

# Crystal Structure of *N,N'*-Bis(dodecyl)diaza-18-crown-6 Complexed Sodium Perchlorate

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(Received: 7 January 1997; in final form: 27 March 1997)

**Abstract.** The crystal structure of a new complex of a diaza-crown ether having two side arms has been determined from X-ray diffraction data. The compound crystallizes in space group  $P\bar{1}$  with cell dimensions  $a = 9.982(1)$ ,  $b = 10.685(1)$ ,  $c = 20.376(2)\text{Å}$ ,  $\alpha = 81.09(1)$ ,  $\beta = 80.92(1)$ ,  $\gamma = 88.43(1)^\circ$ ,  $Z = 2$ . The structure has been solved by direct methods and refined to the final  $R$  value of 0.053 for 3458 observed reflections and 424 parameters. The diaza-18-crown-6 ligand adopts an approximate  $D_{3d}$  conformation. The  $\text{Na}^+$  ion is held inside the molecular cavity of this macrocyclic ligand and a  $\text{ClO}_4^-$  oxygen coordinates with  $\text{Na}^+$ . The average Na—O (18-crown-6) and Na—N bond lengths are 2.426(4) and 2.786(5)Å, respectively; the Na—O ( $\text{ClO}_4^-$ ) bond length is 2.472(4)Å. The mean cavity radius is 1.10 Å and the N···N nonbonding distance is 4.605(6)Å.

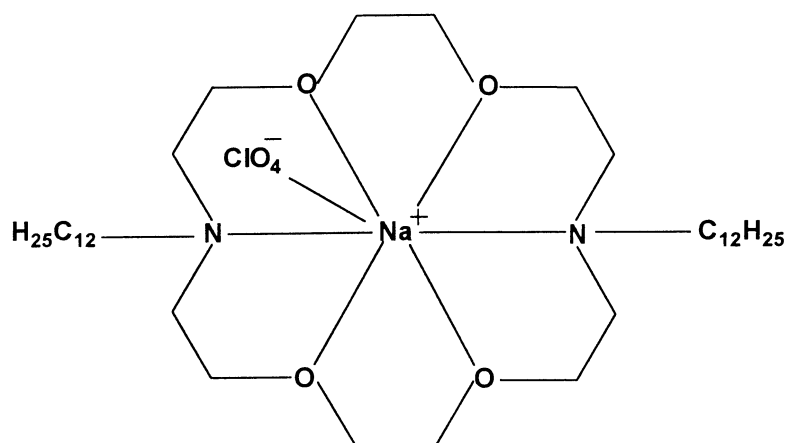
**Key words:** Crown ether, macrocycles, diaza-18-crown-6, sodium complex.

## 1. Introduction

Since Pedersen reported the synthesis and cation complexing characteristics of the crown ethers [1], there has been increasing interest in these compounds as complexing agents for various inorganic and organic cations. Many different modifications of the crown ethers, such as changing the ring size, the kind of substituents, and the type of donor atoms, have been made to enhance their complexing properties [2–6]. The aza-crowns also have complexation properties that are intermediate between those of the all-oxygen crown ligands, which complex strongly with alkaline earth metal ions, and those of the all-nitrogen cyclams which complex strongly with the heavy metal cations [3, 6]. In general, preferential complexation results when the relative sizes of cations and ligand cavities are matched. Cation binding studies [7, 8] have shown that the hole-size relationship fails to explain cation selectivity in flexible macrocycles and lariat ethers. Instead, the key factor is the number of donors and the effective ionic radii [9].

Here we report the crystal structure of a new complex of a diaza-crown ether having two side arms (**1**). The present investigation was carried out to establish the

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(1)

Scheme 1.

three-dimensional structure of this compound and hence obtain the bonding and ligand behaviour in more detail.

## 2. Experimental

The synthesis and purification of the ligand, *N,N'*-di-*n*-dodecyl-4,13-diaza-1,7,10,16-tetraoxa-cyclooctadecane, have been reported previously [10]. The NaClO<sub>4</sub> complex with this ligand (1) was prepared by adding excess NaClO<sub>4</sub>·H<sub>2</sub>O to a methylene chloride solution of the ligand. After being stirred at room temperature for approximately 1 h, the solution was filtered and concentrated in vacuo. Recrystallization from MeCN and then from ethylacetate provided the desired complex with a melting point of 88–89 °C.

