

The coordination chemistry of 1,3-dithiole-2-thione-4,5-dithiolate (dmit) and isologs

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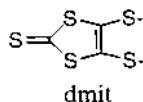
Abstract

The 1,3-dithiole-2-thione-4,5-dithiolate (dmit) ligand and chalcogen atom substituted isologs have been extensively used in organic and coordination chemistry since their first reported synthesis in 1975. Over approximately the past 8 years a number of diverse and interesting dmit coordination complexes have been described. Also, dmit and isologs have been used in a variety of material applications, including in superconducting charge-transfer and radical anion salts. The synthesis, structures and properties of a selection of these compounds will be presented and discussed covering the literature from 1991 to approximately mid-1998. © 1999 Elsevier Science S.A. All rights reserved.

Keywords: Chalcogen; 1,3-Dithiole-2-thione-4,5-dithiolate; Dmit

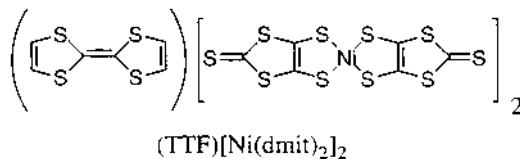
1. Introduction

Over the past 20 years, an enormous amount of research has gone into the synthesis and characterization of 1,3-dithiole-2-thione-4,5-dithiolate (dmit) ligand complexes and related selenium and oxygen substituted isologs.



First accurately reported by Steimecke and coworkers at the University of Leipzig, Germany in the mid 1970's [1], over 700 papers to date have been published which involve the dmit ligand or any of its chalcogen isologs. The acronym 'dmit' stems from the name dimercaptioisotrithione, an older nomenclature system for sulfur-based heterocycles [2,3]. Several reviews have been published devoted to the synthetic aspects of the dmit ligand in organic and coordination chemistry [4–6].

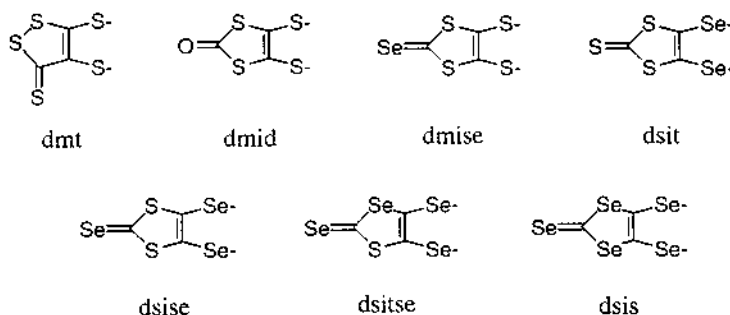
Dmit and related ligand complexes have been used in a variety of applications, but most notably in the assembly of highly electrically conducting radical anion salts and charge-transfer complexes. The discovery of the first molecular superconductor containing a transition metal complex, (TTF)[Ni(dmit)₂]₂ (TTF = tetrathiafulvalene), by Cassoux and coworkers in Toulouse, France initiated a significant effort into the search for more of these fascinating materials.



Currently, eight superconducting complexes have been reported based on the dmit ligand [7–17]. Due to the sulfur-rich, fully conjugated and planar structure of the ligand, and in combination with square planar coordinating metal centers, these complexes are able to stack in a 'poker-chip' like style due to their full planarity, and with a large amount of intermolecular overlap due to the large sulfur atomic

orbitals. This can result in an extensively delocalized system required for highly conducting materials. Several reviews and book chapters have been written solely on the conducting and structural properties of $M(\text{dmit})_2$ anion radical salts and charge-transfer complexes which can be consulted for an in depth understanding of this area [18–22]. The research effort for the assembly of highly conducting systems based on dmit ligand complexes continues to be a major focus. There are a number of other applications that are being investigated such as in chemosensors [23–25], transistors [26,27], magnetic [28–35], non-linear optical [36–41] and in dual electron and ion conducting materials [42]. Due to the broad array of applications seen for the dmit complexes, interdisciplinary collaborations and interactions by chemists, physicists and materials scientists are needed to fully understand their properties.

In 1992, the first comprehensive review appeared on the coordination and organic chemistry of dmit [5]. This review covered the literature from the first report of the synthesis of dmit in 1975 along with the chemistry of the 1,2-dithiole-3-thione-4,5-dithiolate (dmt) isomer of dmit and the 1,3-dithiole-2-one-4,5-dithiolate (dmid) ligand. Since the publication of this review, an enormous amount of work has continued in this area especially in the assembly of a large number of bis- and tris-chelate complexes along with novel mixed-ligand and multimetallic cluster compounds. Also, a large amount of work has gone into the investigation of selenium-rich isologs of dmit which were not covered in the 1992 review. The structures of the selenium containing compounds of focus of 1,3-dithiole-2-selone-4,5-dithiolate (dmise), 1,3-dithiole-2-thione-4,5-diselenolate (dsit), 1,3-dithiole-2-selone-4,5-diselenolate (dsise), 1,3-thiaselenole-2-selone-4,5-diselenolate (dsitse) and 1,3-diselenole-2-selone-4,5-diselenolate (dsis) along with dmt and dmid are shown below:



The synthesis and coordination chemistry of dmit, dmt, dmid and the selenium-enriched ligands will be covered from the years 1991 to approximately mid 1998. The organic chemistry of these ligands was recently thoroughly reviewed by Svenstrup and Becher and will not be covered [6]. Also, a large number of papers report using $(R_4N)_2[Zn(\text{dmit})_2]$ ($R = \text{Et}$ or Bu), dmitSnR_2 ($R = \text{alkyl}$ group) or other coordination compounds simply as ligand delivery systems or

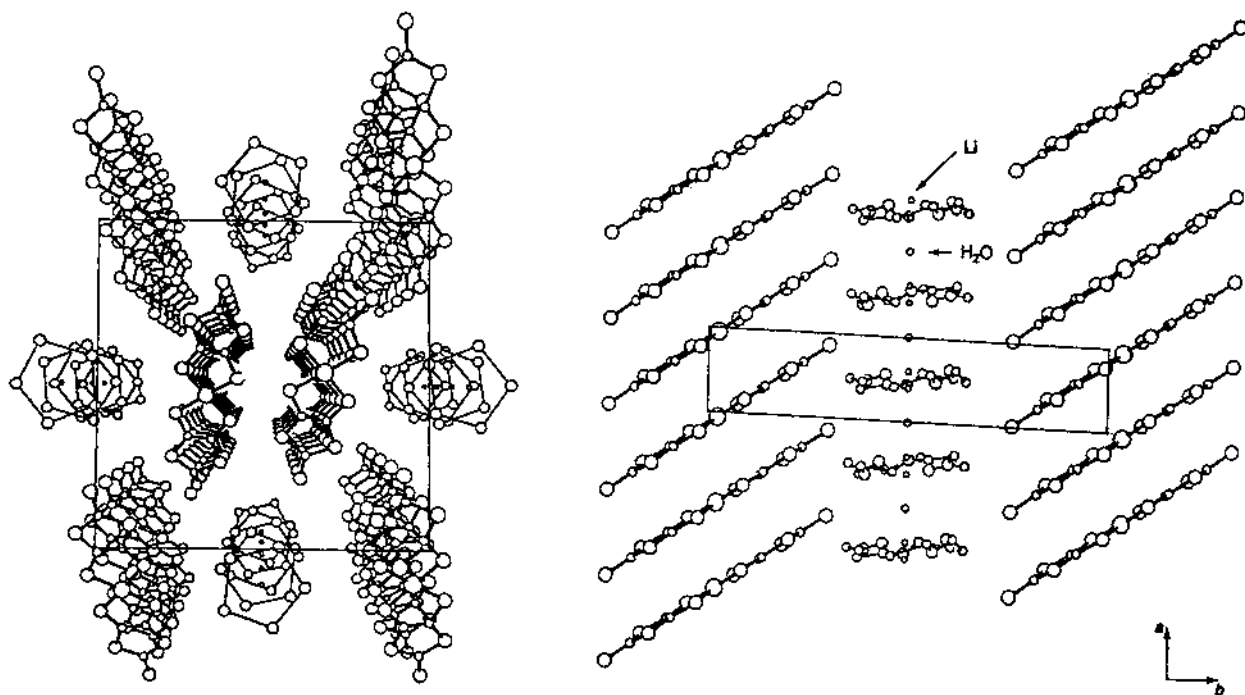


Fig. 1. Views of the crystal packing of $\text{Li}_{0.6}(\text{15-crown-5-ether})[\text{Ni}(\text{dmit})_2]_2 \cdot \text{H}_2\text{O}$ down the a axis (left) and in the a – b plane (right) showing stacks of $\text{Ni}(\text{dmit})_2$ units separated by stacks of crown ether moieties [42]. Reprinted by permission from Nature 394 (1998) 159 copyright 1998 Macmillan Magazines Ltd.

Table 1

Closed-shell bis-chelate complexes of the general formula $(A)_n[Ni(dmit)_2]_m$

A	<i>n</i>	<i>m</i>	Ref.
–		1	[24,126–132]
H ₄ N	2	1	[133]
H ₄ N	1	1	[133,134]
H ₄ N	<i>x</i>	1	[134]
H ₄ N/18-crown-6	1	3	[135]
Me ₄ N	2	1	[136–139]
Me ₄ N	1	1	[136,138,139]
Me ₄ N	1	2	[134,140–145]
Me ₃ HN	2	1	[133,134,137]
Me ₃ HN	1	1	[133,134,137]
Me ₃ HN	1	2	[133,134,137,146]
Me ₃ HN	2	5	[133,134,137]
Me ₂ H ₂ N	2	1	[133,134]
Me ₂ H ₂ N	1	1	[133, 134]
Me ₂ H ₂ N	1	2	[133,134,137]
MeH ₃ N	2	1	[133,134]
MeH ₃ N	1	1	[133,134]
MeH ₃ N	2	5	[133,134,137]
Et ₄ N	2	1	[147–149]
Et ₄ N	1	1	[147]
Et ₄ N	0.5	1	[149]
Et ₃ HN	2	1	[131]
Et ₃ HN	1	1	[131]
Et ₃ HN	<i>x</i>	1	[131]
Et ₂ H ₂ N	2	1	[131]
Et ₂ H ₂ N	1	1	[131]
Et ₂ H ₂ N	<i>x</i>	1	[131]
Pr ₃ HN	2	1	[131]
Pr ₃ HN	1	1	[131]
Pr ₃ HN	<i>x</i>	1	[131]
Pr ₂ H ₂ N	2	1	[129]
Pr ₂ H ₂ N	1	1	[131]
Pr ₂ H ₂ N	<i>x</i>	1	[131]
Bu ₃ HN	2	1	[131]
Bu ₃ HN	1	1	[131]
Bu ₃ HN	<i>x</i>	1	[131]
Bu ₂ H ₂ N	2	1	[131]
Bu ₂ H ₂ N	1	1	[131]
Bu ₂ H ₂ N	<i>x</i>	1	[131]
Bu ₄ N	2	1	[39–41,128,147,149–157]
Bu ₄ N	1	1	[23–25,36,39,40,126,128,147,151,152,154,158–161]
Bu ₄ N	0.29	1	[24,137,149,162]
Bu ₄ N/ γ -cyclodextrin	1	1	[154,163]
Me ₂ Et ₂ N	2	1	[164]
α -Me ₂ Et ₂ N	1	2	[140,164–172]
γ -Me ₂ Et ₂ N	1	2	[172]
Me ₃ (C ₁₂ H ₂₅)N	2	1	[139]
Me ₃ (C ₁₂ H ₂₅)N	1	1	[139]

Table 1 (Continued)

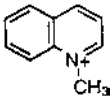
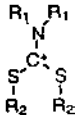
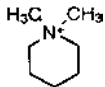
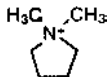
A	<i>n</i>	<i>m</i>	Ref.
Me ₃ (C ₁₆ H ₃₃)N	2	1	[173–175]
Me ₃ (C ₁₈ H ₃₇)N	2	1	[139]
Me ₃ (C ₁₈ H ₃₇)N	1	1	[139]
Me ₂ (C ₁₈ H ₃₇) ₂ N	2	1	[139]
Me ₂ (C ₁₈ H ₃₇) ₂ N	1	1	[139]
Me ₂ (C ₁₂ H ₂₅) ₂ N	2	1	[139,176–181]
Me ₂ (C ₁₂ H ₂₅) ₂ N	1	1	[139,178]
MeO(CH ₂) ₂ H ₃ N	1	3	[182]
<i>N</i> -Alkylpyridinium			
			
