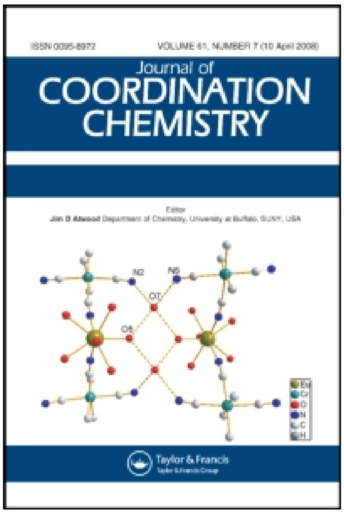

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Molecular design of mononuclear complexes of acyclic Schiff-base ligands

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Molecular design of mononuclear complexes of acyclic Schiff-base ligands

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Data on molecular design of various types of mononuclear complexes of Schiff bases are generalized. Factors for variation of fine structure of Schiff-base ligand systems (metallocycle sizes and their number, nature of donor centers, aldehyde and amine fragments, co-ligands) are examined. A systematization of Salen complexes is offered. Electronic configurations of *p*- and *d*-metals in the coordination and organometallic compounds are taken into account in a series of chelate structures. Non-standard structures are emphasized, including various ligand chelates and molecular complexes.

Keywords: Schiff bases; Mononuclear complexes; Di- and oligodentate azomethine ligands with N.C-, N.N-, N.P-, N.O-, N.S-, N.Se-, N.Te-donor centers; C-, N-, O-, S-, Se-, Te- Salen chelates; Standard and nonstandard structures

1. Introduction

Schiff bases and their structural analogues, as ligating compounds containing acyclic and cyclic imine C=N bonds, are of great importance in modern coordination chemistry [1–6]. Interest in metal complexes of these ligands [7–10] is related to wide variability of their fine structure (rational design [5]) and obtaining of polyfunctional materials. Among them, we note luminescent complexes [11, 12], magnetoactive [13–15] and liquid crystal [16, 17] structures, chemosensors [18–20] and other useful metal-containing azomethine compounds [21]. Bioinorganic and biomimetics, containing a C=N bond, are widely represented [22–24]. Practically all coordination compounds of Schiff bases were synthesized using chemical and electrochemical techniques [2].

The present review will focus on progress in the area of design of mononuclear complexes of Schiff bases, based, in general, on publications of the 21st century. All factors forming mononuclear coordination compounds of Schiff bases are taken into account. Thereupon, a systematization of various types of Schiff-base complexes is offered.

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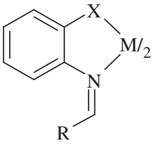
2. Schiff-base complexes with five-membered metallocycles

In early publications cited in a monograph [2] and reviews [5, 7, 9, 25], the five-membered metallocyclic chelate structures of types **1**–**3** were described.



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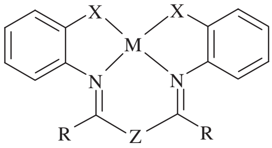

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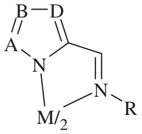
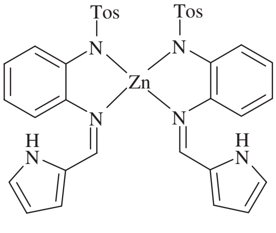
1, 2: X = NR^1, O, S; Z = $(CH_2)_n$, Ar;
R = Ar, Het; n = 0–6; R^1 = Ar, Tos;
Tos = $–SO_2C_6H_4Me$-p; n = 1–6



3: A, B, D = CR^1, N;
R = Alk, Ar, Het; R^1 = H, Alk

Metal-chelates of type **1** are widely represented by complexes with heterocyclic coordinatively-active R-substituents [2, 5, 10]. These functional groups may or may not participate in a metal ligand interaction [5, 25, 26]. Thus, the Schiff-base ligand in **1** (X = NTos, R = 2-Pyrrolyl) is bidentate or tridentate giving the complexes **4** [27] or **5** [28], correspondingly. Salen type complexes **2** (X = O, NTos; Z = Het = 2, 6-Py) were considered in a review [5].



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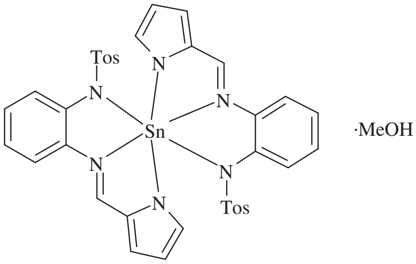

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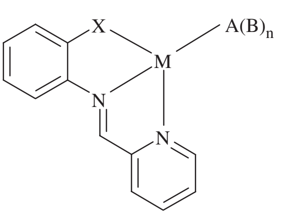
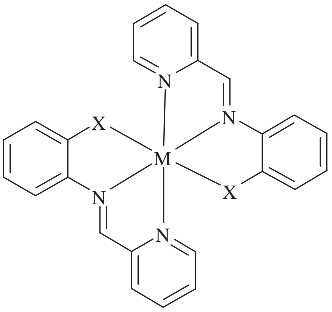
The pyridine (**1**: R = 2-Py) substituent when X = O generally participates in bonding with metal, increasing its coordination number (C.N.). As a result, complexes containing an anionic (A) and organic (B) fragment in **6** are formed, as well as usual chelate structures **7**.



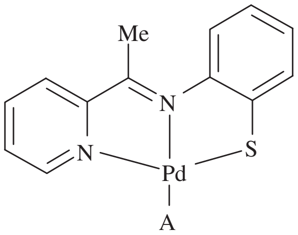
6: X = NTos, O;
A = Cl, ONO_2, OCOMe;
B = Me, Bu, Ph



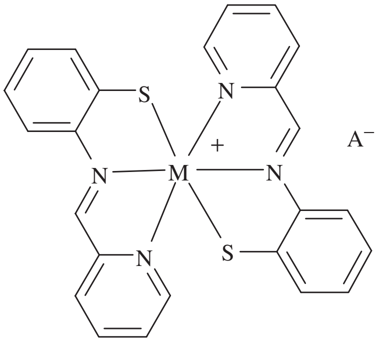
7: X = NTos, O; M = Fe, Co, Ni, Cu, Zn, Cd

Among **6**, recently described complexes of Cu(II) ($X = \mathrm{NTos}$) [29] and Sn(IV) [30], Fe(II) [31], Pd(II) [32], Cu(II) [33, 34] ($X = O$) have been described. Coordination compounds of type **7** have been synthesized and structurally characterized in the case of $X = \mathrm{NTos}$ for Co(II) and Cu(II) [29]; $X = O$, $M = \mathrm{Fe(II)}$ [35], Ni(II) [36], Zn(II) and Cd(II) [37]. Iron(III) forms a cationic complex containing perchlorate [35].

Additionally to **1** ($X = S$) [5, 10, 38], the Pd(II) coordination compounds **8**, characterized by X-ray analysis and containing OCOMe or Me fragments, have been described [32].

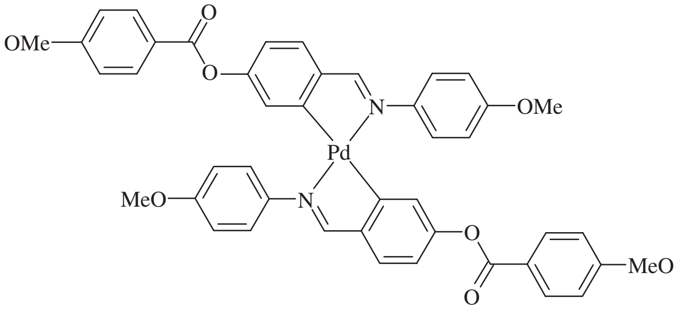


8: A = OCOMe, Me



9: M = Co, A = Cl; M = Fe, A = BPh_4

Co(II) [39] and Fe(III) [40] form cationic complexes of type **9**. Structure **10** is similar to **1**, with five-membered C,N-metal-chelate structures [41] occuring as a result of cyclopalladation [2]. Structure **11** (of type **2** where $n = 0$) [42] seems doubtful without X-ray data.



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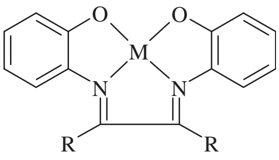
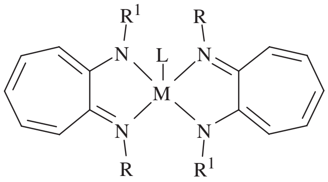
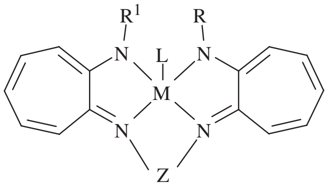


11

Complexes of type **3** are examined in monographs [2, 26] and reviews [9, 25, 43]. In addition to the metallocycles above, five-membered metallocycles are formed in coordination compounds containing non-standard aldehyde fragments, e.g. aminotropolonimines **12**, **13** [10, 18, 44–46], Schiff bases of pyridine-2-aldehyde of type **14** [47], **15** [48] and 2,6-diformylpyridinede [49–53].



12: R = *i*-Pr, *t*-Bu, Bz, Ar;
R^1 = Alk, ArTos, NaphtTos;
ArTs = $-SO_2C_6H_4Me$-*p*;
Napht Ts = $-SO_2C_{10}H_6NMe_2$;
L = Cl, NO

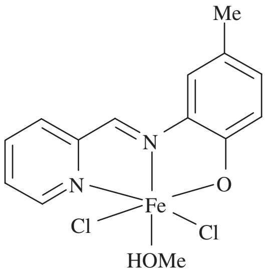


13: Z = $(CH_2)_n$; *n* = 2–6; L = Cl, NO

Ligands for complexes of types **12** and **13** were obtained through stepwise syntheses [18, 45, 46]. Complexes **12** and **13** were prepared from metal chlorides in THF; their structures are proved by X-ray [45] and are of interest as luminophors and fluorescent sensors [18, 46].

The structurally characterized indium compounds **13** [M = In, R = *i*-Pr, L = Cl, Z = $(CH_2)_3$] should be noted. Chloride exchange in **13** results in a series of tricyclic metal-chelates with L = Me, CH_2SiMe_3, OBu [45]. Structures of analogous complexes of gallium(III) are proved by ^1H and ^{13}C NMR spectroscopy [46].

An iron(III) complex with coordinated pyridine nitrogen is a characteristic metal-chelate of type **14** [47].



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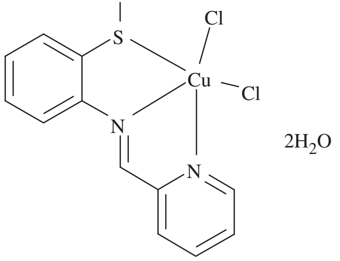
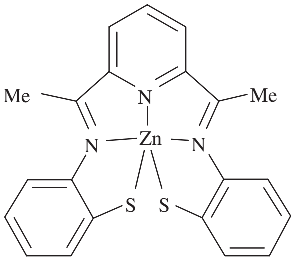
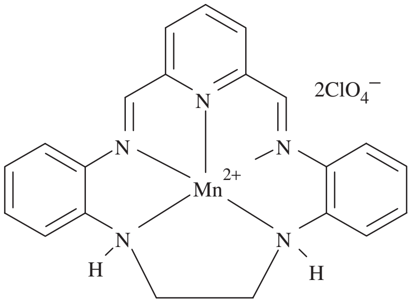
The complex of pyridine-2-aldimine is represented as adduct **15** with CuN_2SCl_2 coordination [48].





15 **16**

Structurally characterized complex **16** of a Schiff base of 2,6-pyridine dicarbonyl derivatives with N_3S_2 chelate environment was described for the first time in the 1970's [49]. More recently, cationic complexes with MN_5 – **17** [50] and MN_6 – **18** [51] were reported.



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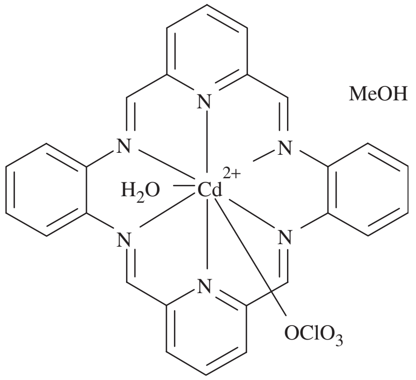
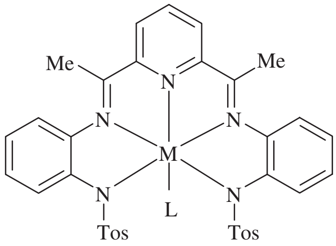
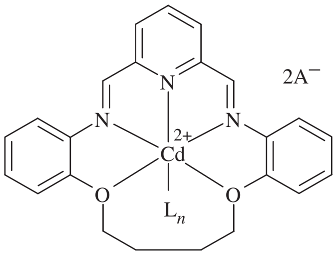


18

Complex **19** possesses a neutral tetrachelate N_5 ligand environment [52].



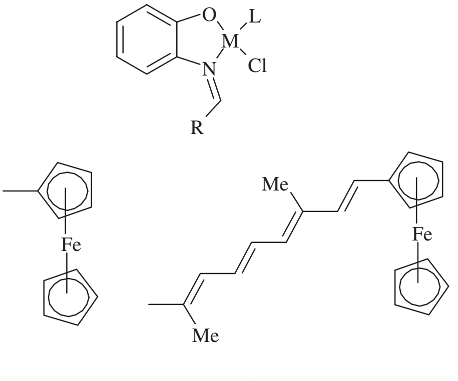
19: M = Zn, L = none; M = Cd, L = H_2O



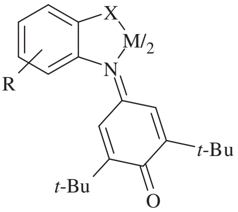
20: L = H_2O, $n = 2$, A = ONO_2, ClO_4

Cationic cadmuim chelates with various anions are represented by Schiff-base complexes depicted as **20** [53]. Among non-standard aldehyde moieties, the ferrocenyl **21** [5, 54] and *p*-quinone containing **22** [55–57] fragments should be noted.

21: R =

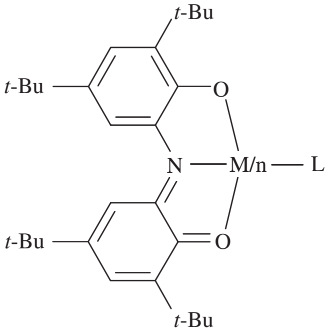


M = Pt, Pd, Ir, Ru; L = PR^1_3; R^1 = Et, *n*-Bu, Ph

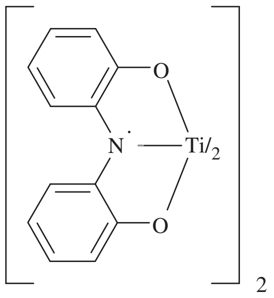


22: X = NTos, O, S; R = H, Me, OMe, Cl, Br, NO_2, 2,3 (5,6) benzo; M = Zn, Cd, Hg

A series of tricyclic complexes **23** was synthesized and structurally characterized [57–60].



23: M = Al, In, Ga, Ti, Ge, Sn, Co, V, Cu, Zn, Ln; n = 1–4; L = none, Py

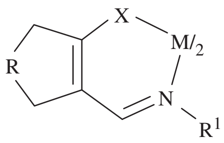


24

The chelates **24** of Al(III) and Ga(III), including anion radical fragments, are paramagnetic [58]. A ferromagnetic exchange is typical for similar complexes of Ti(IV), Ge(IV) and Sn(IV) [57, 60]. Chelates of sulfur-containing Schiff bases of type **1** ($X = S$) are discussed in a monograph [2] and reviews [5, 10]. For instance, a structurally characterized cobalt(III) complex [CoL_2Cl] [39] is of interest as a model of nitrilehydratase ($HL = $ N-2-mercaptophenyl-2′-pyridylmethylenimine).

3. Metal chelates with six-membered metallocycles

Such coordination compounds are mainly represented by the metal-chelate structure **25**.

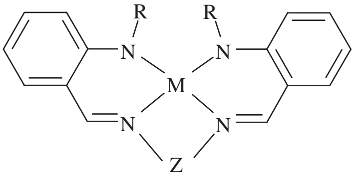


25: R = cyclo-C_4H_4, C_6H_6, Het; R^1 = Alk, Ar, Het; X = NR^2, O, S, Se; R^2 = H, Alk, Tos;

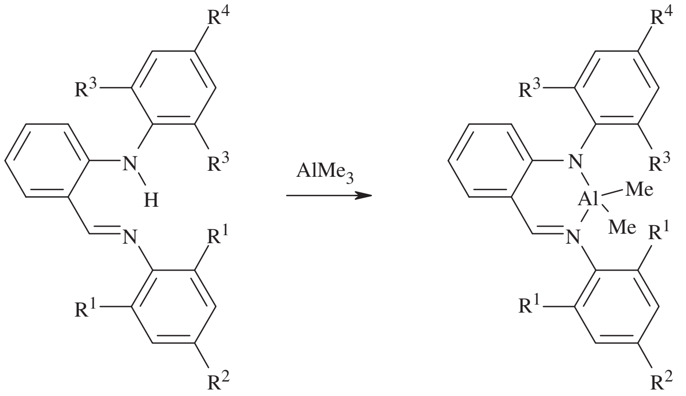
These complexes contain the aromatic aldehyde fragments (R), oxygen donor atoms (X) and coordinatively active or non-active N-substituents (R^1). Despite the fact that a majority of Schiff-base complexes incorporating six-membered metallocycles are represented by N,O-ligand environment [1–11], the metal-chelates with MN_n and $MN_nS(Se)_m$ coordination sphere are of importance. One of the first examples concerning the synthesis of bicyclic complexes of type **25** ($R = $ cyclo-C_4H_4, $X = NTos$, $R^1 = Ar$) has been reported [61].

