Sulfur and Selenium Complexes of the Hydrido-tri(3,5-dimethyl-1-pyrazolyl)borate Nitrosylmolybdenum Fragment, [Tp*Mo(NO)]. An X-Ray Crystal Structure Determination of Tp*Mo(NO)(Se₅)

Max Herberhold^{a,*}, Guo-Xin Jin^a, Arnold L. Rheingold^b

^a Laboratorium für Anorganische Chemie, Universität Bayreuth, D-95440 Bayreuth

^b Department of Chemistry, University of Delaware, Newark, Delaware, 19716, U. S. A.

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Molybdenum-chalcogenido Complexes, Nitrosyl Compounds, X-Ray

The reactions of hydrido-tri(3,5-dimethyl-1-pyrazolyl)borate nitrosylmolybdenum diiodide, Tp*Mo(NO)I₂ (**2**), with the oligochalcogenides (NH₄)₂S₁₀ and (NEt₄)₂Se₆ in THF solution lead to mononuclear *cyclo*-pentachalcogenido complexes, Tp*Mo(NO)(E₅) (E = S (**3a**), Se (**3b**)). In the presence of either H₂S or H₂Se (generated by slow "in situ" hydrolysis of Al₂E₃ in moist THF solution) **2** is converted into binuclear chalcogenido-bridged products, Tp₂Mo₂(NO)₂(μ-E)₂ (E = S (**4a**), Se (**4b**)) which are more conveniently obtained from **3a,b** by dechalcogenation with tri(*n*-butyl)phosphane (1:4.5). The new chalcogen complexes **3a,b** and **4a,b** were characterized by IR, NMR and mass spectroscopy and compared with the related chalcogen compounds derived from pentamethylcyclopentadienyl nitrosylmolybdenum diiodide, Cp*Mo(NO)I₂. The molecular structure of Tp*Mo(NO)(Se₅) (**3b**) has been determined; the complex contains a six-membered MoSe₅ metallacycle in the chair conformation and a linearly coordinated nitrosyl ligand (angle Mo–N–O 178.9(17)°).

Introduction

Two earlier publications [1,2] from our group described mononuclear [1] and binuclear [2] chalcogen complexes of the group 6 metals which are formally derived from the halfsandwich fragment [Cp*M(NO)] (Cp* = η⁵-pentamethylcyclopentadienyl):

Cp*M(NO)(E₅) [1]

E = S ; M = Cr, Mo, W
E = Se ; M = Cr, W

Cp₂M₂(NO)₂(μ-E₂)₂ [2]

E = S ; M = Cr, Mo
E = Se ; M = Cr, Mo

In the present communication, we report on selected sulfur and selenium complexes of molybdenum which are derived from the related (isolobal) fragment [Tp*M(NO)]. The reactions are summarized in Scheme 1. Surprisingly, the binuclear products are different in the Cp* and Tp* series.

Scheme 1.

* Reprint requests to Prof. Dr. M. Herberhold.

Table I. IR and NMR spectroscopic data.

Complex	IR (CsI pellets)		NMR (δ values, CDCl ₃ solution) ^a										
	$\nu(\text{B-H})$ [cm ⁻¹]	$\nu(\text{NO})$ [cm ⁻¹]	¹¹ B	¹ H	¹³ C{ ¹ H}								
				CH ₃	CH ₃	CH	Me	Me	CH	CMe	CMe		
3a Tp*Mo(NO)(S ₅)	2547	1679	-7.65	A: 2.17	2.49	5.89	12.7	14.8	106.5	145.1	151.3		
				B: 2.32	2.38	5.84	12.5	14.4	106.4	144.5	151.7		
3b Tp*Mo(NO)(Se ₅)	2548	1665	-7.14	A: 2.15	2.55	6.01	13.1	15.8	108.6	145.6	154.8		
				B: 2.22	2.33	5.79	12.8	16.4	107.9	146.1	154.3		
4a Tp* ₂ Mo ₂ (NO) ₂ (μ -S) ₂	2549	1670	-9.18	A: 2.48	2.63	5.74	12.9	15.3	108.4	144.9	153.2		
				B: 2.32	2.42	6.09	12.6	15.5	107.1	145.1	152.7		
4b Tp* ₂ Mo ₂ (NO) ₂ (μ -Se) ₂	2548	1656	-9.02	A: 2.45	2.53	5.82	12.7	16.0	106.4	152.0	153.9		
				B: 2.30	2.34	5.69	12.5	16.2	107.1	145.2	152.8		
5 Tp* ₂ Mo ₂ (NO) ₂ I ₂ (μ -O) (Ref. [7])	2544	1624	-9.10	A:	^b	5.78							
				B:	^b	5.97							

^a The signals (singlets) of the two equivalent pz* substituents (**B**) possess double intensity as compared with those of the remaining pz* group (**A**); ^b Two broad signals are observed for the six CH₃ groups at $\delta(^1\text{H})$ 2.37 and 1.83, intensity ratio 5:1.