R ₁ = CH ₃ , R ₂ = H (Mepy)	2	1	[156]
R ₁ = C ₁₂ H ₂₅ , R ₂ = H	2	1	[179]
R ₁ = C ₁₈ H ₃₇ , R ₂ = H	2	1	[160,181,183–185]
R ₁ = C ₁₈ H ₃₇ , R ₂ = H	1	1	[26,27,160,186–188]
R ₁ = CH ₃ , R ₂ = -N(CH ₃) ₂	1	1	[189]
R ₁ = CH ₃ , R ₂ = -CH=CH-C ₆ H ₄ N(CH ₃) ₂	1	1	[189]
R ₁ = CH ₃ , R ₂ = -(CH=CH)-C ₆ H ₄ N(CH ₃) ₂	1	1	[189]
<i>N</i> -Methylquinolinium	2	1	[156]
	1	1	[190]
2-Dialkylamino-1,3-dithioanylinium			
			
R ₁ = CH ₃ , R ₂ = CH ₃	1	1	[191,192]
MDA (R ₁ = CH ₃ , R ₂ = -CH ₂ -)	1	1	[191–193]
	1	2	[192,193]
EDA (R ₁ = C ₂ H ₅ , R ₂ = -CH ₂ -)	1	1	[191,192,194]
	1	3	[194]
PDA (R ₁ -R ₁ = -(CH ₂) ₅ -; R ₂ = -CH ₂ -)	1	1	[192]
<i>N,N</i> -Dimethylpiperidinium	1	2	[119,169,170,172]
			
<i>N,N</i> -Dimethylpyrrolidinium	2	1	[195]
			
(Dmp)	1	1	[195,196]
	1	2	[119,195,196]

Table 1 (Continued)

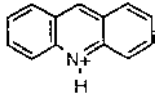
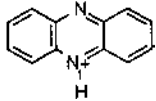
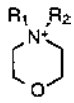
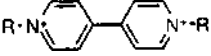
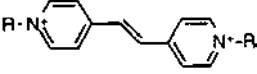
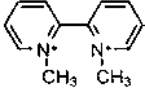
A	<i>n</i>	<i>m</i>	Ref.
Acridinium	1	1	[197,198]
	1	3	[197,198]
Phenazinium	1	1	[197]
 (PheH)	1	3	[197]
Morpholinium			
			
Hmorph ($R_1 = R_2 = H$)	2	3	[182,199]
dmm ($R_1 = R_2 = CH_3$)	2	1	[195]
	1	1	[195]
	1	2	[195]
Hmemorph ($R_1 = H, R_2 = CH_3$)	1	2	[182]
Imidazolium			
			
R = H	1	2	[200]
R = $C_{12}H_{15}$	2	1	[201]
R = $C_{12}H_{15}$	1	1	[201]
Viologen			
			
MV ($R = CH_3$)	1	1	[147,152,157,202]
	2	1	[152]
OV ($R = C_8H_{17}$)	1	1	[157,202,203]
StV ($R = C_{18}H_{37}$)	1	1	[157,202,203]
			
R = CH_3	1	1	[204]
R = C_4H_9	1	1	[204]
R = C_5H_{11}	1	1	[204]
R = C_3H_6CN	1	1	[204]
R = C_4H_8CN	1	1	[204]
R = $CH_2C_6H_4CN$	1	1	[204]
MQ	1	1	[157,202,205]
			

Table 1 (Continued)

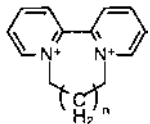
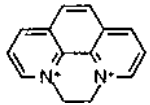
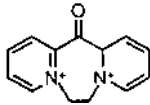
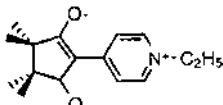
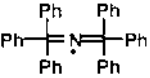
A	<i>n</i>	<i>m</i>	Ref.
2,2'-Bipyridinium			
			
DQ (<i>n</i> = 10)	1	1	[147,152,157,202,203]
	2	1	[152]
PQ (<i>n</i> = 11)	1	1	[147,157,202,205]
BQ (<i>n</i> = 12)	1	1	[147,157,202,205]
DP	1	1	[157,202,203]
			
DPK	1	1	[157,202,203]
			
Me ₃ N(CH ₂) ₄ NMe ₃	1	2	[206]
Me ₃ N(CH ₂) ₄ NMe ₃	1	5	[206]
Gua	1	2	[207–209]
Me ₂ Gua	1	2	[207–209]
Me ₄ Gua	1	2	[208–210]
Me ₆ Gua	1	2	[208–210]
<i>p</i> -EPYNN	1	1	[34,211,212]
			
Me ₂ Et ₂ P	1	2	[213]
MePh ₃ P	1	3	[214,215]
Ph ₄ P	1	3	[162,216–218]
Ph ₄ P	1	1	[216,217]
PhCH ₂ Ph ₃ P	2	1	[219]
PhCH ₂ Ph ₃ P	1	1	[219]
PhCH ₂ Ph ₃ P	1	3	[219,220]
PPN	2	1	[219,221]
	1	1	[219,221]
	0.25	1	[221]
	<i>x</i>	1	[220]
Me ₃ S	2	1	[219]
Me ₃ S	1	1	[219]
Me ₃ S	1	2	[162,218,219]

Table 1 (Continued)

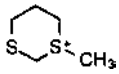
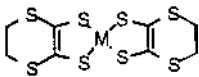
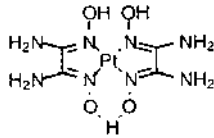
A	<i>n</i>	<i>m</i>	Ref.
Et ₃ S	2	1	[219]
Et ₃ S	1	1	[219]
Et ₃ S	<i>x</i>	1	[220]
MeEt ₂ S	2	1	[219]
MeEt ₂ S	1	1	[219]
MeEt ₂ S	<i>x</i>	1	[220]
Me ₂ EtS	2	1	[219]
Me ₂ EtS	1	1	[219]
S-Methyl-1,3-dithianium	2	1	[195]
 (smdt)	1	1	[195,196]
	1	2	[195,196]
Ph ₄ As	0.25	1	[137]
Cu(cyclam)(CH ₃ CN) ₂	1	2	[28,29]
Ni(cyclam) (CH ₃ CN) ₂	1	2	[29]
Co(Cp) ₂	2	1	[126–128,159]
Co(Cp) ₂	1	1	[126–128,159,222,223]
Co(Cp) ₂	1	3	[159]
Fe(Cp) ₂	1	1	[224]
Fe(Cp)C ₆ H ₅	2	1	[41,225]
Re(CO) ₅	2	1	[226]
M(ddd _t) ₂			
			
M = Pt	1	1	[227]
	1	2	[220]
M = Ni	1	1	[227]
	1	<i>x</i>	[220]
Pt(H ₂ dag)(Hdag)	1	1	[228]
			
Li	2	1	[229]
Li	0.5	1	[229]
Li	0.27	1	[229]
Li	<i>x</i>	1	[199]
Li/(12-crown-4) _{1.5}	2	7	[230]
Li/(15-crown-5) _{1.66}	0.6	2	[42]
Na	2	1	[229]
Na	1	1	[159]
Na	0.5	1	[229]
Ph ₂ I	1	1	[131]
Ph ₂ I	<i>x</i>	1	[131]
vinPh ₂	<i>x</i>	1	[131]

Table 2

Open-shell bis-chelate complexes of the general formula $(A)_n[Ni(dmit)_2]_m$

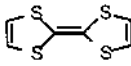
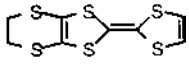
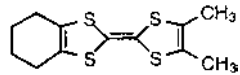
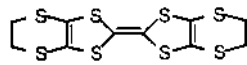
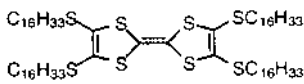
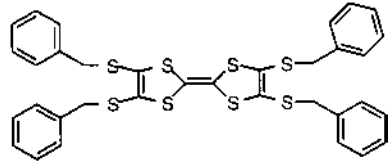
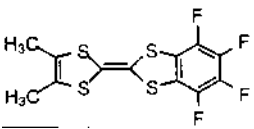
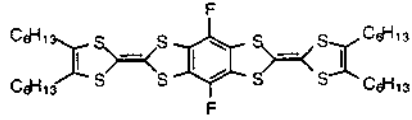
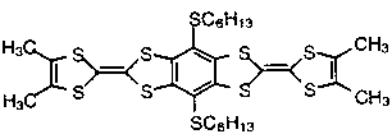
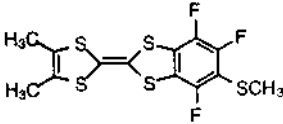
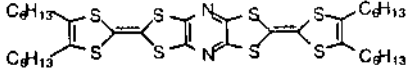
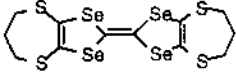
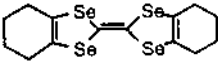
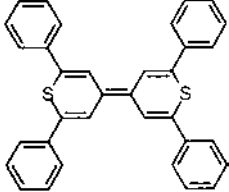
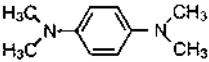
A	<i>n</i>	<i>m</i>	Ref.
TTF	1	2	[30,142,231–238]
			
EDT-TTF	1	1	[16,35,171,239–245]
2	2	2	[246]
			
DMC-TTF			
	1	<i>x</i>	[247]
BEDT-TTF	1	1	[148]
			
THT-TTF	1	1	[148]
			
TBT-TTF	1	1	[148,248,249]
			
D1	<i>x</i>	1	[131,220,250]
			
D2	<i>x</i>	1	[131]
			
D3	<i>x</i>	1	[131]
			

Table 2 (Continued)

A	<i>n</i>	<i>m</i>	Ref.
D4	<i>x</i>	1	[131]
			
D5	<i>x</i>	1	[131]
			
BPDT-TseF	1	1	[251,252]
	1	2	[251,252]
OMTSeF	1	1	[253]
			
DIPSPH ₄	1	1	[254]
			
TMFD	3	1	[255]
			

precursors to organic chemistry on the ligand. These papers will only be referenced, not discussed [43–109].

2. Synthesis of 1,3-dithiole-2-thione-4,5-dithiolate isologs

The synthesis of the root ligand, dmit, first described in 1975 by Steimecke [1] and recently improved in 1996 [81], is the key starting procedure for the synthesis of dmt ('Steimecke rearrangement') [1,5,110], dmld [5,111] and also the mono selenium

Table 3

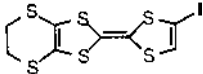
Bis-chelate complexes of the general formula $(A)_n[Pd(dmit)_2]_m$

A	<i>n</i>	<i>m</i>	Ref.
–		1	[127–129,132]
H ₄ N	2	1	[133]
H ₄ N	1	1	[133,134]
Me ₄ N	2	1	[136,138]
Me ₄ N	1	1	[136, 138]
Me ₄ N	1	2	[14,136,138,143,213,256–264]
Me ₃ HN	2	1	[133,134]
Me ₃ HN	1	1	[133,134]
Me ₂ H ₂ N	2	1	[133,134]
Me ₂ H ₂ N	1	1	[133,134]
MeH ₃ N	2	1	[133,134]
MeH ₃ N	1	1	[133,134]
Et ₃ HN	2	1	[131]
Et ₃ HN	1	1	[131]
Et ₃ HN	<i>x</i>	1	[131]
Et ₂ H ₂ N	2	1	[131]
Et ₂ H ₂ N	1	1	[131]
Et ₂ H ₂ N	<i>x</i>	1	[131]
Pr ₃ HN	2	1	[131]
Pr ₃ HN	1	1	[131]
Pr ₃ HN	<i>x</i>	1	[131]
Pr ₂ H ₂ N	2	1	[131]
Pr ₂ H ₂ N	1	1	[131]
Pr ₂ H ₂ N	<i>x</i>	1	[131]
Bu ₄ N	2	1	[40,128,151,265]
Bu ₄ N	1	1	[36,40,128]
Bu ₄ N	1	2	[258]
Bu ₃ HN	2	1	[131]
Bu ₃ HN	1	1	[131]
Bu ₃ HN	<i>x</i>	1	[131]
Bu ₂ H ₂ N	2	1	[131]
Bu ₂ H ₂ N	1	1	[131]
Bu ₂ H ₂ N	<i>x</i>	1	[131]
Me ₂ Et ₂ N	2	1	[15,256]
Me ₂ Et ₂ N	1	2	[15, 170, 257]
Me ₂ (C ₁₀ H ₂₁) ₂ N	2	1	[266]
Me ₂ (C ₁₂ H ₂₅) ₂ N	2	1	[176,179,180,266]
Me ₂ (C ₁₄ H ₂₉) ₂ N	2	1	[266]
Me ₂ (C ₁₈ H ₃₇) ₂ N	1	1	[266]
Me(C ₁₀ H ₂₁) ₃ N	1	1	[266]
Me(C ₁₂ H ₂₅) ₃ N	1	1	[266]
Me(C ₁₄ H ₂₉) ₃ N	1	1	[266]
Me(C ₁₆ H ₃₃) ₃ N	1	1	[266]
Me(C ₁₈ H ₃₇) ₃ N	1	1	[266]
<i>N</i> -Dodecylpyridinium ^a	2	1	[179]
<i>N</i> -Octadecylpyridinium ^a	2	1	[267,268]
(CH ₃) ₂ NC(SCH ₃) ₂ ^a	1	3	[191]
MDA ^a	2	1	[191,269]
MDA ^a	1	1	[191,269]