3.1. *Nitrogen and nitrogen-chalcogen donors*

Tricyclic chelates **26** (R = H) have been described [62]. A series of similar complexes **25** and **26** were mentioned in a monograph [2] and reviews [5, 6]. Among them a series of complexes of type **26** (R = Tos; M = Ni(II) [63, 64], Cu(II) [65] and Zn(II) [66]) were described as well as recently synthesized and structurally characterized dialkylaluminum complexes with *N*,*N*-donor azomethine ligands **27** [67].

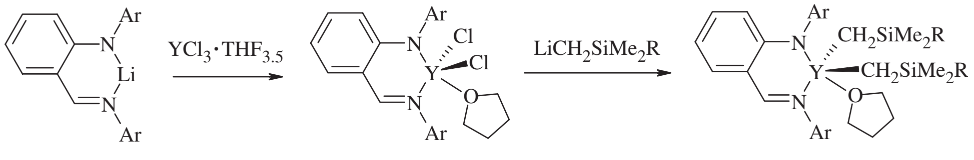


26: R = H, Alk, Tos;
Z = $(CH_2)_n$



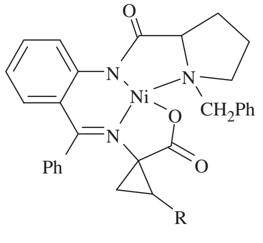
27: $R^1 = R^3 = i\text{-Pr}, R^2 = R^4 = H$ (a);
$R^1 = R^3 = Me, R^2 = R^4 = H$ (b);
$R^1 = i\text{-Pr}, R^2 = R^3 = H, R^4 = Me$ (c);
$R^1 = R^3 = H, R^2 = R^4 = Me$ (d)

A series of yttrium THF adducts based on similar ligand systems in which chloride **28** or methylsiliconhydrocarbon **29** fragments are bounded with the metallacenter [68] was isolated.

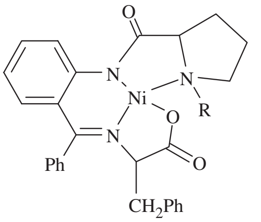


28 **29**

The azomethine moiety constitutes a part of the N_3O-tetradentate Schiff base forming tricyclic structures **30** [69, 70].

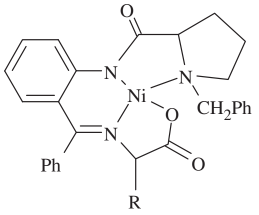


30: R = NO_2, OCOEt

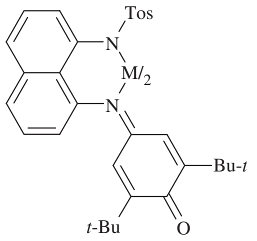


31: R = Ph, CH_2Ph, CH_2Py

Analogous structural features are characteristic for complexes of type **31** [71–73] and **32** [74].

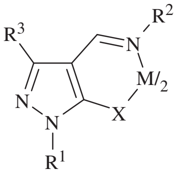


32: R = $CH_2CBrOCOEt$, Ph, NHC_6H_4OCOMe-*p*

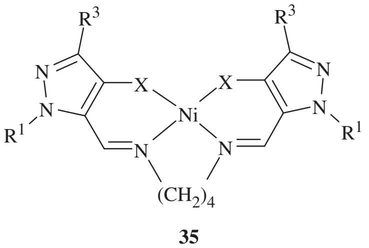


33

Among non-standard structures of complexes of Schiff bases with MN_4 coordination, the metal-chelates obtained from azomethines of mono-N-tosylated 1,8-naphthylenediamine and *p*-quinone **33** [75], 4-imino-5-aminopyrazole **34** (X = NH) [76] and **35** (X = NH) [77], as well as 7-formylindole **36** [78] should be emphasized.



34: X = NR, O, S, Se



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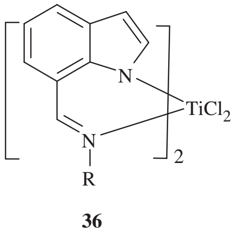
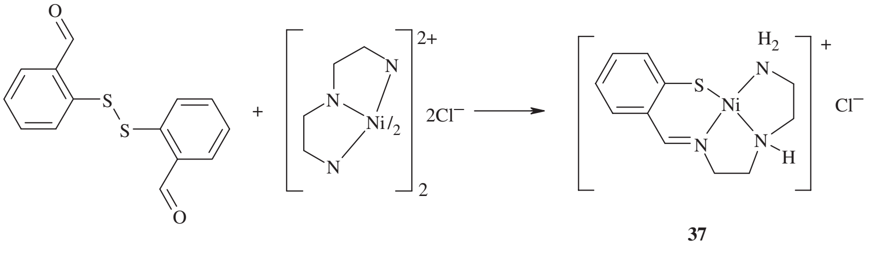


36

Chalcogen-nitrogen ligand environments occur in a number of coordination compounds [2, 5, 6, 10, 43]; we note template syntheses leading to the formation of metal-chelates of the type MN_mS_n **37** [79] and **38** [80, 81].



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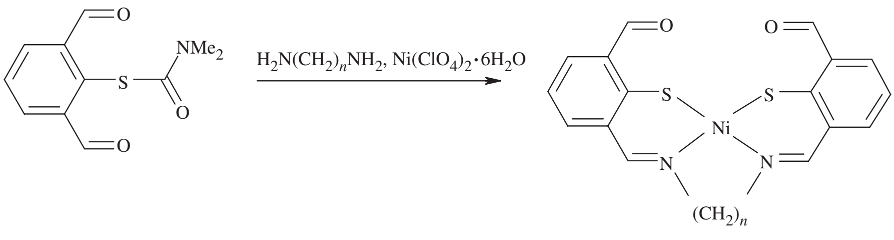

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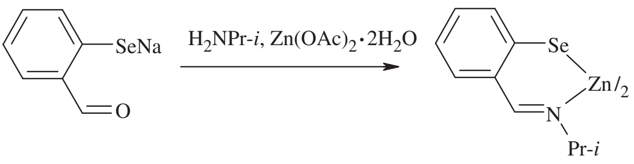
Complexes of selenium- and tellurium-containing aromatic Schiff bases are represented by sporadic structures **39** [5, 10, 82] and **40** [5, 83].



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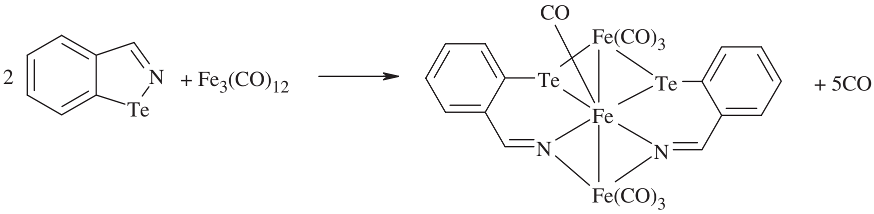

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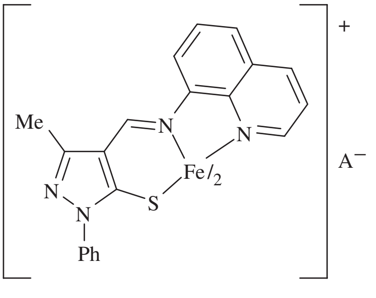
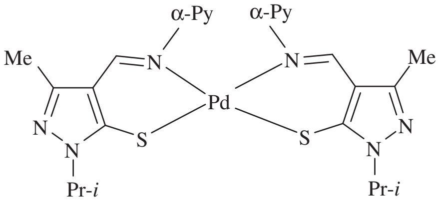
In contrast, a number of S- and Se-containing azomethine metal-chelates are observed among complexes of heterocyclic Schiff bases [1, 2, 5, 6, 10], including the unusual cationic complexes of iron **41** [84], as well as *cis*- (**42**) and *trans*- (**43**) structures of palladium chelates, obtained from imine derivatives of 5-thiopyrazole with coordinative **42** [85] and sterically hindered **43** [86] substituents at nitrogen atom of the C=N bond.



41: A = $FeCl_4$, ClO_4



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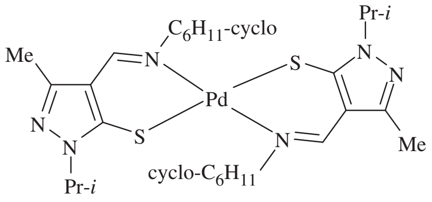
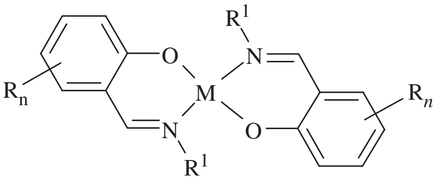


43

Analogous copper complexes are of interest as biomimetic models of active centers of "blue" copper proteins [22, 87–90 and references therein].

3.2. *N,O-donor centers*

Interest in salicylideneimine complexes and their derivatives continues into the present century. Thus, a series of complexes of type **44** with widely varying substituents R and R^1, as well as metallocenters, has been obtained.

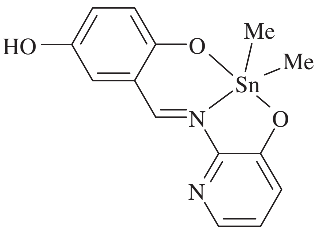


44: *n* = 1–2

A series of complexes of type **44** were structurally characterized (when $R = H$) for Co(II) ($R^1 = CH_2Ph$) [91], Fe(II) ($R^1 = i$-Pr) [92], Cu(II) ($R^1 = Ph$ [93], *p*-Tol [94, 95], PhCHMe [96]), Zn(II) ($R^1 =$ cyclohexyl [97], and CH_2Ph [98]). Metal chelates of similar type when the aldehyde fragment contains: $R = Me$, $R^1 = i$-Pr, $M = Zn(II)$ [99]; $R = t$-Bu, $R^1 = Me$, $M = Cu(II)$ [100], Zn(II) [101, 102]; $R^1 = i$-Pr, cyclohexyl [96], $R^1 = H$, $R = Me$, *t*-Bu, $M = Co(II)$ [103]; R, $R^1 = t$-Bu $M = Cu(II)$ [96] were synthesized and studied by X-ray diffraction.

R(N)-Substituted complexes are represented by *bis*-chelates **44** with diethylamino ($R = NEt_2$) [104] and nitro ($R = NO_2$) [105–107] substituents. The former include Co(II), Cu(II) ($R^1 = C_6H_4NO_2$-*p* [104]) complexes and the latter are represented by coordination compounds of Co(II) ($R^1 =$ cyclohexyl) [105] and Zn(II) ($R^1 =$ cyclohexyl [106], PhChMe [107]).

R(O)-Substituted coordination compounds are well known and represented by *bis*-chelates of Schiff bases with hydroxy [108] and alkoxy [109–112] substituents. Additionally, metal chelates of type **44** ($M = Zn(II)$, $R = OEt$, $R^1 = PhCHMe$ [110], $R = OMe$, $R^1 = PhCHMe$ [107], $R^1 = CH_2CH_2OH$ [111]) and compounds **45** [108] and **46** [109] were described and structurally characterized.



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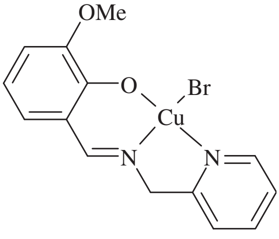

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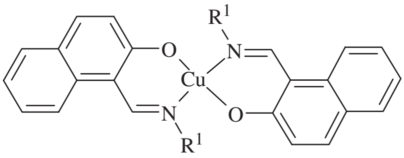
Schiff bases with naphthalene **47** ($R^1 = HCEt_2, C_8H_9CHMe$) [96, 112] and phenylanthracene **48** [113] are known.



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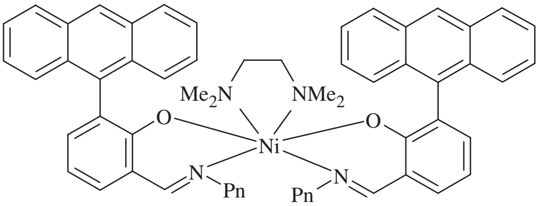

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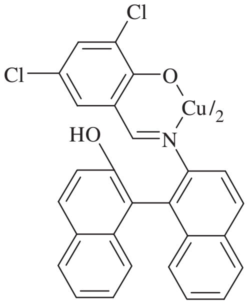

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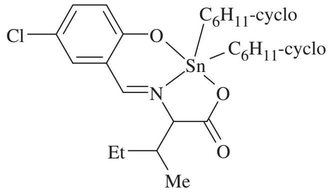
An important place among Schiff-base metal chelates is occupied by complexes of arylhalogensubstituted salicylideneimines – monosubstituted ones ($R = Cl, Br$; $n = 1$) are represented by chloro- and bromo-derivatives **44** ($M = Cu(II)$, $R^1 = PhCHMe$ [110], $R^1 =$ *i*-Pr [114], $R^1 =$ cyclohexyl [115], $R^1 = CH_2CH_2OH$ [116]; $M = Zn(II)$, $R =$ PhCHMe [107], PhCHEt [119], $R^1 =$ cyclohexyl [117]; $M = Ni(II)$, $R^1 =$ *i*-Pr, $R^1 = PhCHMe$ [118]).

Similar coordination compounds **44** ($R = Cl, n = 2, M = Ni(II)$, $R^1 =$ *i*-Pr, PhCHMe [118]; $M = Cu(II)$, $R^1 = PhCHMe$ [110]; $M = Zn(II)$, $R^1 = PhCHMe$ [107], PhCHEt [119]; $M = Cu(II)$, $R^1 = 2'$-hydroxy-1,1′-binaphthyl **49** [120]) are known for dichlorosubstituted ligands. For monochlorosubstituted salicylaldehyde complex **50** [121], as well as *bis*-chelate **51** [122], were reported. Structurally characterized *bis*-chelates of type **44** with $R = Br$ are represented by complexes of Ni(II) ($R^1 =$ cyclohexyl [123, 124]); Cu(II) ($R^1 =$ cyclohexyl [125], MeCHEt [126], MeCHPh [126]); Zn(II) ($R^1 =$ cyclohexyl [127]).



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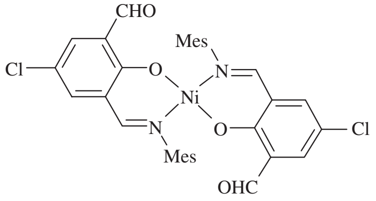
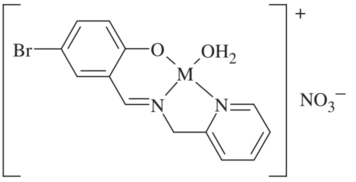
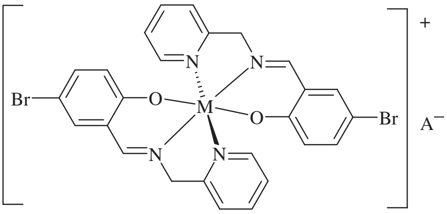


51

In addition to the complexes above, tetracoordinated compounds **52**, containing coordinatively active pyridine substituents at the nitrogens of the azomethine fragments (M = Cu(II) [128] and M = Zn(II) [129]), are reported.

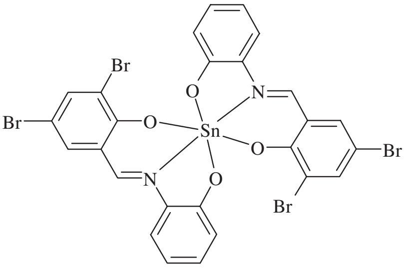


52: M = Cu, Zn



53: M = Fe(III), Co(III); A= ClO_4, NO_3

Hexacoordinated chelates are formed by dibromo-substituted (in an aldehyde moiety) azomethines and O-donor fragments **55** [130, 131]. Coordination compounds with phenolic N-substituent **54** [28] and adducts with 1,10-phenanthroline, containing a mono-O-coordinated ethanosulfonate fragment **55** (M = Ni(II) [132], M = Zn(II) [133], belong to this type of metal chelate.



