An Efficient, Easy Route for the Synthesis of 2,2′:6′,2′′-Terpyridine 1′-Oxides

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2,6-Dibromopyridine *N*-oxide and 4-nitro-2,6-dibromopyridine *N*-oxide were treated with tributyl(pyridin-2-yl)stannane under the conditions of the Stille coupling reaction. Derivatives of 2,2′-bipyridine 1-oxides as well as 2,2′:6′,2′′-terpyridine 1′-oxides were synthesised. Oxidation of 2,2′:6′,2′′-terpyridine 1′-oxide with *m*-CPBA gave 2,2′:6′,2′′-terpyridine 1,1′-dioxide. The crystal structures of 4-nitro-6-bromo-2,2′-bipyridine 1-oxide and 2,2′:6′,2′′-terpyridine 1,1′-dioxide were determined by X-ray structure analysis.

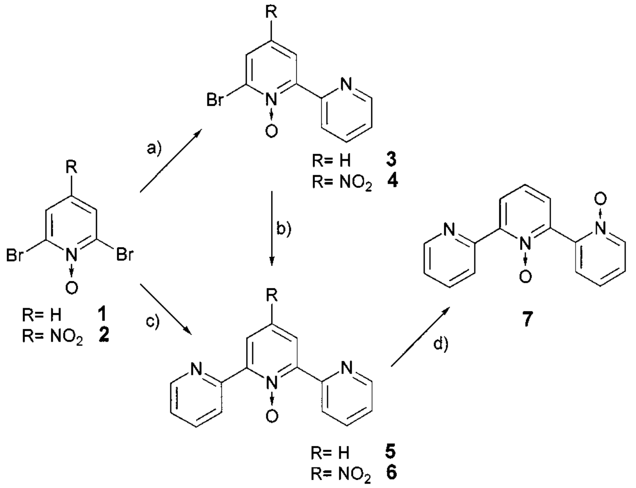
Introduction

In general oligopyridines and 2,2′:6′,2′′-terpyridine (tpy) in particular, have received considerable attention in supramolecular chemistry.[1] One of the interesting fields in supramolecular chemistry relates to the photochemical properties of homo- and heterometallic complexes using the metal-binding domains of oligopyridines.[2,3] Particular interest rests in 4′-substituted tpy-ligands and many substituents can be directly inserted using Kröhnke methodology.[4] The work of most groups has been concerned with attaching functional groups directly to the C(4′) of 2,2′:6′,2′′-terpyridine[5−16].

Though electrophilic reactions on pyridine rings do not normally occur, oxidation of the ring nitrogen to form pyridine *N*-oxide facilitates electrophilic attack at C(4) of the pyridine ring. Using standard methods, 2,2′:6′,2′′-terpyridine was oxidised with *m*-CPBA in dichloromethane to give 2,2′:6′,2′′-terpyridine 1-oxide and 2,2′:6′,2′′-terpyridine 1,1′′-dioxide, respectively.[17] The exhaustive oxidation of 2,2′:6′,2′′-terpyridine with hydrogen peroxide in acetic acid yielded 2,2′:6′,2′′-terpyridine 1,1′,1′′-trioxide.[18] All these *N*-oxides were further treated with nitric acid in sulfuric acid to give 4-nitropyridine *N*-oxides. To the best of our knowledge, there is no report of selective oxidation of the nitrogen of the central pyridine ring.

Results and Discussion

Metal mediated cross couplings such as the Suzuki reaction[19] or Stille coupling[20] reaction have found wide applications in the synthesis of aromatic and heterocyclic compounds. We have already applied this methodology for the synthesis of 4′-functionalised-2,2′:6′,2′′-terpyridines.[12−16] The advantage of this method is that many functional groups such as nitro, carboxylate, carbonyl or cyano groups do not react under the prescribed reaction conditions. There exists, however, to the best of our knowledge, no report of using pyridine *N*-oxides. We would now like to report a Stille coupling reaction with 2,6-dibromopyridine *N*-oxides (Scheme 1).