Results and Discussion

1. Synthesis and Spectroscopy

A versatile starting material for the preparation of the new chalcogen complexes is the dicarbonyl nitrosylmolybdenum compound Tp*Mo(NO)(CO)₂ (**1**), which is oxidatively decarbonylated by iodine in solution (*e. g.* in methylcyclohexane) to give the diiodo nitrosylmolybdenum derivative Tp*Mo(NO)I₂ (**2**) [3].

The direct reaction of **2** with excess chalcogen – in the soluble form of either ammonium polysulfide, (NH₄)₂S_x ($x \sim 10$), or tetraethylammonium hexaselenide, (NEt₄)₂Se₆ – in polar solvents leads to the chalcogen-rich *cyclo*-pentasulfido and -pentaselenido compounds **3a** and **3b**, respectively. The mononuclear sulfur complex Tp*Mo(NO)(S₅) (**3a**) corresponds closely to Cp*Mo(NO)(S₅) [1] and can be obtained similarly by prolonged photolysis of **1** in the presence of *cyclo*-octa-sulfur, S₈, in acetonitrile solution. The photochemical route appears not to be suitable for the synthesis of the mononuclear selenium complex **3b**. It is remarkable to note that the counterpart of **3b**, [Cp*Mo(NO)(Se₅)], was not isolated in our studies with the [Cp*Mo(NO)] system [2].

When **2** is exposed to a limited concentration of H₂S or H₂Se, – generated in situ by hydrolysis of Al₂E₃ (E = S, Se) in moist THF – the dinuclear, chalcogen-bridged complexes Tp*₂Mo₂(NO)₂(μ -E)₂ (E = S (**4a**), Se (**4b**)) are isolated after chro-

matography over silica in addition to an oxo-bridged side-product, [Tp*Mo(NO)I]₂(μ -O) (**5**). The binuclear compounds **4a, b** take up chalcogen from either Na₂S₄ or (NEt₄)₂Se₆ to give the chalcogen-rich mononuclear moieties **3a, b**. On the other hand, complexes **3a, b** are cleanly converted into **4a, b** by dechalcogenation with tri(ⁿbutyl)phosphane, PⁿBu₃. This is in contrast to the corresponding dechalcogenation of Cp*Mo(NO)(S₅) using PⁿBu₃ which leads to the formation of a dinuclear product with two disulfido bridges [2]. Attempts to obtain more chalcogen-rich bridges – such as found in Cp*₂Mo₂(NO)₂(μ -E)₂ (E = S, Se) [2], Cp*₂Re₂(L)₂(μ -E)(μ -E₂) (E = S, Se; L = O [4], N^tBu [5]) and Cp*₂Co₂(μ -S₂)₂ [6] – by applying stoichiometric control of the PⁿBu₃ concentration were not successful. It appears that the voluminous Tp* tripod ligand allows only two monochalcogenido bridges in **4a, b**.

The oxo-bridged complex **5** is known [7]; it is apparently formed by partial hydrolysis of **2** under the reaction conditions:

In the IR spectra of **3** - **5** (Table I) a bond of medium intensity near 2250 cm⁻¹ corresponds to the $\nu(\text{B-H})$ vibration, and two absorptions at 1380

1	Tp*Mo(NO)(CO) ₂	1655 [3]	Cp*Mo(NO)(CO) ₂	1639 [1]
2	Tp*Mo(NO)I ₂	1700 [3]	Cp*Mo(NO)I ₂	1641 [1]
3a	Tp*Mo(NO)(S ₅)	1678	Cp*Mo(NO)(S ₅)	1639 [1]
3b	Tp*Mo(NO)(Se ₅)	1665	{CpCo[P(OMe) ₂ O] ₃ }Mo(NO)(S ₅)	1667 [5]
4a	Tp ₂ Mo ₂ (NO) ₂ (μ-S) ₂	1670	Cp ₂ Mo ₂ (NO) ₂ (μ-S ₂) ₂	1652, 1645 [2]
4b	Tp ₂ Mo ₂ (NO) ₂ (μ-Se) ₂	1656	Cp ₂ Mo ₂ (NO) ₂ (μ-Se ₂) ₂	1641, 1615 [2]
5	Tp ₂ Mo ₂ (NO) ₂ (μ-O)	1624		