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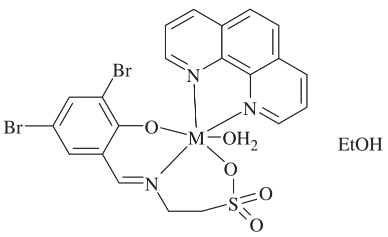
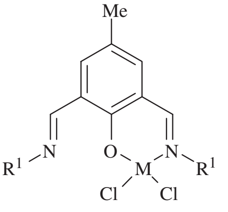
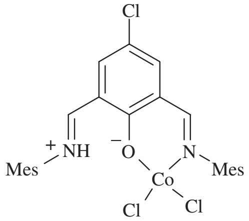


55: M = Ni, Zn

A series of coordination compounds incorporating *bis*-azomethine ligands **56** (M = Co(II), R^1 = Mesyl [134]; Zn(II), R^1 = MeCHPh [135]), **57** [136] and **58** [137] are characterized structurally.



56: M = Co, Zn;
R^1 = Mes, CH(Me)Ph



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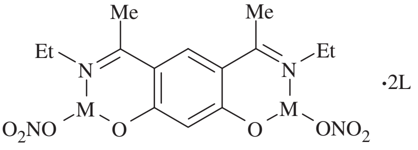
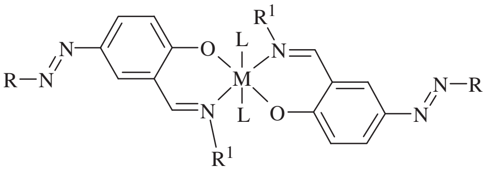


58: M = Cu, UO_2; L = bipy, 1,10-phen

A few structures of Schiff-base complexes with aldehyde comprising azo-fragments **59** (M = Ni, R = –4-$C_6H_4OCH_2Et$, R^1 = *n*-C_5H_{11}, L = none [138]; M = Cu, R = Ph, R^1-R^1 = –$(CH_2)_3$–, L = none [139a], M = Ni, Cu; R = Ph, R^1-R^1 = –*o*-$C_6H_4O(CH_2)_4OC_6H_4$-*o*–, L = none [139b], M = Ni, R = 4-Py, R^1 = $(CH_2)_{10}$Et; L = Py [139c]) and **60** M = Ni [140a], M = Cu [140b] are described.



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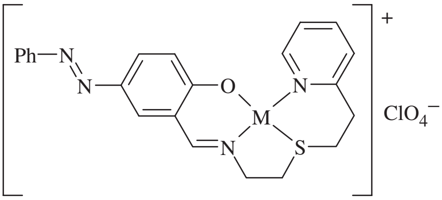
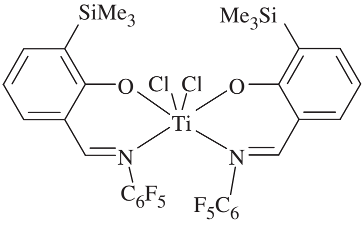


60

The structures of Ti(IV) complex including an aldehyde fragment containing $SiMe_3$-groups and pentafluorophenyl substituents at the nitrogen of C=N groups **61** [141] are unusual.



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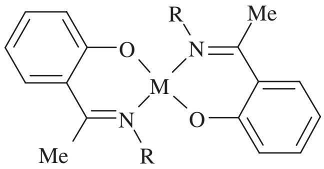
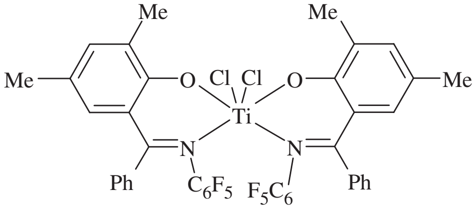


62: M = Ni, Pd; R = 2,4-di-*i*-PrC_6H_3, Mes

In addition to aldimine derivatives, chelates obtained from keto-imines, e.g. compounds of type **62** (M = Ni(II), R = 2,6-di-*i*-propylphenyl [142], M = Pd(II), R = Me, R^1 = Mesyl [143]) are known. Similar structure is attributed to a *bis*-chelate of Ti(IV) **63**, including a $N_2O_2Cl_2$ ligand environment [144].



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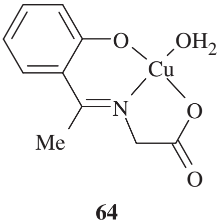

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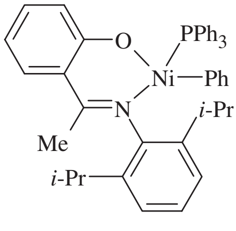
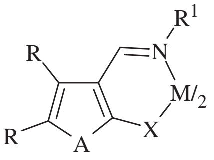
Enol-imine structures with a coordinatively active O-substituent **64** [145] and organometallic center **65** [142] are of interest.



65

Complexes derived from aniles based on various carbonyl functionalized heterocyclic compounds are widely represented [1, 2, 4–6, 10]. Among coordination compounds incorporating a pyrazole fragment **34**, **35**, **41**–**43**, complexes of five-membered fundamental heterocycles (pyrrol, furan and thiophene) **66**, **67** have been reported [5, 6, 10, 25]. This set of donor centers forms the coordination sphere in complexes including exotic aldehyde moeties **66**–**70**.



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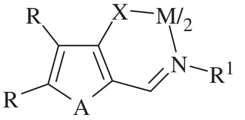
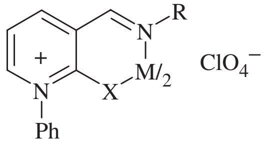


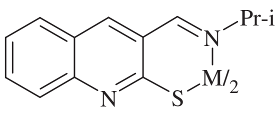
67

A = NR^2, O, S; R = Alk, Ar, C_4H_4-cyclo

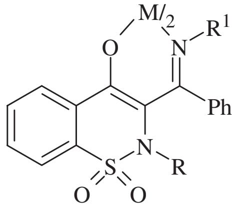


68: X = O, S; M = Cu, Zn;
R = $(CH_2)_n$, Ar

Schiff-base metal chelates, including six-membered heterocyclic systems, are described for azines **68** and **69** [10, 25, 146] and their thiodioxide-derivatives **70** [147–149]. Structures of **70** (R = Et, R^1 = antipyrine) [147, 148] were unambiguously proved by X-ray diffraction data.



69: M = Zn, Cd

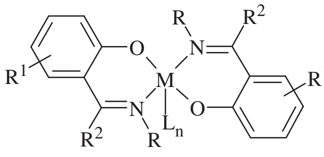


70: R = Alk; R^1 = Ar, Antipyrine;
M = Co, Ni, Cu, Zn

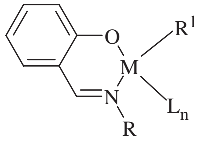
Complexes **70** have been prepared using traditional chemical (starting from acetate of corresponding metals) and electrochemical procedures.

3.3. *Amine components*

3.3.1. Coordinatively non-active substituents. Coordination compounds with coordinatively non-active substituents at the N-atom of the imine bond are represented by metal-chelates **71** and **72** with saturated acyclic and cyclic hydrocarbons and their derivatives, as well as aromatic fragments.



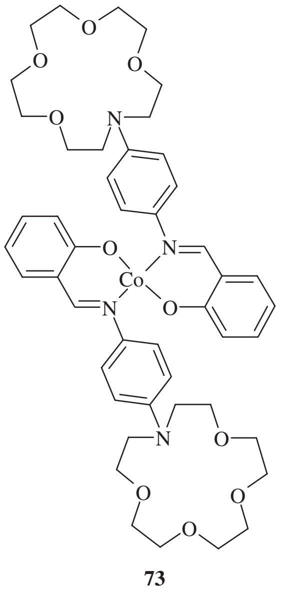
71: L = none, Cl



72: L = Py, PPh$_3$; R^1 = Me, Ph.

In a series of structurally characterized complexes, R-substituents (R^1 = R^2 = H and L = none) **71** such as alkyl (e.g., n-propyl [M = Zn(II)] [150], allyl [M = Pd(II)] [151], chloropropyl [M = Ni(II)] [152] and methylphenyl [M = Cu(II)] [153] are described. The series of similar *bis*-chelates of aromatic amines **71** are widely represented: R = C_6H_4Me-*p*, R^1 = H, M = Cu(II) [154]; R = C_6H_4Me-*p*, R^1 = OMe-6, M = Co(II) [155]; R = $C_6H_3(CH_3)_2$-3,5, R^1 = H, M = Cu(II) [156]; R = C_6H_3(*i*-C_3H_7)$_2$-2,6, R^1 = H, M = Ni(II) [113]; R = C_6H_4(HC = CH_2)-*p*, R^1 = H, M = Pd(II) [151]; C_6H_4(COMe)-*p*, R^1 = H, M = Cu(II) [157]; R = C_6H_4(NMe$_2$)-*p*, R^1 = H, M = Zn(II) [158]; R = C_6H_4(OMe)-*p*, R^1 = H, M = Cu(II) [159]; R = C_6H_4(F)-*p*, R^1 = OMe-3, M = Cu(II) [160].

A few complexes with exotic fragments in N–Ar–R-substituents are known, e.g. 4-(1-aza-15-crown-5, R = H) **73** [161, 162], 4-(benzenesulfonamide) **74** [163] and 4-(4,4,5,5-tetramethyl(1.3.2-dioxoboran) **75** [164].



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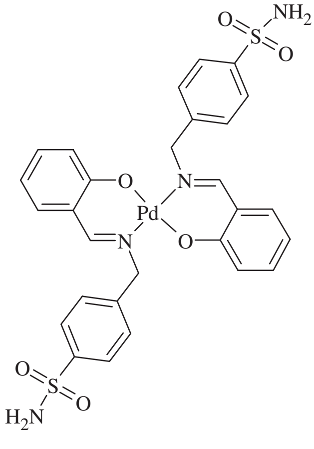

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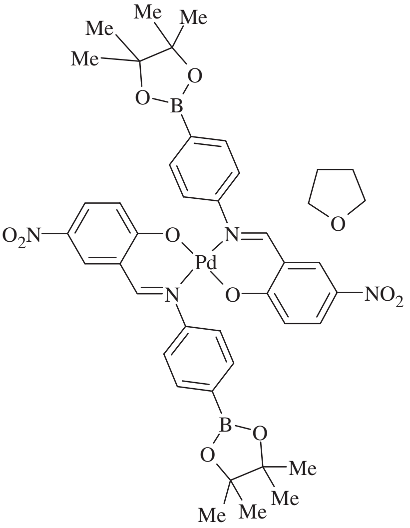

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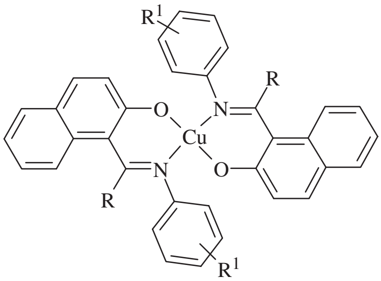

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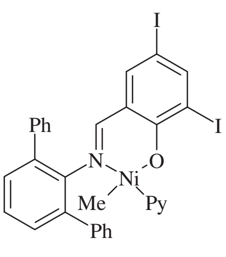
Complexes of naphthalenecarbonyl derivatives **76** ($R = H$, $R^1 =$ iodo-2 [165]; $R = Me$, $R^1 = (i\text{-}C_3H_7)_2$ [142]) were reported.

Among the compounds of type **71** ($L = Cl_n$) with N-aryl substituents are the monochloride aluminum ($R = C_6H_3(i\text{-}C_3H_7)_2$, $R = 4\text{-Me}$, $M = Al(III)$, $n = 1$ [166]) and dichloride titanium(IV) ($R =$ allyl, $R^1 = 6\text{-Ph}$, $M = Ti(IV)$, $n = 2$ [167], $R = CF_5$, $R^1 = H$, Me, *i*-Pr, *t*-Bu, $SiMe_3$, $SiEt_3$ [142, 168], $R = CF_5$, $R^1 =$ Me, Et, *i*-Pr, *t*-Bu, $R^2 =$ H, Ph, $M = Ti(IV)$, $n = 2$ [145]) complexes. These complexes are of interest as catalysts [168].

Metal chelates of type **71** with aryl substituents at nitrogen of C=N are classic in coordination chemistry [1, 2, 5, 6], but data on similar complexes incorporating cycloalkane fragments are scarce. Among them, salicylideneiminate structures ($L =$ none) with N-cyclopropyl ($M = Mn(II)$ [169], Cu(II) [170], Zn(II) [171–174], cyclobutyl ($M = Cu(II)$ [170]) and cyclohexyl ($M = Co(II)$ [175]) moieties are known. Cyclopropyl and cyclohexyl fragments were introduced into complexes ($M = Cu(II)$ of 3-hydroxy-2-naphthylimine [170].

Complexes of type **72** are represented by a limited number of complexes containing methyl and pyridine, **77** and **78** [176], or phenyl and phosphine **79** [177] fragments at nickel.



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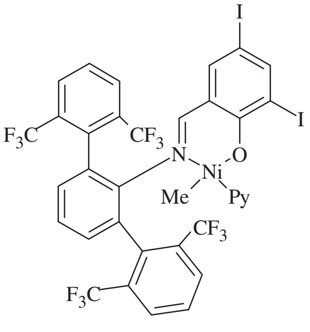

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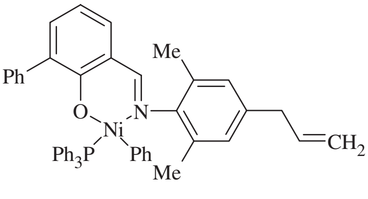

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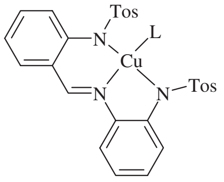
3.3.2. Coordinatively active substituents. Coordination compounds of Schiff bases with coordinatively active fragments at nitrogen of C=N– are systematized taking into account the types of additional donor centers (N, P, O, S), their number, electronic configuration of metals and character of aldehyde moiety. Aspects of this are examined in reviews [5, 6] and monograph [2].

3.3.2.1. *N-containing substituents.* Compounds of type **80** ($L = H_2O$ [178], L = 2-aminopyridine [179]) of *o*-tosylaminobenzaldehyde and mono-N-tosylated *o*-phenylenediamine were obtained and structurally characterized.



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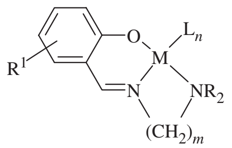
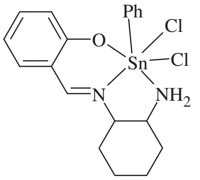


81

Most of the examined chelates have been synthesized from aniles of salicylaldehyde. A series of complexes with alkylmonoamine fragments of type **81** increasing the coordination number of the metallacenter by one, includes compounds of the following metals: Al(III) ($R = Me, R^1 = \text{di-}t\text{-Bu}, L = Me, m = 1, n = 2$ [180]); Ni(II) ($R = Me, R^1 = 1,2\text{-cyclo-}C_4H_4, L = NCS, m = 1$ [181], $R = Et, R^1 = 2,4\text{-}di\text{-}Cl, L = N_3, m = n = 1$ [182]); Cu(II) ($R = R^1 = Me, R^1 = Cl, L = OCOMe$, $m = n = 1$ [183], $R = Me, R^1 = 2,4\text{-}di\text{-}Br, L = NCS, m = n = 1$ [184], $R = Et, R^1 = H, L = N_3, m = n = 1$ [185], $R = Et, R^1 = 2,4\text{-}di\text{-}Cl$, $L = N_3, m = n = 1$ [186]) и Zn(II) ($R = Me, R^1 = 3,5\text{-}di\text{-}t\text{-Bu}$, $L = Cl$, $L^1 = MeOH$, $n = 1$ [187]). Amine fragments are included in cyclohexyl **82** [188] and ethylenedinitrile **83** [189] substituents.



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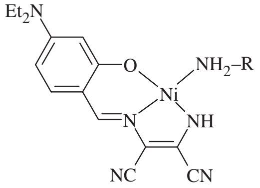
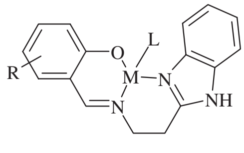
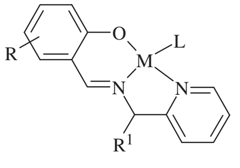


83

Nitrogens containing heterocycles, azoles and azines are involved in Schiff-base chelates as mono-N-donor fragments [1, 5, 25]. In particular, complexes **84** of Cu(II) (R = 4-Br, L = NCS [190]; $R = 5\text{-}NO_2$, L = Cl [191]) and Zn(II) (R = 4-Br, L = Cl [192]) include benzimidazole substituents.