Table 3 (Continued)

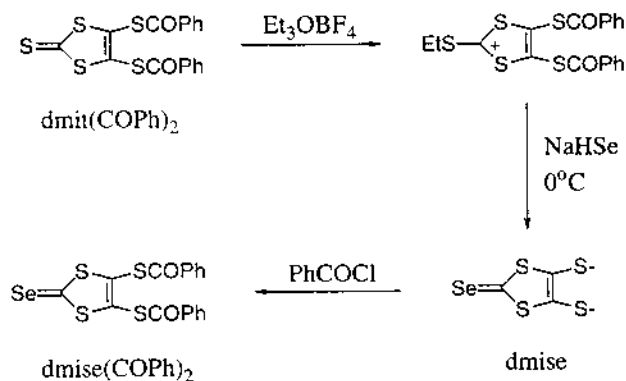
A	<i>n</i>	<i>m</i>	Ref.
MDA ^a	1	2	[269]
EDA ^a	1	1	[191]
Dmp ^a	1	2	[119]
1,3-Didodecylimidazolium ^a	2	1	[201]
MV ^a	1	1	[157,202]
OV ^a	1	1	[157,202]
StV ^a	1	1	[157,202]
MQ ^a	1	1	[157,202]
DQ ^a	1	1	[157,202]
PQ ^a	1	1	[157,202]
BQ ^a	1	1	[157,202]
DP ^a	1	1	[157,202]
DPK ^a	1	1	[157,202]
Me ₄ P	1	2	[213,260–263,270]
Me ₂ Et ₂ P	1	2	[213,263]
MePh ₃ P	1	3	[271]
Ph ₄ P	1	3	[271]
PhCH ₂ Ph ₃ P	2	1	[219]
PhCH ₂ Ph ₃ P	1	1	[219]
PPN ^a	2	1	[219]
PPN ^a	1	1	[219,221]
PPN ^a	0.25	1	[221]
Me ₃ S	2	1	[219]
Me ₃ S	1	1	[219]
Me ₂ EtS	2	1	[219]
Me ₂ EtS	1	1	[219]
MeEt ₂ S	2	1	[219]
MeEt ₂ S	1	1	[219]
MeEt ₂ S	1	2	[219]
MeEt ₂ S	<i>x</i>	1	[220]
Et ₃ S	2	1	[219]
Et ₃ S	1	1	[219]
Me ₄ As	1	2	[141,213,260,261,263,272]
Me ₂ Et ₂ As	1	2	[263]
Me ₄ Sb	2	1	[262]
Me ₄ Sb	1	2	[213,260–263]
Me ₂ Et ₂ Sb	1	2	[213,263]
MeEt ₂ Sb	2	1	[213]
Co(Cp) ₂	2	1	[127,128]
Co(Cp) ₂	1	1	[127,128]
Fe(C ₅ Me ₅) ₂	1	1	[224]
Fe(Cp)(C ₆ H ₆)	2	1	[41,225]
Fe(Cp)(PhMe)	2	1	[224]
Fe(Cp)(PhMe)	1	1	[224]
Ni(ddd _t) ₂ ^a	1	<i>x</i>	[219]
Pt(ddd _t) ₂ ^a	1	<i>x</i>	[219]
Cs	1	2	[141,142,258,272–275]
TTF ^b	1	2	[30,142,233,258]

Table 3 (Continued)

A	<i>n</i>	<i>m</i>	Ref.
EDT-TTF ^b	1	1	[242]
	1	2	[170]
	2	2	[133,276–279]
	2	3	[279]
I EDT-TTF	1	1	[171,280]
			
DMC-TTF ^b	1	<i>x</i>	[247]
D1 ^b	<i>x</i>	1	[131]
D2 ^b	<i>x</i>	1	[131,220,250]
D3 ^b	<i>x</i>	1	[131]
D4 ^b	<i>x</i>	1	[131,220]
D5 ^b	<i>x</i>	1	[131]

^a See Table 1.^b See Table 2.

ligand, dmise [112,113]. This method for the synthesis of dmise(COPh)₂ includes the alkylation of the thiocarbonyl group of the dmit starting compound dmit(COPh)₂, followed by the reaction of the formed dithiolium salt with sodium hydrogen selenide:



In 1986 Nigrey described the synthesis of the two selenium isologs of dmit, 1,3-dithiole-2-thione-4,5-diselenolate (dsit), starting from the vinylene trithiocarbonate molecule (1,3-dithiole-2-thione) where the vinylene moiety was lithiated followed by reaction with elemental selenium [114].

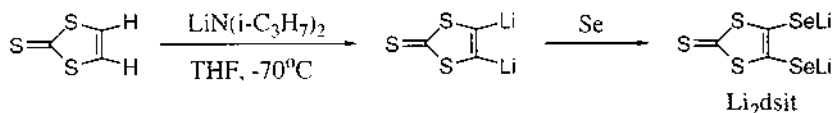
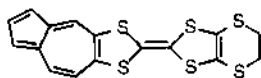


Table 4

Bis-chelate complexes of the general formula (A)_n[Pt(dmit)₂]_m

A	<i>n</i>	<i>m</i>	Ref.
–		1	[129,132]
H ₄ N	2	1	[133]
H ₄ N	1	1	[133,134]
H ₄ N	1	<i>x</i>	[134]
Me ₄ N	2	1	[136,138]
Me ₄ N	1	2	[136,138,141,258,281]
Me ₃ HN	2	1	[133,134,282]
Me ₃ HN	1	1	[133,134,282]
Me ₃ HN	1	<i>x</i>	[134]
Me ₃ HN	1	3	[133,134,282]
Me ₂ H ₂ N	2	1	[133,134]
Me ₂ H ₂ N	1	1	[133,134]
Me ₂ H ₂ N	1	<i>x</i>	[134]
MeH ₃ N	2	1	[133,134]
MeH ₃ N	1	1	[133,134]
MeH ₃ N	1	<i>x</i>	[134]
Bu ₄ N	2	1	[150,151,283]
Me ₂ (C ₁₂ H ₂₅) ₂ N	2	1	[176,179,180,267,284]
<i>N</i> -Dodecylpyridinium ^a	2	1	[179]
<i>N</i> -Octadecylpyridinium ^a	2	1	[267]
MV ^a	1	1	[157,202]
OV ^a	1	1	[157,202,203]
StV ^a	1	1	[157,202]
MQ ^a	1	1	[157,202,205]
DQ ^a	1	1	[157,202,203]
PQ ^a	1	1	[157,202,205]
BQ ^a	1	1	[157,202,205]
DP ^a	1	1	[157,202]
DPK ^a	1	1	[157]
Fe(Cp)(C ₆ H ₆)	2	1	[41,225]
Fe(C ₅ Me ₅) ₂	1	1	[224]
PPN ^a	1	1	[221]
DMC-TTF ^b	1	<i>x</i>	[247]
AET	2	1	[285]

^a See Table 1.^b See Table 2.

The air and moisture sensitivity of this 1,2-diselenolate prevents its use as a starting reagent for functionalization. However, as has been shown by Steimecke [1] for dmit, complexation of the dichalcogenolates with zinc(II) followed by the addition of tetrabutylammonium bromide results in the formation of a stable

Table 5

Bis-chelate complexes of the general formula (A)_n[M(dmit)₂]

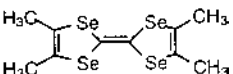
M	A	<i>n</i>	Ref.
MoO	Bu ₄ N	2	[286]
OsN	Bu ₄ N	2	[287]
ReO	Ph ₄ P	1	[288]
TcN	Bu ₄ N	2	[289]
Au	Me ₄ N	1	[290]
	Me ₄ N	0.5	[290,291]
	Me ₃ PrN	<i>x</i>	[291]
	Me(C ₁₀ H ₂₁) ₃ N	1	[266,292–300]
	Et ₄ N	1	[301]
	Et ₄ N	0.5	[291]
	Et ₄ N/(TCNQ)	1	[291,301]
			
	Bu ₄ N	1	[302,303]
	Bu ₄ N	0.5	[291]
	Bu ₄ N	<i>x</i>	[291]
	EtBu ₃ N	<i>x</i>	[291]
	Et ₂ Bu ₂ N	<i>x</i>	[291]
	Et ₃ BuN	<i>x</i>	[291]
	(C ₁₀ H ₂₁) ₃ HN	1	[304]
	PPN ^a	1	[305]
	<i>N</i> -Octadecylpyridinium ^a	1	[267]
	Fe(C ₅ Me ₅) ₂	1	[303]
	Fe(C ₆ Me ₅) ₂	0.33	[303]
	Fe(Cp) ₂	0.33	[303]
	Me ₃ S	<i>x</i>	[291]
	Bu ₃ S	<i>x</i>	[291]
	Me ₃ Te	1	[306]
	Li	0.5	[302]
	Na	0.5	[302]
	K	0.5	[302]
	TTF ^b	2	[307]
	TTF ^b	1.5	[308]
	BEDT-TTF ^b	1	[308]
	TMTSF	1.5	[308]
			
Cd	Et ₄ N	2	[309–311]
	Me ₃ (PhCH ₂)N	2	[310,311]
	Me ₃ (C ₁₆ H ₃₃)N	2	[310]
	Me ₃ (C ₁₈ H ₃₇)N	2	[311]
	<i>N</i> -Hexadecylpyridinium ^a	1	[310,311]
Co	Et ₄ N	1.2	[309,311]
	Bu ₄ N	1	[24]
	Fe(Cp)(C ₆ H ₆)	1	[41,225]
	Fe(Cp)(PhMe)	2	[224]
	Fe(Cp)(PhMe)	1	[224]

Table 5 (Continued)

M	A	<i>n</i>	Ref.
Cu	Et ₄ N	2	[148]
	Et ₃ (C ₁₆ H ₃₃)N	2	[173]
	Bu ₄ N	2	[151,265,312]
	Bu ₄ N	1	[24,151]
	MQ ^a	1	[202,205]
	PQ ^a	1	[202,205]
	BQ ^a	1	[202,205]
	DP ^a	1	[202]
	DPK ^a	1	[202]
	Fe(Cp)(C ₆ H ₆)	2	[41,225]
	Fe(Cp)(PhMe)	2	[224]
	Fe(Cp)(PhMe)	1	[224]
	THT-TTF ^b	1	[148]
	TBT-TTF ^b	1	[148,313]
Fe	Me ₄ N	1	[314]
	Me ₄ N	0.53	[314]
	Me ₄ N	0.4	[314]
	Et ₄ N	1	[309,311]
	Bu ₄ N	1	[314]
	Bu ₄ N	0.66	[314]
	Bu ₄ N	0.33	[314]
	Bu ₄ N	0.05	[314]
	Fe(C ₅ Me ₅) ₂	1	[314]
	TTF ^b	1	[314]
Hg	Et ₄ N	2	[309,311]
	Na	2	[315]
Mn	Et ₄ N	1.2	[309,311]
Sb	Et ₄ N	1	[316]
	1,4-Dimethylpyridinium	1	[316]
Se	PPN ^a	2	[317]
	Ph ₄ As	2	[317]
	Me ₃ Te	2	[317]
Sn	Et ₄ N	1.5	[309,311]
Te	PPN ^a	2	[317]
	Me ₃ Te	2	[317]
	Co(Cp) ₂	2	[317]
Zn	Et ₄ N	2	[147,148,156,318–322]
	Pr ₄ N	2	[323]
	Bu ₄ N	2	[38,147,153,155,288,324–326]
	Me ₃ (FcCH ₂)N (Fc = ferrocene)	2	[322]
	1,4-Dimethylpyridinium	2	[316,320]
	MV ^a	1	[147]
	MQ ^a	1	[205]
	DQ ^a	1	[147]
	PQ ^a	1	[147,205]
	BQ ^a	1	[147,205]

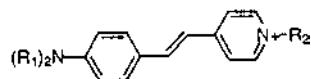
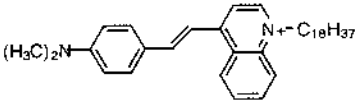
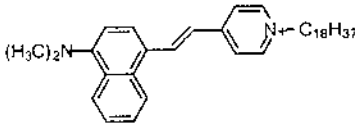
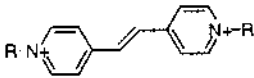
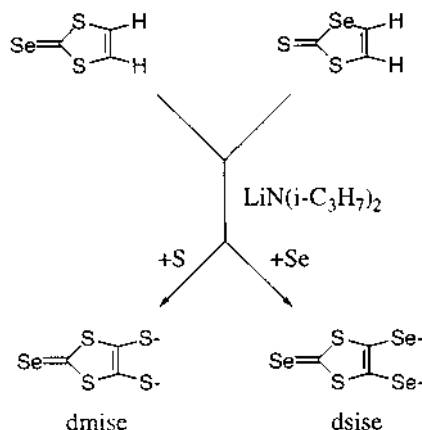


Table 5 (Continued)

M	A	n	Ref.
	$R_1 = \text{CH}_3, R_2 = \text{C}_{18}\text{H}_{37}$	2	[37,38,327]
	$R_1 = \text{C}_2\text{H}_5, R_2 = \text{C}_{16}\text{H}_{33}$	2	[38]
	$R_1 = \text{C}_2\text{H}_5, R_2 = \text{C}_{18}\text{H}_{37}$	2	[38]
	$R_1 = \text{C}_4\text{H}_9, R_2 = \text{C}_{18}\text{H}_{37}$	2	[328]
		2	[37]
		2	[38]
			
	$R = \text{CH}_3$	1	[204]
	$R = \text{C}_4\text{H}_9$	1	[204]
	$R = \text{C}_5\text{H}_{11}$	1	[204]
	$R = (\text{CH}_2)_3\text{CN}$	1	[204]
	$R = (\text{CH}_2)_4\text{CN}$	1	[204]
	$R = p\text{-CH}_2\text{PhCN}$	1	[204]
	Na	2	[315]
	BEDT-TTF ^b	1	[148]
	THT-TTF ^b	1	[148]
	TBT-TTF ^b	1	[148,329]

^a See Table 1.^b See Table 2.

zinc bis-chelate which can be used as an ‘off-the-shelf-reagent’ in the synthesis of derivatives, e.g. the dibenzoyl compound $\text{dsit}(\text{COPh})_2$ [115]. Applying Nigrey’s method on the various known vinylene trichalcogenocarbonates (see e.g. [116,117]) allows one to prepare all dmit isologs. Starting from the heterocycles 1,3-dithiole-2-selone and 1,3-thiaselenole-2-thione, the carbon–carbon double bond is lithiated followed by the insertion of sulfur or selenium into the formed carbon lithium bond leads to the formation of dmise [118,119], or to the three selenium atom containing compound 1,3-dithiole-2-selone-4,5-diselenolate (dsise) [118,120], respectively.