Scheme 1. a) Tributyl(pyridin-2-yl)stannane (1 mol), Pd(PPh$_3$)$_4$ (1 mol %), toluene, 110 °C, 16 h, **3** (66%), **4** (60%). b) Tributyl(pyridin-2-yl)stannane (1 mol), Pd(PPh$_3$)$_4$ (1 mol %), toluene, 110 °C, 16 h, **5** (65%), **6** (75%). c) Tributyl(pyridin-2-yl)stannane (2 mol), Pd(PPh$_3$)$_4$ (2 mol %), toluene, 110 °C, 16 h, **5** (71%), **6** (73%). d) **5** (1mol), *m*-CPBA (1 mol), dichloromethane, room temp., 20 h, 35%.

2,6-Dibromopyridine *N*-oxide **1**[13] was treated with 1 equiv. tributyl(pyridin-2-yl)stannane[21] in the presence of 0.01 equiv. of Pd(PPh$_3$)$_4$ for 16 h in toluene to give 6-bromo-2,2′-bipyridine 1-oxide **3** in 66% yield as a colourless microcrystalline solid. Compound **3** has recently been prepared by a different method.[22] The reaction of **3** with 1 equiv. tributyl(pyridin-2-yl)stannane in the presence of the catalyst yielded **5** in 65% yield as a colourless microcrystalline solid. However, if **1** was treated with 2 equiv. tributyl(pyridin-2-yl)stannane in the presence of 0.02 equiv. of Pd(PPh$_3$)$_4$ under the same conditions, 2,2′:6′,2′′-terpyridine 1′-oxide **5** was obtained directly in 71% yield.

This is the first example of a tpy ligand containing *N*-oxide at the central pyridine ring. Due to the symmetry of the molecule, one would expect only six signals in the ^{1}H

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NMR spectrum of 2,2′:6′,2′′-terpyridine 1′-oxide **5** which is exactly the case. In $CDCl_3$ solution, in comparison with 2,2′:6′,2′′-terpyridine, significant shifts of protons of the central pyridine rings were observed, while the protons at the terminal pyridine rings were not especially affected. The protons $H^{3'}$ were shifted to high field and observed as a doublet at $\delta = 8.07$ ($\Delta\delta = 0.38$) while proton $H^{4'}$ was also shifted to high field and observed as a triplet at 7.46 ($\Delta\delta = 0.50$). In the electron ionisation mass spectrum of **5**, parent ion peaks were observed at $m/z = 249$ followed by a fragment peak at $m/z = 233$ (tpy). All the analytical and spectroscopic results provide good evidence for the product.

Once again, 4-nitro-2,6-dibromopyridine *N*-oxide **2**[13] was treated under the same conditions with 1 equiv. tributyl(pyridin-2-yl)stannane to give 6-bromo-4-nitro-2,2′-bipyridine 1-oxide **4** in 60% yield as yellow needles. Its structure in the solid state[23] is presented here for the first time (Figure 1). Compound **4** is not planar and the interplanar angle of the two pyridine rings is 32°.

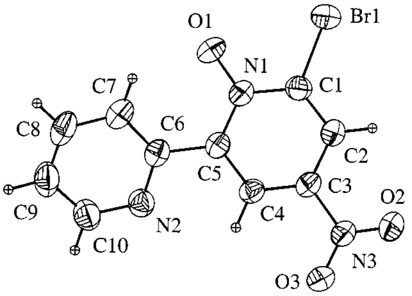

Figure 1. Solid state structure of compound **4**. The CCDC number is 153154.