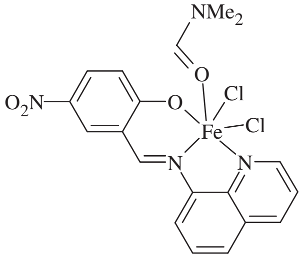


84: M = Cu(II), Zn(II)

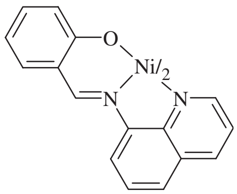


85

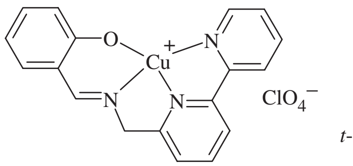
Azine-containing Schiff bases form metal chelates **85** where pyridine substituent is coordinated with various metals: Al(III) ($R = 3,5$-*di*-*t*-Bu, $R^1 = H, L = Me_2$ [180]), Fe(III) ($R = R^1 = H, L = 2Cl$-MeOH [193]), Cu(II) ($R = 4$-$NO_2, R^1 = H, L =$ NCS-MeOH [194]; $R = R^1 = H, L = H_2O$ [195]; $R = 3$-OMe, $R^1 = H, L = Cl$ [109], R = cyclo-C_4H_4, $R^1 = H, L = ONO_2$ [196], $R = H, R^1 = Py, L = Cl$ [197]) and Zn(II) ($R = 4$-NO_2, $R^1 = H, L =$ NCS-MeOH [198], $R = R^1 = H, L = H_2O$ [199], R = cyclo-C_4H_4, $R^1 = H$, $L = ONO_2$ [200]). Similar complexes bearing 8-aminoquinoline **86** [201], **87** [202] and 2,2-bipyridine N-substituents **88** [203] are known.



86



87



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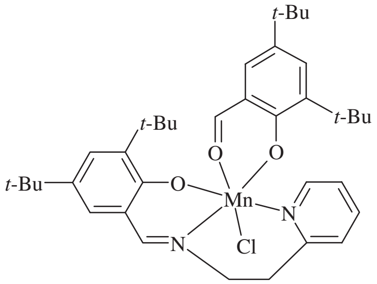
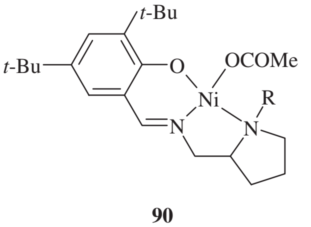


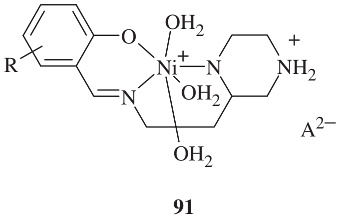
89

Pyridine substituent is coordinated with Mn(III) in the complex of type **89** [204] comprising a mixed ligand (salicylaliminato-salicylaldehydato) environment. In contrast, in a series of salicylidene bis-chelates where nitrogen of azine heterocycles directly bond with the C=N-fragment, it does not bind with the metallacenter [2, 5, 6, 205, 206].

Additional coordination with a metallacenter is characteristic for complexes containing hydrogenated five- (pyrrolidine **90**: $R = CH_2C_6H_5$, 1-$CH_2C_{10}H_7$ [207]) and six-membered nitrogen heterocycles, e.g. piperazine **91** ($R = H, NO_2, Br$; $A = SO_4, 2BPh_4$) [208].

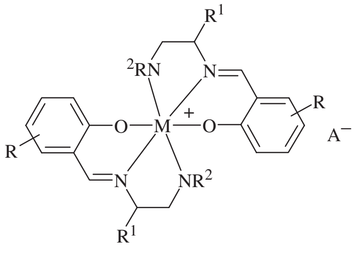


90

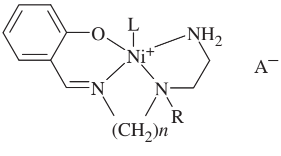


91

Monoamine substituents increase coordination number by two in complexes of type **92**: Co(II) ($R = R^1 = H, R^2 = CH_2CH_2OH, A = NO_3^-$ [209], $R = R^1 = H$, $R^2 = CH_2CH_2OH$, $A = Cl^-$ [210]) and Fe(III) ($R = 3\text{-OMe}, R^1 = Me, R^2 = H, A = PF_6^-$ [211], $R = 5\text{-Br}, R^1 = H, R^2 = Et, A = PF_6^-$ [130]).

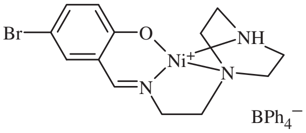


92: M = Co, Fe

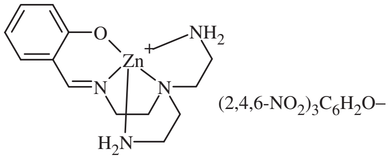


93

The increase in coordination number by two units took place in complexes of type **93** as a result of coordination of N-substituents possessing two amine fragments: Ni(II) ($R = H, L = 0, X = NH, n = 1, A = ClO_4^-$ [211], $R = Me, L = 0, X = NH, n = 2, A = I^-$ [211]; Cu(II) ($R = H, L = H_2O, X = O, n = 1, A = Cl^-$ [212–214]). A similar situation is observed in the metal chelate **94** [208].

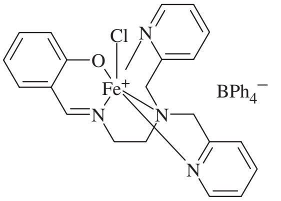


94

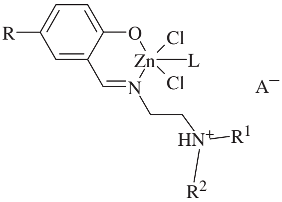


95

Augmentation of coordination number by three is characteristic for complexes of types **95** [215] and **96** [216], which include three donor N-atoms in the substituent at the azomethine nitrogen.

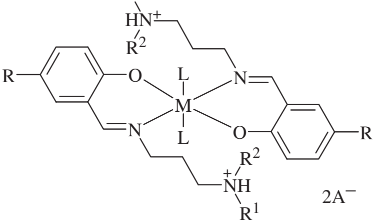


96



97

Quaternized aminofragments do not take part in additional coordination as observed in complexes of types **97** and **98**.



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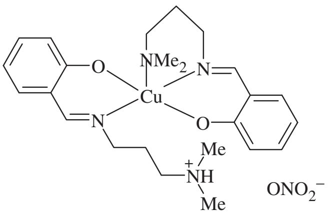
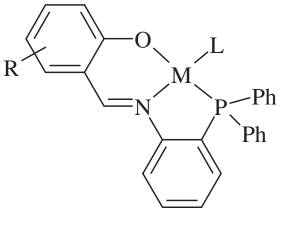


99

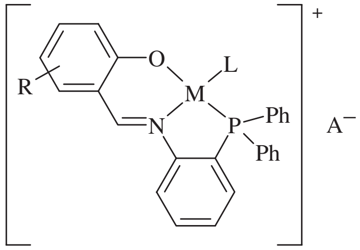
Metal chelates of type **97** are represented only by complexes of Zn(II) ($R = R^1 = H$, $R^2 = CH_2CH_2OH, L = none$ [217], $R = 4\text{-}NO_2, R^1 = R^2 = Me, L = none$ [218], $R = 4\text{-}NO_2$, $R^1 = R^2 = Me$, $L = H_2O$ [218], $R = 2,4\text{-}di\text{-}Cl$, $R^1 = R^2 = Me$, $L = none$ [219]).

A wider number of metals is reported for *bis*-chelates **98**: Co(II) ($R = R^1 = H$, $R^2 = cyclohexyl$, $A = NCS, L = none$ [220]), Ni(II) ($R = R^1 = H, R^2 = cyclohexyl$, $A = NO_3$, $L = none$ [221], $R = OH, R^1 = R^2 = Me, A = Cl, L = none$ [222], $R = R^1 = H$, $R^2 = cyclohexyl$, $A = N_3, L = none$ [223], $R = R^1 = H, R^2 = cyclohexyl$, $A = NCS$, $L = none$ [224], $R = NO_2, R^1 = H, R^2 = cyclohexyl$, $A = NO_3, L = MeOH$ [225], $R = C_4H_4, R^1 = H, R^2 = cyclohexyl$, $A = Cl^-, L = H_2O$ [226]) and Cu(II) ($R = H$, $R^1 = R^2 = Me, A = ClO_4^-, L = none$ [227], $R = C_4H_4, R^1 = H, R^2 = cyclohexyl$, $A = Cl^-$, $L = H_2O$ [228]). Thus, in the presence of nonquaternized and quaternized N-substituents only nonquaternized are coordinated **99** [229].

3.3.2.2. *P-containing substituents.* In contrast to similar compounds with N-donors (Section 3.3.2.1), metal chelates of Schiff bases incorporating phosphorus-containing donor substituents are represented by a limited number of palladium and platinum complexes **100** and **101** [230, 231].

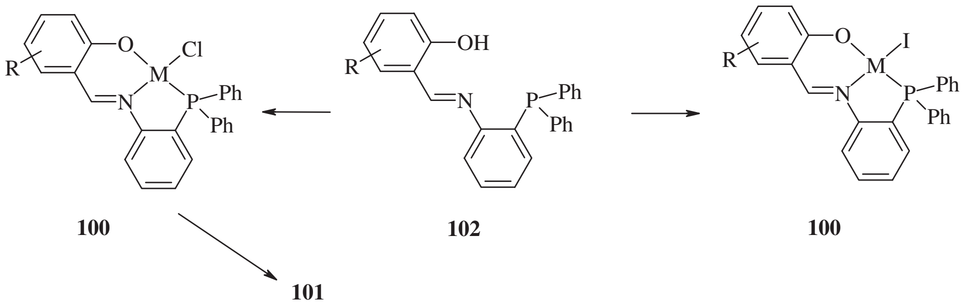


100: M = Pd, Pt;
L = Me, Cl, I



101: M = Pd(II), Pt(II)
$L = PPh_3, P(p\text{-}Tol)_3, P(CH_2CH_2CN)_3$

Synthesis of ligands **102** is described [232, 233], while coordination compounds are synthesized according to the following scheme [231]:

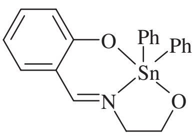


100 **102** **100**

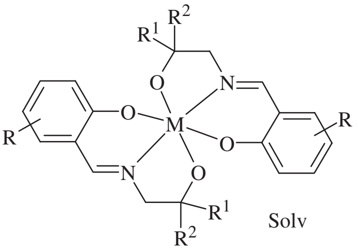
101

The complexes were characterized by IR and ^{31}P NMR spectroscopies and the structures of some complexes were established by X-ray analysis, e.g. **100** ($R = H, L = Me$ [230]; $R = 4\text{-}OMe, L = Cl$ [231]; $R = 4\text{-}OMe, L = I$ [231]; $R = 4\text{-}Br$, $L = Cl$ [231]), **101** ($R = 4\text{-}Cl, L = Pp\text{-}Tol_3, A = ClO_4^-$ [231]; $R = 4\text{-}Br$, $L = PPh_3$, $A = ClO_4^-$ [231]).

3.3.2.3. *O-containing substituents.* Among coordination compounds with O-containing substituents, complexes comprising alcohol fragments at N-atom of C=N are the most common and include structures of several types: with deprotonated O-coordinated alcohol of types **103–105**; non-deprotonated O-coordinated alcohol substituent of types **106–107**; with non-coordinated alcohol fragments of type **108**; mixed-coordinated ones of type **109**, including coordinated and non-coordinated, non-deprotonated alcohol O-substituents. Recent references include complexes with O-coordinated metal chelates of types **103** [234] and **104** [235–240].



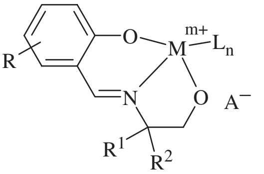
103



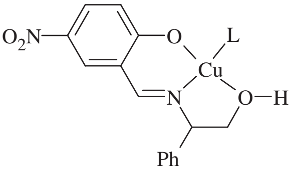
104

Chelates of type **104** are represented by the following compounds: Ti(IV) ($R = R^1 = R^2 = H$, Solv = none [235]; $R = H, R^1 = R^2 = Me$, Solv = none [236]), Mn(IV) ($R = R^1 = R^2 = H$, Solv = MeCN [237]; $R = R^1 = H, R^2 = Ph$, Solv = none [238]; $R = R^1 = H, R^2 = CH_2Ph$, Solv = DMF [239]; $R = C_4H_4, R^1 = R^2 = H$, Solv = none [240]).

A series of complexes are similar to the cationic chelate depicted as **105** ($M = Ni$, $R = H, R^1 = Me, R^2 = CH_2OH, m = 1, L = Py, n = 3, A = MeCOO^-$ [241]), ($M = Cu$, $R = H, R^1 = R^2 = CH_2OH, m = 0, A = Cl, L =$ none [237]; $R = H, R^1 = R^2 = CH_2OH$, $M = A =$ none, $L = Br$ [153]) and ($M = Zn, R = R^1 = R^2 = H, m = 0, A = MeCOO^-$, $L = H_2O$ [242]).

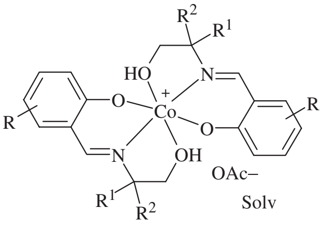


105



106

Non-deprotonated O-bonded alcohol moieties constitute a part of structures **106** ($L = Cl$ [243], Br [244]) and **107** ($R = H, C_4H_4$; $R^1 = R^2 = H, Me$ [240]).



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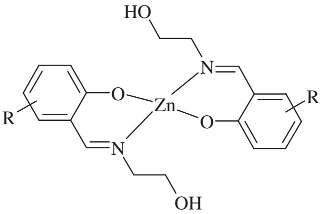
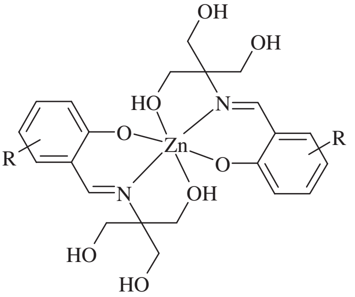


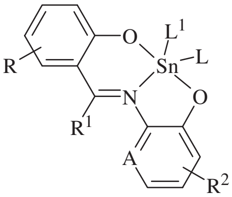
108

Alcoholiminate complexes of Schiff bases with non-coordinated hydroxyl are shown as **108** [245]. The complex including both coordinated **106** and non-coordinated **108**, non-deprotaned alcohol fragments is represented by **109** (R = H, Me, OH) [245].



109

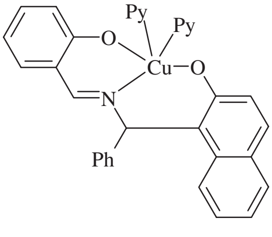
The phenolic substituents at the nitrogen of imine, unlike alcohol moieties, undergo deprotonation in all known compounds [5, 7, 246], e.g. in chelates of types **110**, **111** [247–250] and **112**–**114** [28, 251–253].



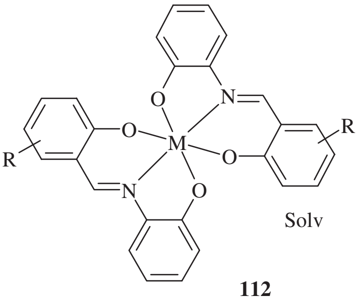
110

The structurally characterized complexes **110** (A = CH) are presented by *bis*-chelates of Sn(IV) (R = R^1 = R^2 = H, L = L^1 = *n*-Bu [247]; R = 5-Me, R^1 = R^2 = H, L = L^1 = n-Bu [247]; R = 5-OMe, R^1 = H, R^2 = 4-NO_2, L = L^1 = *n*-Bu [248]; R = 5-OMe, R^1 = H, R^2 = 4–NO_2, L = L^1 = Ph [248]).

3-Hydroxypyridyl **110** (R = H, Br; R^1 = R^2 = H, A = N) [249] and 2-hydroxynaphthyl **111** [250] substituents are phenolic components in similar complexes.

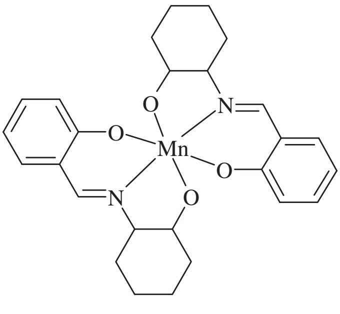


111

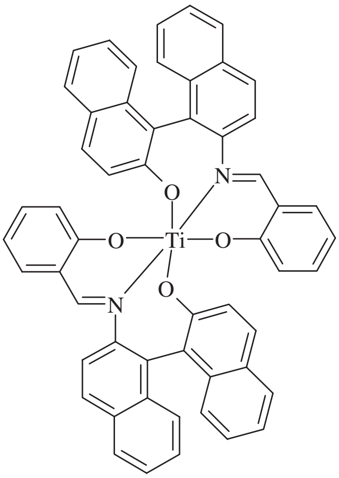


112

Complexes of Sn(IV) (R = H, Solv = MeCN [28]) and Mn(IV) (R = 5-Cl, Solv = THF [251]) of type **112** include tetra-chelate structures. The metal chelates with cyclohexanol **113** [237] and 2-hydroxy-1,1′-binaphthyl **114** [252, 253] substituents belong to the same type of compounds. The last is of interest since the coordination unit is formed by six- and seven-membered metallocycles [252, 253].