Table II. Nitrosyl stretching frequencies, $\nu(\text{NO})$ [cm⁻¹] (CsI pellets).

and 1360 cm⁻¹ indicate the $\delta(\text{CH}_3)$ deformations of the two different CH₃*substituents in the Tp* ligand. The nitrosyl stretching frequency, $\nu(\text{NO})$, depends on the nature of the tridentate ligand; thus, an increasing amount of charge density is transferred to the [Mo(NO)(S₅)] fragment when Tp* is replaced by either {CpCo[P(OMe)₂O]₃} or Cp*. The binuclear complexes **4a**, **b** and **5** possess only a single $\nu(\text{NO})$ absorption which is consistent with a centrosymmetric structure (Table II).

As expected, the ¹H and ¹³C NMR spectra (Table I) indicate that two of the three pyrazolyl rings (pz*) are equivalent. This has been confirmed in the X-ray crystallographic structure determination of **3b**.

2. Molecular structure of Tp*Mo(NO)(Se₅) (**3b**)

The molecular geometry of **3b** is shown in Fig. 1; the relevant bond distances and angles are summarized in Table III.

Complex **3b** is monomeric with the central metal Mo in a distorted octahedral coordination sphere. To a first approximation the molecule contains

Fig. 1. Molecular structure of Tp*Mo(NO)(Se₅) (**3b**).Table III. Selected bond distances and angles for Tp*Mo(NO)(Se₅) (**3b**).

Bond distances [Å]			
Mo(1)–Se(1)	2.512(3)	B(1)–N(2)	1.501(31)
Mo(1)–Se(5)	2.471(3)	B(1)–N(4)	1.525(32)
Mo(1)–N(1)	2.207(20)	B(1)–N(6)	1.530(28)
Mo(1)–N(3)	2.291(17)	N(1)–N(2)	1.380(24)
Mo(1)–N(5)	2.178(16)	N(3)–N(4)	1.382(24)
Mo(1)–N(7)	1.769(18)	N(5)–N(6)	1.353(24)
		N(7)–O(7)	1.169(24)
Bond angles [°]			
Se(1)–Se(2)	2.323(3)		
Se(2)–Se(3)	2.316(3)		
Se(3)–Se(4)	2.298(4)		
Se(4)–Se(5)	2.322(4)		
Se(1)–Mo(1)–Se(5)	100.4(1)	Mo(1)–N(7)–O(7)	178.9(17)
Se(1)–Mo(1)–N(1)	167.6(4)	Mo(1)–Se(1)–Se(2)	103.6(1)
Se(1)–Mo(1)–N(3)	86.7(4)	Mo(1)–Se(5)–Se(4)	106.1(1)
Se(1)–Mo(1)–N(5)	91.4(5)	Se(1)–Se(2)–Se(3)	101.8(1)
Se(1)–Mo(1)–N(7)	91.9(6)	Se(2)–Se(3)–Se(4)	106.1(1)
		Se(3)–Se(4)–Se(5)	99.4(1)
Se(5)–Mo(1)–N(1)	90.2(4)		
Se(5)–Mo(1)–N(3)	85.2(4)	N(2)–B(1)–N(4)	110.0(16)
Se(5)–Mo(1)–N(5)	164.5(5)	N(2)–B(1)–N(6)	109.5(17)
Se(5)–Mo(1)–N(7)	94.6(5)	N(4)–B(1)–N(6)	110.0(19)
N(1)–Mo(1)–N(3)	87.8(6)	N(1)–N(2)–B(1)	121.7(18)
N(1)–Mo(1)–N(5)	77.0(6)	N(3)–N(4)–B(1)	121.1(16)
N(1)–Mo(1)–N(7)	93.5(8)	N(5)–N(6)–B(1)	118.4(15)
N(3)–Mo(1)–N(5)	85.5(6)		
N(3)–Mo(1)–N(7)	178.6(8)		
N(5)–Mo(1)–N(7)	95.0(7)		