The reaction of **4** with 1 equiv. tributyl(pyridin-2-yl)stannane in the presence of 0.01 equiv. of $Pd(PPh_3)_4$ yielded **6** in 75% yield as a yellow microcrystalline solid. However, if **2** was treated with 2 equiv. tributyl(pyridin-2-yl)stannane in the presence of the catalyst under the same conditions, 4′-nitro-2,2′:6′,2′′-terpyridine 1′-oxide **6** was obtained directly in 73% yield.

Due to the symmetry of the molecule, one expected and observed, only five signals in the ^1H NMR spectrum of 4′-nitro-2,2′:6′,2′′-terpyridine 1′-oxide **6**. Compound **6** was compared with 4′-nitro-2,2′:6′,2′′-terpyridine.[13] In $CDCl_3$ solution protons $H^{3'}$ were shifted to high field and were observed as a doublet at $\delta = 9.00$ ($\Delta\delta = 0.16$) while protons H^6 ($\delta = 8.82$, $\Delta\delta = 0.06$) and H^3 ($\delta = 8.71$, $\Delta\delta = 0.07$) were shifted to low field, respectively. In the electron ionisation mass spectrum of **6**, parent ion peaks were observed at $m/z = 294$ followed by fragments at $m/z = 278$ [M − O], 248 [M − NO_2], and finally at $m/z = 232$ [tpy]. All the analytical and spectroscopic data are in agreement with the proposed formulation and structure.

By using standard methods, 2,2′:6′,2′′-terpyridine 1′-oxide **5** was oxidised with 1 equiv. *m*-CPBA in dichloromethane to give 2,2′:6′,2′′-terpyridine 1,1′-dioxide **7** in 35% yield as colourless orthorhombic plates.

Due to the asymmetry of the molecule, one expected and observed all eleven signals in the ^1H NMR spectrum of 2,2′:6′,2′′-terpyridine 1,1′-dioxide **7**. In $CDCl_3$ solution proton $H^{4'}$ was observed as a triplet at almost the same value ($\delta = 7.44$, $\Delta\delta = 0.02$) as in 2,2′:6′,2′′-terpyridine 1′-oxide **5**. In the Maldi-TOF mass spectrum of **7**, parent ion peaks were observed at $m/z = 265$. All the analytical and spectroscopic results are consistent with the formulated product.

The structure of **7** in the solid state[24] is presented here for the first time (Figure 2). While 2,2′:6′,2′′-terpyridine is almost a planar molecule,[13] 2,2′:6′,2′′-terpyridine 1,1′,1′′-trioxide was assumed not to be planar. The lack of planarity for the trioxide is due to the steric interaction of the oxygen atoms and probable repulsion of their lone pairs of electrons.[25] This assumption was confirmed by this novel compound. The two pyridine *N*-oxide rings are almost vertical and show an interplanar angle of 87.66° while the interplanar angle between the central pyridine *N*-oxide ring and the terminal pyridine ring is 38.47°. The interplanar angle between the terminal pyridine *N*-oxide ring and the terminal pyridine ring is 70.99°.

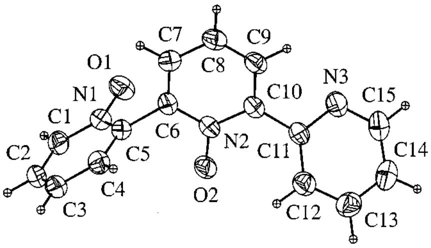

Figure 2. Solid state structure of compound **7**. The CCDC number is 153155.

2,2′:6′,2′′-Terpyridine oxides are also versatile ligands for the reaction with transition metals. While 2,2′:6′,2′′-terpyridine reacts readily with iron(II) at room temperature, the reaction of 2,2′:6′,2′′-terpyridine *N*-oxides is complicated with the increasing number of oxygen atoms. Coordination of two $tpyO_3$ ligands by all six oxygen atoms was assumed to result in the formation of highly distorted pseudo octahedral systems. 2,2′:6′,2′′-Terpyridine 1,1′,1′′-trioxide was treated with transition metals like Cu^{II}, Ni^{II}, Fe^{III}, Co^{II},[25] and Mn^{II-IV} [26] to obtain metal complexes. Europium(III) complexes of this ligand are also known.[27] Interestingly, the three oxygen atoms of the $tpyO_3$ ligands stabilise iron(III) complexes[25] while the iron(III) complexes of 2,2′:6′,2′′-terpyridines are labile.