Starting compounds for the synthesis of 1,3-thiaselenole-2-selone-4,5-diselenolate (dsitse) [121], the four selenium atom isologs of dmit, are 1,3-thiaselenole-2-selone or 1,3-diselenole-2-thione [116,117]. The addition of elemental sulfur in the last step of these reactions for preparing various diselenium dmit-isologs with exocyclic thiolate functions results in scrambling and rearrangement processes which leads to a mixture of products. The 1,3-diselenole-2-selone-4,5-diselenolate (dsis), the selenium analog of dmit is also available according to Nigrey's method [122], starting from the 1,3-diselenole-2-selone [116,117]. However, a much faster method for the

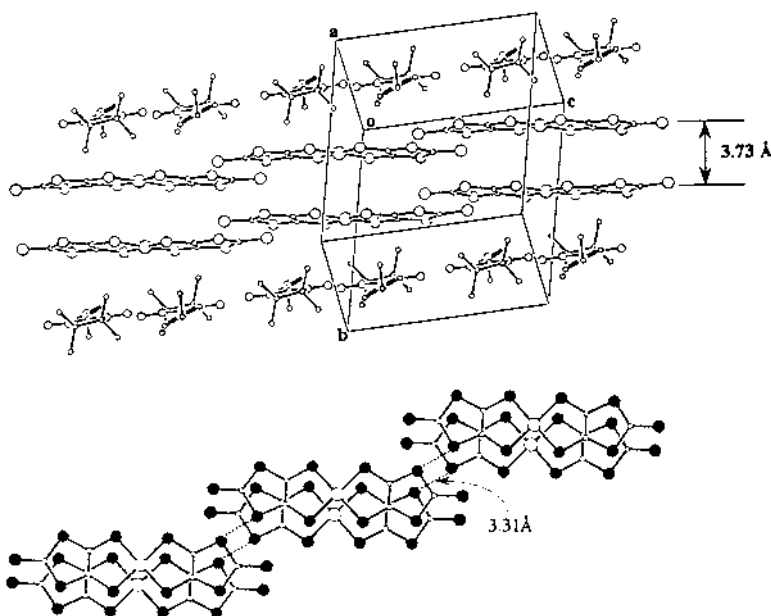


Fig. 2. Figures showing one-dimensional chains of $p\text{-EPYNN}$ and one-dimensional ladder chains of $\text{Ni}(\text{dmit})_2$ units [34]. Reprinted by permission from H. Imai, T. Inabe, T. Otsuka, T. Okuno, K. Awaga, Phys. Rev. B 54 (1996) R6838. Copyright 1996 by the Am. Phys. Soc.

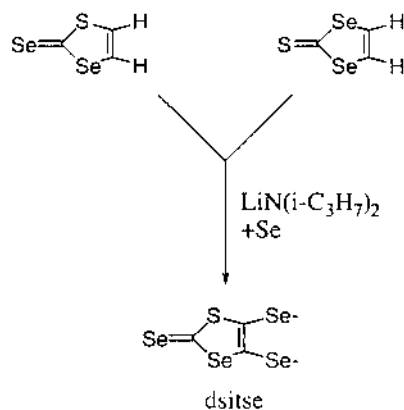
Table 6

Tris-chelate complexes of the general formula $(A)_n[M(dmit)_3]$

M	A	<i>n</i>	Ref.
MoO	Bu ₄ N	2	[326]
Mo	Bu ₄ N	2	[286,326,335]
	Bu ₄ N	0.8	[326]
	Bu ₄ N	0.4	[286,335]
	Bu ₄ N	0.3	[286,335]
	Bu ₄ N	0.05	[286,335]
	Fe(C ₅ Me ₅) ₂	1	[286,326,335]
Nb	Bu ₄ N	1	[326,336]
	Bu ₄ N	0.05	[326]
	TTF ^a	1	[326]
Re	—	—	[288]
	Bu ₄ N	1	[288,326]
Sb	Et ₄ N	1	[320]
	1,4-Dimethylpyridinium	1	[320]
V	Bu ₄ N	2	[326]
	<i>N</i> -Methylphenazinium (NMP)	2	[326]
	Fe(Cp) ₂	1	[326]
	Fe(C ₅ H ₄ Me) ₂	1	[326]
	Fe(C ₅ Me ₅) ₂	1	[326]
	Ni(Cp) ₂	1	[326]
	TTF ^a	2	[326]
	BEDT-TTF ^a	1.5	[334]
W	Bu ₄ N	2	[326,335]
	Bu ₄ N	0.8	[335]
	Bu ₄ N	0.4	[335]
	Bu ₄ N	0.3	[326,335]
	Bu ₄ N	0.2	[335]
	Bu ₄ N	0.15	[335]
	Ph ₄ P	2	[333]
	Fe(C ₅ Me ₅) ₂	1	[326,335]

^a See Table 2.

synthesis of dsis(COPh)₂ is the electrochemical [123,124] or chemical reduction [125] of carbon diselenide.



3. Chelate complexes of 1,3-dithiole-2-thione-4,5-dithiolate and isologs

3.1. 1,3-Dithiole-2-thione-4,5-dithiolate

Amongst the dithiolate ligands of interest in this review, dmit continues to be the most extensively studied in the assembly of bis- and tris-chelate complexes. The most commonly combined ‘closed-shell’ cations with $M(\text{dmit})_2$ anions are of the alkylammonium family, but other classes such as sulphonium, phosphonium and metallocenium cations have been investigated. A number of ‘open-shell’ TTF and bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF)-based donors have also been combined with $M(\text{dmit})_2$ anions. The major driving force for the study of metal bis-dmit salts, in particular where the metal center is Ni, Pd or Pt, continues to be the search for highly electrically conducting and potentially superconducting non-integer oxidation state (NIOS) complexes. Tables 1 and 2 summarize $M(\text{dmit})_2$ bis-chelate salts of Ni, while Tables 3 and 4 summarize the Pd and Pt-based complexes, respectively. Table 5 lists bis-dmit salts for all other metals.

Since the previous review [5], five new $M(\text{dmit})_2$ (where $M = \text{Ni}$ or Pd) NIOS systems have been synthesized, via electrocrystallization, which behave as superconductors at low temperatures and/or under high pressures. The donor-acceptor complex α -(EDT-TTF)[Ni(dmit)₂] is the first ambient pressure superconductor based on the $M(\text{dmit})_2$ anions with a $T_c = 1.3$ K [16,35,171,239–245]. Another donor-acceptor complex α' -(TTF)[Pd(dmit)₂]₂, undergoes a superconducting transition at 5.93 K under 24 kbar pressure [330–332]. The anion-radical salt complexes with a closed-shell cation β -(Me₄N)[Pd(dmit)₂]₂ ($T_c = 6.2$ K at 6.5 kbar)

Table 7
Bis-chelate complexes of the general formula (A)_n[M(dmt)₂]

M	A	<i>n</i>	Ref.
Ni	Bu ₄ N	2	[151]
	MV ^a	1	[157]
	OV ^a	1	[157,202,203]
	StV ^a	1	[157]
	MQ ^a	1	[157]
	DQ ^a	1	[157,202,203]
	PQ ^a	1	[157]
	BQ ^a	1	[157]
	DP ^a	1	[157]
	DPK ^a	1	[157]
Se	PPN ^a	2	[317]
	Me ₃ Te	2	[317]
Te	PPN ^a	2	[317]
	Me ₃ Te	2	[317]
	Co(Cp) ₂	2	[317]

^a See Table 1.

Table 8

Bis-chelate complexes of the general formula (A)_n[M(dmid)₂]

M	A	<i>n</i>	Ref.
Cu	Me ₄ N	1	[338,342,343]
	Et ₄ N	2	[339]
	Et ₄ N	1	[343]
	Et ₄ N	0.65	[343]
	Bu ₄ N	1.2	[343]
	Bu ₄ N	1.1	[338,342,343]
	Bu ₄ N	1	[343]
	Bu ₄ N	0.9	[343]
	Bu ₄ N	0.8	[343]
	Bu ₄ N	0.7	[343]
	Et ₃ (C ₁₆ H ₃₃)N	2	[344]
Ni	–		[339]
	Me ₄ N	2	[339]
	Me ₄ N	1	[338,342,343]
	Me ₄ N	0.65	[343]
	Et ₄ N	2	[339]
	Et ₄ N	2 (I _{2,3})	[339]
	Et ₄ N	2 (I _{7,8})	[339]
	Et ₄ N	1.1	[338,342,343]
	Et ₄ N	0.17	[339]
	Et ₃ (C ₁₆ H ₃₃)N	2	[344]
	Bu ₄ N	1.2	[338,342,343]
	Bu ₄ N	1.1	[338,342,343]
	Bu ₄ N	1	[338,340–343,345–347]
	Bu ₄ N	0.9	[340]
	Bu ₄ N	0.6	[338,346,347]
	Bu ₄ N	0.44	[346,347]
	Bu ₄ N	0.35 (TTF ^a)	[338,347]
	Bu ₄ N	0.03	[338]
	Bu ₄ N	<i>x</i>	[340,341]
	MV ^b	1	[157,202]
	OV ^b	1	[157,202]
	StV ^b	1	[157,202]
	MQ ^b	1	[157]
	DQ ^b	1	[157,202]
	PQ ^b	1	[157]
	BQ ^b	1	[157]
	DP ^b	1	[157]
	DPK ^b	1	[157]
	Fe(C ₅ Me ₅) ₂	1	[33]
Zn	Et ₄ N	2	[322]
	Et ₃ (C ₁₆ H ₃₃)N	2	[344]
	Bu ₄ N	2	[338,342,343,345]
	Me ₃ (FcCH ₂)N ^c	2	[322]

^a See Table 2.^b See Table 1.^c See Table 5.

[14,136,138,143,213,256,257,259–263] and $(\text{Me}_2\text{Et}_2\text{N})[\text{Pd}(\text{dmit})_2]_2$ ($T_c = 4\text{K}$ at 2.4 kbar) [15,170,257] all display superconducting behavior under high pressures. The most recent superconductor synthesized is the complex $\beta'-(\text{Me}_2\text{Et}_2\text{P})[\text{Pd}(\text{dmit})_2]_2$ with a $T_c = 4.0\text{--}1.8\text{K}$ at 6.9–10.4 kbar [17].

Nakamura and coworkers have reported the electrocrystallization and characterization of a novel molecular metal which displays both electronic and ionic conduction [42]. The complex $\text{Li}_{0.6}(\text{15-crown-5-ether})[\text{Ni}(\text{dmit})_2]_2 \cdot \text{H}_2\text{O}$ which is shown in Fig. 1 is composed of stacks of $\text{Ni}(\text{dmit})_2$ units which provide pathways for electronic conduction and are separated by parallel stacks of 15-crown-5-ether moieties in a channel-like formation which facilitates ion conduction. The salt has a room temperature conductivity of 240 S cm^{-1} . Temperature dependent magnetic susceptibility and NMR measurements were used to prove the existence of Li^+ movement within the crown ether channels [42].

