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Synthesis, characterization and crystallographic determination of symmetrical Schiff bases and their Zn(II) metal complexes

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Abstract

Two new tetradentate symmetrical Schiff bases derived from 2,2'-diamino-4,4'-dimethyl-1,1'-biphenyl or its dibromo substituted analog and 3,5-diiodosalicylaldehyde were synthesized. The reactions of these Schiff bases with zinc acetate generated the corresponding Zn(II)-complexes in which the zinc atom is bonded through the two azoimine nitrogen atoms in addition to the two oxygen atoms of the deprotonated hydroxyl groups. The Schiff bases and their Zn(II) complexes were characterized using UV-Vis, ^{1}H- and ^{13}C-NMR, IR spectroscopy and elemental analysis. The structures of the two Schiff bases as well as the two zinc(II) complexes were further approved by single crystal X-ray analysis.

Introduction

Schiff bases have great affinity to bind metal ions for a wide range of applications in the areas of industry, biology and inorganic chemistry [1–3]. Treatment of the Schiff bases with metals such as copper, zinc, cobalt, nickel, aluminum, ruthenium, vanadium and iron produced a variety of useful metal complexes [1–5]. The latter exhibited excellent biological activity for anti-malarial, anti-viral, anti-tumor, anti-fungal and anti-inflammatory properties [6–8]. Some of these compounds are used as corrosion inhibitors [9, 10], as catalysts in polymers [11] and dyes [12, 13]. The metal's oxidation state is important in the design of these complexes which enables them to contribute significant roles in optimal doses and bio-availabilities of the administered agents [1–3, 14]. Molecular docking study of two nickel complexes with 2-hydroxy-4-methoxybenzaldehyde and 2-amino-2-methylpropanol Schiff bases showed that they have inhibitory against coronavirus and molecular targets of human angiotensin [15]. Similar docking study on cobalt(III) complexes of substituted 2-hydroxybenzylidene-4-hydroxybenzohydrazides showed that these complexes bind to the major protease SARS-CoV-2 and the molecular targets of human angiotensin (ACE-2) [16].

The biphenyl-based Schiff bases are an important class of Schiff bases with interesting applications in several fields. A biphenyl salicylhydrazone Schiff base is found to be promising analytical tool for detecting copper ions. The compound with low toxicity, showed outstanding cell permeation and is highly selective chemosensor for biological systems [17]. Oxovanadium(V) complexes of tridentate Schiff bases based on biphenyl and hydroxyl-salicylaldehyde are active for hydrogen peroxide mediated oxygenation of organic sulfides to the corresponding sulfoxides [18]. The bidentate Schiff base ligand 4-chloro-2-[1-(4-phenylphenyl)ethylideneamino]phenol and its metal complexes (Co, Ni, Cu, Zn) were found to have efficiency to decolorize methylene blue

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dye [19]. Palladium complexes of naphthyl- and biphenyl-salicylaldimine Schiff bases were reported to have good catalytic activity for Suzuki-Miyaura coupling reactions [20].

In our lab, we investigated the metal complexes of tetradentate Schiff bases based on biphenyl backbone derived from 2,2'-diamino-4,4'-dimethyl-1,1'-biphenyl or its dibromo analog (2,2'-diamino-4,4'-dimethyl-6,6'-dibromo-1,1'-biphenyl) and substituted salicylaldehydes were reported and reacted with metal acetates (Cu, Co, Ni, Mn, Fe, Zn) to form the corresponding metal-Schiff base complexes with different compositions [21–27]. For all metals used, the Schiff bases were coordinated to the metal in a tetradentate fashion through the two imine nitrogen atoms in addition to the deprotonated hydroxyl groups [21–27]. In some case, a solvent molecule is found to coordinate to the metal [26]. The biological activities of these Schiff bases and their complexes were tested as antimicrobial and anticancer agents and proved to have interesting activates [21–27]. Recently, we also performed an assessment and molecular modeling studies of Schiff bases with the potential to serve as promising candidates for the management of diabetes mellitus [28].

As a continuation of our efforts in the area of metal–Schiff bases complexes, two new iodo-substituted biphenyl Schiff bases are synthesized and their zinc(II) complexes are reported. The Schiff bases and the Zn(II) complexes are spectroscopically characterized in addition to their molecular structure determination.