a mirror plane which passes through the atoms Mo(1) and Se(3), the pz* ring including N(3) and N(4), and the nitrosyl group N(7)–O(7). The cyclo-Se₅ chelate ligand adopts the chair conformation; the selenium atoms Se(1), Se(2), Se(4) and Se(5) are arranged in a plane with an average deviation of only 0.001 Å. An analogous chair conformation was observed earlier for the halfsandwich cyclo-pentasilfido complexes Cp*V(O)(S₅) [8], Cp*W(NO)(S₅) [1], CpCo(PMe₃)(S₅) [9] and {CpCo[P(OMe)₂O]₃}Mo(NO)(S₅) [5]. The bite

angle Se(1)–Mo–Se(5) ($100.4(1)^\circ$) in **3b** is found in the expected range between 89 and 113° (*cf.* refs. [1,8]) and may be compared with the corresponding value in $\text{Tp}^*\text{Mo}(\text{NO})[\text{S}(\text{CH}_2)_2\text{CONH}(\text{CH}_2)_2\text{S}]$ ($105.13(4)^\circ$) [10] and $\text{Tp}^*\text{Mo}(\text{Cl})(\text{S}_4)$ (94.65° *av*) [11]. It is remarkable to note that the reductive sulfuration of $\text{Tp}^*\text{MoO}_2\text{Cl}$ using $\text{B}_2\text{S}_3/\text{S}_8$ in the presence of H_2S led to a *cyclo-tetrasulfido* derivative, $\text{Tp}^*\text{Mo}(\text{Cl})(\text{S}_4)$ [11].

The nitrosyl ligand in **3b** is linearly coordinated (Mo(1)–N(7)–O(7) $178.9(17)^\circ$). The Mo(1)–N(3) bond *trans* to the nitrosyl group is longer ($2.291(17)$ Å) than the two other Mo(1)–N bonds ($2.207(20)$ and $2.178(16)$ Å), as expected as a result of the *trans*-influence. The Mo atom is displaced out of the equatorial plane (containing N(1), N(5), Se(1) and Se(5) with an average deviation of 0.037 Å) towards the nitrosyl ligand by 0.152 Å.

Experimental

All reactions and manipulations were carried out under dry argon using Schlenk techniques, as described earlier [1,2]. The starting nitrosylmolybdenum complexes $\text{Tp}^*\text{Mo}(\text{CO})_2\text{NO}$ (**1**), $\text{Tp}^*\text{Mo}(\text{NO})\text{I}_2$ (**2**) [3] and the oligochalcogenides $(\text{NH}_4)_2\text{S}_x$ ($x \sim 10$) [1], $(\text{NEt}_4)_2\text{Se}_6$ [12] were obtained by methods described previously.

The EI-MS data were obtained on a Varian MAT 8500, the FD-MS data on a Varian MAT 311A instrument; the most intense peak of the isotope pattern is listed.

1. Syntheses

$\text{Tp}^*\text{Mo}(\text{NO})(\text{S}_5)$ (**3a**)

a) By sulfuration: A concentrated solution of ammonium polysulfide, $(\text{NH}_4)_2\text{S}_{10}$, in 5 ml of methanol was added dropwise to the dark-brown solution of 0.67 g (1.00 mmol) $\text{Tp}^*\text{Mo}(\text{NO})\text{I}_2$ (**2**) in 70 ml chloroform. The colour of the reaction mixture changed to green and then to dark-red. The mixture was stirred for 2 h at room temperature before the solvents were removed under reduced pressure. The crude product was chromatographed over silica. Elution with CH_2Cl_2 afforded a dark-red zone from which 0.40 g (68.7%) **3a** was isolated. Recrystallization from a chloroform / toluene / hexane mixture at -25°C gave black needles. EI-MS: *m/e* 585 (M^+ , 22%), 521 (M^+-2S , 20%), 489 (M^+-3S , 100%), 457 (M^+-4S , 68%), 425 ($\text{Tp}^*\text{Mo}(\text{NO})^+$, 46%).

b) By photo-induced decarbonylation in the presence of S_8 : An orange solution containing 0.48 g (1.00 mmol) $\text{Tp}^*\text{Mo}(\text{CO})_2\text{NO}$ (**1**) and 0.26 g (1.00 mmol) S_8 in 100

ml acetonitrile was irradiated at 15°C for 5 h with a high-pressure mercury arc (Hanovia 450 W). Carbon monoxide was liberated and the colour of the solution gradually changed to dark-red. After evaporation of the solvent the product was isolated by column chromatography over silica (elution with CH_2Cl_2) and purified by recrystallization from a chloroform/toluene/hexane mixture at -25°C . Black needles, yield 0.12 g (20.5%).