Intensity data were collected from a crystal of dimensions 0.24 × 0.16 × 0.64 mm on an Enraf-Nonius CAD-4 diffractometer [11] using graphite-monochromated MoK<sub>α</sub> radiation (λ = 0.71073 Å) using the ω – 2θ scan technique. Accurate cell dimensions were obtained by the least-squares analysis of the setting angles of 21 reflections in the range 10 ≤ θ ≤ 18°. Three control reflections, which were monitored every two hours, showed slight decrease in intensity during data collection. Orientation control was performed every 200 reflections. Data were corrected for Lp effects and for absorption using the ψ-scan data.

The structure was solved by direct methods using the program SIR in the MoIEN program package [12], and refined by full-matrix least squares minimizing

$\sum w(F_o - F_c)^2$ . The refinement was done with anisotropic temperature factors for all non-H atoms. At this stage, high thermal motion was noted for the perchlorate oxygen atoms and the first three carbon atoms of the dodecyl unit attached to N(13). It was not possible to resolve a disorder model for oxygen atoms but resolution of the disorder for carbon atoms was possible. The C(13')–C(14') and C(14')–C(15') distances were restrained to the values found in the dodecyl unit attached to N(4) by using restrained least-squares techniques in MoIEN. The coordinates of C(13'), C(14') and C(15') atoms were fixed during the full-matrix refinement. All H-atoms were then geometrically located 0.95 Å from their parent atoms and not refined; displacement parameters were fixed at 1.3  $U_{eq}$  of the parent C atom.

Final refinement converged to  $R = 0.053$  for 3458 observed reflections and 424 parameters. Crystal data and a summary of intensity data collection and structure refinement are given in Table I.

### 3. Results and Discussion

The molecular structure of the compound (**1**) is illustrated in Figure 1 which shows the numbering scheme employed. Table II lists the final atomic parameters. Bond lengths, bond angles and torsion angles are given in Tables III and IV, respectively. In this crystal structure, the macrocyclic ligand 1,7,10,16-tetraoxa-4,13-diaza cyclooctadecane (diaz-18-crown-6) adopts an approximate  $D_{3d}$  conformation which is most commonly observed for 18-crown-6 and diaza-18-crown-6 [13–15], with C—O—C—C and O—C—C—O torsion angles near 180° and 70°. Slight differences in binding requirements lead to deviations from this idealized geometry (see Table IV). Some torsion angles are very different from the staggered one, especially around the nitrogen atoms. The macrocycle oxygen atoms are on average 0.151(3) Å alternately above and below the oxygen mean plane. The distances of N(4) and N(13) from the mean plane of the oxygen atoms are –1.026(4) and –2.248(4) Å, respectively. The sodium ion is situated –0.133(2) Å below the mean plane of the four ring oxygen atoms.

The mean values of the bond lengths and angles in the ligand are similar to those found in related ligands [14,16,17]. The values are C—O 1.416(6), C—N 1.466(6), C—C 1.490(7) Å; O—C—C 109.0(5), N—C—C 112.9(5), C—O—C 114.1(4) and C—N—C 112.2(4)°. The N···N nonbonding distance is 4.605(6) Å and the N—Na—N angle is 111.5(1)°. The O···O and N···O contacts also have standard values (see Table III).

In the complex, the sodium ion is heptacoordinated. The average values for the metal–donor atom distances are 2.426(4) (Na<sup>+</sup>···O) and 2.786(5) Å (Na<sup>+</sup>···N). The coordination shell of the sodium ion is completed by a perchlorate oxygen, O(4), which is located over the plane in an apical position. In addition to metal–donor distances, a complex's mean cavity radius  $R$ , as defined by Mathieu *et al.* [16] can be determined from the structural data. This value is calculated by averaging the differences between the metal–donor distance and the donor's covalent radius,

Table I. Crystal data and summary of intensity data collection and structure refinement.