Besides electrical conductivity, a number of other applications for $\text{M}(\text{dmit})_2$ complexes have been investigated. For instance in molecule-based magnets, the first spin ladder was synthesized of composition $(p\text{-EPYNN})[\text{Ni}(\text{dmit})_2]$ (where $p\text{-EPYNN} = p\text{-N-ethylpyridinium } \alpha\text{-nitronyl nitroxide}$, see Table 1) [34,211]. Within the crystal lattice, the radical cation $p\text{-EPYNN}$ units are arranged in one-dimensional chains with ferromagnetic interactions. The chains of $\text{Ni}(\text{dmit})_2$ moieties in a ladder formation exhibits coexistent antiferromagnetic interactions. Views of the crystal structure are shown in Fig. 2.

The Langmuir–Blodgett (LB) technique has been utilized to assemble well organized films of $\text{M}(\text{dmit})_2$ salts with cations containing long hydrophobic alkyl chains [158,173,177,178,181,183–188,267,284,292–300,313,329]. In one instance, LB films of $[\text{Me}(\text{C}_{10}\text{H}_{21})_3\text{N}][\text{Au}(\text{dmit})_2]$ have exhibited room temperature electrical conductivities in the range of $30\text{--}50\text{ S cm}^{-1}$ [292–294]. Films of $\text{M}(\text{dmit})_2$ salts have also been applied in gas sensor [23–25,284] and field-effect transistor devices [26,27].

$\text{Zn}(\text{dmit})_2$ anions have been combined with a series of dyes in films which exhibit non-linear optical (NLO) properties. The LB technique has been applied to align the chromophores in a non-centrosymmetric manner to potentially maximize the second harmonic generation (SHG) properties [37,38,328].

Metal tris-chelate complexes of dmit are primarily composed of Group 5–7 metals along with antimony as shown in Table 6. They tend to show distorted octahedral or trigonal prismatic geometries [286,288,320,326,333–336]. Mo, V

Table 9
Tris-chelate complexes of the general formula $(\text{A})_n[\text{M}(\text{dmit})_3]$

M	A	<i>n</i>	Ref.
Mo	–		[333]
	Ph_4P	2	[333]
W	–		[333]
	Ph_4P	2	[333]

Table 10

Bis-chelate complexes of the general formula (A)_n[M(dmise)₂]

M	A	<i>n</i>	Ref.
Au	Bu ₄ N	1	[118,348,349]
Cd	Bu ₄ N	2	[118,348,349]
Cu	Bu ₄ N	2	[118,348]
Hg	Bu ₄ N	2	[118,348,349]
Ni	Me ₄ N	2	[136,138]
	Me ₄ N	1	[113,136,138]
	Me ₄ N	0.5	[113,136,138,350,351]
	Me ₃ HN	0.5	[170,351–353]
	Me ₂ H ₂ N	0.5	[353]
	MeH ₃ N	0.5	[353]
	Bu ₄ N	2	[113,118,119,348,349,353]
	Bu ₄ N	1	[113,118,119,343,349,351,353]
	<i>N,N</i> -Dimethylpiperidinium ^a	0.5	[354]
	EDT-TTF ^b	1	[170,172,351,352]
Pd	Me ₄ N	2	[136,138]
	Me ₄ N	1	[136,138]
	Me ₄ N	0.5	[136,138,261,262,351,355]
	Bu ₄ N	2	[113,118,348,349,353]
	Bu ₄ N	1	[113]
	Me ₄ As	2	[262]
	Me ₄ As	0.5	[261,262,355,356]
	Me ₄ P	2	[261]
	Me ₄ P	0.5	[261,262,355,356]
	Me ₂ Et ₂ P	2	[213]
	Me ₂ Et ₂ P	1	[213]
	Me ₄ Sb	2	[262]
	Me ₄ Sb	0.5	[261,262,355]
	Cs	0.5	[170,172,353]
Pt	Me ₄ N	2	[136,138]
	Me ₄ N	0.5	[136,138]
Zn	Bu ₄ N	2	[118,119,121,348,349]

^a See Table 1.^b See Table 2.

and W tris-chelate complexes which have low oxidation potentials, have been oxidized by I₂, [Fe(Cp)₂]PF₆ and (TTF)₃(BF₄)₂ to yield a number of NIOS complexes with conductivities as high as 10^{−1} S cm^{−1} [286,326,335]. The NIOS complex (BEDT-TTF)₃[V(dmit)₃]₂ has been characterized by X-ray crystallography and shows semiconducting behavior [334]. A review on bulky metal complexes with dmit and isologs has been written by Matsubayashi and can be referred to for a more in depth discussion of these types of compounds [337].

3.2. 1,2-Dithiole-3-thione-4,5-dithiolate

A much smaller amount of work has been reported with M(dmt)₂ complexes. In comparison with M(dmit)₂ salts, they are considerably less soluble and tend

Table 11

Bis-chelate complexes of the general formula (A)_n[M(dsit)₂]

M	A	n	Ref.
Ni	Me ₄ N	2	[136,138]
	Me ₄ N	1	[136,138]
	Me ₄ N	0.5	[136,138]
	Bu ₄ N	2	[358,359]
	Bu ₄ N	1	[358]
Pd	Me ₄ N	2	[204,222,224]
	Me ₄ N	1.5	[204]
	Me ₄ N	1	[204,222]
	Me ₄ N	0.5	[136,138,270,357]
Pt	Me ₄ N	2	[136,138]
	Me ₄ N	1.5	[138]
	Me ₄ N	0.5	[136,138]
Zn	Bu ₄ N	2	[121,348]

to be difficult to crystallize [5]. Since 1991, no new tris-dmt salts have been reported and only a few bis-chelate complexes. These are listed in Table 7. Some of the complexes have been assembled combining Ni(dmt)₂ with viologen-based cations for applications in electrical conductivity and dioxygen activation [157,202,203].

3.3. 1,3-Dithiole-2-one-4,5-dithiolate

As shown in Table 8, many new NIOS complexes based on M(dmtd)₂ have been synthesized via chemical and/or electrochemical oxidation but no X-ray structures

Table 12

Bis-chelate complexes of the general formula (A)_n[M(dsise)₂]

M	A	n	Ref.
Au	Bu ₄ N	1	[118,348,349]
Cd	Bu ₄ N	2	[118,348,349]
Cu	Bu ₄ N	2	[118,349]
Hg	Bu ₄ N	2	[118,348,349]
Ni	Me ₄ N	2	[136,138]
	Me ₄ N	1	[136,138]
	Me ₄ N	0.5	[136,138]
	Bu ₄ N	2	[118,349]
	Bu ₄ N	1	[118,349]
Pd	Me ₄ N	2	[136,138]
	Me ₄ N	0.5	[136,138]
	Bu ₄ N	2	[118,348,349]
Pt	Me ₄ N	2	[136,138]
	Me ₄ N	1.5	[138]
	Me ₄ N	0.5	[136,138]
Zn	Bu ₄ N	2	[118,120,121,348,349]

Table 13

Bis-chelate complexes of the general formula (A)_n[M(dsis)₂]

M	A	<i>n</i>	Ref.
Au	Bu ₄ N	1	[348,360]
Cd	Bu ₄ N	2	[360]
Cu	Bu ₄ N	2	[312]
Fe	Me ₄ N	1	[314]
	Bu ₄ N	1	[314]
	Bu ₄ I	0.05	[314]
	Fe(C ₅ Me ₅) ₂	0.7	[314]
	TTF ^a	0.28	[314]
Ni	Me ₄ N	2	[136,138]
	Me ₄ N	0.5	[136,138]
	Bu ₄ N	2	[360]
Pd	Me ₄ N	2	[136,138]
	Me ₄ N	1	[138]
	Me ₄ N	0.5	[136,138]
	Bu ₄ N	2	[348,360]
Pt	Me ₄ N	2	[136,138]
	Me ₄ N	1.5	[138]
	Me ₄ N	0.5	[136,138]
	Me ₄ N	0.66	[138]
	Bu ₄ N	2	[348,360]
Zn	Bu ₄ N	2	[121,348]
	Ph ₄ P	2	[360]

^a See Table 2.

have been reported. This may be due to the fact that the terminal one group does not facilitate crystallization [5,338]. A conductivity as high as 10^{-2} S cm⁻¹ was reached with (Et₄N)_{0.17}[Ni(dmid)₂] [339]. NIOS M(dmid)₂ complexes have also been studied by X-ray photoelectron spectroscopy (XPS) and for applications in electrical switching and memory devices [340,341].

Table 9 lists the tris-chelate complexes of dmid. Group 6 metals Mo and W have been utilized in the formation of tris-chelate complexes including the neutral [W(dmid)₃] compound [333].

Table 14

Tris-chelate complexes of the general formula (A)_n[M(dsis)₃]

M	A	<i>n</i>	Ref.
W	Bu ₄ N	2	[361]
	Bu ₄ N	0.6	[361]
	Bu ₄ N	0.1	[361]
	Fe(C ₅ Me ₅) ₂	0.5	[361]
	TTF ^a	0.5	[361]

^a See Table 2.

Table 15

Mixed-ligand chelate complexes of the general formula (A)_n[M(L₁)(L₂)]

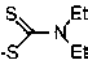
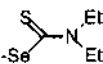
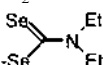
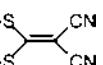
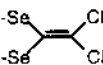
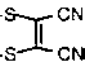
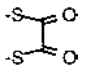
M	A	<i>n</i>	L ₁	L ₂	Ref.
Au	–		dmit	C ₆ F ₅	[362] ^a
	–		dmit	Cl	[362] ^a
	–		dmit	Br	[362] ^a
	Bu ₄ N	1	dmit	Ph ₃ P	[307]
	Bu ₄ N	1	dmit	MePh ₂ P	[307]
Co	Bu ₄ N	1	dmit	Me ₃ P	[307]
	–		dmit	Cp	[159,318]
	–		dmit	C ₅ Me ₅	[318]
	–		dmid	Cp	[318]
Cu	Et ₄ N	1	dmit	C ₅ Me ₅	[318]
	Bu ₄ N	1	dmit	Ph ₃ P	[363]
			dsit	Et ₂ dtc	[364]
					
	Bu ₄ N	1	dsit	Et ₂ tsc	[364]
					
	Bu ₄ N	1	dsit	Et ₂ dsc	[364]
					
	Bu ₄ N	2	dsit	<i>i</i> -mnt	[364]
					
	Bu ₄ N	2	dsit	<i>i</i> -mns	[364]
					
	Bu ₄ N	1	dsis	Et ₂ dtc	[312]
	Bu ₄ N	1	dsis	Et ₂ tsc	[312]
	Bu ₄ N	1	dsis	Et ₂ dsc	[312]
	Bu ₄ N	2	dsis	<i>i</i> -mnt	[312]
	Bu ₄ N	2	dsis	<i>i</i> -mns	[312]
	Bu ₄ N	2	dsis	dmit	[312]
	Bu ₄ N	2	dsis	mnt	[312]
					
	Bu ₄ N	2	dsis	dto	[312]
					
Hg	–		dmit	dppe ^b	[365] ^a
	–		dmit	dppf ^c	[365] ^a

Table 16

Mixed dmit and dmise ligand chelate complexes with the general formula $(A)_n[M(C_3S_xSe_y)_2]$

M	A	n	x	y	Ref.
Cu	Bu ₄ N	2	4.4	0.6	[369]
Ni	Bu ₄ N	2	4.7	0.3	[369]
	Bu ₄ N	1	4.7	0.3	[369]
	Bu ₄ N	2	4.67	0.33	[370]
	—		4.7	0.3	[369]
	Bu ₄ N	2	4.4	0.6	[369]
Cu/Ni	Bu ₄ N	1	4.4	0.6	[369]
	Bu ₄ N	2	4.4	0.6	[369]

conductivity of 10 S cm^{−1}. The goal of substituting for a larger Se atom is to increase dimensionality observed in the crystal lattice in order to enhance the electrical properties [352,353]. Kato et al. electrocrystallized a series of 1:2 Pd(dmise)₂ salts with tetramethylammonium, phosphonium, arsonium and antimonium cations for comparative studies of their crystal structures, electrical conductivities and electronic structures [262].

3.5. 1,3-Dithiole-2-thione-4,5-diselenolate

The M(dsit)₂ salts shown in Table 11 are primarily assembled from Group 10 metals. The Zn(II) complex (Bu₄N)₂[Zn(dsit)₂] was also made and studied using ⁷⁷Se-NMR and compared to other Zn(II) based selenium containing dmit isologs [348]. The first synthesis and crystal structure of a NIOS complex with dsit, (Me₄N)[Pd(dsit)₂]₂, was reported by Faulmann et al. in 1993 [357]. Tris-dsit compounds have not been reported.

3.6. 1,3-Dithiole-2-selone-4,5-diselenolate

All Group 10–12 metals except Ag have been used in the assembly of bis-dsise complexes as shown in Table 12. The first NIOS complexes were reported in 1993 and (Me₄N)[Ni(dsise)₂]₂, a semiconductor, has the highest room temperature conductivity of 12 S cm^{−1} [136,138].