Experimental section

Materials and methods

Chemicals and reagent grade solvents were obtained from commercial sources and used as received. The nuclear magnetic resonances spectra were recorded on Bruker AC 400 MHz spectrometer with residual solvent as a reference. The infrared spectra were measured on a Bruker FT-IR-4100 spectrometer. Electronic absorption spectra were recorded on PS-2600 Pasco spectrophotometer in DMSO solution. CHN elemental analyses were carried out on Perkin Elmer 240 instrument at the Institute of Organic and Macromolecular Chemistry of the Friedrich Schiller University in Jena, Germany. The compounds 2,2'-diamino-4,4'-dimethyl-6,6'-dibromobiphenyl [29] and 2,2'-diamino-4,4'-dimethyl-1,1-biphenyl [25] were prepared according to reported methods.

General procedure for the preparation of Schiff bases (SB1, SB2)

A 20 mL ethanol solution of 2,2'-diamino-4,4'-dimethyl-1,1'-biphenyl (0.466 g, 2.20 mmol) or 2,2'-diamino-4,4'-dimethyl-6,6'-dibromo-1,1'-biphenyl (0.816 g, 2.20 mmol) and 3,5-diiodosalicylaldehyde (2.09 g, 4.40 mmol) was heated with stirring at 80 ºC for 4 h. During the reaction, the corresponding Schiff bases were precipitated, collected by simple filtration, washed with cold ethanol and dried in vacuum. The obtained solid was recrystallized from a methanol solution for two days to give pure crystalline compounds.

3,5-Diiodosalicylideneamino-4,4'-dimethyl-1,1'-biphenyl (SB1) Orange (61%). M.p. = >242ºC. IR (KBr): ν3463 (O-H); ν2919 (C-H); ν1601 (C=N); ν1436 (C=C); ν1346 (C-O); ν658 (C-I). ^{1}H-NMR (400 MHz, $CDCl_3$): δ 13.49 (s, 2 H, O*H*); 8.36 (s, 2 H, C*H*=N); 6.84–7.94 (m, 6 H, Ar-*H*); 2.38 (s, 6 H, C*H*$_3$). ^{13}C-NMR (100 MHz, $CDCl_3$): δ 161.06 (*C*=N); 119.51–159.39 (Ar); 21.24 (*C*H$_3$). UV–Vis. in DMSO: λ_{max} (ε_{max}): 365 nm (6.51×10^3 $M^{-1}cm^{-1}$). Anal. Calc. for $C_{28}H_{20}I_4N_2O_2$: C, 36.84; H, 2.18; N, 3.03%. Found: C, 36.36; H, 2.18; N, 3.01%.

3,5-Diiodosalicylideneamino-6,6'-dibromo-4,4'-dimethyl-1,1'-biphenyl (SB2) Orange (70%). M.p. = >270 °C. IR (KBr,): ν3453 (O-H); ν2951 (C-H); ν1607 (C=N); ν1440 (C=C); ν1350 (C-O); ν713 (C-Br); ν660 (C-I) cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$): δ 12.91 (s, 2 H, O*H*); 8.42 (s, 2 H, C*H*=N); 6.89–7.96 (m, 4 H, Ar-*H*); 2.37 (s, 6 H, C*H*$_3$). ^{13}C-NMR (100 MHz, $CDCl_3$): δ 161.18 (*C*=N); 118.40-159.75 (Ar); 21.19 (*C*H$_3$). UV–Vis. in DMSO: λ_{max} (ε_{max}): 374 nm (2.75×10^3 $M^{-1}cm^{-1}$). Anal. Calc. for $C_{28}H_{18}I_4Br_2N_2O_2$: C, 31.08; H, 1.68; N, 2.59%. Found: C, 31.01; H, 1.61; N, 2.58%.