Formula	C <sub>36</sub> H <sub>74</sub> N <sub>2</sub> O <sub>8</sub> NaCl
Color/Shape	colourless/needle
Formula wt.	721.44
Space group	$P\bar{1}$
Temp., °C	23
Cell Constants(No. reflections > $\theta$ used)	21 reflections > 10°
$a$ , Å	9.982(1)
$b$ , Å	10.683(1)
$c$ , Å	20.376(2)
$\alpha$ , deg	81.09(1)
$\beta$ , deg	80.92(1)
$\gamma$ , deg	88.43(1)
Cell vol, Å <sup>3</sup>	2120.2
Formula units/unit cell	2
$D_{calc}$ , g cm <sup>-3</sup>	1.13
$\mu_{calc}$ , cm <sup>-1</sup>	1.4
Diffractometer/Scan	Enraf Nonius CAD4/ $\omega-2\theta$
Range of relative transm. factors, %	98.2–99.9
Radiation, graphite monochromator	MoK $\alpha$ $\lambda = 0.71073$ Å
Max. crystal dimensions, mm	0.16 × 0.24 × 0.64
Scan width	0.6 + 1.24 tan $\theta$
Standard reflections	3 measured every 120 min.
Decay of standards	3.5%
Reflections measured	7094
2 $\theta$ range, deg	2–48.1
Range of $h, k, l$	$-11 \leq h \leq 11, -12 \leq k \leq 0, -23 \leq l \leq 23$
Reflections observed	3458 [ $F_o \geq 3\sigma(F_o)$ ]
Corrections applied	Lorentz-pol., linear decay, empirical absorption
Structure solution	Direct methods
Treatment of hydrogen atoms	located and not refined except H6
No. of parameters varied	424
Weights	$4F_o/\sigma(F_o)$
GOF	1.16
$R = \sum   F_o  -  F_c   / \sum  F_o $	0.0535
$R_w$	0.0531
Largest feature final diff. map, e Å <sup>-3</sup>	0.56

1.4 Å(O) and 1.5 Å(N).  $R$  is equal to 1.10 Å and corresponds nicely to the effective ionic radius, 1.12 Å, for heptacoordinated Na<sup>+</sup> [9].

In the perchlorate ion, the O—Cl—O bond angles are close to the tetrahedral value (average 109.23(3)Å) and the Cl—O bond distances average 1.409(4)Å. The dodecyl groups attached to N(4) and N(13) are oriented so that the torsion angles

Table II. Fractional atomic coordinates and equivalent isotropic displacement parameters ( $\text{\AA}^2$ ).
$$B_{\text{eq}} = \frac{8\pi^2}{3} \sum_i \sum_j U_{ij} a_i^* a_j^* \mathbf{a}_i \cdot \mathbf{a}_j$$