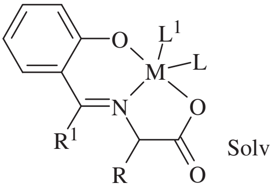


113

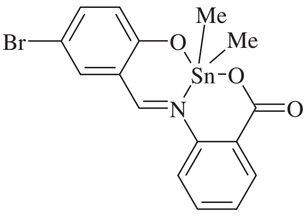


114

Schiff-base complexes obtained as the result of deprotonation of the hydroxyl fragment of organo-acid substituents at the azomethine fragment **115** [254–257], are widely represented, e.g. by a series of tin(IV) coordination compounds ($R = R^1 = H$, $L = L^1 = Ph$, Solv = none [254]; $R = H$, $R^1 = Me$, $L = L^1 = Me$, Solv = H_2O [255]; $R = Me$, $R^1 = H$, $L = L^1 =$ *n*-Bu, Solv = H_2O [255]; $R = H$, $R^1 = Me$, $L = L^1 = Ph$, Solv = none [254]; $R = H$, $R^1 =$ *i*-Pr, $L = L^1 =$ *n*-Bu, Solv = none [254]; $R = H$, $R^1 =$ *i*-Pr, $L = L^1 = Ph$, Solv = none [254, 256]; $R = H$, $R^1 =$ *sec*-Bu, $L = L^1 =$ *n*-Bu, Solv = none [254]; $R = H$, $R^1 =$ *n*-decyl, $L = L^1 = = Ph$, Solv = CH_2Cl_2 [254]; $R = H$, $R^1 = Ph$, $L = L^1 = Ph$, Solv = none [254]; $R = H$, $R^1 =$ 3-indolyl, $L = L^1 =$ *n*-Bu, Solv = none [257]).



115

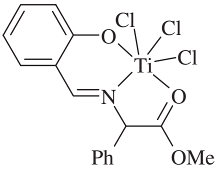


116

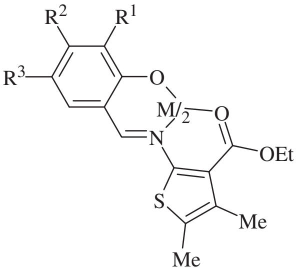
Tin(IV) complexes of type **116** [258] can also be attributed to similar complexes.

A series of copper complexes of type **115** containing various N- and O-donor bases as L and L^1 is described and structurally characterized [259–264] ($R^1 = Me$, CH_2Ph; $L\text{-}L^1 = 2,2'$-bipy, Solv = none [259, 260]; $R^1 = CH_2C_6H_4OH$-*p*, L = 4-Me-py, L^1 = none, Solv = none [261]; $R^1 = CH_2C_6H_4OH$-*p*, $L\text{-}L^1 = 2,2'$-bipy, 1,10-phen; Solv = MeOH [262]; $R^1 = CH_2Ph$, L = piperidine, L^1 = none, Solv = none [263]; $R^1 = CH_2C_6H_4OH$-*p*, L = py, $L^1 = H_2O$, Solv = none [264]).

A limited number of Schiff-base complexes where the N-substituent is coordinated through the carbonyl fragment of an organo-acid group **117** [265] and **118** [266] are known. Structures of coordination compounds of types **117** and **118** (M = Ni(II), $R^1 = R^2 = H$, $R^3 = Cl$) are proved by X-ray diffraction [265, 266].

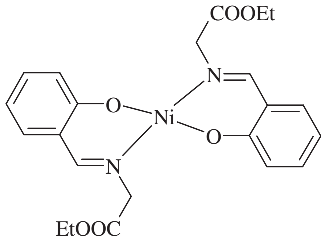


117



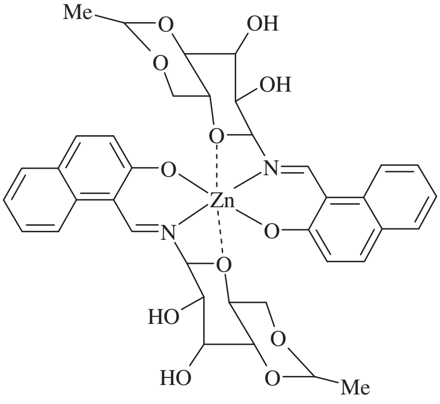
118: R^1-R^3 = H, Me, Cl;
M = Co, Ni, Cu, Zn, Pd

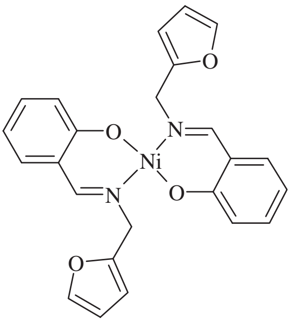
On the other hand, in *bis*-chelate **119** the carboxyl substituents do not participate in coordination with Ni(II) [267].



119

Carbohydrate functionalized Schiff bases can either bind or not bind the metallacenter through the carbohydrate oxygen [268, 269], e.g. pentacoordinated (trigonal bipiramid) and hexacoordinated (pseudo-BCT geometry) structures of type **120** [268] are examined as an example of coordinated carbohydrate fragments.



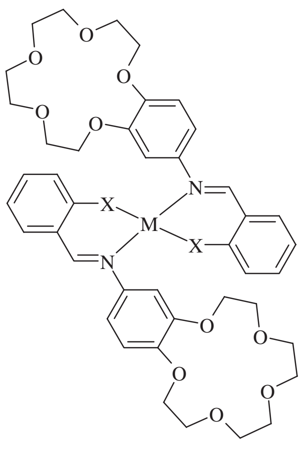


120

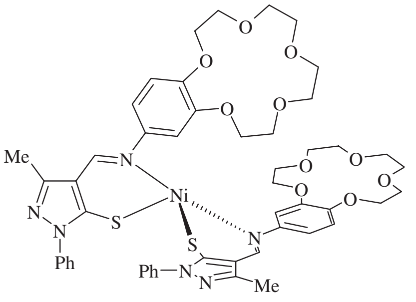
121

Zn–O distances in **120** are 2.713 and 2.703 Å [268].

The oxygen of the furyl substituent is not coordinated in a *bis*-chelate of type **121** [270]. Coordination compounds of 3*d*-metals with non-coordinated benzo-15-crown-5-oxygen fragment **122** [271–275] were described and structurally characterized as well as **123** derived from 4-amino-5-mercaptopyrazole [276].



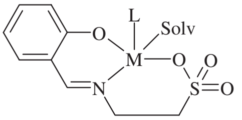
122: X = NTos, O;
M = Co, Ni, Cu, Zn



123

Coordination of Li^+, Na^+ and K^+ through oxygens of the crown-ether cavity was confirmed by ^1H and ^7Li NMR spectroscopy [277] and X-ray diffraction [275, 278].

O-coordination is characteristic for the SO_3-substituent in complexes of Ni(II) and Cu(II) **124** (L = 2, 2′-bipy, 1, 10-phen, Solv = H_2O, EtOH) [279–281], while such coordination is not observed in **125** [282].



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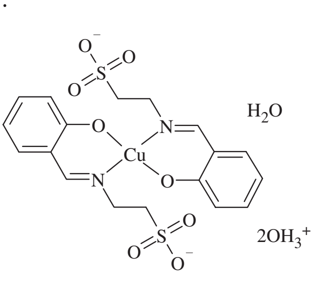
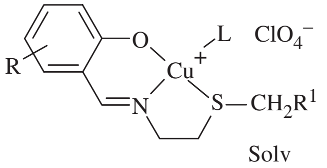


125

3.3.2.4. *S-containing substituents*. A series of Schiff-base metal chelates with S-coordinated functional groups at the N-atom of the imine has been structurally characterized, e.g. complexes with methylsulfide fragments and their derivatives **126** [283–285], thiophenol moieties **127** [286, 287], **128** [288, 289] as well as thioether **129** [290].



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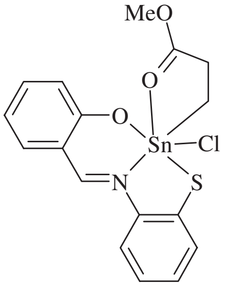
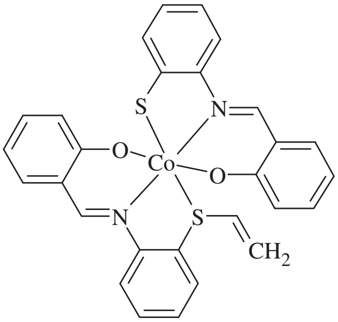


127

The following coordination compounds belong to type **126** Cu(II) ($R = R^1 = H$, $L = 2,2$-bipy, Solv = none [283]; $R = R^1 = H$, $L = 1,10$-phen, Solv = MeOH [284]; $R = R^1 = H$, 1,10-2,9-*di*-Me-phen, Solv = none [284]; $R = 3$-OMe, $R^1 = H$, $L = 1,10$-phen, Solv = none [283]; $R = 3$-OMe, $R^1 = H$, $L = 1,10$-2,9-*di*-Me-phen [283]). A metal chelate including heteroaromatic substituent **126** ($R = 5$-Br, $R^1 = CH_2$-2-Py, L = none [285]) at sulfur represents the same class of coordination compounds. 2.9-Dimethyl-1, 10-phenanthroline (R = H) [290] and 1,10-phenantrene-6,7-quinoxaline ($R = 6,7$-cyclo-C_2N_2) [290] are co-ligands in the complexes of type **129**.



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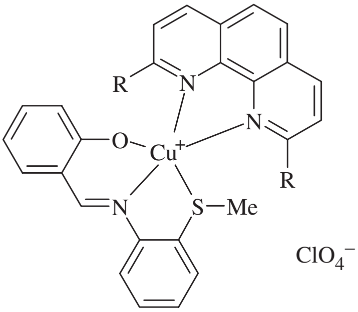
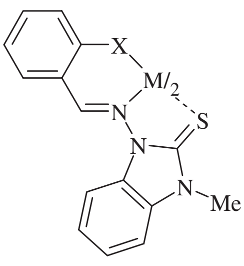


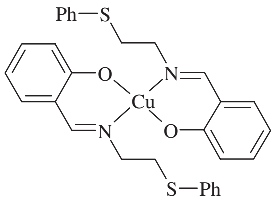
129: R = H, Me

Thione is coordinated in complexes derived from 1-amino-2-thiobenzimidazole **130** [291].



130: X = NTos, O;
M = Co, Ni, Cu, Zn, Cd

The thioether sulfur are not involved in coordination in **131** [284] and **132** [292].



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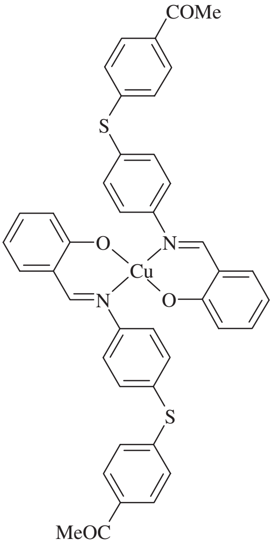
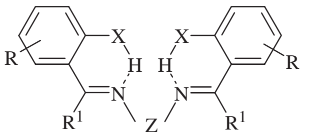


132

3.3.3. Salen complexes and their analogues. An important aspect of molecular design of the amine fragment in Schiff-base coordination compounds consists of creation of internitrogen aliphatic, aromatic or heterocyclic bridges for preparation of structures comprising various metal-chelate ring sizes and combinations of metallocycles. Such compounds are Salen complexes [1, 2, 4–8, 10, 293, 294]. Most of these compounds have been synthesized from tetradentate ligands **133** leading to tricyclic chelates of type **134**.



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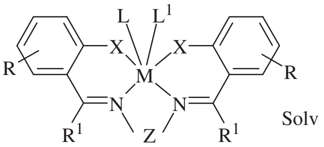


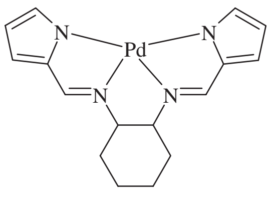
134

$X = NR^2$, O, S; $Z = (CH_2)n$, Ar; L, L^1 = none, anions, N-bases;
R = H, NO_2, Hal, C_4H_2-cyclo; R^1 = H, Alk, Ar; R^2 = H, Alk, Ar, Tos;
Solv = none, CH_2Cl_2, toluene

The N, O, S, Se and Te-containing fragments are bridges Z. This allows distinguishing in the following discussion a classical C-Salen and their analogues, aza-, oxo- and chalcogen-containing Salen complexes.

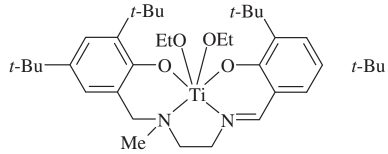
3.3.3.1. *Classic C-Salen metal chelates.* The systematization of this section takes into account the size of interazomethine bridge (n), metallocenter (M), nature of aldehyde (X, R) and amine fragments, as well as anion or co-ligand components.

Metal chelates **134** ($X = NR, n = 2$) are examined in a monograph [2], reviews [5, 6, 10] and in the present review (Section 3.1: **26, 35, 38**). Analogous to a review [293], a metal chelate of type **135**, containing two five-membered metallocycles PdN_4 [295], can be attributed to this type of complex.



135

Metal chelates of type **134** obtained from salicylaldehyde ($X = O$), ethylenediamine [$Z = (CH_2)_2$] and their derivatives are widely reflected in recent references. A great number of *p*- and *d*-metals form this type of coordination compounds, e.g. Al(III) ($R = bis$-3, 5-*di*-*tert*-Bu, $R^1 = H$, $L = OPh$, $L^1 =$ none, Solv = none [296]; $R = bis$-3, 5-di-*tert*-Bu, $R^1 = H$, $L = OMes$, $L^1 =$ none, Solv = none [296]; $R = 3$-*tert*-Bu, 5-NO_2 or 5-Cl, $R^1 = H$, $L = Et$, $L^1 =$ none, Solv = none [296]), Ga(III) ($R = bis$-3, 5-*di*-*tert*-Bu, $R^1 = H$, $L = Cl$, $L^1 =$ none, Solv = none [297]), Sn(IV) ($R = bis$-3, 5-*di*-*tert*-Bu, $R^1 = H$, $L = Cl$, $L^1 =$ none, Solv = $CHCl_3$ [298]; $R = bis$-3, 5-*di*-*tert*-Bu, $R^1 = H$, $L = L^1 = CF_3SO_3$, Solv = CH_2Cl_2 [298]; $R = 3$, 5-*di*-Cl, $R^1 = H$, $L = Cl$, $L^1 =$ none, Solv = CH_2Cl_2 [299]; $R = H$, $R^1 = Me$, $L = Cl$, $L^1 = Ph$, Solv = none [300]), Ti(IV) ($R = 3$-*tert*-Bu, $R^1 = H$, $L = L^1 = Cl$, Solv = Meptol [301]; $R = bis$-3, 5-*di*-*tert*-Bu, $R^1 = H$, $L = L^1 = Cl$, Solv = toluene [301]), as well as chelate **136** [302]. Cationic complexes of aluminum(III) of type **137** ($M = Al$, $R = bis$-3, 5-*di*-*tert*-Bu, $R = R^1 = R^2 = H$, $L = L^1 = MeOH$, $A = BF_4^-$, Solv = H_2O, MeOH [303]) were described.



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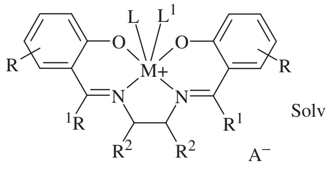
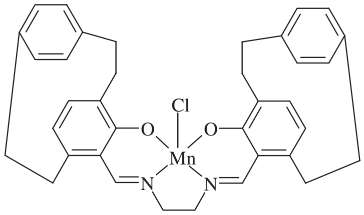


137

A series of structurally characterized complexes of type **134** is reported for Mn(III) ($Z = CH_2–CH_2$) ($R = R^1 = H$, $L = H_2O$, $L^1 = Cl$, Solv = H_2O [304, 305]; $R = Br$, $R^1 = H$, $L = H_2O$, $L^1 = H_2O$, Solv = none [306]; $R = H$, $R^1 = Me$, $L = N_3$, $L^1 =$ none, Solv = none [307]; $R = H$, $R^1 = Me$, $L = NCS$, $L^1 =$ none, Solv = none [308]; $R = H$, $R^1 = Me$, $L = Cl$, $L^1 =$ none, Solv = none [308]; $R =$ cyclo-C_4H_4, $R^1 = Me$, $L = Cl$, $L^1 =$ none [308]). Similar structure is attributed to complexes incorporating the uncommon aldehyde (R) **138** [309], **139** [310], **140** [311], amine **141** [312] and co-ligand (L) **142** [313] fragments, as well as complex **143** [314].