3.7. 1,3-Thiaselenole-2-selone-4,5-diselenolate

The diselenolato ligand dsitse has four selenium atoms and one sulfur atom at a thiole position. First reported in 1992 by Poleschner and associates, only two bis-chelates have been described: (Bu₄N)₂[Zn(dsitse)₂] and (Ph₄P)₂[Zn(dsitse)₂] [121]. They were characterized by IR, ¹³C- and ⁷⁷Se-NMR and X-ray crystallography.

Table 17

Cluster complexes of the general formula (A)_n[M_x(L₁)_y(L₂)_z]

M	x	A	n	L ₁	y	L ₂	z	Ref.
Au	2	–		μ-dmit	1	Me ₃ P	2	[307]
	2	–		μ-dmit	1	Me ₂ PhP	2	[307]
	2	–		μ-dmit	1	MePh ₂ P	2	[307]
	2	–		μ-dmit	1	Ph ₃ P	2	[307,319]
	2	–		μ-dmit	1	CH ₂ Ph ₃ P	2	[307]
	2	–		μ-dmit	1	CH ₂ Ph ₂ MeP	2	[307]
	2	–		μ-dmit	1	CH ₂ PhMe ₂ P	2	[307]
	2	–		μ-dmit	1	μ-dppe ^a	1	[375]
	2	–		dmit	1	Ph ₃ As	1	[319]
	4	–		μ-dmit	2	μ-(Ph ₂ PCH ₂ PPh ₂)	2	[319]
	2	Bu ₄ N	2	μ-dmit	1	C ₆ F ₅	2	[307]
	2	Bu ₄ N	1	μ-dmit	1	μ-CH ₂ PPh ₂ CH ₂	1	[375]
	2	Bu ₄ N	1	μ-dmit	1	μ-S ₂ CNEt ₂	1	[375]
	2	Bu ₄ N	1	μ-dmit	1	μ-S ₂ CN(CH ₂ Ph) ₂	1	[375]
	2	PPN ^b	2	μ-dmit	1	C ₆ F ₅	2	[307]
	2	PPN ^b	2	μ-dmit	1	Cl	2	[307]
	2	PPN ^b	2	μ-dmit	1	Br	2	[307]
	2	PPN ^b	1	μ-dmit	2			[375]
	2	PPN ^b	1	μ-dmit	1	μ-S ₂ CNMe ₂	1	[375]
	2	PPN ^b	1	μ-dmit	1	μ-S ₂ CN(CH ₂ Ph) ₂	1	[375]
	2	PPN ^b	1	μ-dmit	1	μ-CH ₂ PPh ₂ CH ₂	1	[375]
	2	Ag	2	μ-dmit	2			[375]
	2	TTF ^c	1	μ-dmit	1	μ-CH ₂ PPh ₂ CH ₂	1	[375]
	2	TTF ^c	1	dmit	1	C ₆ F ₅	2	[307]
	2	TTF ^c	3	dmit	1	Br	2	[307]
Cu	2	Bu ₄ N	2	dmit	2	μ-tto	1	[215,218,371,372]
	2	Bu ₄ N	0.33	dmit	2	μ-tto	1	[371]
	2	Bu ₄ N	2	dmid	2	μ-tto	1	[215,372]
	2	Bu ₄ N	2	dsit	2	μ-tto	1	[215,372]
	4	Bu ₄ N	2	dmit	3			[376–378]
	4	Bu ₄ N	0.2	dmit	3			[377,378]
	4	Mepy ^b	2	dmit	3			[376–378]
	4	Fe(C ₅ Me ₅) ₂	1.2	dmit	3			[377,378]
	4	Bu ₄ N	2	dmt	4			[379]
	4	Bu ₄ N	2	dsis	3			[377,378]
	4	Bu ₄ N	0.16	dsis	3			[377,378]
	4	Fe(C ₅ Me ₅) ₂	0.5	dsis	3			[377,378]
Mn	2	Me ₄ N	4	dmit	4			[32]
Nb	2	Bu ₄ N	2	dmit	4	μ-S ₂	2	[336]
Ni	2	Me ₄ N	2	dmit	2	μ-tto	1	[374]
	2	Et ₄ N	2	dmit	2	μ-tto	1	[374]
	2	Bu ₄ N	2	dmit	2	μ-tto	1	[215,373]
	2	Bu ₄ N	0.33	dmit	2	μ-tto	1	[374]
	2	Bu ₄ N	x	dmit	2	μ-tto	1	[374]
	2	TTF ^c	0.57	dmit	2	μ-tto	1	[374]
	2	Bu ₄ N	2	dmise	2	μ-tto	1	[215,374]
	2	Bu ₄ N	2	dsit	2	μ-tto	1	[215,374]
	3	Et ₄ N	2	dmit	2	μ-edt ^d	4	[380]
	3	Ph ₄ As	2	dmit	2	μ-pdt ^e	2	[380]

Table 17 (Continued)

M	x	A	n	L ₁	y	L ₂	z	Ref.
Re	2	Et ₄ N	2	dmit	5			[381]
	2	Et ₄ N	0.65	dmit	5			[381]
	2	Et ₄ N	0.5	dmit	5			[381]
	2	Et ₄ N	0.3	dmit	5			[381]
	2	Bu ₄ N	2	dmit	5			[381]
	2	Ph ₄ P	2	dmit	5			[381]
	2	Ph ₄ P	0.2	dmit	5			[381]
	2	Ph ₄ P	0.1	dmit	5			[381]
	2	TTF ^c	1	dmit	5			[381]
	2	Et ₄ N	2	dmise	5			[382]
	2	Et ₄ N	0.5	dmise	5			[382]
	2	Et ₄ N	0.15	dmise	5			[382]
	2	Ph ₄ P	2	dmise	5			[382]
	2	TTF ^c	1	dmise	5			[382]
WS	2	Ph ₄ P	2	dmit	2	μ-S	2	[333]

^a See Table 16.^b See Table 1.^c See Table 2.^d edt = Ethane-1,2-dithiol.^e pdt = Propane-1,2-dithiol.

3.8. 1,3-Diselenole-2-selone-4,5-diselenolate

Tables 13 and 14 list bis- and tris-dsis compounds, respectively. The all selenium analog of dmit (dsis) has been primarily coordinated with metals of Groups 10–12. Also, Tanaka and Matsubayashi assembled Fe(dsis)₂-based NIOS complexes by I₂ or Fe(Cp)₂⁺ oxidation with (TTF)_{0.28}[Fe(dsis)₂] having the highest conductivity of 4.0×10^{-5} S cm⁻¹ [314]. The complex (Me₄N)_{0.5}[Ni(dsis)₂] has the highest conductivity of a M(dsis)₂-based complex (1 S cm⁻¹) [138].

Douki and Matsubayashi has synthesized a series of W(dsis)₃ complexes and examined their UV–vis, IR, ESR, electrochemical and electrical conductivity properties. The NIOS complex [Fe(Cp*)₂]_{0.5}[W(dsis)₃] showed the highest conductivity of 1.5×10^{-3} S cm⁻¹ [361].

4. Mixed ligand and cluster complexes

Since the publication of the previous review on the coordination chemistry of dmit [5], an area that has exploded in research effort is that of the synthesis and characterization of novel mixed-ligand and cluster complexes containing dmit and its isologs. As will be discussed, dmit-based ligands have shown the ability to coordinate in different fashions to yield a large number of interesting and unexpected coordination complexes.

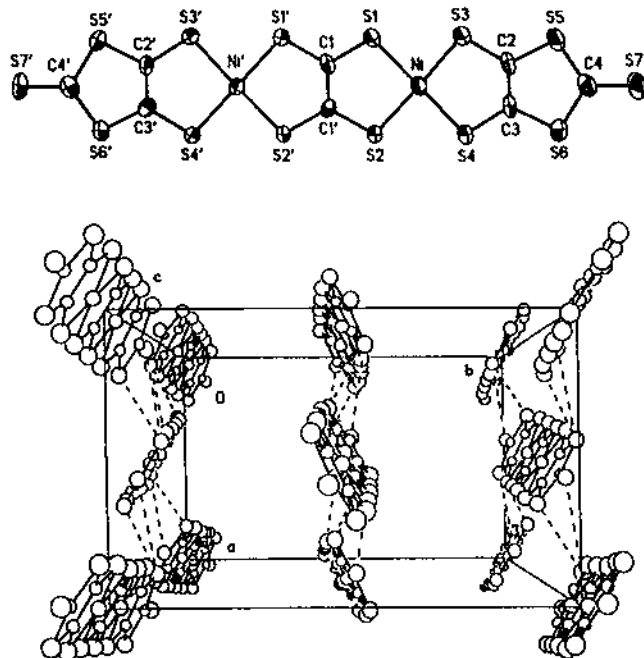


Fig. 3. Structure of the dianion of the bimetallic complex $(\text{Bu}_4\text{N})_2\{\text{tto}[\text{Ni}(\text{dmit})_2]\}_2$ (top) and a view of the packing diagram of the dianions down the *c* axis (bottom) [373]. Reproduced with permission from *Inorg. Chem.* 36 (1997) 958. Copyright 1997 Am. Chem. Soc.

Tables 15 and 16 list mixed-ligand complexes composed of metals coordinating two ligands of differing composition with at least one of the ligands being dmit or an isolog.

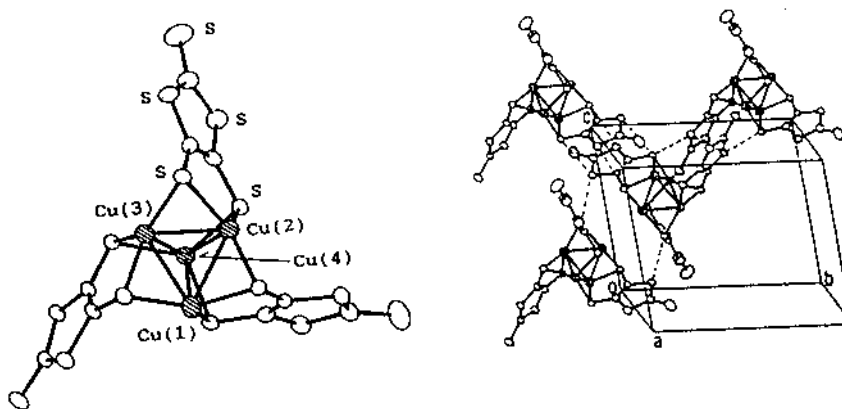


Fig. 4. Molecular structure of the cluster anions of $(\text{Mepy})_2[\text{Cu}_4(\text{dmit})_3]$ (left) with shaded copper atoms to show the pyramidal arrangement [378]. A view of the packing arrangement is on the right. Reprinted by permission from *Phosphorus, Sulfur, and Silicon* 67 (1992) 339. Copyright 1992 Gordon and Breach Publishers.

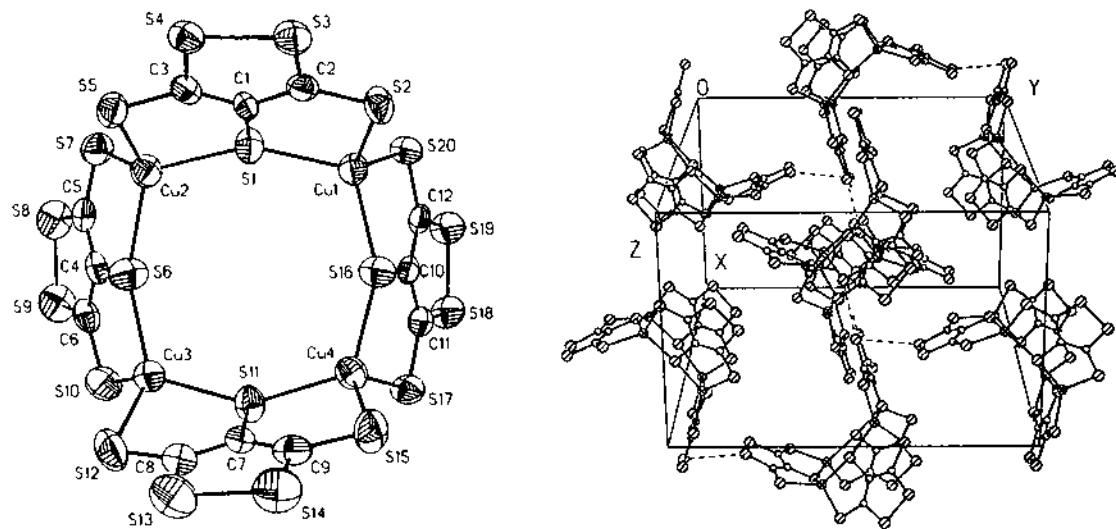


Fig. 5. Figures showing the dianion cluster of $(\text{Bu}_4\text{N})_2[\text{Cu}(\text{dmt})]_4$ (left) and the packing arrangement of the dianions (right) [379]. Reproduced with permission from *Inorg. Chem.* 35 (1996) 793. Copyright 1996 Am. Chem. Soc.