Atom	$x/a$	$y/b$	$z/c$	$B_{\text{eq}}$
Cl	0.2650(1)	0.2955(1)	0.91235(7)	5.41(3)
Na	0.2011(2)	0.1663(2)	1.09416(9)	4.59(4)
O(1)	0.0134(3)	0.3017(3)	1.0773(2)	5.25(8)
O(2)	0.1366(4)	0.3094(3)	0.8918(2)	8.1(1)
O(3)	0.3015(4)	0.4111(4)	0.9286(2)	9.4(1)
O(4)	0.2593(4)	0.1998(3)	0.9702(2)	7.2(1)
O(5)	0.3590(5)	0.2586(4)	0.8604(2)	11.1(1)
O(7)	0.4261(3)	0.2352(3)	1.1068(2)	5.82(9)
O(10)	0.3660(3)	0.0009(3)	1.0794(2)	5.45(8)
O(16)	0.0045(3)	0.0458(3)	1.0826(2)	4.61(7)
N(4)	0.2033(4)	0.3912(3)	1.1493(2)	4.81(9)
N(13)	0.1437(4)	-0.0412(4)	1.1932(2)	5.4(1)
C(1')	0.1298(5)	0.3849(4)	1.2173(3)	5.6(1)
C(2')	0.1021(6)	0.5093(5)	1.2458(3)	6.0(1)
C(2)	-0.0002(5)	0.4261(4)	1.0939(3)	5.5(1)
C(3')	0.0299(6)	0.4879(5)	1.3177(3)	6.0(1)
C(3)	0.1373(5)	0.4710(4)	1.0987(2)	5.4(1)
C(4')	-0.0214(6)	0.6086(5)	1.3438(3)	6.1(1)
C(5')	-0.1009(6)	0.5914(5)	1.4135(3)	6.0(1)
C(5)	0.3465(5)	0.4257(5)	1.1444(3)	6.7(1)
C(6)	0.4370(5)	0.3687(5)	1.0907(3)	7.0(2)
C(6')	-0.1574(6)	0.7126(5)	1.4364(3)	6.3(1)
C(7')	-0.2400(6)	0.6971(5)	1.5056(3)	6.2(1)
C(8')	-0.2992(6)	0.8189(5)	1.5277(3)	6.5(1)
C(8)	0.5188(5)	0.1695(5)	1.0638(3)	7.1(2)
C(9')	-0.3798(6)	0.8039(5)	1.5975(3)	6.7(2)
C(9)	0.4990(5)	0.0319(5)	1.0869(3)	7.0(2)
C(10')	-0.4402(6)	0.9254(6)	1.6189(3)	7.0(2)
C(11')	-0.5204(7)	0.9110(6)	1.6889(3)	8.2(2)
C(11)	0.3228(6)	-0.1206(5)	1.1132(3)	6.6(1)
C(12')	-0.5839(7)	1.0319(7)	1.7078(4)	10.1(2)
C(12)	0.2666(6)	-0.1187(5)	1.1852(3)	6.8(2)
C(13')	0.129	-0.018	1.264	10.7(2)
C(14')	0.007	0.042	1.290	10.4(2)
C(14)	0.0246(5)	-0.1031(5)	1.1785(3)	6.1(1)
C(15')	0.010	0.076	1.362	9.0(2)
C(15)	-0.0678(5)	-0.0135(5)	1.1446(2)	5.7(1)
C(16')	-0.1205(7)	0.1270(6)	1.3877(3)	8.0(2)
C(17')	-0.1254(6)	0.1776(5)	1.4527(3)	6.9(2)
C(17)	-0.0745(5)	0.1263(4)	1.0417(2)	5.4(1)
C(18)	-0.1090(5)	0.2491(5)	1.0677(3)	5.8(1)
C(18')	-0.2614(6)	0.2291(6)	1.4784(3)	7.4(2)

Table II. Continued.

$$B_{\text{eq}} = \frac{8\pi^2}{3} \sum_i \sum_j U_{ij} a_i^* a_j^* \mathbf{a}_i \cdot \mathbf{a}_j$$

Atom	$x/a$	$y/b$	$z/c$	$B_{\text{eq}}$
C(19')	-0.2678(6)	0.2808(5)	1.5441(3)	7.1(2)
C(20')	-0.4043(6)	0.3339(6)	1.5701(3)	7.5(2)
C(21')	-0.4081(6)	0.3859(6)	1.6353(3)	7.2(2)
C(22')	-0.5444(6)	0.4407(6)	1.6611(3)	7.6(2)
C(23')	-0.5456(6)	0.4984(6)	1.7239(3)	7.9(2)
C(24')	-0.6813(7)	0.5538(7)	1.7484(4)	9.7(2)

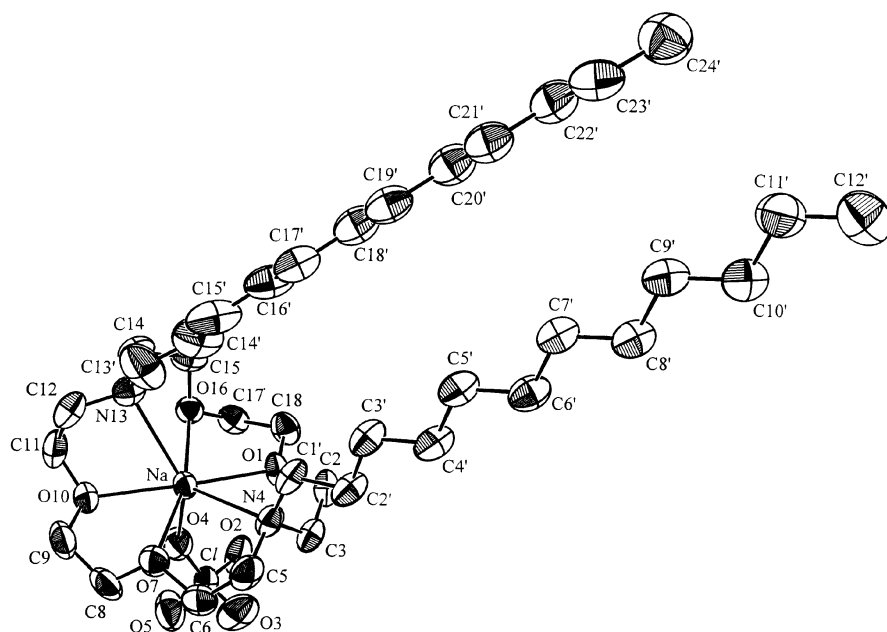


Figure 1. Drawing of the compound with 50% probability displacement ellipsoids and the numbering scheme. H atoms are not shown for clarity.