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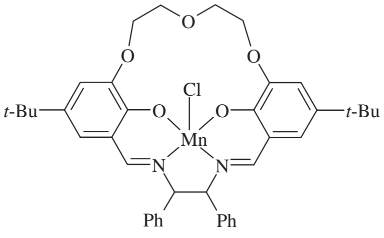

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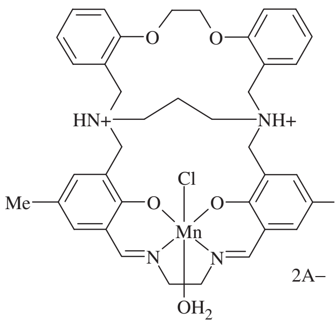

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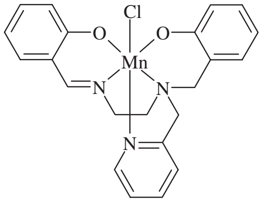

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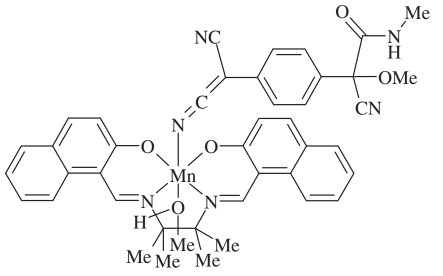

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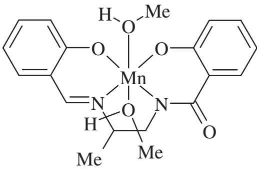
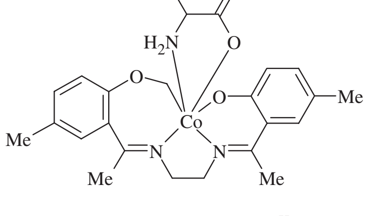


143

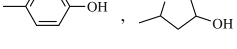
Manganese Salen chelates of type **137** are represented by complexes with varying fine structure ($R = NO_2, R^1 = R^2 = H, L = H_2O, L^1 = MeOH$, $A = ClO_4^-$, Solv = none [315]; $R = OH, R^1 = R^2 = H, L = L^1 = Me, A = Cl$ [316]; $R = 3, 5$-*di*-Cl, $R^1 = R^2 = H, L = H_2O$, $L^1 = MeOH$, $A = ClO_4^-$, Solv = none [317]; R = cyclo-C_4H_4, $R^1 = R^2 = H, L = L^1 = MeOH$, $A = ClO_4^-$, Solv = none [318]; $R = Br, R^1 = H, R^2$ = di-Me, $L = L^1 = MeOH$, $A = ClO_4^-$, Solv = none [319]).

The same structural approach is applied for iron(III) chelates of type **134** ($R =$ 5-COOH, $R^1 = H, L = Cl, L^1 = 0$, Solv = none [320]; $R = H, R^1 = Ph, L = Cl, L^1$ = none, Solv = Et_2O [321]; $R = 3, 5$-*bis*-Mes, $R^1 = H, L = Cl, L^1$ = none, Solv = MeCN [322]) and **137** ($R = 3, 5$-*bis*-Mes, $R^1 = H, R^2 = Mes, L = H_2O, L^1$ = none, $L = ClO_4^-$, Solv = CH_2Cl_2, H_2O [322]).

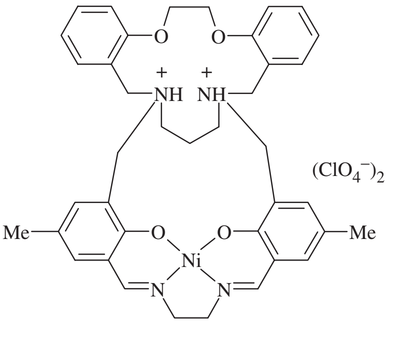
Cobalt Salen compounds are represented by complexes of type **134** (R = cyclo-$C_4H_4, R^1 = H, L = L^1$ = none, Solv = none [323]; $R = R^1 = Me, L = i$-Pr, L^1 = none, Solv = H_2O [324]) and **137** ($R = R^1 = Me, R^2 = H, L = Py, L' = H_2O, L = ClO_4^-$, Solv = CH_2Cl_2 [325]; $R = R^1 = Me, L = L^1$ = 1-Me-Im [326]), as well as products including unusual N, O-bidentated co-ligands (L–L′) **144** [327].



144: R =

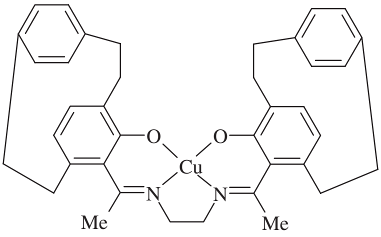


An extensive number of nickel Salen ($Z = CH_2–CH_2$) compounds are displayed by a series of complexes of type **134** ($R = R^1 = H, L = L^1 =$ none, Solv = none [328, 329]; Solv = $CHCl_3$ [330]; Solv = DMF [331]; R = 5-OH, $R^1 = H$, $L = L^1 =$ none, Solv = EtOH [329]; R = 5-$O(CH_2)_3Et$, $R^1 = H$, $L = L^1 =$ Solv = none [332]; R = 5-Cl, $R^1 = Ph$, $L = L^1 =$ Solv = none [333]; R = H, $R^1 = CH_2OMe$, $L = L^1 =$ Solv = none [334]; R = 3-*tert*-Bu, $R^1 = CH_2OMe$, $L = L^1 =$ Solv = none [334]; R = 5-NO_2, $R^1 = CH_2OMe$, $L = L^1 =$ Solv = none [334]; R = cyclo-C_4H_4, $R^1 = CH_2OMe$, $L = L^1 =$ Solv = none [334]; R = 3-CH_2OMe, 5-Me, $R^1 = R^3 = R^4 = Ph$, $L = L^1 =$ Solv = none [335]; R = H, $R^1 = Me$, $L = L^1 =$ Solv = none [336]). Among similar complexes a chelate **145** (Solv = H_2O, MeCN) with uncommon aldehyde should be mentioned [337].



145

A great number of copper(II) chelates with ethylenediamine fragment **134** are known, e.g. (R = H, 5-NO_2, 5-OMe, 5-Br, $R^1 = H$, $L = L^1 =$ Solv = none [338]; R = *bis*-3, 5-*di*-*tert*-Bu, $R^1 = H$, $L = L^1 =$ Solv = none [339]; R = 5-OMe, $R^1 = H$, $L = L^1 =$ Solv = none [340]; R = cyclo-C_4H_4, $R^1 = H$, $L = L^1 =$ Solv = none [341]; R = H, $R^1 = Me$, $L = L^1 =$ Solv = none [336]; R = H, $R^1 = Me$, $L = L^1 =$ Solv = none [342]; $R = R^1 = H$, $R^3 = R^4 =$ *tert*-Bu, $L = L^1 =$ Solv = none [343]; R = 3-CH_2-piperidine, 5-*tert*-Bu, $R^1 = H$, $L = L^1 =$ Solv = none [344]; R = H, 5-NO_2, 5-OMe, 5-Br, $R^1 = H$, $R^3 = R^4 = Ph$, $L = L^1 =$ Solv = none [345]). The copper complex incorporating the paracyclophanyl moiety in the keto fragment is depicted by **146** [346], and the metal chelate **147** includes an unusual amine component [347].



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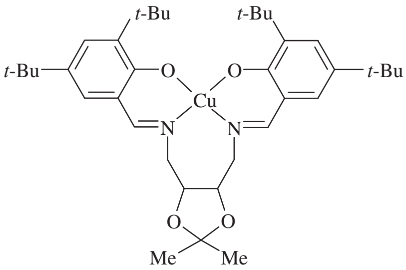
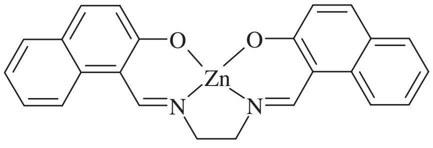


147

Formation of tetracoordinated complexes (C.N. = 4) from rigidly planar Salen of type **134** $\{Z = (CH_2)_2\}$ is non-typical for zinc(II). In this respect, structure **148** [348] is surprising. Nevertheless, the formation of a tetragonal-pyramidal polyhedron **149** is unequivocal [349].



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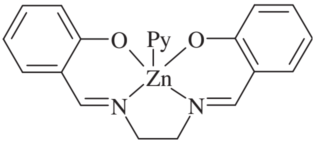
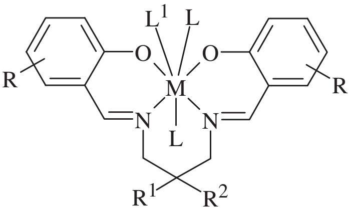


149

Few examples of complexes of type **134** for molybdenum ($Z=(CH_2)_n$, $n=2$, 3; R = H, 3,5-*di-tert*-Bu; R^1 = H, Me; L, L^1 = =O, =N-*tert*-Bu; Solv = none [350]) and platinum(II) (R = H, R^1 = Me, L = L^1 = Solv = none [351]) are known.

In coordination compounds of types **134** and **138** and their analogues based on ethylenediamine ($n=2$), tricyclic 5,6,6-membered metal-chelate structures are realized. Expansion of metallocycle is achieved due to increase of the size of the internitrogen bridge. Thus, three six-membered metallocycles are formed in complexes of propylenediamine derivatives [$n=3, Z=(CR^1R^2)(CH_2)_2$] **150** and **151**. Complexes incorporating various *p*- and *d*-metals (M), L and L^1 fragments, anions (A) and solvents (Solv) of this type have been synthesized and



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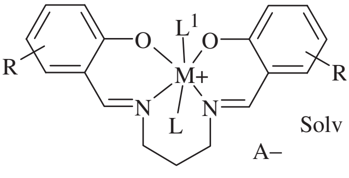
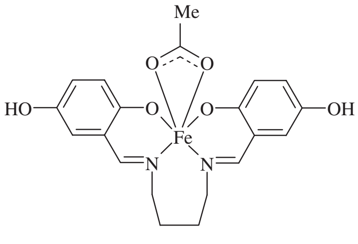


151

are represented by coordination compounds of aluminum(III) **150** (R = *bis*-3, 5-*di-tert*-Bu, $R^1=R^2$ = Me, L = Et, L^1 = Solv = none [352]), titanium(IV) (R = 3-*tert*-Bu, $R^1=R^2$ = H, L = L^1 = Cl, Solv = none [353]), iron(III) (R = $R^1=R^2$ = H, L = NCS, $L^1=H_2O$, Solv = none [354]; R = cyclo-C_4H_4, $R^1=R^2$ = H, L = L^1 = Solv = none [355]), cobalt(II) (R = Me, $R^1=R^2$ = H, L = L^1 = Solv = none [356]; R = $R^1=R^2$ = H, L = NCS, $L^1=H_2O$, Solv = none [357]; R = cyclo-C_4H_4, $R^1=R^2$ = H, L = L^1 = Solv = none [358]), nickel(II) (R = Me, $R^1=R^2$ = H, L = L^1 = Solv = none [359]; R = cyclo-C_4H_4, $R^1=R^2$ = H, L = L^1 = none, Solv = MeOH [360]; R = 3-OMe, $R^1=R^2$ = H, L = $L^1=H_2O$, Solv = none [361]; R = 3, 5-NO_2, $R^1=R^2$ = H, L = L^1 = 3, 4-*di*-Me-pyr, Solv = $C_4H_8O_2$ [362]), copper(II) (R = $R^1=R^2$ = H, L = L^1 = Solv = none [363]; R = Me, $R^1=R^2$ = H, L^1 = Solv = none [364]; R = cyclo-C_4H_4, $R^1=R^2$ = H, L = L^1 = Solv = none [365]; R = 3-OMe, $R^1=R^2$ = Me, L = L^1 = none, Solv = H_2O [340]; R = 4-OMe, $R^1=R^2$ = Me, L = L^1 = none, Solv = EtOH [340]) and zinc(II) (R = cyclo-C_4H_4, $R^1=R^2$ = H, L = L^1 = Solv = none [349, 366]).

Cationic complexes **151** have a few structures of compounds of aluminum(III) (R = *bis*-3, 5-*di-tert*-Bu, L = L^1 = THF, A = BH_4^-, Solv = THF [367]) and manganese (R = H, L = $L^1=H_2O$, A = Cl^-, Solv = none [311]; R = H, L = $L^1=H_2O$, A = Br^-, Solv = none [368]).

One seven-membered and two six-membered metallocycles are formed in similar butylenediamine Salen-compounds [$Z=(CH_2)_4$], e.g. complexes of iron(III) **152** [369] and copper(II) **153** (R = H [370], R = 3-OMe [361]).



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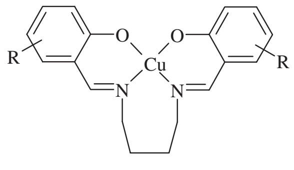
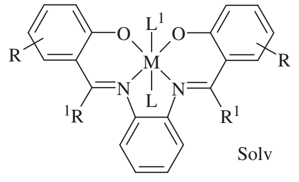


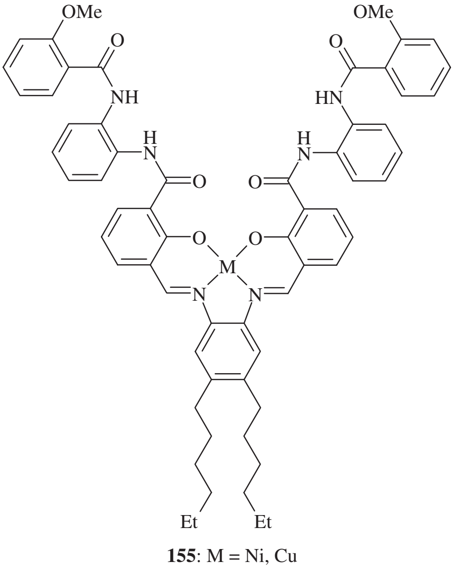
153

In Ar-Salen complexes of type **154**, the aromatic moieties (Ar) are internitrogen bridges.

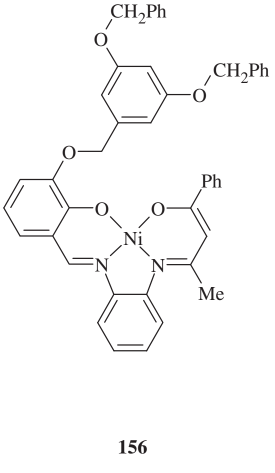


154

A series of Ar-Salen coordination compounds **154** are known, e.g. Al(III) [296], Sn(IV) ($R = R^1 = H, L = (CH_2)_2Et$, $L^1 = Cl$, Solv = none [299]; R = 3,5-*di-tert*-Bu, $R^1 = H$, $L = L^1 = Cl$, Solv = none [298]; R = *bis*-3,5-*di-tert*-Bu, $R^1 = H$, $L = L^1 = Br$, Solv = CH_2Cl_2 [298]; R = 3,5-*di*-Cl, $R^1 = H$, $L = L^1 = Cl$, Solv = THF [299]), Mn(III) ($R = R^1 = H$, L = NCS, L^1 = Solv = none [371]; $R = R^1 = H$, L = MeOH, $L^1 = ONO_2$, Solv = MeCN [372]; $R = R^1 = H$, $L = H_2O$, $L^1 = OClO_3$, Solv = none [371]), Co(II) (R = 5-NO_2, $R^1 = H$, $L = L^1 = H_2O$, Solv = DMF [373]), Ni(II) ($R = R^1 = H$, $L = L^1$ = Solv = none [374, 375]; R = H, R^1 = Me, $L = L^1$ = Solv = none [376]), Co(II) ($R = R^1 = H$, $L = L^1$ = Solv = none [377]; R = H, R^1 = Me, $L = L^1$ = Solv = none [378]; R = 3-Ph, $L = L^1$ = Solv = none [379]; R = 5-COOH, $L = L^1$ = none, Solv = H_2O [380]) and Zn(II) (R = 3, 5-*di*-Br, L = Py, L^1 = none, Solv = DMF [381]). Ar-Salen chelates including uncommon amine **155** [382] and aldehyde **155**, **156** [382, 383] fragments have similar structures.

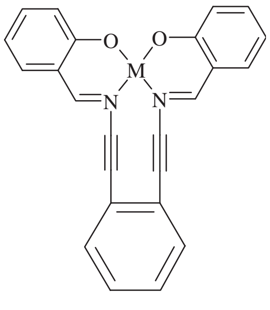


155: M = Ni, Cu



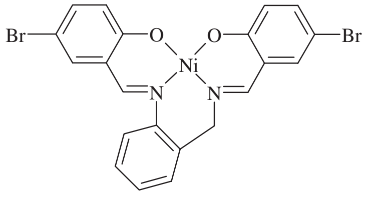
156

Nickel(II) and copper(II) Salen-complexes **157** containing acetylene and aromatic fragments in the internitrogen bridge were reported [384].