Cerrada et al. have reported their investigations of Au(III) mixed-ligand complexes [307,362]. For example, $(\text{Bu}_4\text{N})[\text{Au}(\text{dmit})\text{L}]$ (where $\text{L} = \text{Me}_3\text{P}$, MePh_2P , Ph_3P) are all air and moisture stable and the complex when $\text{L} = \text{Ph}_3\text{P}$, was characterized by X-ray crystallography and displays trigonal coordination [307].

Matsubayashi et al. [366], Olk et al. [312,364] and Kato et al. [367] have reported mixed-ligand complexes from ligand-exchange reactions. Bis-dmit or dmit isolog complexes have shown the ability to ligand-exchange in solution with other bis-dmit or dmit isologs [366,367] or bis-1,1-dichalcogenolate complexes [312,364]. The formation of the mixed-ligand complexes can be followed by UV–vis and EPR spectroscopy. The complexes $(\text{TTF})[\text{Ni}(\text{dmit})(\text{mnt})]$ and $(\text{EDT-TTF})[\text{Ni}(\text{dmit})(\text{mnt})]$ synthesized by electrocrystallization, have shown to display metallic-like conductivity behavior down to 20–30 K [367].

Table 16 lists mixed dmit and dmise ligand chelate complexes synthesized by the addition of a metal ion to a stirring mixture of the ligands by You and coworkers [369,370]. Crystals obtained for X-ray crystallography and EPR measurements contain non-integral amounts of sulfur and selenium. The $\text{Se}/\text{S}=\text{C}$ bond length was determined to vary with respect to the stoichiometric ration of Se/S . The neutral compound $[\text{Ni}(\text{C}_3\text{S}_{4.4}\text{Se}_{0.6})_2]$ exhibited a pressed pellet conductivity of 0.29 S cm^{-1} [369].

Reynolds and coworkers assembled a series of Cu(II) and Ni(II) bimetallic complexes bridged by a tto (tto = tetrathiooxalate) ligand and capped with a dmit,

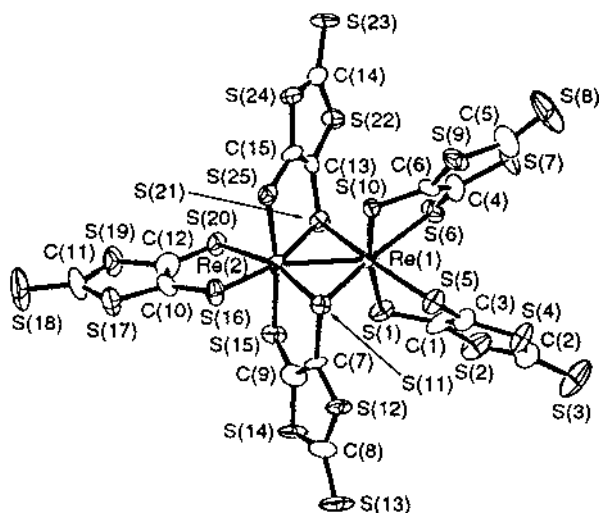


Fig. 6. Figure showing the dinuclear cluster anion of $(\text{Ph}_4\text{P})_2[\text{Re}_2(\text{dmit})_5]$ with an Re_2S_{10} core. The geometry about each Re atom is distorted octahedral [381]. Reproduced by permission of the R. Soc. Chem.

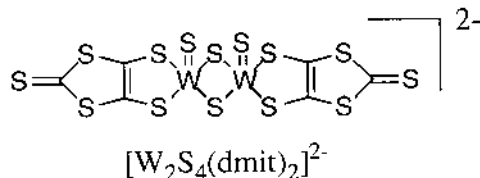


Fig. 7. A proposed structure of the dianion of the complex $(\text{Ph}_4\text{P})_2[\text{W}_2\text{S}_4(\text{dmit})_2]$ [333].

dmid, dmise or dsit ligand [215,218,371–374] as listed in Table 17. They were designed to extend conjugation and increase intermolecular interactions to have enhanced electrical conductivity properties. The Cu(II) bimetallic systems were assembled using a biphasic, three-solvent method and showed tetrahedrally distorted square planar coordination [215,218,371,372]. The Ni(II) d^8 bimetallic complexes all displayed square planar coordination [215,373,374]. The NIOS complex $(\text{Bu}_4\text{N})_x\{\text{tto}[\text{Ni}(\text{dmit})_2]\}$ obtained by electrocrystallization of the dianion $(\text{Bu}_4\text{N})_2\{\text{tto}[\text{Ni}(\text{dmit})_2]\}$ exhibited a pressed pellet conductivity of 0.5 S cm^{-1} [374] (Fig. 3).

Tables 17 and 18 lists mixed-ligand chelate complexes composed of a metal coordinated to three to six ligands with at least one ligand being dmit or an isolog. The complexes containing three ligands consist of metals from Groups 4–6, 8, 11 and 14. Fourmigué and coworkers assembled Mo and Ti mixed tris-ligand complexes and found that their geometry varies with the electron count of the complex [31,396]. An antiferromagnetic ground state was observed in the neutral compound $[\text{Cp}^*\text{Mo}(\text{dmit})_2]$, with a transition temperature of 8 K [31]. Zeltner et al. have examined tris-ligand complexes of Hf, Ti, V and Zr of the general formula Cp_2ML ($\text{L} = \text{dmit}, \text{dmt}, \text{dmid}, \text{dmise}, \text{dsit}, \text{dsise}, \text{dsitse}$ and dsis) [385,395,398]. The authors studied the complexes with IR, UV–vis, EPR and NMR. The ruthenium complex $[\text{Cp}^*\text{Ru}(\text{dmit})(\text{NO})]$ was assembled and X-ray analysis showed the complex exhibits C_s symmetry [389]. Neutral tris-ligand tin complexes containing dmit were assembled by Wardell and associates [321,391]. The crystal structure of $[\text{MePhSn}(\text{dmit})]$ showed a distorted trigonal bipyramidal geometry and extensive $\text{S}\cdots\text{S}$ intermolecular interactions [321]. These same authors have also assembled tetrakis-ligand tin, lead and antimony complexes [321,388,390,392,394]. Crystal structures of the tin salts $(\text{Bu}_4\text{N})[\text{Me}_2\text{Sn}(\text{dmit})\text{Cl}]$ and $(\text{Et}_4\text{N})[\text{Ph}_2\text{Sn}(\text{dmid})\text{Cl}]$ both exhibit a trigonal bipyramidal coordination geometry [324,392] while in $(\text{Bu}_4\text{N})_2[\text{Sn}(\text{dmit})_2\text{I}_2]$, the coordination is in an octahedral-like fashion [394]. The Sb(V) salts $(\text{Et}_4\text{N})[\text{Ph}_2\text{Sb}(\text{dmit})_2]$ and $(\text{Et}_4\text{N})[(p\text{-tolyl})_2\text{Sb}(\text{dmit})_2]$ which were the first organoantimony-dmit compounds to be characterized by X-ray crystallography, have a distorted octahedral coordination environment. The aryl groups are *cis* with respect to each other.

Baker et al. have synthesized pentakis-ligand tungsten complexes and obtained a crystal structure for the neutral compound $[\text{W}(\text{dmit})(\text{CO})_2(\text{Et}_3\text{P})_2]$ [24]. The CO ligands are *cis* to each other and *trans* to the dmit ligand and the coordination about the tungsten is nearly trigonal prismatic. The compounds were also studied by ^{31}P -NMR and IR spectroscopy [399].

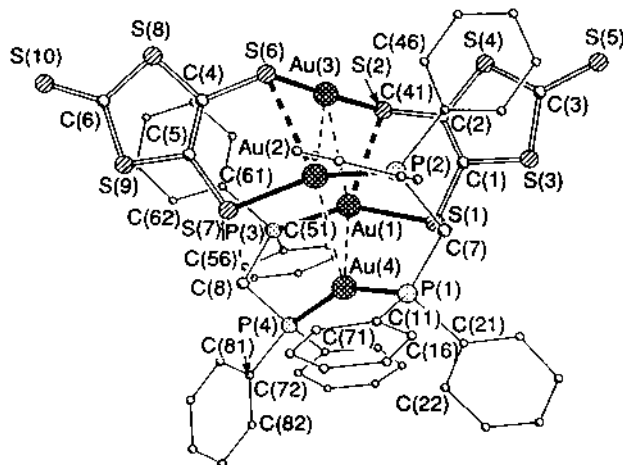


Fig. 8. Molecular structure of the neutral cluster complex $[\text{Au}_4(\mu\text{-dmit})_2(\mu\text{-dppm})_2]$ with a coplanar trapezium core of gold atoms [319]. Reproduced by permission of the R. Soc. Chem.

Many new cluster complexes based on dmit or an isolog with Cu, Mn, Nb or Re have been reported since the previous review and are listed in Table 17. The complex $(\text{Mepy})_2[\text{Cu}_4(\text{dmit})_3]$ reported by Matsubayashi and Yokozawa consists of four Cu and three dmit ligands with a central Cu_4S_6 cluster core [376–378] (Fig. 4). The Cu atoms are arranged in a distorted tetrahedron. One of the Cu atoms of each cluster is coordinated to a terminal thione sulfur of an adjacent cluster. The dms analog has also been synthesized and both complexes have been studied by IR, UV–vis and cyclic voltammetry [376–378].

The complex $(\text{Bu}_4\text{N})_2[\text{Cu}_4(\text{dmt})_4]$ synthesized by Reynolds and coworkers is the only cluster complex containing the dmt ligand [379] (Fig. 5). The cluster consists of a planar, eight-membered ring core of four Cu and four S atoms. The coordination about each Cu atom is distorted tetrahedral. Nonbonding intermolecular $\text{S}\cdots\text{S}$ interactions are observed from intermolecular thiole–thiole contacts [379].

Matsubayashi and associates have also prepared Re-based cluster complexes containing dmit or dmise [381,382]. A crystal structure of $(\text{Ph}_4\text{P})_2[\text{Re}_2(\text{dmit})_5]$ was obtained and contains a Re_2S_{10} core of C_2 symmetry (Fig. 6). The coordination about the Re atoms is a distorted octahedron [381]. The dmise analog was also prepared and the complexes were characterized by XPS, cyclic voltammetry, IR, MS, EPR and UV–vis. The complexes were chemically and electrochemically oxidized to yield several NIOS compounds where the complex $(\text{TTF})[\text{Re}_2(\text{dmit})_5]$ exhibited a high conductivity of $4.9 \times 10^{-2} \text{ S cm}^{-1}$. Yang et al. reported a bimetallic tungsten complex of the composition $(\text{Ph}_4\text{P})_2[\text{W}_2\text{S}_4(\text{dmit})_2]$ [333] characterized by IR, elemental analysis and UV–vis (Fig. 7).

Tables 17 and 18 contains assembled anionic gold complexes while Table 19 lists cationic gold chelate complexes. Laguna and coworkers have synthesized

heteroleptic complexes containing 2–4 gold atoms [307,319,375]. A common structural feature of the Au(I) complexes is the dmit ligand acts as a bridging ligand between two gold atoms forming a diauracycle. This has been observed by X-ray studies of compounds such as $[\text{Au}_2(\mu\text{-dmit})(\text{Ph}_3\text{P})_2]$ [307] and $(\text{Bu}_4\text{N})[\text{Au}_2(\mu\text{-dmit})(\mu\text{-CH}_2\text{PPh}_2\text{CH}_2)]$ [375]. In $[\text{Au}_2(\mu\text{-dmit})(\text{Ph}_3\text{P})_2]$, one Au atom is linearly coordinated while the other is pseudotrigonal [307] and in $(\text{Bu}_4\text{N})[\text{Au}_2(\mu\text{-dmit})(\mu\text{-CH}_2\text{PPh}_2\text{CH}_2)]$, the coordination about each Au atom is slightly bent [375]. The authors observed that oxidation of one of the gold atoms can lead to the formation of a monometallic Au(III) complex. The compound $(\text{PPN})_2[\text{Au}_2(\mu\text{-dmit})(\text{C}_6\text{F}_5)_2]$ was oxidized by TCNQ (see Table 5) to yield $(\text{PPN})[\text{Au}(\text{dmit})(\text{C}_6\text{F}_5)_2]$ and confirmed by X-ray crystallography [307]. The coordination about the Au(III) center is square planar. The tetrametallic Au(I)

