

Structure of Dioxo[2-(salicylidenamino)phenolato(2-)-O,N,O']- [tris(dimethylamino)phosphine oxide]tungsten(VI)

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Abstract. $[W(C_{13}H_9NO_2)_2O_2(C_6H_{18}N_3OP)]$, $M_r = 606.27$, monoclinic, $P2_1/n$, $a = 16.037(1)$, $b = 16.367(1)$, $c = 19.098(2)$ Å, $\beta = 113.65(1)^\circ$, $V = 4591.8(7)$ Å³, $Z = 8$, $D_x = 1.754$ g cm⁻³, $\lambda(Mo K\alpha) = 0.71073$ Å, $\mu = 52.3$ cm⁻¹, $F(000) = 2384$, $T = 295$ K, $R = 0.042$ for 5270 observed reflections with $I > 2.5\sigma(I)$. The structure contains two crystallographically independent molecules. Each W atom is octahedrally coordinated by the three coordinating atoms of the Schiff base and one of the oxo groups [$W=O = 1.723(5)$ and $1.730(5)$ Å] in the equatorial plane with the other oxo group [$W=O = 1.702(5)$ and $1.703(5)$ Å] and the O of the tris(dimethylamino)phosphine oxide ligand in apical positions.

Experimental. Data were collected on an Enraf-Nonius CAD-4F diffractometer for a block-shaped (0.5 × 0.5 × 0.5 mm) yellow crystal glued on top of a glass fibre. Unit-cell parameters and their e.s.d.'s were derived from a least-squares treatment of 25 SET4 reflections ($11 < \theta < 14^\circ$). The cell was tested for higher metrical symmetry (Spek, 1988). Intensity data for 8656 reflections (h 0:19, k 0:19, l -22:20; $\theta < 25^\circ$; Zr-filtered Mo $K\alpha$ radiation) were collected in the ω -2 θ scan mode with $\Delta\omega = (0.70 + 0.35\tan\theta)^\circ$. Two reference reflections (222; $\bar{2}\bar{2}\bar{2}$) indicated no decay during 138 h of X-ray exposure. The intensity data were corrected for Lp and absorption with *DIFABS* (Walker & Stuart, 1983). The variance $\sigma^2(I)$ was calculated based on counting statistics plus an instability constant term $(0.04I)^2$ as derived from the excess variance in the reference reflections (McCandlish, Stout & Andrews, 1975). The space group was derived from the observed systematic absences. The structure was solved with standard Patterson and Fourier techniques with *SHELXS86* (Sheldrick, 1986) and refined on F by blocked full-matrix least squares with *SHELX76* (Sheldrick, 1976). H atoms were introduced on calculated positions and refined with fixed geometry with respect to

the atoms to which they are attached. Refinement with weights based on $w^{-1} = \sigma^2(F)$ converged at $R = 0.042$ [$wR = 0.033$; $S = 3.97$; 573 parameters; 5270 reflections with $I > 2.5\sigma(I)$, $(\Delta/\sigma)_{\max} = 0.5$]. The refined parameter set included a scale factor, coordinates and anisotropic thermal parameters for the non-H atoms and coordinates and five common isotropic U 's for the H atoms. C(215) and C(219) are probably disordered. A final difference Fourier map did not show residual peaks higher than -0.90 and 0.98 e Å⁻³. Scattering factors from Cromer & Mann (1968) and anomalous-dispersion terms from Cromer & Liberman (1970) were used. Final parameters are given in Table 1* with bond lengths and angles in

* Lists of structure factors, anisotropic thermal parameters, H-atom parameters and a full list of bond lengths and angles have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 52447 (57 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

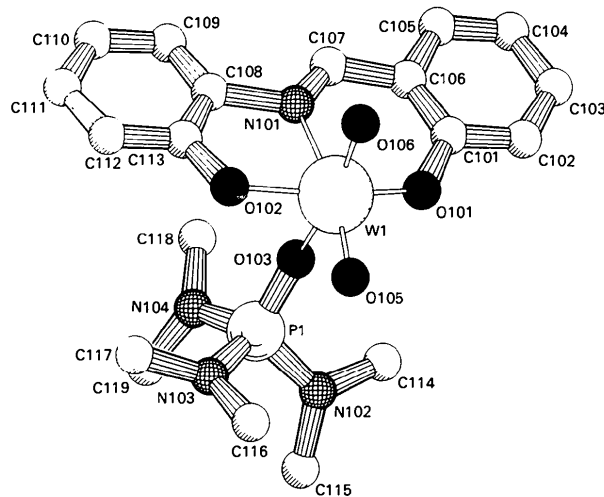


Fig. 1. View of one of the two independent molecules with numbering scheme.