$\text{Tp}^*\text{Mo}(\text{NO})(\text{Se}_5)$ (**3b**)

A DMF solution (30 ml) containing 0.63 g (0.94 mmol) $\text{Cp}^*\text{Mo}(\text{NO})\text{I}_2$ (**2**) and 0.69 g (0.94 mmol) $(\text{NEt}_4)_2\text{Se}_6$ [12] was stirred at 70°C for 2 h. Work-up of the black reaction product by column chromatography on silica (elution with CH_2Cl_2) gave 0.46 g (60.1%) **3b**. Black prismatic crystals (mp. $260 - 261^\circ\text{C}$) were obtained upon crystallization from chloroform/hexane at -25°C . EI-MS: *m/e* 819 (M^+ , 35%), 661 (M^+-2Se , 58%), 631 ($\text{Tp}^*\text{MoSe}_3^+$, 32%), 583 (M^+-3Se , 30%), 553 ($\text{Tp}^*\text{MoSe}_2^+$, 30%), 503 (M^+-4Se , 100%), 473 (Tp^*MoSe^+ , 64%).

$\text{Tp}_2^*\text{Mo}_2(\text{NO})_2(\mu-\text{S})_2$ (**4a**)

a) By desulfurization: A dark-red solution of 0.24 g (0.41 mmol) $\text{Tp}^*\text{Mo}(\text{NO})(\text{S}_5)$ (**3a**) and 0.37 g (1.83 mmol) tri(*n*-butyl)phosphane, P^nBu_3 , in 100 ml CH_2Cl_2 was stirred at room temperature for 40 min. The dark-brown reaction mixture was brought to dryness in a vacuum and the residue separated by column chromatography on silica. Elution with CH_2Cl_2 produced a dark red-brown zone which contained 0.16 g (85.7%) **4a**. Recrystallization from either $\text{CHCl}_3/\text{hexane}$ (-25°C) or $\text{C}_6\text{H}_6/\text{hexane}$ (room temperature) gave a pure product. EI-MS: *m/e* 910 (M^+ , 24%), 880 (M^+-NO , 4%), 814 ($\text{M}^+-\text{pz}^*\text{H}$, 8%), 784 ($\text{M}^+-\text{pz}^*\text{H}-\text{NO}$, 8%), 521 ($\text{Tp}^*\text{Mo}(\text{NO})\text{S}_3^+$, 13%), 489 ($\text{Tp}^*\text{Mo}(\text{NO})\text{S}_2^+$, 15%), 457 ($\text{Tp}^*\text{Mo}(\text{NO})\text{S}^+$, 100%), 425 ($\text{Tp}^*\text{Mo}(\text{NO})^+$, 40%), FD-MS: *m/e* 909.7 (M^+), correct isotope pattern.

b) By sulfuration: Hydrogen sulfide, H_2S , was slowly generated by adding a solution of 0.23 g (12.78 mol) water in 10 ml THF to a reaction mixture which contained 0.32 g (0.48 mmol) $\text{Tp}^*\text{Mo}(\text{NO})\text{I}_2$ (**2**) and 0.64 g (4.27 mmol) Al_2S_3 in 80 ml THF. The colour changed immediately from dark-brown to green and then gradually to red-brown. The reaction mixture was stirred for 2 d, then filtered and the filtrate brought to dryness. Work-up by column chromatography over silica gave two products: 0.14 g (67.4%) $\text{Tp}_2^*\text{Mo}_2(\text{NO})_2(\mu-\text{S})_2$ (**4a**, dark red-brown, eluted with CH_2Cl_2), and 0.06 g (22.6%) $\text{Tp}_2^*\text{Mo}_2(\text{NO})_2\text{I}_2(\mu-\text{O})$ (**5** [7], green, eluted with $\text{CH}_2\text{Cl}_2/\text{THF}$ ($3:1$)). Both compounds were recrystallized from $\text{CHCl}_3/\text{hexane}$ at -25°C . EI-MS of **5**: *m/e* 1117 (M^+ , 20%), 990 (M^+-I , 17%), 974 ($\text{Tp}_2^*\text{Mo}_2(\text{NO})_2\text{I}^+$, 52%), 847 ($\text{Tp}_2^*\text{Mo}_2(\text{NO})_2^+$, 20%), 679 ($\text{Tp}^*\text{Mo}(\text{NO})\text{I}_2^+$, 14%),

649 ($\text{Tp}^*\text{MoI}_2^+$, 38%), 552 ($\text{Tp}^*\text{Mo}(\text{NO})\text{I}^+$, 100%), 522 (Tp^*MoI^+ , 98%).