In conclusion, we have established a methodology for preparing the 2,2′-bipyridine 1-oxides **3−4** (where the more hindered pyridine ring is selectively oxidised) and the 2,2′:6′,2′′-terpyridine 1′-oxides **5−6** (where the central pyridine ring is selectively oxidised) as well as 2,2′:6′,2′′-terpyridine 1,1′-dioxide **7**. Starting from different stannyl compounds and using this methodology, even higher oligopyridines such as 2,2′:6′,2′′:6′′,2′′′-quaterpyridines or 2,2′:6′,2′′:6′′,2′′′:6′′′,2′′′′-quinquepyridines, selectively oxidised at the central pyridine rings, should be accessible. In consideration of the versatile reactivity of pyridine *N*-oxides and 4-nitro-pyridine *N*-oxides (reduction, nucleophilic substitution e.g. by bromo, chloro, alkoxy and thiol groups) a series of new ligands for reaction with transition metals are accessible. The coordination chemistry of these ligands is currently under investigation.

Experimental Section

General: See ref.[13]

General Procedure for Stille Coupling Reactions: 2,6-Dibromopyridine *N*-oxides **1** and **2** (200 mg), tributyl(pyridin-2-yl)stannane (1 or 2 mol equivalents), and $Pd(PPh_3)_4$ (0.01 or 0.02 mol equivalent) were heated under nitrogen in toluene (50 mL) for 16 h. Upon cooling to room temp. saturated ammonium chloride (20 mL) was added. The reaction mixture was stirred for a further 30 min. The mixture was filtered over Celite. The precipitate was washed with dichloromethane (50 mL) and the organic phase was separated. The aqueous phase was extracted with toluene (3 × 25 mL). The combined organic phases were dried ($MgSO_4$) and the solvent was removed. Concentrated hydrochloric acid (30 mL) was added to the residue and extracted with dichloromethane (3 × 30 mL). The aqueous phase was cautiously neutralised with solid sodium hydroxide. The oligopyridines were then extracted with dichloromethane (3 × 30 mL) and dried ($MgSO_4$). The solvent was removed and the product purified on silica gel with dichloromethane/ethyl acetate: 9:1. All compounds were recrystallised from ethanol.

6-Bromo-2,2′-bipyridine 1-Oxide (3): Yield 130 mg (66%). m.p. 89 °C. − ^{1}H NMR ($CDCl_3$): δ = 8.82 (dd, $^{3}J = 8.10$, $^{4}J = 1.50$, 1 H, $H^{6'}$), 8.68 (dd, $^{3}J = 7.80$, $^{4}J = 1.80$, 1 H, $H^{3'}$), 8.12 (dd, $^{3}J = 7.90$, $^{4}J = 2.10$, 1 H, H^{3}), 7.80 (ddd, $^{3}J = 8.10$, $^{3}J = 7.80$, $^{4}J = 1.80$, 1 H, $H^{4'}$), 7.68 (dd, $^{3}J = 7.90$, $^{4}J = 2.10$, 1 H, H^{5}), 7.35 (ddd, $^{3}J = 8.10$, $^{3}J = 7.80$, $^{4}J = 1.50$, 1 H, $H^{5'}$), 7.18 (t, $^{3}J = 7.90$, 1 H, H^{4}).