Table 1. Final coordinates and equivalent isotropic thermal parameters and their e.s.d.'s in parentheses

$$U_{eq} = (1/3)\sum_i \sum_j U_{ij} a_i^* a_j^* a_i \cdot a_j$$

	x	y	z	U_{eq} (Å ²)
W(1)	0-50837 (2)	0-28627 (2)	0-65782 (2)	0-0406 (1)
P(1)	0-4106 (1)	0-3087 (2)	0-4583 (1)	0-0449 (9)
O(101)	0-4627 (4)	0-3907 (3)	0-6703 (3)	0-052 (2)
O(102)	0-4966 (4)	0-1745 (4)	0-6188 (3)	0-056 (2)
O(103)	0-4153 (3)	0-3109 (3)	0-5388 (3)	0-046 (2)
O(105)	0-6008 (3)	0-3134 (3)	0-6385 (3)	0-056 (2)
O(106)	0-5526 (4)	0-2593 (4)	0-7518 (3)	0-037 (2)
N(101)	0-3692 (5)	0-2438 (5)	0-6508 (4)	0-093 (4)
N(102)	0-4193 (4)	0-4031 (4)	0-4332 (4)	0-052 (3)
N(103)	0-4875 (5)	0-2574 (5)	0-4405 (4)	0-100 (4)
N(104)	0-3154 (5)	0-2638 (5)	0-4060 (4)	0-061 (3)
C(101)	0-3965 (6)	0-4136 (5)	0-6948 (4)	0-045 (3)
C(102)	0-3981 (6)	0-4924 (6)	0-7202 (5)	0-055 (4)
C(103)	0-3340 (7)	0-5192 (6)	0-7447 (5)	0-069 (4)
C(104)	0-2642 (6)	0-4679 (7)	0-7415 (6)	0-080 (5)
C(105)	0-2611 (6)	0-3901 (6)	0-7171 (5)	0-065 (4)
C(106)	0-3271 (6)	0-3633 (5)	0-6932 (5)	0-047 (3)
C(107)	0-3125 (6)	0-2786 (7)	0-6678 (5)	0-072 (5)
C(108)	0-3520 (7)	0-1536 (6)	0-6208 (5)	0-059 (4)
C(109)	0-2781 (7)	0-1082 (7)	0-6102 (5)	0-070 (4)
C(110)	0-2735 (7)	0-0305 (7)	0-5824 (6)	0-079 (5)
C(111)	0-3434 (8)	-0-0009 (6)	0-5653 (6)	0-076 (5)
C(112)	0-4173 (7)	0-0467 (6)	0-5773 (5)	0-067 (4)
C(113)	0-4228 (6)	0-1263 (6)	0-6053 (5)	0-054 (4)
C(114)	0-3859 (6)	0-4674 (6)	0-4659 (5)	0-067 (4)
C(115)	0-4256 (7)	0-4245 (7)	0-3614 (5)	0-080 (5)
C(116)	0-5806 (6)	0-2934 (8)	0-4631 (6)	0-089 (5)
C(117)	0-4823 (8)	0-1691 (7)	0-4289 (7)	0-099 (6)
C(118)	0-2422 (6)	0-2564 (7)	0-4312 (5)	0-089 (5)
C(119)	0-2880 (7)	0-2551 (6)	0-3230 (5)	0-080 (4)
W(2)	1-02108 (2)	0-23193 (2)	0-68692 (2)	0-0412 (1)
P(2)	0-9410 (2)	0-2754 (2)	0-4860 (1)	0-068 (1)
O(201)	0-9537 (4)	0-1329 (4)	0-6460 (3)	0-069 (3)
O(202)	1-0343 (4)	0-3487 (4)	0-7007 (3)	0-066 (3)
O(203)	0-9600 (4)	0-2690 (4)	0-5676 (3)	0-052 (3)
O(205)	1-1216 (3)	0-2131 (3)	0-6764 (3)	0-061 (3)
O(206)	1-0421 (4)	0-2091 (4)	0-7795 (3)	0-073 (3)
N(201)	0-8815 (5)	0-2773 (6)	0-6715 (4)	0-071 (4)
N(202)	0-8379 (6)	0-2992 (9)	0-4357 (5)	0-137 (6)
N(203)	1-0010 (7)	0-3490 (7)	0-4758 (5)	0-120 (5)
N(204)	0-9720 (8)	0-1901 (8)	0-4543 (5)	0-120 (5)
C(201)	0-8738 (6)	0-1053 (6)	0-6423 (5)	0-051 (4)
C(202)	0-8590 (7)	0-0228 (7)	0-6358 (6)	0-074 (5)
C(203)	0-7773 (9)	-0-0083 (8)	0-6314 (7)	0-099 (6)
C(204)	0-7096 (8)	0-0396 (9)	0-6321 (6)	0-091 (6)
C(205)	0-7224 (7)	0-1218 (8)	0-6385 (5)	0-076 (5)
C(206)	0-8064 (7)	0-1560 (6)	0-6456 (5)	0-058 (4)
C(207)	0-8126 (7)	0-2474 (8)	0-6568 (5)	0-085 (6)
C(208)	0-8831 (7)	0-3722 (7)	0-6866 (5)	0-060 (4)
C(209)	0-8178 (8)	0-4213 (8)	0-6868 (6)	0-081 (5)
C(210)	0-8356 (7)	0-5023 (8)	0-7057 (6)	0-083 (5)
C(211)	0-9236 (8)	0-5332 (6)	0-7246 (5)	0-080 (5)
C(212)	0-9913 (7)	0-4815 (6)	0-7240 (5)	0-067 (4)
C(213)	0-9721 (7)	0-4007 (6)	0-7047 (5)	0-055 (4)
C(214)	0-798 (1)	0-369 (1)	0-459 (1)	0-18 (1)
C(215)	0-781 (1)	0-249 (2)	0-385 (1)	0-57 (3)
C(216)	0-980 (1)	0-390 (2)	0-405 (1)	0-31 (2)
C(217)	1-0902 (9)	0-366 (1)	0-5277 (9)	0-160 (9)
C(218)	0-998 (1)	0-191 (1)	0-392 (1)	0-26 (2)
C(219)	0-954 (2)	0-119 (1)	0-476 (1)	0-45 (3)

