y, z+1$ . These bonds form a double linkage joining adjacent molecules along  $c$  thereby generating an infinite chain of hydrogen-bonded molecules in the  $z$  direction. The parallel chains are themselves joined by hydrogen bonds. H(3)

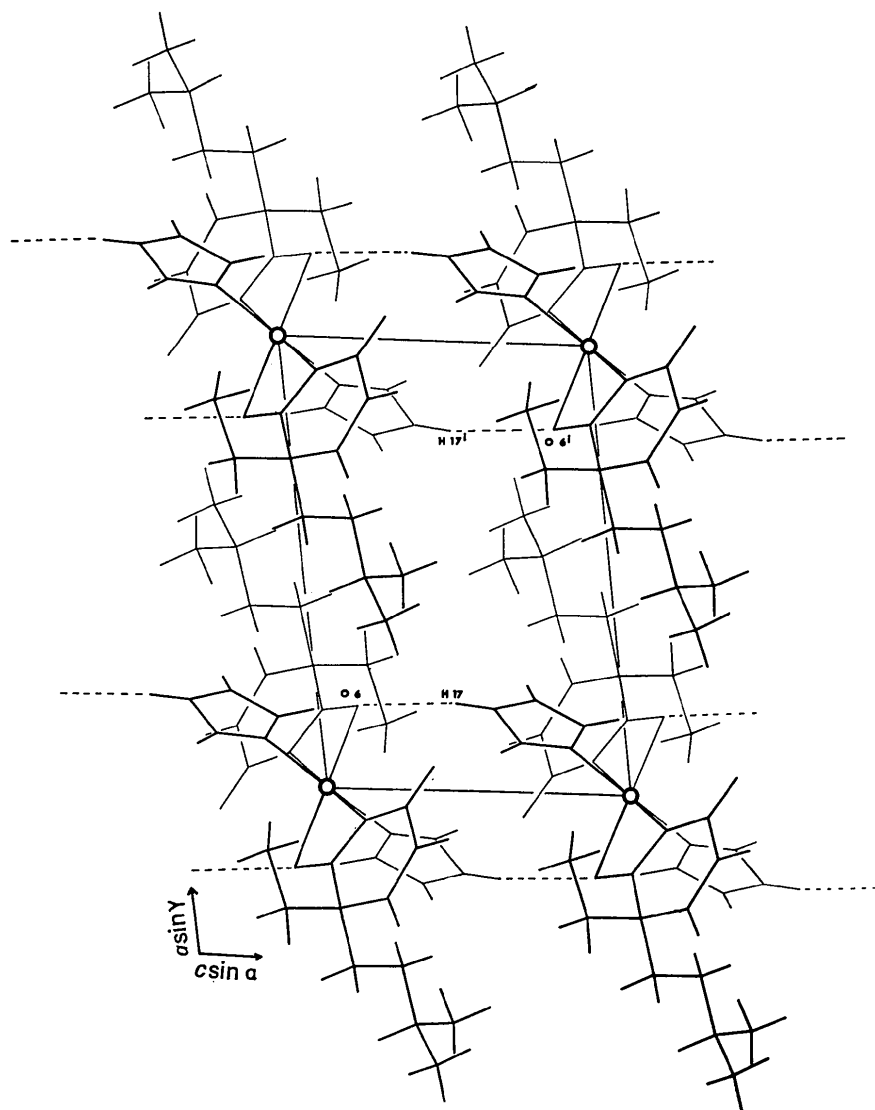


Fig. 4. The [010] projection of the structure. The intermolecular hydrogen bonding is represented by dotted lines.

and O(3) of the molecule at  $x, y, z$  are linked to O(2<sup>1</sup>) and H(3<sup>1</sup>) respectively of the molecule at  $x, y-1, z-1$ . These bonds occur as a double bridging linkage in the centre of the  $yz$  plane and approximately parallel to the short diagonal of the  $bc$  face.

There are thus only two crystallographically distinct hydrogen bonds, N(17)–H(17)···O(6) and N(3)–H(3)···O(2<sup>1</sup>). However, each molecule has associated with it a total of eight hydrogen bonds which emanate as four centrosymmetric pairs.

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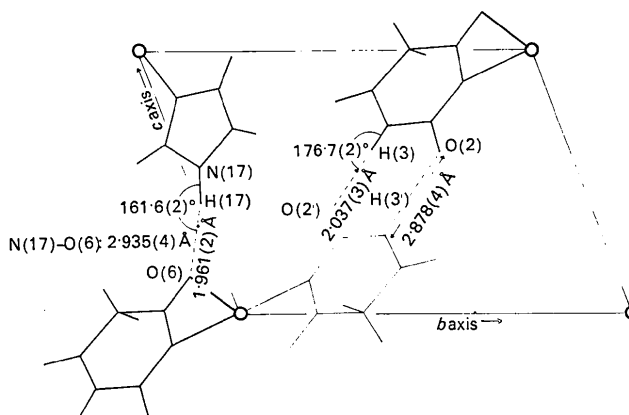


Fig. 5. The geometry of the hydrogen-bond interactions.

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## Structure du Sulfate d'Ammonium et de Glycinium $NH_4NH_3CH_2COOHSO_4$

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The crystal structure of ammonium glycine sulphate (NGS) has been solved from three-dimensional data collected on a Weissenberg camera with  $Cu K\alpha$  radiation. The crystal of NGS is monoclinic, space group  $P2_1/c$  with  $a = 8.262$  (7),  $b = 10.073$  (8),  $c = 8.632$  (8) Å,  $\beta = 92.66$  (5)° and  $Z = 4$ . The structure has been refined to give an  $R$  index of 0.079 for 922 independent reflexions. The glycine molecule is a monoprotonated ion. Thus the chemical formula should be written  $NH_4^+(NH_3CH_2COOH)^+SO_4^{2-}$ . The glycine ion is not planar.

### Introduction

Il semble que la présence de très courtes liaisons hydrogène O–H···O (ou O–H···F), soit entre deux groupements glycine, soit entre un groupement glycine et un atome d'oxygène (ou de fluor) de l'anion, joue un rôle important dans le comportement ferroélectrique des composés de glycine, en particulier pour le sulfate et

fluorobéryllate de triglycine et le nitrate de diglycine (Sato, 1968; Warkusz & Łukaszewicz, 1973).

Nous avons préparé de nouveaux composés contenant un seul groupement glycine: il s'agit des sulfates et fluorobéryllates de glycine et d'ammonium, de glycine et de potassium (Vilminot & Cot, 1973).

L'objet de cette publication est la présentation de la structure à 22°C de l'un d'entre eux: le sulfate double