6-Bromo-4-nitro-2,2′-bipyridine 1-Oxide (4): Yield 120 mg (60%). m.p. 171 °C. − IR (KBr): 1556, 1451, 1389, 1345, 1275, 740. − ^{1}H NMR ($CDCl_3$): δ = 9.11(d, $^{4}J = 3.30$, 1 H, H^{3}), 8.84 (md, $^{3}J = 8.36$, 1 H, $H^{6'}$), 8.78 (m, 1 H, $H^{3'}$), 8.52 (d, $^{4}J = 3.30$, 1 H, H^{5}), 7.88 (ddd, $^{3}J = 8.10$, $^{3}J = 7.56$, $^{4}J = 1.76$, 1 H,$H^{4'}$), 7.44 (ddd, $^{3}J = 8.10$, $^{3}J = 7.56$, $^{4}J = 1.30$, 1 H, $H^{5'}$). − ^{13}C NMR ($CDCl_3$): δ = 149.77, 147.72, 141.25, 136.70, 135.55, 125.54, 125.39, 125.17, 123.47, 120.90. − MS (Maldi-TOF): *m*/*z* = 296. − $C_{10}H_6BrN_3O_3$ (296.1): calcd. C 40.57, H 2.04, N 14.19; found C 40.52, H 1.84, N 14.09.

2,2′:6′,2′′-Terpyridine 1′-Oxide (5): Yield 140 mg (71%). m.p. 157 °C. − ^{1}H NMR ($CDCl_3$): δ = 8.76 (m, 2 H, H^{6}), 8.69 (d, $^{3}J = 8.08$, 2 H, H^{3}), 8.07 (d, $^{3}J = 7.84$, 2 H, $H^{3'}$), 7.81 (ddd, $^{3}J = 8.30$, $^{3}J = 7.80$, $^{4}J = 1.80$, 2 H, H^{4}), 7.46 (t, $^{3}J = 8.08$, 1 H, $H^{4'}$), 7.55 (ddd, $^{3}J = 8.10$, $^{3}J = 7.80$, $^{4}J = 1.80$, 2 H, H^{5}). − ^{13}C NMR ($CDCl_3$): δ = 150.51, 149.42, 148.23, 135.97, 127.69, 125.73, 125.41, 124.06. − MS (EI, rel. int): *m*/*z* = 249 (100, M), 233 (45, tpy). − $C_{15}H_{11}N_3O \cdot H_2O$ (267.3): calcd. C 67.41, H 4.90, N 15.72; found C 67.75, H 4.80, N 15.33.

4′-Nitro-2,2′:6′,2′′-terpyridine 1′-Oxide (6): Yield 145 mg (73%). m.p. 256−257 °C. − IR (KBr): 1525, 1336, 1276, 742. − ^{1}H NMR ($CDCl_3$): δ = 9.00 (s, 2 H, $H^{3'}$), 8.82 (m, 2 H, H^{6}), 8.71 (md, $^{3}J = 8.08$, 2 H, H^{3}), 7.90 (ddd, $^{3}J = 8.30$, $^{3}J = 7.80$, $^{4}J = 1.80$, 2 H, H^{4}), 7.47 (ddd, $^{3}J = 8.10$, $^{3}J = 7.80$, $^{4}J = 1.20$, 2 H, H^{5}). − ^{13}C NMR ($CDCl_3$): δ = 149.77, 149.55, 148.55, 142.30, 136.48, 125.40, 125.08, 121.40. − MS (EI, rel. int): *m*/*z* = 294 (100, M), 278 (18, M − O), 248 (50, M − NO_2), 232 (29, tpy), 220 (21), 144 (18), 128 (33), 78 (55, py). − $C_{15}H_{10}N_4O_3 \cdot H_2O$ (312.3): calcd. C 57.69, H 3.87, N 17.94; found C 57.58, H 3.65, N 17.95.