157: M = Ni, Cu

Chelates with three 6,6,6- **158** [385] and the 6,6,7-membered **159** [386–389] metallocycles are known for Ar-Salen complexes. Metal-chelates of type **159** have been isolated and structurally characterized for iron(II) (R = cyclo-C_8H_6, Solv = CH_2Cl_2 [386]), cobalt(II) (R = H, Solv = MeOH [387, 388]) and copper(II) (R = 3, 5-*di*-Br, Solv = CH_2Cl_2 [389]).



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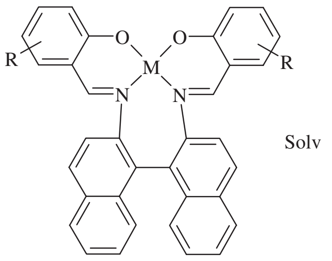
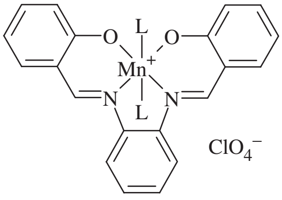


159

M = Fe(II), Co(II), Cu(II)

Cationic Ar-Salen **160** (L = imidazole) [390] and Heteroaryl-Salen with pyridine internitrogen fragment **161** (R = 3, 5-*di*-*tert*-Bu [391, 392]; R = H, C≡CR^1, R^1 = H, 4-pentylphenyl, $SiMe_3$ [392]) complexes are represented by a few compounds.



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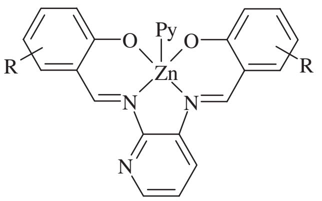
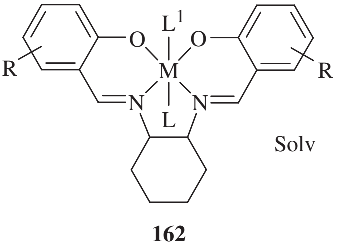


161

Salen complexes with 1,2-diaminocyclohexane internitrogen bridges are represented by metal chelates of type **162** [343, 393–401], e.g. by complexes of titanium(IV) (R = 3, 5-*di*-*tert*-Bu, L = L^1 = Cl, Solv = toluene [393]; R = 3, 5-*di*-Cl, L = L^1 = Cl, Solv = MeCOMe [393]), manganese(III) (R = 3, 5-*di*-*tert*-Bu, L = Cl, L^1 = Solv = none [394]), cobalt(II) (R = H, L = L^1 = Solv = none [343]; R = 3, 5-*di*-*tert*-Bu, L = Cl, L^1 = none, Solv = C_6H_6 [395]), nickel(II) (R = 3-tert-Bu, L = L^1 = Solv = none [396]; R = 3, 5-*di*-*tert*-Bu, L = L^1 = Solv = none [397]), copper(II) (R = 5-Me, L = L^1 = Solv = none [398];

R = 5-*tert*-Bu, $L = L^1$ = none, Solv = H_2O [399]; R = 5-*tert*-Bu, $L = L^1$ = none, Solv = CH_2Cl_2 [399]) and zinc(II) (R = H, L = Py, L^1 = Solv = none [400]; R = 3, 5-*di-tert*-Bu, L = Py, L^1 = Solv = none [400]; R = 5-Cl, L = Py, L^1 = Solv =none [401]; R = 5-Br, L = Py, L^1 = Solv = none [401]).



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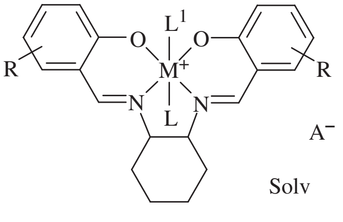
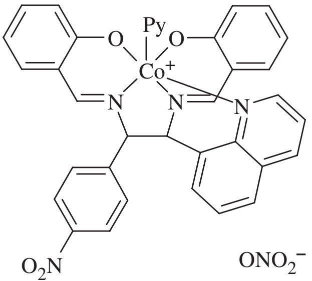


163

Schiff-base complexes of manganese(III) (R = 3-OMe, $L = L^1 = H_2O$, A = Cl, Solv = H_2O [402]; R = 3, 5-*di-tert*-Bu, $L = L^1$ = 2-phenyl-3-methylaziridine, A = MeCOO or A = CH_2ClCOO, Solv = CH_2Cl_2 [403]; R = 3, 5-*di*-Br, $L = L^1 = H_2O$, A = ClO_4, Solv = H_2O [404]) are of type **163**. A non-standard structure with additional coordination of quinoline **164** [405] has been revealed for a cationic cobalt(III) complex.



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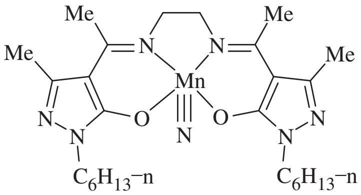
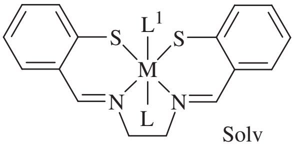


165

A few structures for Salen ethylenediamine complexes of type **165**, e.g. manganese(II) [406], are known. Similar metal chelates obtained from *o*-chalcogenbenzaldehyde derivatives of type **166** widely occur [5, 6, 10, 407].



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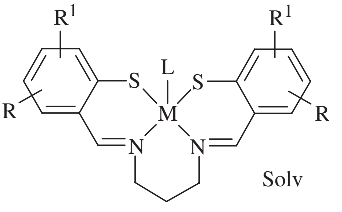


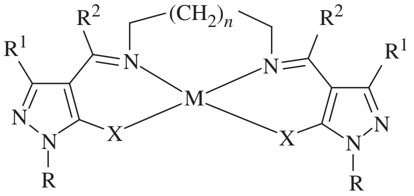
167

Omitted in previous reviews [5, 6, 10, 407], complexes of iron(II) **166** (L = CO, L^1 = Py, Solv = none [408]; L = Cl, L^1 = none, Solv = none [409]) should be mentioned, as well as the compounds of nickel(II) ($L = L^1$ = none, Solv = CH_2Cl_2 [410]) and copper(II) ($L = L^1$ = Solv = none [410]).

Complexes of type **167** derived from propylenediamine Salen ligands are represented by tricyclic (6.6.6.) cobalt(III) ($R = R^1$ = 3, 5-*di-tert*-Bu, L = MeCOO, Solv = $CHCl_3$, Et_2O [411]), nickel(II) ($R = R^1$ = H, L = none, Solv = $C_4H_8O_2$

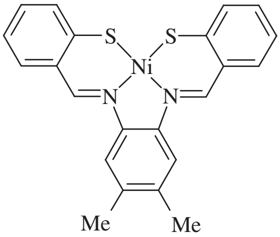
[412, 413]; $R = 3\text{-CHO}, R^1 = 5\text{-Me}$, L = none, Solv = none [80]; $R = 3\text{-(COMe)}_2$, $R^1 = 5\text{-}tert\text{-Bu}$, L = none, Solv = none [414]; $R = R^1 = H$, L = none, Solv = none [415]) and palladium(II) ($R = 3\text{-CHO}, R = 5\text{-}tert\text{-Bu}, R^1 = H$, L = none, Solv = none [416]) complexes.

Salen complexes containing alkyl internitrogen bridges (Z) and heterocyclic aldehyde fragments are widely represented, e.g. by pyrazole derivatives **168** [10, 407, 417–419]. They contain two six-membered metallocycles and one of different size depending on the value of n [417–419].



168: R, R^1, R^2 = H, Alk, Ar;
M = Co, Ni, Zn, n = 1–6

Salen complexes including tricyclic (6.5.6) **169** [420] and (6.7.6) **170** [421], **171** [422] chelate rings derived from aromatic diamines are isolated and structurally characterized.



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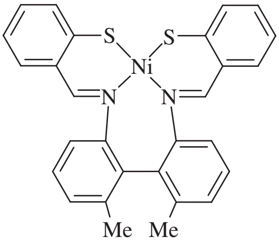

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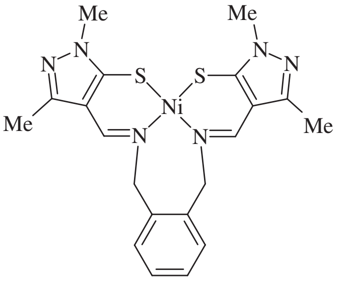
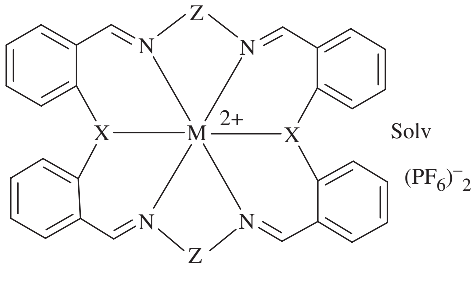
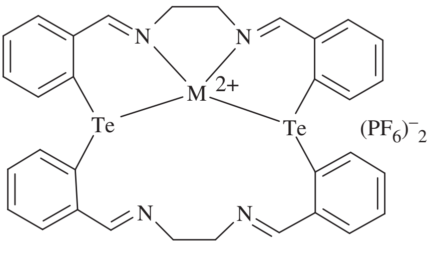


171

A series of cobalt(II), nickel(II) and palladium(II) Salen-like complexes of types **172** and **173** with selenium [423] and tellurium [424] donor centers has been described.



172: M = Co^{2+}, Ni^{2+}, Pd^{2+};
Z = $(CH_2)_n$, n = 2, 3;
X = S, Se;
Solv = none, H_2O

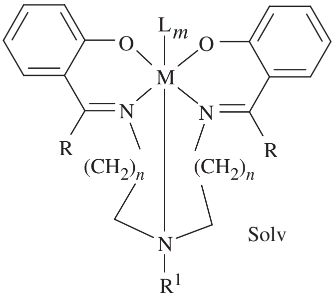


173: M = Ni^{2+}, Pd^{2+}

Six (**172**: $n = 2$, two five- and four six-membered) and three (**173**, two six- and one five-membered) metallocycles are realized in these metal chelates.

Salen complexes containing N–, O–, S–, as well as Te-heteroatoms in the diimine bridge, are widely reported. Introduction of coordinatively-active heteroatoms into this bridge generally leads to increase of coordination number and quantity of metallocycles.

3.3.3.2. *Aza-Salen complexes.* This type involves complexes containing either one **174** [425–430] or two **175** [426, 431–435] nitrogen atoms in the interazomethine fragment.



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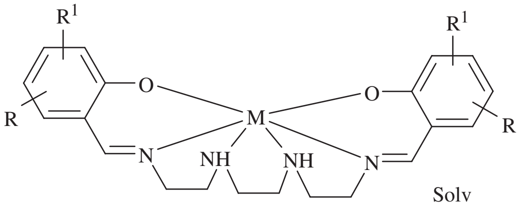
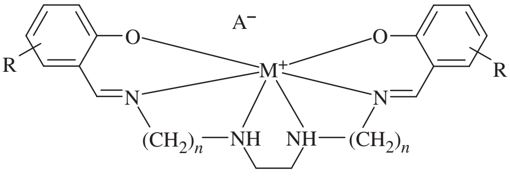


175

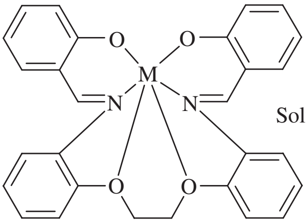
Coordination compounds of type **174** include chelates containing two four- ($n = 0$) and two six- or two five- and two six- ($n = 1$) membered metallocycles, e.g. complexes of tin(IV) ($R = R^1 = H, n = 0, L = Me, m = 2$, Solv = H_2O, MeOH, DMSO [425]), manganese(III) ($R = H, R^1 = Me, n = 1, L = OCOMe$, $m = 1$, Solv = none [426]), iron(II) ($R = H, R^1 = Me, n = 1, L = Solv = 0$ [427]), iron(III) ($R = H, R^1 = Me$, $n = 1, L = Cl, m = 1$, Solv = none [428]) and copper(II) ($R = R^1 = H, n = 1, L =$ none, Solv = toluene [429]; $R = Me, R^1 = H, n = 1$, L = Solv = none [430]; $R = H, R^1 =$ $(CH_2)_3SiMe_3$, $n = 1$, L = Solv = none [429]).

When two nitrogen atoms are included in the internitrogen bridge, structures of type **175** with two six- and three five-membered metallocycles are formed, e.g. complexes of iron(II) ($R = H, R^1 = 5\text{-}NO_2$, Solv = none [431]; $R = 3\text{-OMe}, R^1 = 5\text{-}NO_2$, Solv = none [432]; $R = 3\text{-OEt}, R^1 = 5\text{-}NO_2$, Solv = THF [432]), nickel(II) ($R = 3\text{-CHO}, R^1 = 5\text{-Me}$, Solv = none [433]) and zinc(II) ($R = R^1 = H$, Solv = H_2O [434]; $R = H, R^1 =$ 3-OMe, L = Solv = none [434]), as well as X-ray characterized cationic aza-Salen complexes **176** [426, 435] containing two four-membered, one five-membered and two six-membered ($n = 1$) or three five-membered and two six-membered ($n = 2$) metal-chelate rings [426, 435]. The cationic oxorhenium(V) mononitric methyl Salen complex incorporating two five- and two six-membered metallocycles was synthesized and structurally characterized [426, 435].

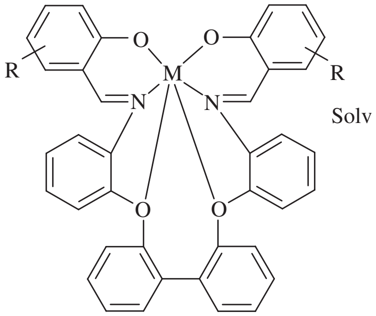


176: M = Mn, Fe; R = H, OMe, Cl, Br; n = 1,2; A = Cl, ClO_4, PF_6

3.3.3.3. *Oxo-Salen metal chelates.* This type of Schiff-base complex is represented by pentachelate compounds containing three five-, two six- **177** [289] or two five-, two six- and one seven-membered **178** metallocycles [436].

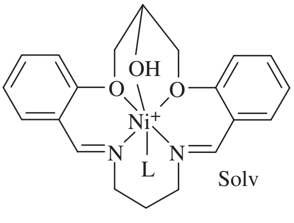


177: M = Ni(II), Co(II), Zn(II)



178

Complexes of cobalt(II), nickel(II) (Solv = MeCOOH [289]) and zinc(II) (Solv = MeOH [289]) belong to type **177**. Structures **178** include coordination compounds of manganese(II) (R = H, Solv = CH_2Cl_2 [436]) and cobalt(II) (R = H, Solv = CH_2Cl_2 [436]; R = cyclo-C_4H_4, Solv = MeCOOEt [436]). Nickel(III) complexes **179** (L = ONO_2^-, Br^-; Solv = none, MeCN [437, 438]) with three coordinated oxygen atoms are known, as well as copper(II) chelates with non-coordinated oxygens **180** (R = H, cyclo-C_4H_4, Solv = CH_2Cl_2 [436]) and **181** [439].



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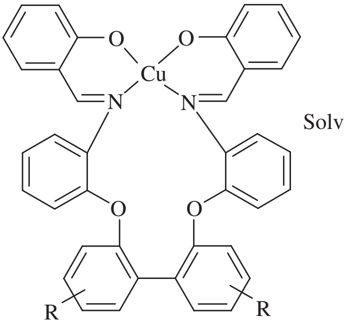

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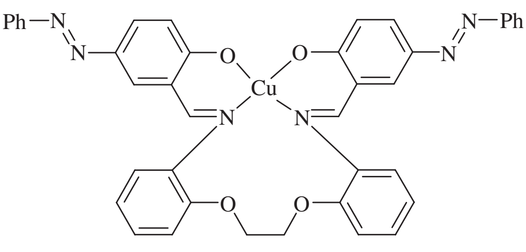
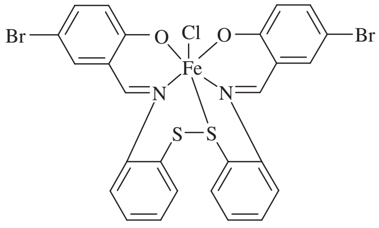


181

3.3.3.4. *Chalcogen-Salen complexes.* Salen complexes with chalcogen donor atoms in the bridging fragments are represented by tetra and pentachelate structures **182** [440] and **183** [435, 441], respectively.



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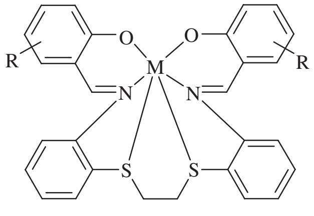
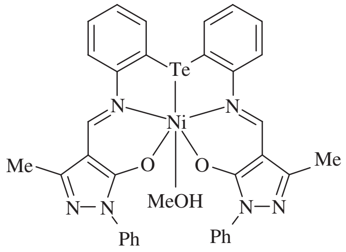
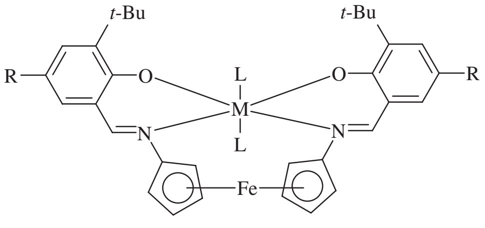


183: M = Ni(II), Zn(II)

Salen complexes with selenium atoms in the internitrogen bridge are unknown, but a tellurium-containing Salen derived with two five- and two six-membered metal chelates was recently synthesized and structurally characterized **184** [442].