Table 2. Bond distances (Å) and angles (°) for the non-H atoms

W(1)—O(101)	1-912 (5)	W(2)—O(201)	1-931 (6)
W(1)—O(102)	1-956 (6)	W(2)—O(202)	1-929 (7)
W(1)—O(103)	2-199 (5)	W(2)—O(203)	2-175 (5)
W(1)—O(105)	1-723 (5)	W(2)—O(205)	1-730 (5)
W(1)—O(106)	1-702 (5)	W(2)—O(206)	1-703 (5)
W(1)—N(101)	2-290 (9)	W(2)—N(201)	2-263 (9)
P(1)—O(103)	1-509 (6)	P(2)—O(203)	1-467 (6)
P(1)—N(102)	1-641 (7)	P(2)—N(202)	1-59 (1)
P(1)—N(103)	1-636 (9)	P(2)—N(203)	1-60 (1)
P(1)—N(104)	1-627 (8)	P(2)—N(204)	1-67 (1)
O(101)—C(101)	1-37 (1)	O(201)—C(201)	1-33 (1)
O(102)—C(113)	1-36 (1)	O(202)—C(213)	1-34 (1)
N(101)—C(107)	1-22 (1)	N(201)—C(207)	1-14 (1)
N(101)—C(108)	1-57 (1)	N(201)—C(208)	1-58 (1)
N(102)—C(114)	1-43 (1)	N(202)—C(214)	1-47 (2)
N(102)—C(115)	1-46 (1)	N(202)—C(215)	1-32 (3)
N(103)—C(116)	1-50 (1)	N(203)—C(216)	1-43 (2)
N(103)—C(117)	1-46 (1)	N(203)—C(217)	1-40 (2)
N(104)—C(118)	1-44 (1)	N(204)—C(218)	1-40 (2)
N(104)—C(119)	1-47 (1)	N(204)—C(219)	1-31 (2)
C(101)—C(102)	1-38 (1)	C(201)—C(202)	1-37 (1)
C(101)—C(106)	1-38 (1)	C(201)—C(206)	1-38 (1)
C(102)—C(103)	1-36 (1)	C(202)—C(203)	1-38 (2)
C(103)—C(104)	1-38 (2)	C(203)—C(204)	1-34 (2)
C(104)—C(105)	1-35 (1)	C(204)—C(205)	1-36 (2)
C(105)—C(106)	1-38 (1)	C(205)—C(206)	1-41 (2)
C(106)—C(107)	1-46 (1)	C(206)—C(207)	1-51 (2)
C(108)—C(109)	1-34 (2)	C(208)—C(209)	1-32 (2)
C(108)—C(113)	1-36 (1)	C(208)—C(213)	1-41 (2)
C(109)—C(110)	1-37 (2)	C(209)—C(210)	1-37 (2)
C(110)—C(111)	1-39 (2)	C(210)—C(211)	1-40 (2)
C(111)—C(112)	1-36 (2)	C(211)—C(212)	1-38 (2)
C(112)—C(113)	1-40 (1)	C(212)—C(213)	1-37 (1)
O(101)—W(1)—O(102)	154-2 (3)	O(201)—W(2)—O(202)	154-1 (3)
O(101)—W(1)—O(103)	80-2 (2)	O(201)—W(2)—O(203)	82-3 (2)
O(101)—W(1)—O(105)	101-6 (3)	O(201)—W(2)—O(205)	101-5 (3)
O(101)—W(1)—O(106)	96-8 (3)	O(201)—W(2)—O(206)	94-7 (3)
O(101)—W(1)—N(101)	82-3 (3)	O(201)—W(2)—N(201)	81-9 (3)
O(102)—W(1)—O(103)	81-7 (2)	O(202)—W(2)—O(203)	81-1 (2)
O(102)—W(1)—O(105)	96-9 (3)	O(202)—W(2)—O(205)	97-8 (3)
O(102)—W(1)—O(106)	95-4 (3)	O(202)—W(2)—O(206)	96-3 (3)
O(102)—W(1)—N(101)	75-6 (3)	O(202)—W(2)—N(201)	75-0 (3)
O(103)—W(1)—O(105)	91-0 (2)	O(203)—W(2)—O(205)	88-7 (2)
O(103)—W(1)—O(106)	163-8 (3)	O(203)—W(2)—O(206)	165-3 (3)