2,2′:6′,2′′-Terpyridine 1,1′-Dioxide (7): To a solution of 2,2′:6′,2′′-terpyridine 1′-oxide (**5**) (100 mg, 0.40 mmol) in dichloromethane (20 mL) at room temperature was added *m*-CPBA (90 mg, 75%, 0.40 mmol) in dichloromethane (20 mL). The solution was stirred for 20 h. The organic phase was then extracted with 10% Na_2CO_3 (2 × 30 mL) and dried ($MgSO_4$). The solvent was removed and the product purified on silica gel with dichloromethane/ethyl acetate: 9:1. − Yield 37 mg (35%). m.p. 179−180 °C. − ^{1}H NMR ($CDCl_3$): δ = 8.90 (d, H^{6}, $^{3}J = 8.10$, 1 H), 8.73 (m, $H^{6''}$, 1 H), 8.36 (dd, $^{3}J = 7.30$, $^{4}J = 1.50$, 1 H, $H^{3''}$), 8.31 (dd, $^{3}J = 8.10$, $^{4}J = 2.04$, 1 H, H^{3}), 7.77 (ddd, $^{3}J = 8.10$, $^{3}J = 7.80$, $^{4}J = 1.80$, 1 H, H^{5}), 7.57 (dd, $^{3}J = 7.80$, $^{3}J = 2.04$, 2 H, $H^{3',5'}$), 7.44 (t, $^{3}J = 7.80$, 1 H, $H^{4'}$), 7.35 (m, 3 H). − ^{13}C NMR ($CDCl_3$): δ = 149.50, 149.28, 147.90, 143.49, 143.34, 140.11, 136.06, 128.56, 128.26, 127.54, 126.37, 125.89, 124.84, 124.68, 124.30. − MS (Maldi-TOF): *m*/*z* = 265. − $C_{15}H_{11}N_3O_2 \cdot H_2O$ (283.3): calcd. C 63.60, H 4.62, N 14.83; found calcd. C 63.50, H 4.70, N 14.66.

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[1] J.-M. Lehn, *Supramolecular Chemistry* VCH, Weinheim, **1995**.

[2] V. Balzani, A. Juris, M. Venturi, S. Campagna, S. Serroni, *Chem. Rev.* **1996**, *96*, 759−833.

[3] J.-P. Collin, P. Gaviña, V. Heitz, J.-P. Sauvage, *Eur. J. Inorg. Chem.* **1998**, 1−14.

[4] F. Kröhnke, *Synthesis* **1976**, 1−24.

[5] K. Potts, *Bull. Soc. Chim. Belg.* **1990**, *99*, 741−768.

[6] E. C. Constable, M. D. Ward, *J. Chem. Soc., Dalton Trans.* **1990**, 1405−1409.

[7] E. C. Constable, A. M. W. Cargill Thompson, D. A. Tocher, M. A. M. Daniels, *New J. Chem.* **1992**, *16*, 855−867.

[8] R. Ziessel, *Synthesis* **1999**, 1839−1865.

[9] G. R. Pabst, J. Sauer, *Tetrahedron* **1999**, *55*, 5067−5088.

[10] R.-A. Fallahpour, E. C. Constable, *J. Chem. Soc., Perkin Trans. 1* **1997**, 2263−2264.

[11] R.-A. Fallahpour, M. Neuburger, M. Zehnder, *Polyhedron* **1999**, *18*, 2445−2454.

[12] R.-A. Fallahpour, *Eur. J. Inorg. Chem.* **1998**, 1205−1207.

[13] R.-A. Fallahpour, M. Neuburger, M. Zehnder, *New J. Chem.* **1999**, *23*, 53−61.

[14] R.-A. Fallahpour, M. Neuburger, M. Zehnder, *Synthesis* **1999**, 1051−1055.

[15] R.-A. Fallahpour, *Synthesis* **2000**, 1138−1142.

[16] R.-A. Fallahpour, *Synthesis* **2000**, 1665−1667.

[17] R. P. Thummel, Y. Jahng, *J. Org. Chem.* **1985**, *50*, 3635−3636.

