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Silver(I) complexes of 1,3-diallylbenzimidazolin-2-one: A versatile ligand for discrete binuclear assemblies and a polymeric ladder and helix

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ABSTRACT

1,3-Diallylbenzimidazolin-2-one (**2**) reacts with silver tetrafluoroborate and hexafluorophosphate salts to furnish discrete binuclear complexes, whereas reaction with silver perchlorate and triflate salts provided 1D coordination polymers. Within each of the structures the ligand adopted a different coordination mode.

1. Introduction

We have long been involved in the design and synthesis of heterocyclic ligands for use in coordination, organometallic and metallosupramolecular chemistry [1]. The use of silver salts for the preparation of diverse metallosupramolecular assemblies has proved particularly popular in recent years [2]. We have championed the use of the silver-alkene interaction as a useful synthon in supramolecular chemistry [3]. Recently, we reported that 1,3-diallylurea (**1**) (Scheme 1) reacts with silver salts to produce 1D polymeric ladders and helicates [4]. This ligand has considerable conformational flexibility that hinders the control of the species produced. We were interested in using a less flexible ligand and now describe the use of 1,3-diallylbenzimidazolin-2-one (**2**) for the construction of both discrete and polymeric silver assemblies.

2. Experimental

2.1. General

Melting points were determined using an Electrothermal melting point apparatus and are uncorrected. Elemental analyses were performed by the Campbell microanalytical laboratory at the University of Otago. Solvents were purified according to standard procedures. Other reagents were obtained from commercial sources and used as supplied.

2.2. Preparation of 1,3-diallylbenzimidazolin-2-one (*2*)

A mixture of 2-hydroxybenzimidazole (2.68 g, 20.0 mmol), allylbromide (3.5 ml, 40.0 mmol), K_2CO_3 (5.52 g, 40 mmol) in acetone (50 ml) was refluxed overnight. The reaction mixture was cooled to room temperature and the solid material was filtered off. The solvent was removed *in vacuo* to give a yellow oily liquid. The oily liquid was washed with 50 ml of 1 M NaOH solution and the organic layer was extracted with 3 × 40 ml diethyl ether. The organic extracts were combined and dried over $MgSO_4$. Diethyl ether was evaporated under reduced pressure to give the crude product as a light yellow oily liquid. Purification of the crude product with silica gel column chromatography using 20:80 ethyl acetate:petroleum ether gave pure **2** as an oily liquid (3.0 g, 70%). ^{1}H NMR (500 MHz, $CDCl_3$): δ 4.53 (4H, d, J = 5.0 Hz, H5), 5.20–5.24 (4H, m, H7), 5.87–5.94 (2H, m, H6), 6.99 (2H, d, J = 9.0 Hz, H1), 7.07 (2H, d, J = 9.0 Hz, H2). ^{13}C NMR (126 MHz, $CDCl_3$): δ 43.79 C5, 108.48 C2, 117.79 C7, 121.47 C1, 129.56 C3, 132.29 C6, 154.02 C4. IR (cm^{-1}) 3489, 3064, 3014, 2985, 2918, 1863, 1710, 1645, 1619, 1493, 1439, 1403, 1349, 1327, 1301, 1274, 1198, 1173, 1135, 1077, 1022, 990, 926, 830, 755, 735, 692. ESI-MS: found MNa^+ = 237.1001; $C_{13}H_{14}N_2ONa$ requires MNa^+ = 237.0998.

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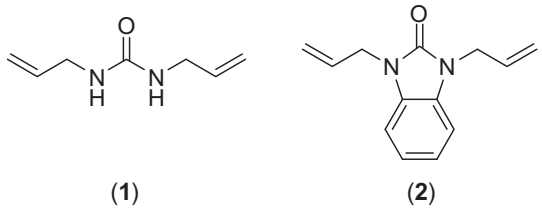
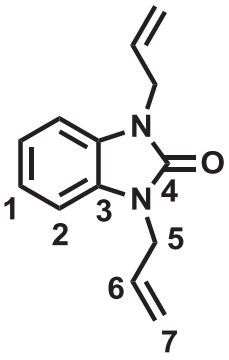


(1) (2)

Scheme 1.



2.3. Preparation of silver(I) complexes **3–6**

1,3-Diallylbenzimidazolin-2-one (0.0218 g, 0.10 mmol) was dissolved in 1 ml acetone and was added to the appropriate silver(I) salt (0.10 mmol) also dissolved in 1 ml acetone. The solution was left in darkness at room temperature, and diethyl ether was allowed to diffuse into the solution. This enabled the isolation of colourless crystals suitable for single crystal X-ray structure analysis.

Data for complex **3**: Yield 0.0127 g, 31%. M.p. 179–181 °C. IR (cm^{-1}): 2985, 2926, 1649, 1611, 1489, 1448, 1431, 1411, 1329, 1277, 1205, 1094, 1047, 1028, 1001, 957, 889, 760, 685. Elem. *Anal.* Calc. for $C_{13}H_{14}N_2O{\cdot}AgBF_4$: C, 38.18; H, 3.45; N, 6.85. Found: C, 38.57; H, 3.32; N, 6.88%.