General procedure for the synthesis of zn(II) complexes (ZnSB1, ZnSB2)

To a solution of the Schiff base (1.30 mmol) in 10 mL of absolute ethanol, a solution of zinc(II) acetate hydrate (0.286 g, 1.30 mmol) dissolved in 3 mL of absolute ethanol was added dropwise at room temperature under inert atmosphere. The reaction mixture was refluxed for 6 h. During this time, the corresponding complexes were precipitated, collected by filtration, washed with cold ethanol and dried under vacuum. The complexes were recrystallized from MeOH solution.

ZnSB1 Yellow (62%). M.p.= 280–282 °C. IR (KBr): ν2921 (C-H); ν1599 (C=N); ν1425 (C=C); ν1309 (C-O); ν673

(C-I); ν538 (Zn-O); ν428 (Zn-N) cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$): δ 8.18 (s, 2 H, C*H*=N); 6.81–8.02 (m, 8 H, Ar-*H*); 2.31 (s, 6 H, C*H*$_3$). ^{13}C-NMR (100 MHz, $CDCl_3$): δ 170.71 (*C*=N), 119.39-167.39 (Ar); 21.02 (*C*H$_3$). UV–Vis. in DMSO: λ_{max} (ε_{max}): 402 nm (5.25×10^3 $M^{-1}cm^{-1}$). Anal. Calc. for $C_{28}H_{18}I_4N_2O_2Zn$: C, 34.06; H, 1.84; N, 2.84%. Found: C, 34.01; H, 1.79; N, 2.81%.

ZnSB2 Yellow (50%). M.p.= 290–292 °C. IR (KBr): ν2921 (C-H); ν1591 (C=N); ν1423 (C=C); ν1309 (C-O); ν705 (C-Br) ; ν679 (C-I); ν546 (Zn-O); ν414 (Zn-N) cm^{-1}. ^{1}H-NMR (400 MHz, $CDCl_3$): δ 8.20 (s, 2 H, C*H*=N); 6.81–8.02 (m, 6 H, Ar-*H*); 2.30 (s, 6 H, C*H*$_3$). ^{13}C-NMR (100 MHz, $CDCl_3$): δ 169.15 (*C*=N), 119.06–167.51 (Ar); 21.03 (*C*H$_3$). UV–Vis. in DMSO: λ_{max} (ε_{max}): 407 nm (2.75×10^3 $M^{-1}cm^{-1}$). Anal. Calc. for $C_{28}H_{16}I_4Br_2N_2O_2Zn$: C, 29.36; H, 1.41; N, 2.45%. Found: C, 29.28; H, 1.38; N, 2.44%.

Crystal structure determination of SB1, SB2, ZnSB1 and ZnSB2

X-ray diffraction measurements were performed on a CCD-type diffractometer (Rigaku XtaLAB P200) with Mo Kα radiation (λ = 0.71075 Å) at 100 K in a stream of cooled nitrogen gas. The crystal structures were solved using a direct method using the SHELXS-2014 program [30] and refined by a full-matrix least-squares method on F^2 using the SHELXL-2014 program [31]. Crystallographic data and parameters of **SB1, SB2, ZnSB1** and **ZnSB2** are shown in Table 1.

Table 1 Crystallographic data for **SB1, SB2, ZnSB1** and **ZnSB2**.