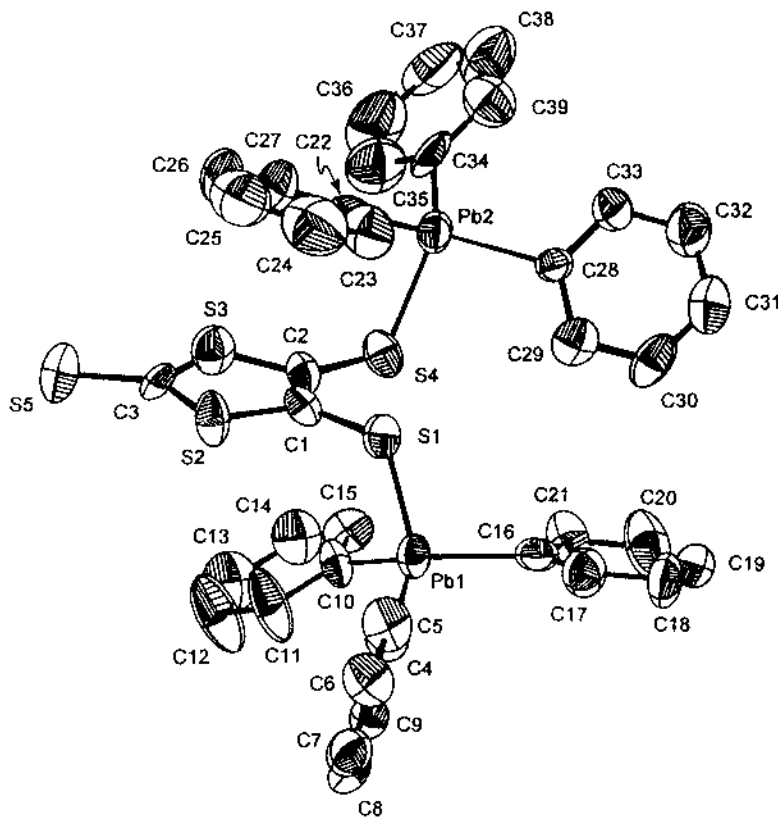


Fig. 9. Molecular structure of the neutral complex $[(\text{Ph}_3\text{Pb})_2(\mu\text{-dmit})]$ [388]. Reprinted from Polyhedron, 15, S.M.S.V. Doidge-Harrison, J.T.S. Irvine, G.M. Spencer, J.L. Wardell, syntheses and properties of the lead 1,3-dithiole-2-thione-4,5-dithiolate (dmit) compounds: $\text{Ph}_2\text{Pb}(\text{dmit})$, $[\text{Q}][\text{Ph}_2\text{Pb}(\text{dmit})\text{I}]$ [$\text{Q} = \text{NEt}_4$ or 1,4-Me₂Pyridinium], $(\text{Ph}_3\text{Pb})_2(\text{dmit})$ and $\text{Pb}(\text{dmit})$, 1807. Copyright 1996, with permission from Elsevier Science.

Table 18

Mixed-ligand chelate complexes of the general formula $(A)_n[M(L_1)(L_2)(L_3)(L_4)(L_5)(L_6)]$

M	A	n	L ₁	L ₂	L ₃	L ₄	L ₅	L ₆	Ref.
Au	–		dmit	C ₆ F ₅	py				[362]
	–		dmit	C ₆ F ₅	MePh ₂ P				[383]
	–		dmit	C ₆ F ₅	Ph ₃ P				[383]
	–		dmit	C ₆ F ₅	Ph ₃ As				[383]
	Bu ₄ N	1	dmit	C ₆ F ₅	Br				[362]
	Bu ₄ N	1	dmit	C ₆ F ₅	C ₆ F ₅				[307]
	PPN ^a	1	dmit	C ₆ F ₅	Cl				[362]
	PPN ^a	1	dmit	C ₆ F ₅	SCN				[362]
Er	K	2	dmit	phen ^b	phen ^b	Cl	Cl	Cl	[384]
	K	2	dmit	phen ^b	phen ^b	Cl	Cl	Cl/I _{3.0}	[384]
Gd	K	2	dmit	phen ^b	phen ^b	Cl	Cl	Cl	[384]
Hf	–		dmit	Cp	Cp				[385]
	–		dmt	Cp	Cp				[385]
	–		dmid	Cp	Cp				[385]
	–		dmise	Cp	Cp				[385]
La	K	2	dmit	phen ^b	phen ^b	Cl	Cl	Cl	[384]
	K	2	dmit	phen ^b	phen ^b	Cl	Cl	Cl/I _{3.5}	[384]
Mo	–		dmit	dmit	C ₅ Me ₅				[31]
	–		dmit	Cp	Cp				[386]
	–		dmid	Cp	Cp				[386]
	Ph ₄ P	1	dmit	dmit	C ₅ Me ₅				[31]
Nd	K	2	dmit	phen ^b	phen ^b	Cl	Cl	Cl	[384]
Ni	–		dmit	dmit	Piperidine	Piperidine			[387]
	Bu ₄ N	1	dmit	dmit	Piperidine	Piperidine			[387]
Os	Bu ₄ N	1	dmit	CH ₂ SiMe ₃	CH ₂ SiMe ₃	N			[287]
Pb	–		dmit	Ph	Ph				[388]
	Et ₄ N	1	dmit	Ph	Ph	I			[388]
	1,4-Dimethyl-pyridinium	1	dmit	Ph	Ph	I			[388]
Ru	–		dmit	C ₅ Me ₅	NO				[389]
Sb	Et ₄ N	1	dmit	dmit	Ph	Ph			[390]
	Et ₄ N	1	dmit	dmit	<i>p</i> -Tolyl	<i>p</i> -Tolyl			[390]
	1,4-Dimethyl-pyridinium	1	dmit	dmit	Ph	Ph			[390]
Sm	K	2	dmit	phen ^b	phen ^b	Cl	Cl	Cl	[384]

Table 18 (Continued)

M	A	<i>n</i>	L ₁	L ₂	L ₃	L ₄	L ₅	L ₆	Ref.
Sn	–		dmit	Me	Me				[321,383]
	–		dmit	Me	Me	Pyridine			[321]
	–		dmit	Me	Ph				[321]
	–		dmit	Et	Et				[321]
	–		dmit	Bu	Bu				[321]
	–		dmit	Oct	Oct				[321]
	–		dmit	CH ₂ CH ₂ CO ₂ Me	CH ₂ CH ₂ CO ₂ Me				[391]
	–		dmit	CH ₂ CH ₂ CO ₂ Pr ⁱ	CH ₂ CH ₂ CO ₂ Pr ⁱ				[391]
	–		dmit	Ph	Ph				[321]
	–		dmit	Ph	Ph	2,2'-Bipyridine			[321]
	–		dmit	Ph	Ph	DMF			[321]
	–		dmit	<i>o</i> -MeOPh	<i>o</i> -MeOPh				[321]
	Et ₄ N	1	dmit	dmit	CH ₂ CH ₂ CO ₂ Me				[391]
	Et ₄ N	1	dmit	Bu	Bu	Br			[321]
	Et ₄ N	1	dmit	Ph	Ph	Br			[321]
	Et ₄ N	1	dmit	Ph	Ph	SCN			[321]
	Et ₄ N	1	dmit	Ph	Ph	Cl			[392]
	Et ₄ N	1	dmit	Ph	Ph	SCN			[392]
	Bu ₄ N	1	dmit	dmit	Bu				[393]
	Bu ₄ N	1	dmit	dmit	Ph				[393]
	Bu ₄ N	2	dmit	dmit	Br	Br			[394]
	Bu ₄ N	2	dmit	dmit	I	I			[393]
	Bu ₄ N	1	dmit	Me	Me	Cl			[324]
	Bu ₄ N	1	dmit	Ph	Ph	Br			[321]
	1,4-Dimethyl-pyridinium	1	dmit	dmit	CH ₂ CH ₂ CO ₂ Me				[391]
	1,4-Dimethyl-pyridinium	1	dmit	Ph	Ph	I			[321]
	MV ^a	1	dmit	Ph	Ph	Cl		Cl	[321]
	Me ₃ NCH ₂ SnMe ₃	1	dmit	Me	Me	I			[321]
	(<i>p</i> -Me ₂ NC ₆ H ₄) ₃ C	1	dmit	Ph	Ph	Cl			[321]
	Me ₃ SO	1	dmit	Ph	Ph	I			[321]
Ti	–		dmit	Cp	Cp				[318,385,395,396]
	–		dmit	C ₅ Me ₅	C ₅ Me ₅				[397]
	–		dmt	Cp	Cp				[385,395]
	–		dmid	Cp	Cp				[385,395,396]
	–		dmise	Cp	Cp				[385,395]

Table 18 (Continued)

M	A	n	L ₁	L ₂	L ₃	L ₄	L ₅	L ₆	Ref.
V	—	1	dsit	Cp	Cp				[395,398]
	—		dsit	C ₅ H ₄ Me	C ₅ H ₄ Me				[398]
	—		dsit	C ₅ H ₄ Et	C ₅ H ₄ Et				[398]
	—		dsise	C ₅ H ₄ Me	C ₅ H ₄ Me				[398]
	—		dsise	C ₅ H ₄ Et	C ₅ H ₄ Et				[398]
	—		dsitse	C ₅ H ₄ Me	C ₅ H ₄ Me				[398]
	—		dsitse	C ₅ H ₄ Et	C ₅ H ₄ Et				[398]
	—		dsis	C ₅ H ₄ Me	C ₅ H ₄ Me				[398]
	—		dsis	C ₅ H ₄ Et	C ₅ H ₄ Et				[398]
	Bu ₄ N		dmit	dmit	Cp				[396]
	—		dmit	Cp	Cp				[385]
	—		dmt	Cp	Cp				[385]
	—		dmid	Cp	Cp				[385]
	—		dmise	Cp	Cp				[385]
	—		dsit	C ₅ H ₄ Me	C ₅ H ₄ Me				[398]
W	—	1	dsise	C ₅ H ₄ Me	C ₅ H ₄ Me				[398]
	—		dsitse	C ₅ H ₄ Me	C ₅ H ₄ Me				[398]
	—		dsis	C ₅ H ₄ Me	C ₅ H ₄ Me				[398]
	—		dmit	CO	CO	dppe ^c			[399]
	—		dmit	Cp	Cp				[400]
	—		dmit	C ₅ H ₄ Bu ^t	C ₅ H ₄ Bu ^t				[400]
	—		dmit	C ₅ H ₄ SiMe ₃	C ₅ H ₄ SiMe ₃				[400]
	—		dmid	Cp	Cp				[400]
	TCNQF ^d		dmit	Cp	Cp				[400]
	TCNQF ^d		dmit	C ₅ H ₄ Bu ^t	C ₅ H ₄ Bu ^t				[400]
Y Zr	—	2	dmit	Cp	Cp				[400]
	K		dmit	phen ^b	phen ^b	Cl	Cl	Cl	[384]
	—		dmit	Cp	Cp				[345,385]
	—		dmt	Cp	Cp				[385]
	—		dmid	Cp	Cp				[385]
	—		dmise	Cp	Cp				[385]

^a See Table 1.^b phen = 11,10-phenanthroline.^c See Table 16.^d TCNQF = 7,7,8,8-tetracyano-1,2,4,5-tetrafluoroquinodimethane.

Table 19

Gold chelate complexes of the general formula $[\text{Au}_x(\mu\text{-dmit})(\text{L}_1)(\text{L}_2)(\text{L}_3)](\text{Q})$

<i>x</i>	L_1	L_2	L_3	Q	Ref.
4	$\mu\text{-dmit}$	$\mu\text{-(Ph}_2\text{PCH}_2\text{PPh}_2)^{\text{a}}$	$\mu\text{-(Ph}_2\text{PCH}_2\text{PPh}_2)^{\text{a}}$	–	[319]
3	Ph_3P	Ph_3P	Ph_3P	ClO_4	[307,319]
2	MePh_2P	MePh_2P		ClO_4	[307]
2	$\text{CH}_2\text{Ph}_3\text{P}$	$\text{CH}_2\text{Ph}_3\text{P}$		ClO_4	[307]

^a dppm = $\text{Ph}_2\text{PCH}_2\text{PPh}_2$.

complex $[\text{Au}_4(\mu\text{-dmit})_2(\mu\text{-dppm})_2]$, is composed of a core of four coplanar gold atoms in the shape of a trapezium [319] (Fig. 8).

Au-dmit complexes have been used in electrocrystallization experiments. The compound $[\text{Au}_2(\mu\text{-dmit})(\text{Ph}_3\text{P})_2]$ was electrooxidized in the presence of Bu_4NClO_4 and yielded $[\text{Au}_2(\text{dmit})_2(\text{Ph}_3\text{P})]$ which exhibited a pressed pellet conductivity of $1.3 \times 10^{-5} \text{ S cm}^{-1}$ [307].

Similar to the diauracycles previously mentioned, a bimetallic complex of composition $[(\text{Ph}_3\text{Pb})_2\text{dmit}]$ has been reported by Doidge-Harrison et al. [388] (Fig. 9). The compound is composed of a dmit ligand bridging two lead atoms each having three bound phenyl groups. The geometry about each lead atom is distorted tetrahedral [388].

5. Conclusions

In conclusion, dmit and isologs have been used in the assembly of anionic and cationic bis, tris, mixed-ligand and cluster complexes. They have also been combined with a large variety of counterions leading to diverse crystal packing arrangements and materials exhibiting a wide array of physical properties. Based on the current amount of interest and number of researchers exploring dmit and related isologs, new and fascinating developments in the chemistry of these ligands are expected.

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