Data for complex **4**: Yield 0.0134 g, 29%. M.p. 109–111 °C. IR (cm^{-1}): 2921, 2850, 1649, 1612, 1489, 1449, 1431, 1413, 1329, 1277, 1206, 1189, 1049, 1024, 1002, 957, 822, 758, 686. Elem. *Anal.* Calc. for $C_{13}H_{14}N_2O{\cdot}AgPF_6$: C, 33.43; H, 3.02; N, 6.00. Found: C, 33.50; H, 3.21; N, 6.10%.

Data for complex **5**: Yield 0.0378 g, 90%. M.p. 139–141 °C. IR (cm^{-1}): 2978, 2922, 1645, 1610, 1489, 1446, 1403, 1328, 1276, 1204, 1066, 1002, 957, 759, 684. Elem. *Anal.* Calc. for $C_{13}H_{14}N_2O{\cdot}AgClO_4$: C, 37.04; H, 3.35; N, 6.64. Found: C, 37.18; H, 3.22; N, 6.59%.

Data for complex **6**: Yield 0.0319 g, 68%. M.p. 69–71 °C. IR (cm^{-1}): 3010, 2927, 1668, 1619, 1490, 1437, 1400, 1359, 1224, 1196, 1157, 1081, 1026, 944, 753, 632. Elem. *Anal.* Calc. for $C_{13}H_{14}N_2O.AgSO_3CF_3{\cdot}H_2O$: C, 34.37; H, 3.30; N, 5.73%. Found: C, 34.51; H, 3.12; N, 5.74%.

2.4. X-ray crystallography

The crystal data and details of the data collections and refinements for the X-ray structures are listed in Table 1. Measurements were made with a Bruker APEX II instrument using Mo Kα (λ = 0.71073 Å) radiation. The structures were solved by direct methods using SHELXS [5] and refined on F^2 using all data by full-matrix least-squares procedures with SHELXL [5]. Non-hydrogen atoms

Table 1
Crystal data and structure refinement for **3–6**.

Complex	**3**	**4**	**5**	**6**
Empirical formula	$C_{26}H_{28}Ag_2B_2F_8N_4O_2$	$C_{26}H_{32}Ag_2F_{12}N_4O_4P_2$	$C_{13}H_{14}AgClN_2O_5$	$C_{14}H_{16}AgF_3N_2O_5S$
Formula weight	817.88	970.24	421.58	489.22
T (K)	112	113	113	113
Crystal system	monoclinic	triclinic	triclinic	monoclinic
Space group	$P2_1/n$	$P\bar{1}$	$P\bar{1}$	$P2_1/n$
a (Å)	8.3701(3)	8.5059(5)	8.7979(3)	8.9647(3)
b (Å)	16.7017(5)	9.5738(6)	8.8951(3)	10.7915(4)
c (Å)	10.0532(3)	10.4942(6)	9.3445(3)	18.1858(6)
α (°)	90	92.382(4)	72.488(2)	90
β (°)	96.754(2)	105.339(3)	84.987(2)	96.909(2)
γ (°)	90	101.690(4)	82.233(2)	90
V (Å^3)	1395.63(8)	802.94(8)	690.15(4)	1746.57(10)
Z	2	1	2	4
ρ_{calc} (mg/mm^3)	1.946	2.007	2.029	1.860
μ (mm^{-1})	1.491	1.430	1.680	1.333
F(000)	808	480	420	976
Crystal size (mm^3)	0.49 × 0.40 × 0.15	0.48 × 0.44 × 0.39	0.45 × 0.32 × 0.28	0.26 × 0.16 × 0.14
2θ range for data collection (°)	4.76–55.0	6.24–50.1	4.58–55.0	5.34–53.8
Index ranges	$-10 \leqslant h \leqslant 10$, $-21 \leqslant k \leqslant 21$, $-13 \leqslant l \leqslant 13$	$-10 \leqslant h \leqslant 10$, $-11 \leqslant k \leqslant 11$, $-12 \leqslant l \leqslant 12$	$-11 \leqslant h \leqslant 11$, $-11 \leqslant k \leqslant 11$, $-12 \leqslant l \leqslant 12$	$-11 \leqslant h \leqslant 11$, $-13 \leqslant k \leqslant 13$, $-23 \leqslant l \leqslant 23$
Reflections collected	31314	14991	15575	37451
Independent reflections (R_{int})	3192 (0.0366)	2842 (0.0540)	3170 (0.0355)	3755 (0.0715)
Completeness	100	99.6	100	99.5
Data/restraints/parameters	3192/0/227	2842/0/227	3170/0/199	3755/2/243
Goodness-of-fit (GOF) on F^2	1.050	0.996	1.063	1.059
Final R indexes [$I \geqslant 2\sigma(I)$]	R_1 = 0.0186, wR_2 = 0.0485	R_1 = 0.0355, wR_2 = 0.0864	R_1 = 0.0208, wR_2 = 0.0538	R_1 = 0.0346, wR_2 = 0.0809
Final R indexes [all data]	R_1 = 0.0216, wR_2 = 0.0497	R_1 = 0.0409, wR_2 = 0.0884	R_1 = 0.0228, wR_2 = 0.0549	R_1 = 0.0489, wR_2 = 0.0845