$\text{Tp}_2^*\text{Mo}_2(\text{NO})_2(\mu\text{-Se})_2$ (**4b**)

a) By deselenization: The dark-brown solution of 0.24 g (0.29 mmol) $\text{Tp}^*\text{Mo}(\text{NO})(\text{Se}_3)$ (**3b**) and 0.26 g (1.31 mmol) tri(*n*-butyl)phosphane, $\text{P}^n(\text{Bu})_3$, in 100 ml CH_2Cl_2 was stirred for 1 h and then worked up as described above for **4a**. The product **4b** (0.12, 75.2%) was obtained as violet prismatic crystals when crystallized from $\text{CHCl}_3/\text{hexane}$ (-25°C). EI-MS: 1004 (M^+ , 2%), 661 ($\text{Tp}^*\text{Mo}(\text{NO})\text{Se}_3^+$, 4%), 503 ($\text{Tp}^*\text{Mo}(\text{NO})\text{Se}^+$, 14%), 473 (Tp^*MoSe^+ , 8%), 96 (pz^*H^+ , 100%). FD-MS: m/e 1004.2 (M^+), correct isotope pattern.

b) By selenization: Moist THF (*i. e.* 0.053 g, 2.96 mmol H_2O in 10 ml THF) was added dropwise at ambient temperature to a suspension of 0.50 g (0.74 mmol) $\text{Tp}^*\text{Mo}(\text{NO})\text{I}_2$ (**2**) and 0.47 g (1.48 mmol) Al_2Se_3 in 100 ml THF. In the course of 2 d the colour became violet. After filtration the solution was brought to dryness and the product mixture was separated by column chromatography on silica. Elution with CH_2Cl_2 developed a violet zone which contained 0.29 g (77.%) **4b**, while subsequent elution with $\text{CH}_2\text{Cl}_2/\text{THF}$ (3:1) gave 0.04 g (9.6%) of green **5**. Recrystallization from $\text{CHCl}_3/\text{hexane}$ at -25°C gave a pure product.

2. Crystal structure determination of $\text{Tp}^*\text{Mo}(\text{NO})(\text{Se}_3)$ (**3b**)

A black prismatic crystal ($0.10 \times 0.16 \times 0.38 \text{ mm}^3$) was grown by slow crystallization from chloroform / toluene / hexane at -25°C . Formula $\text{C}_{15}\text{H}_{22}\text{BMoN}_7\text{OSe}_5$ (817.9). Monoclinic, space group $\text{C}2/c$; $a = 20.876(4)$, $b = 11.309(2)$, $c = 21.792(4) \text{ \AA}$, $\beta = 102.42(3)^\circ$; $V = 5024.4(8) \text{ \AA}^3$, $Z = 8$. $D_{\text{calc.}}$ 2.163 g/cm^3 , $\mu(\text{MoK}\alpha)$ 9.520 mm^{-1} .

Data collection: Diffractometer Siemens P4, $\text{MoK}\alpha$ radiation ($\lambda = 0.71073 \text{ \AA}$), graphite monochromator; 241 K, 2θ scan range $4.0 - 45.0^\circ$, scan type 2θ - θ , index range $-22 \leq h \leq 21$, $0 \leq k \leq 12$, $0 \leq l \leq 23$. Collected reflections 3298, independent reflections 3199 ($R_{\text{int}} = 5.94\%$), observed reflections 1864 ($F > 4.0\sigma(F)$).

Solution and refinement: Siemens SHELXTL PLUS, direct methods, $R = 6.82\%$, $R_w = 8.35\%$, ($w^{-1} = \sigma^2(F) + 0.0020F^2$); largest difference peak 1.82 $\text{e\AA}^{-3}$, largest difference hole $-1.15 \text{ e\AA}^{-3}$, GOF 1.38 [13].

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