[18] F. H. Case, *J. Org. Chem.* **1962**, *27*, 640−641.

[19] N. Miyaura, A. Suzuki, *Chem. Rev.* **1995**, *95*, 2457−2483.

[20] V. Farina, V. Krishnamurthy, W. J. Scott, *Org. Reactions* **1997**, *50*, 1−652.

[21] C. Bolm, M. Ewald, M. Felder, G. Schlingloff, *Chem. Ber.* **1992**, *125*, 1169−1190.

[22] F. Trécourt, B. Gervais, O. Mongin, C. Le Gal, F. Mongin, G. Quéguiner, *J. Org. Chem.* **1998**, *63*, 2892−2897.

[23] Crystal structure determinations of $C_{10}H_6BrN_3O_3$ (**4**): Crystal size 0.06 × 0.08 × 0.16 mm, *M* = 296.08, orthorhombic, space group *Pcab*, *a* = 6.8155(1), *b* = 10.5262(2), *c* = 29.1891(5) Å, *V* = 2094.06 Å^{3}, *F*(000) = 1168, *Z* = 8, D_c = 1.88 g cm^{-3};

unit cell parameters were determined by the least squares method using 25 independent reflections. Data collection was carried out on a four-circle Enraf−Nonius CAD4 diffractometer using monochromated Mo-K_α radiation ($\lambda = 0.71069$ Å), $T = 293$ K. The ω-2θ technique was used to measure 30948 reflections, range $2° \le \theta \le 27.48°$. Three standard reflections monitored every hour during data collection showed a decay of 3.56%. 4389 unique data with $I \ge 3\sigma(I)$ were used to solve and refine the structure. The final $wR(F^2)$ was 0.0361. The usual corrections were applied. The absorption correction was determined by using Ψ-scans; max/min transmission 1.0/0.23. The structure was solved by direct methods using the program SIR92.[28] Anisotropic least squares refinement was carried out on all non-hydrogen atoms using the program CRYSTALS.[29] The hydrogen atoms are in calculated positions with a fixed distance of 0.96 Å. Scattering factors were taken from the International Tables for X-ray Crystallography[30].

[24] Crystal structure determinations of $C_{15}H_{11}N_3O_2$ (**7**): Crystal size 0.05 × 0.12 × 0.18 mm, $M = 265.27$, orthorhombic, space group *P*cab, $a = 9.1823(4)$, $b = 26.139(1)$, $c = 10.0356(3)$ Å, $V = 2408.74$ Å^3, $F(000) = 1104$, $Z = 8$, $D_c = 1.46$ g cm^{-3}; unit cell parameters were determined by the least squares method using 25 independent reflections. Data collection was carried out on a four-circle Enraf−Nonius CAD4 diffractometer using monochromated Mo-K_α radiation ($\lambda = 0.71073$ Å), $T = 293$ K. The ω-2θ technique was used to measure 26257 reflections, range $2° \le \theta \le 27.48°$. Three standard reflections monitored every hour during data collection showed a decay of 4.49%. 5129 unique data with $I \ge 3\sigma(I)$ were used to solve and refine the structure. The final $wR(F^2)$ was 0.0508.

[25] W. M. Reiff, W. A. Baker, Jr., *Inorg. Chem.* **1970**, *9*, 570−576.

[26] M. M. Morrison, D. T. Sawyer, *Inorg. Chem.* **1978**, *17*, 338−339.

[27] A. Musumeci, R. P. Bonomo, V. Gucinotta, A. Seminara, *Inorg. Chim. Acta* **1982**, *59*, 133−140.

[28] SIR92: A. Altomare, G. Cascarano, G. Giacovazzo, A. Guagliardi, M.C. Burla, G. Polidori, M. Camalli, *J. Appl. Cryst.* **1994**, *27*, 435.

[30] *CRYSTALS*, Issue 9, D. Watkin; Chemical Crystallography Laboratory, Oxford, **1990**.

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