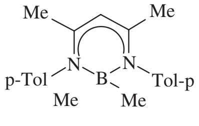
184

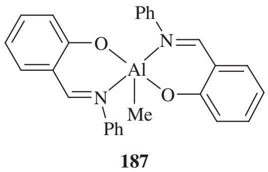
185: R = H, *t*-Bu

Finally, the magnesium(II), titanium(IV) and zirconium(IV) Salen complexes **185** were isolated from 1,1′-diaminoferrocene and aryl-substituted salicylaldehydes and structurally characterized [443].

3.4. *Central atoms*

P- and *d*-elements [1–10, 293, 294, 444] are the most common metals forming Schiff-base coordination compounds and considerable attention is recently paid to organometallic centers. This aspect is examined with some examples in the previous sections but requires to be extended. Thus, a situation with dimethylboron derivatives **186** [445], methylaluminum **187** [446] and dimethylgallium **188** [446, 447] is representative.





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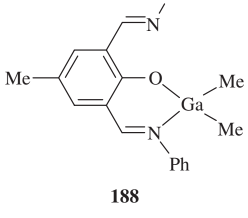
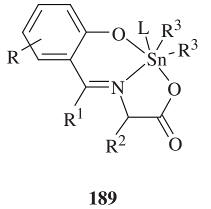
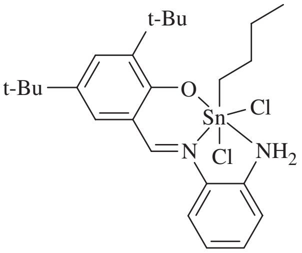




188

189

Chelates of various types incorporating organometallic centers **189** for tin(IV) are known, e.g. R = 3-Me, R^1 = Me, R^2 = H, R^3 = Ph [448]; R = 3,5-*di*-Br, R^1 = H, R^2 = Me, R^3 = cyclohexyl [449]; R = R^2 = H, R^3 = Me, L = 1,10-phen [448]) and **190** [450].



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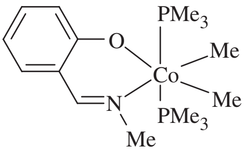
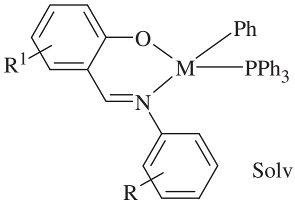


191

Organic fragments constitute a part of the metallocenter in Schiff-base chelates of *d*-metals, e.g. cobalt(III) complexes **191** [451] and a series of complexes of type **192** [452–457] for nickel(II) [452–455] and palladium(II) [454–457].

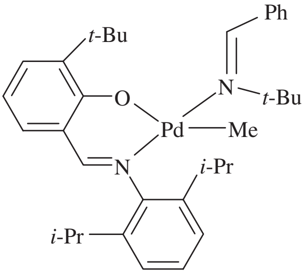


192: M = Ni(II), Pd(II); R = H, 2,6-Me_2, 2,6-*i*-Pr_2, 2,6-*t*-Bu_2;
R^1 = H, OMe, CH_2–CH=CH_2, NO_2, Cl, 9-antracenyl

3.5. *Co-ligands*

The bases (co-ligands) additionally bounded with metal are of considerable importance in the molecular design of azomethine chelates. Coordinatively-unsaturated metal complexes with Schiff bases usually form adducts essentially with inorganic and organic N,O,P-donors [2, 4, 5], e.g. molecular oxygen is a typical co-ligand [458].

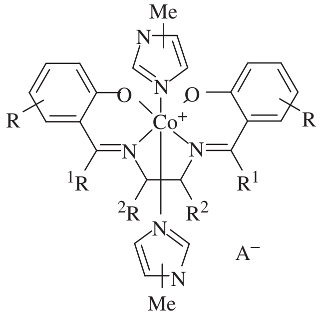
The nitrogen-containing compounds (*N*-bases) are common organic co-ligands, e.g. N-*tert*-butylbenzaldimine in **193** [457].



193

The adduct formation is extremely characteristic for tetra-coordinated Schiff-base chelates and leads to penta- and hexa-coordinated structures, based on previously reviewed data, and augmented in the following discussion.

A series of adducts of Schiff bases with nitrogen-containing five- and six-membered heterocycles was characterized by X-ray diffraction. Salen complexes of cobalt(II) of type **194** with 1-methylimidazole ($R = 5\text{-Me}, R^1 = \text{Me}, R^2 = \text{H}$, Solv = none, $A = ClO_4$ [324]), 2-methylimidazole ($R = R^1 = \text{H}, R^2\text{-}R^2 = 1,2$-cyclohexane, Solv = MeOH, $A = ClO_4$ [459]; $R = 3,5$-*di-tert*-Bu, $R^1 = \text{H}, R^2\text{-}R^2 = 1,2$-cyclohexene, $A = ClO_4$ [459]; $R = 5\text{-OMe}, R^1 = \text{H}, R^2\text{–}R^2 = 1,2$-cyclohexene, $A = ClO_4$ [459]) and copper chelate with imidazole **195** ($R = \text{H}, 5\text{-Cl}$; $A = ClO_4$) [460], as well as nickel(II) compounds with 1-*n*-pentyl-2-aminobenzimidazole **196** [461] and 2,6-diaminopiridine **197** [462], are reported.



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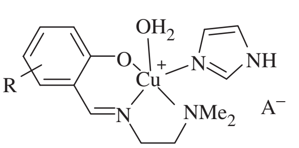

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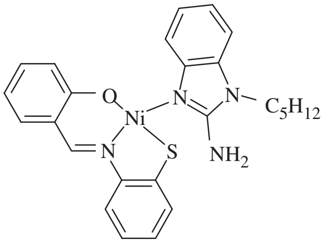

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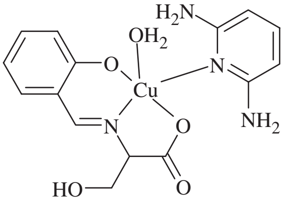

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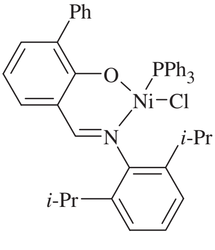
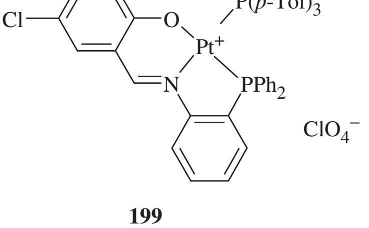


198

Among phosphorus-containing co-ligands, the phosphines **198** [463], **199** [231], diphosphines **200** [464] and 1,3,5-triazaphosphaadamantane **201** ($M = \text{Ni}, \text{Pd}$; $R = 3,5$-*di*-Cl or cyclo-C_4H_4) [465] are highlighted.



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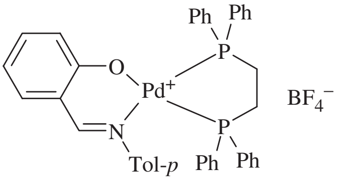

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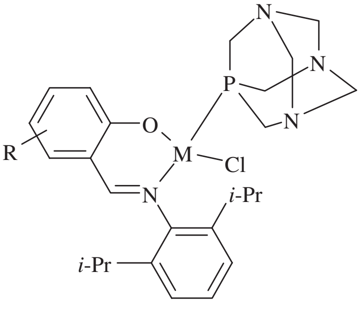

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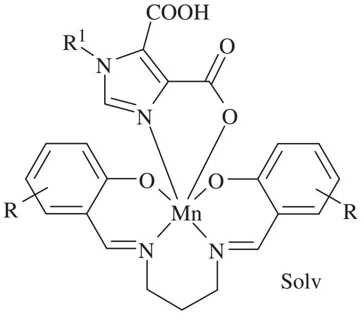


202

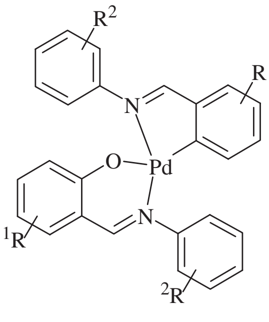
Apart from those in previous [1, 2, 4, 5] and the present review of Schiff-base adducts, the manganese(III) Salen-type coordination compound incorporating the original O,N-containing co-ligand **202** (R = H, *tert*-Bu; $R^1 = H, Me$; Solv = none, H_2O, MeCN) was reported [466].

4. Non-standard structures

In this section, azomethine chelates with mixed ligand environment and adducts based on chelating Schiff bases will be discussed.

4.1. *Mixed ligand environment*

Schiff-base complexes containing in the same molecule various coordination units with N,C- and N,O-ligand environment, can be attributed to this type of coordination compounds, e.g. metal chelates of palladium(II) **203** ($R = 5\text{-}NMe_2$ or $5\text{-}NO_2, R^1 = 4\text{-}NEt_2$ or $4\text{-}NO_2$; $R^2 = 4\text{-}OMe$, Solv = none, CH_2Cl_2 [467]) and **204** ($R = OCOC_6H_4OMe\text{-}p$, $R^1 = NO_2, R^2 = OMe, R^3 = NMe_2$, Solv = none [381]; $R = 4\text{-}NMe_2, R^1 = 4\text{-}NO_2$, $R^2 = OCOPh$, $R^3 = OMe$, Solv = $CHCl_3$ [468]; $R = 4\text{-}NMe_2$, $R^1 = NC_6H_4NO_2\text{-}p, R^2 = 4\text{-}OMe, R^3 = 4\text{-}OMe$, Solv = toluene [381]).



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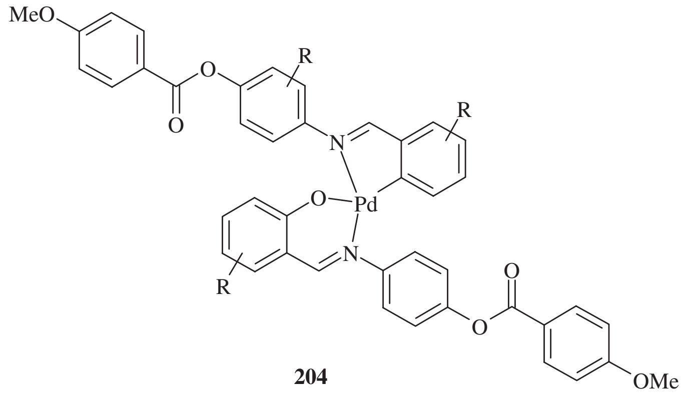
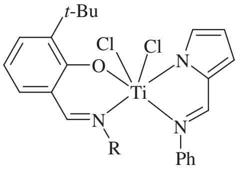


204

Complexes involving N,N- and N,O-ligand environments are of interest, e.g. complexes Ti(IV) and Co(II) of types **205** (R = Ph, C_6F_5 [468]) and **206** [47], respectively.



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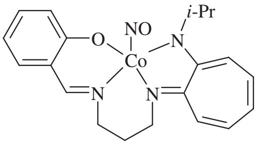

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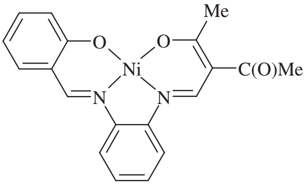
Nickel(II) complexes of type **207** [470] and **208** [471] with two different N,O chelate environments of salicylaliminate and β-ketiminate combined in the same molecule were reported.



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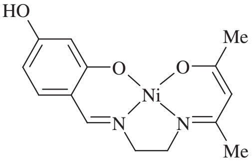
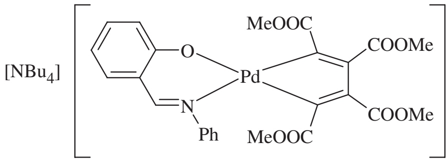


208

The palladium(II) is involved in N,O- and C,C-chelate environments in anionic complex **209** [472].



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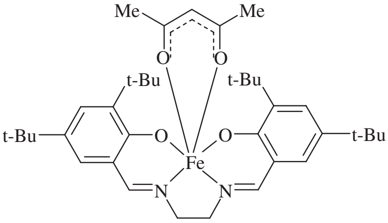
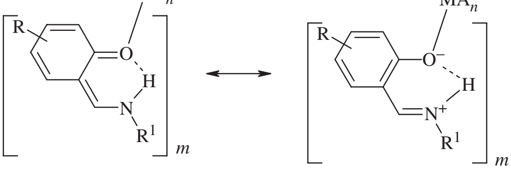


210

Iron(III) complex **210** [465] comprises a mixed chelate N,O and O,O coordination sphere.

4.2. *Adducts*

Besides the classic chelate structures with deprotonated ligands, non-standard adduct structures containing a completely unchanged ligand system were synthesized, e.g. **211** [2, 5, 6, 10, 43, 150, 473–475].



211a **211b**

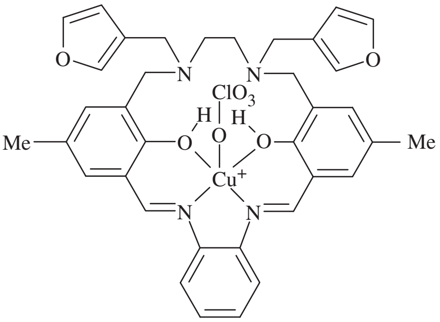
A problem in determination of localization of the coordination bond in such adducts [10] was resolved in favor of O-metal binding **211** (monodentate ligand) by X-ray diffraction [150, 474, 475].

The keto-amine (**a**) tautomeric form has been established for the imine ligand in **211** ($R = \text{cyclo-}C_4H_4, R^1 = Ph, MA_n = ZnCl_2, m = 2$) [474], whereas the zwitter-ionic (**b**) resonance form of the ligand, noncharacteristic for a free o-hydroxyazomethine [6], was discussed [150] ($R = H, R^1 = Pr, MA_n = ZnCl_2, m = 2$).

FTIR, ^1H NMR and X-ray data also indicate the zwitter-ionic (**b**) resonance form of the ligand in **211** ($R = H,\ OMe, R^1 = C_6H_4\text{-}p\text{-}R^2, R^2 = H, Me,\ OMe, Cl, MA_n = ReO_3, m = 1$) [475]. Therefore, the equilibrium [150] is of interest and affords the opportunity to directed synthesis of either metal chelates **71** or adducts **211** which were mentioned previously [5, 10].

$$\mathbf{71} \xrightleftharpoons[\text{NaOH or NEt}_3]{\text{2 HCl}} \mathbf{211}$$

The enol-imine tautomeric form of the Schiff-base ligand in cationic adduct **212** [476], revealed by X-ray analysis, is unexpected.



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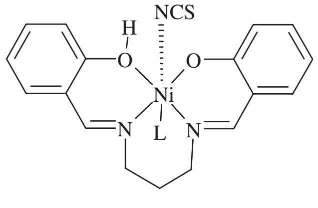
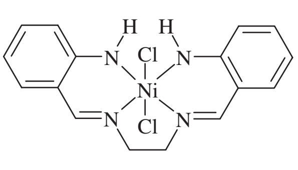


213: L = none, H_2O

A similar situation is observed in Salen complex **213**, unusual in containing protonated and deprotonated hydroxyl groups in the metal-chelate rings [477].

Adducts are also known for complexes of Salen ligands with nitrogen coordination environment **214** [478].



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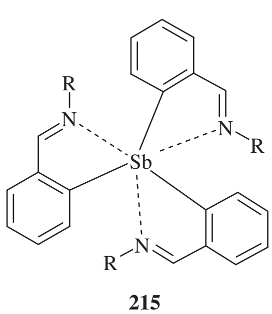

215

Standard binding of metal center and imine nitrogen in the complex of Sb(III) (according to X-ray diffraction data) is missing while a non-standard hypervalent N → Sb-bond **215** [479] is observed.

5. Conclusions

Molecular design of Schiff-base complexes in the 21st century will be focused on directed variation of the binding of azomethine derivatives to allow buildup of different chelate ring size and combinations, including N.C-, N.N-, N.P-, N.O-, N.S-, N.Se- and N.Te-ligand environments. Thereupon, the problem of selectively obtaining tetra-, penta- and hexa-coordinated chelate structures will be resolved, as well as standard and non-standard metal binding with chelating ligands, competitive coordination, stabilization of noncharacteristic for "free" ligand systems tautomeric forms under complexation. Based on the reviewed data, further development of coordination chemistry of acyclic and also macrocyclic Schiff bases is needed [1, 4, 480].

Approach to the molecular design of azomethine complexes in the present review may be used to vary the structure of the other chelating ligand systems, as demonstrated, for example, for chelates of β-diketones and their structural analogues [481].

Acknowledgements

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