O(103)—W(1)—N(101)	75-7 (2)	O(203)—W(2)—N(201)	76-5 (3)
O(105)—W(1)—O(106)	105-2 (3)	O(205)—W(2)—O(206)	106-0 (3)
O(105)—W(1)—N(101)	165-4 (2)	O(205)—W(2)—N(201)	164-2 (3)
O(106)—W(1)—N(101)	88-1 (3)	O(206)—W(2)—N(201)	88-9 (3)
O(103)—P(1)—N(102)	107-4 (3)	O(203)—P(2)—N(202)	112-4 (5)
O(103)—P(1)—N(103)	119-4 (3)	O(203)—P(2)—N(203)	107-7 (4)
O(103)—P(1)—N(104)	106-7 (4)	O(203)—P(2)—N(204)	111-1 (5)
N(102)—P(1)—N(103)	105-2 (4)	N(202)—P(2)—N(203)	105-8 (6)
N(102)—P(1)—N(104)	114-5 (4)	N(202)—P(2)—N(204)	111-9 (6)
N(103)—P(1)—N(104)	103-9 (4)	N(203)—P(2)—N(204)	107-7 (6)
W(1)—O(103)—P(1)	141-7 (3)	W(2)—O(203)—P(2)	161-3 (5)
W(1)—O(101)—C(101)	132-4 (5)	W(2)—O(201)—C(201)	132-5 (6)
W(1)—N(101)—C(107)	130-9 (7)	W(2)—N(201)—C(207)	134-7 (9)
W(1)—N(101)—C(108)	109-4 (6)	W(2)—N(201)—C(208)	110-9 (6)
W(1)—O(102)—C(113)	124-0 (6)	W(2)—O(202)—C(213)	126-5 (6)
C(107)—N(101)—C(108)	119-7 (9)	C(207)—N(201)—C(208)	114 (1)
P(1)—N(102)—C(114)	118-3 (6)	P(2)—N(202)—C(214)	119 (1)
P(1)—N(102)—C(115)	123-2 (6)	P(2)—N(202)—C(215)	122 (1)
C(114)—N(102)—C(115)	114-4 (7)	C(214)—N(202)—C(215)	117 (1)
P(1)—N(103)—C(116)	119-0 (7)	P(2)—N(203)—C(216)	123 (1)
P(1)—N(103)—C(117)	122-5 (8)	P(2)—N(203)—C(217)	124 (1)
C(116)—N(103)—C(117)	115-0 (9)	C(216)—N(203)—C(217)	111 (1)
P(1)—N(104)—C(118)	121-5 (6)	P(2)—N(204)—C(218)	122 (1)
P(1)—N(104)—C(119)	120-4 (7)	P(2)—N(204)—C(219)	120 (1)
C(118)—N(104)—C(119)	114-8 (8)	C(218)—N(204)—C(219)	117 (2)
O(101)—C(101)—C(102)	118-0 (8)	O(201)—C(201)—C(202)	117-9 (9)
O(101)—C(101)—C(106)	124-0 (7)	O(201)—C(201)—C(206)	123-1 (9)
C(102)—C(101)—C(106)	118-0 (9)	C(202)—C(201)—C(206)	119 (1)
C(101)—C(102)—C(103)	121 (1)	C(201)—C(202)—C(203)	120 (1)
C(102)—C(103)—C(104)	120-0 (9)	C(202)—C(203)—C(204)	123 (1)
C(103)—C(104)—C(105)	120 (1)	C(203)—C(204)—C(205)	119 (1)
C(104)—C(105)—C(106)	119-1 (9)	C(204)—C(205)—C(206)	120 (1)
C(101)—C(106)—C(105)	121-5 (8)	C(201)—C(206)—C(205)	119 (1)
C(101)—C(106)—C(107)	126-7 (9)	C(201)—C(206)—C(207)	126 (1)
C(105)—C(106)—C(107)	111-7 (9)	C(205)—C(206)—C(207)	114 (1)
N(101)—C(107)—C(106)	120 (1)	N(201)—C(207)—C(206)	118 (1)
N(101)—C(108)—C(109)	126 (1)	N(201)—C(208)—C(209)	130 (1)
N(101)—C(108)—C(113)	110-5 (9)	N(201)—C(208)—C(213)	108-2 (9)