C(3)—N(4)—C(1')—C(2') and C(14)—N(13)—C(13')—C(14') are  $-58.3(5)$  and  $-49.9(4)^\circ$  respectively.

### Acknowledgements

The authors wish to acknowledge the purchase of the CAD4 diffractometer under Grant DPT/TBAG-1 of The Scientific and Technical Research Council of Turkey.

Table III. Selected geometric parameters ( $\text{\AA}$ ,  $^\circ$ ).

Cl—O(2)	1.408(4)	N(13)—C(14)	1.469(6)
Cl—O(3)	1.399(4)	C(1')—C(2')	1.531(7)
Cl—O(4)	1.431(4)	C(2')—C(3')	1.515(7)
Cl—O(5)	1.397(4)	C(2)—C(3)	1.488(7)
Na—O(1)	2.371(3)	C(3')—C(4')	1.516(7)
Na—O(4)	2.472(4)	C(4')—C(5')	1.501(7)
Na—O(7)	2.447(4)	C(5')—C(6')	1.509(8)
Na—O(10)	2.403(4)	C(5)—C(6)	1.501(7)
Na—O(16)	2.438(4)	C(6')—C(7')	1.505(8)
O(1)—C(2)	1.419(5)	C(7')—C(8')	1.516(8)
O(1)—C(18)	1.413(5)	C(8')—C(9')	1.508(8)
O(7)—C(6)	1.417(6)	C(8)—C(9)	1.481(7)
O(7)—C(8)	1.419(6)	C(9')—C(10')	1.513(8)
O(10)—C(9)	1.414(6)	C(10')—C(11')	1.509(8)
O(10)—C(11)	1.416(6)	C(11')—C(12')	1.501(9)
O(16)—C(15)	1.417(6)	C(11)—C(12)	1.489(7)
O(16)—C(17)	1.416(6)	C(13')—C(14')	1.425
N(4)—C(1')	1.454(7)	C(14')—C(15')	1.565
N(4)—C(3)	1.463(6)	C(14)—C(15)	1.480(7)
N(4)—C(5)	1.470(6)	C(15')—C(16')	1.451(8)
N(13)—C(12)	1.463(7)	C(16')—C(17')	1.500(8)
N(13)—C(13')	1.492(4)	C(17')—C(18')	1.498(9)
C(17)—C(18)	1.501(6)	C(21')—C(22')	1.516(9)
C(18')—C(19')	1.518(8)	C(22')—C(23')	1.503(9)
C(19')—C(20')	1.511(9)	C(23')—C(24')	1.506(9)
C(20')—C(21')	1.511(8)	N(4)—N(13)	4.605(6)
O(1)—O(10)	4.701(5)	O(16)—O(7)	4.846(4)
O(1)—O(7)	4.281(5)	O(16)—O(10)	3.619(5)
O(1)—N(4)	2.841(5)	O(16)—N(13)	2.854(6)
Na—N(4)	2.809(4)		
Na—N(13)	2.764(5)		
O(2)—Cl—O(3)	108.6(3)	C(6)—O(7)—C(8)	113.4(4)
O(2)—Cl—O(4)	109.7(3)	Na—O(10)—C(9)	114.8(3)
O(2)—Cl—O(5)	109.1(3)	Na—O(10)—C(11)	114.5(3)
O(3)—Cl—O(4)	109.5(3)	C(9)—O(10)—C(11)	113.9(4)
O(3)—Cl—O(5)	111.1(3)	Na—O(16)—C(15)	114.0(3)
O(4)—Cl—O(5)	108.8(2)	Na—O(16)—C(17)	107.6(3)
O(1)—Na—O(4)	85.8(1)	C(15)—O(16)—C(17)	114.9(4)
O(1)—Na—O(7)	125.4(1)	C(1')—N(4)—C(3)	113.6(4)
O(1)—Na—O(10)	160.1(1)	C(1')—N(4)—C(5)	112.5(4)
O(1)—Na—O(16)	69.0(1)	C(3)—N(4)—C(5)	111.7(4)
O(4)—Na—O(7)	91.8(1)	C(12)—N(13)—C(13')	103.8(4)
O(4)—Na—O(10)	79.7(1)	C(12)—N(13)—C(14)	112.7(4)
O(4)—Na—O(16)	88.1(1)	C(13')—N(13)—C(14)	112.9(4)

Table III. Continued.

O(7)—Na—O(10)	69.1(1)	N(4)—C(1')—C(2')	117.9(4)
O(7)—Na—O(16)	165.6(1)	C(1')—C(2')—C(3')	112.1(4)
O(10)—Na—O(16)	96.8(1)	O(1)—C(2)—C(3)	107.9(4)
Na—O(1)—C(2)	124.4(3)	C(2')—C(3')—C(4')	113.8(4)
Na—O(1)—C(18)	119.2(3)	N(4)—C(3)—C(2)	113.5(4)
C(2)—O(1)—C(18)	114.2(4)	C(3')—C(4')—C(5')	115.6(5)
Cl—O(4)—Na	142.3(2)	C(4')—C(5')—C(6')	114.4(5)
Na—O(7)—C(6)	110.4(3)	N(4)—C(5)—C(6)	112.6(4)
Na—O(7)—C(8)	105.2(3)	O(7)—C(6)—C(5)	107.9(5)
C(5')—C(6')—C(7')	115.2(5)	C(14')—C(15')—C(16')	109.6(3)
C(6')—C(7')—C(8')	114.9(5)	O(16)—C(15)—C(14)	108.5(4)
C(7')—C(8')—C(9')	115.0(5)	C(15')—C(16')—C(17')	114.7(5)
O(7)—C(8)—C(9)	108.0(5)	C(16')—C(17')—C(18')	113.7(6)
C(8')—C(9')—C(10')	114.8(5)	O(16)—C(17)—C(18)	112.1(4)
O(10)—C(9)—C(8)	108.1(5)	O(1)—C(18)—C(17)	107.5(4)
C(9')—C(10')—C(11')	114.9(6)	C(17')—C(18')—C(19')	114.3(5)
C(10')—C(11')—C(12')	113.9(6)	C(18')—C(19')—C(20')	114.7(6)
O(10)—C(11)—C(12)	112.1(5)	C(19')—C(20')—C(21')	113.8(6)
N(13)—C(12)—C(11)	112.2(5)	C(20')—C(21')—C(22')	114.0(6)
N(13)—C(13')—C(14')	117.1(2)	C(21')—C(22')—C(23')	113.8(6)
C(13')—C(14')—C(15')	112.0	C(22')—C(23')—C(24')	113.1(6)
N(13)—C(14)—C(15)	113.2(4)		
N(3)—Na—N(14)	111.5(1)		

Table IV. Torsion angles about the diaza-18-crown-6 ligand ( $^{\circ}$ )

C(18)—O(1)—C(2)—C(3)	-176.8(4)
C(2)—O(1)—C(18)—C(17)	172.6(4)
C(8)—O(7)—C(6)—C(5)	-173.2(4)
C(6)—O(7)—C(8)—C(9)	178.8(4)
C(11)—O(10)—C(9)—C(8)	-166.6(4)
C(9)—O(10)—C(11)—C(12)	84.5(5)
C(17)—O(16)—C(15)—C(14)	174.7(4)
C(15)—O(16)—C(17)—C(18)	74.5(5)
C(5)—N(4)—C(3)—C(2)	170.2(4)
C(3)—N(4)—C(5)—C(6)	-81.5(5)
C(14)—N(13)—C(12)—C(11)	74.7(5)
C(12)—N(13)—C(14)—C(15)	-143.4(4)
O(1)—C(2)—C(3)—N(4)	-59.5(5)
N(4)—C(5)—C(6)—O(7)	-60.6(5)
O(7)—C(8)—C(9)—O(10)	63.4(6)
O(10)—C(11)—C(12)—N(13)	63.9(6)
N(13)—C(14)—C(15)—O(16)	60.7(5)
O(16)—C(17)—C(18)—O(1)	52.0(5)



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