Table 2. The molecule is shown in Fig. 1. The programs *PLATON* and *PLUTON* (Spek, 1982) were used for the calculation of geometrical data and the plot respectively. All calculations were performed on a MicroVAX-II.

Related literature. Dioxo- and (oxo)peroxotransition-metal complexes can serve as intermediates in catalytic oxidation of organic substances (Sheldon & Kochi, 1981; Sheldon, 1985; Di Furia & Modena, 1985; Khan, 1986; Holm, 1987). In particular, the subgroup 6B metals Cr, Mo and W have attracted great interest over the past twenty years. While

dioxo- and (oxo)peroxomolybdenum complexes containing organic ligands have been investigated rather extensively by X-ray studies (e.g. Hawkins, Dewan & Sharpless, 1986; Dengel, Griffith, Powell & Skapski, 1987), only few structural studies have been performed on corresponding W and Cr complexes (Stomberg & Olsen, 1985; Yang, Dzugan & Goedken, 1985; Venturello, D'Aloisio, Bart & Ricci, 1985). The structure of the corresponding oxoperoxotungsten Schiff-base complex $WO(O_2)-[(2\text{-salicylidenamino})phenolato(2-)] [tris(dimethylamino)phosphine\ oxide]$, and a comparative study of the two compounds will be reported elsewhere (Terheijden, Kraaykamp, Timmer, Meinema & Spek, 1989).

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Structure of Bis(3',4'-dimethyltetrafulvalen-3-yl)ethylenium Hexafluoroarsenate Hemibutyronitrile Solvate

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Abstract. $C_{18}H_{18}S_8^+ \cdot AsF_6^- \cdot \frac{1}{2}C_4H_7N$, $M_r = 715.35$, triclinic, $P\bar{1}$, $a = 7.133$ (3), $b = 14.718$ (3), $c = 14.778$ (3) Å, $\alpha = 68.75$ (2), $\beta = 79.46$ (3), $\gamma = 73.12$ (3)°, $V = 1378.4$ (7) Å³, $Z = 2$, $D_x = 1.724$ g cm⁻³, $\lambda(Mo K\alpha) = 0.71073$ Å, $\mu = 18.62$ cm⁻¹, $F(000) = 724$, $T = 298$ K, final $R = 0.0853$ for 1602 observed unique reflections. The organic cations are planar, stacked along the a axis in a zigzag stack with overlaps 'double bond over ring'. AsF_6^- and the disordered solvent molecules are situated between the organic stacks. Crystals of the compound exhibit semiconducting behaviour.

Experimental. Crystals were prepared by electrochemical oxidation of $C_{18}H_{18}S_8$ in a 4:1 mixture of chlorobenzene and butyronitrile containing $n\text{-Bu}_4NAsF_6$ (0.100 M) (Lerstrup & Bechgaard, 1990). The size of the crystal used was $0.007 \times 0.087 \times 0.20$ mm. The X-ray data were measured with an Enraf-Nonius CAD-4 diffractometer. The lattice parameters were derived from setting angles of 22 reflections, $8.8 < \theta < 15.9^\circ$. Intensity data were collected with ω scans [$\Delta\omega = (0.70 + 0.35\tan\theta)^\circ$] using graphite-monochromated $Mo K\alpha$ radiation, max. counting time was 8 min, $|h| \leq 8$, $|k| \leq 16$, $|l| \leq 16$,