	SB1	SB2	ZnSB1	ZnSB2
CCDC No.	2,321,561	2,321,562	2,321,563	2,321,564
Empirical formula	$C_{28}H_{20}I_4N_2O_2$	$C_{28}H_{18}Br_2I_4N_2O_2$	$C_{28}H_{18}I_4N_2O_2Zn$	$C_{28}H_{16}Br_2I_4N_2O_2Zn$ CH_3OH
Formula weight (g mol^{-1})	924.10	1081.89	987.46	1177.29
Temperature (K)	100(2)	100(2)	100(2)	100(2)
Wavelength (Å)	0.71075	0.71075	0.71075	0.71075
Crystal system	Monoclinic	Monoclinic	Tetragonal	Monoclinic
Space group	$P2_1/n$	$C2/c$	$I4_1/a$	$P2_1/c$
Unit cell dimensions				
a (Å)	8.1269(3)	23.6039(6)	32.9663(4)	9.8306(2)
b (Å)	12.9420(4)	9.2364(2)	32.9663(4)	22.2689(4)
c (Å)	13.6850(4)	14.3181(4)	14.2781(3)	14.9421(3)
α (°)	90	90	90	90
β (°)	92.511(3)	105.951(3)	90	98.3421(18)
γ (°)	90	90	90	90
V (Å^3)	1437.99(7)	3001.37(14)	15517.1(5)	3236.47(11)
Z	2	4	16	4
ρ_{calcd} (g cm^{-3})	2.134	2.394	1.691	2.416
μ (mm^{-1})	4.363	6.850	3.839	7.086
F(000)	860	1992	7328	2176
Crystal size (mm^3)	0.20 × 0.10 × 0.05	0.10 × 0.10 × 0.05	0.50 × 0.10 × 0.10	0.20 × 0.15 × 0.10
Reflections collected	3526	3766	9366	8094
Independent reflections	3198	3518	7189	7269
Refined parameters	167	176	334	374
R_1 [$I > 2\sigma(I)$]	0.0247	0.0261	0.0470	0.0227
wR_2 (all data)	0.0613	0.0622	0.1316	0.0518
Goodness-of-fit on F^2	1.030	1.041	1.020	1.031
Largest diff. peak and hole (eÅ^{-3})	1.73 and − 0.99	2.07 and − 0.44	1.92 and − 0.75	0.90 and − 0.85

Results and discussion

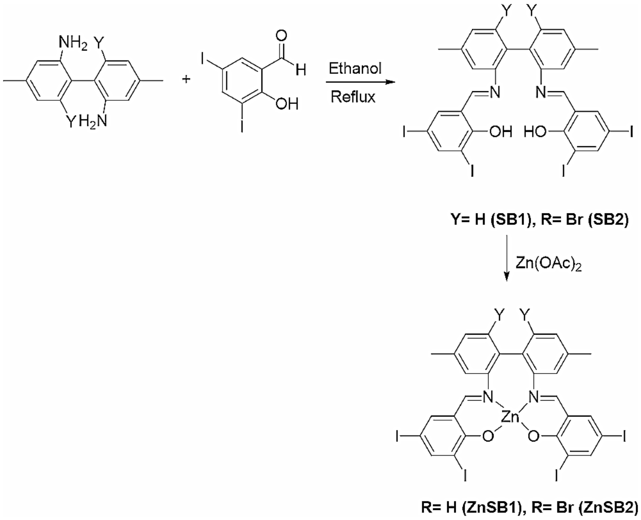
Synthesis

The reactions between 2,2'-diamino-4,4'-dimethyl-1,1'-biphenyl or 2,2'-diamino-6,6'-dibromo-4,4'-dimethyl-1,1'-biphenyl and 3,5-diiodosalicylaldehyde furnished the corresponding Schiff bases as shown in Scheme 1. The subsequent reactions of these Schiff bases with zinc acetate afforded the Zn(II) complexes with moderate yields (Scheme 1).

The obtained Schiff bases **SB1** and **SB2** and the corresponding zinc(II) complexes are stable in air at room temperature and soluble in commonly used organic solvents. The two new Schiff bases and the Zn(II) complexes have been characterized by elemental analysis, IR, ^{1}H-, ^{13}C-NMR spectroscopy as well as X-ray structure determination.

Spectral characterization

The IR spectra of the prepared Schiff bases (Figures S1 and S2) showed bands in the regions 3453–3463, 2919–2951, 1601–1607, 1436–1440 and 1346–1350 cm^{-1} characteristic of the absorptions of O-H, C-H, C = N, C = C and C-O functionalities, respectively. The IR spectra of the zinc(II) complexes (Figures S3 and S4) showed a shift to lower wavenumber in the position of imine (-C = N-) band, indicating bonding of this group to the metal center. In addition, the band of the hydroxyl group disappeared from the spectra of **SB1** and **SB2** indicating bonding to Zn(II)-ion. Moreover, two new bands, in the ranges of 414–428 and 538–546 cm^{-1} (due to M-N and M-O stretching) are observed in the metal complexes' spectra (Figures S3 and S4) represented further evidence of oxygen and nitrogen coordination to zinc. These data are consistent with those published for similar complexes [22–26].



Scheme 1 Synthesis of symmetrical Schiff bases **SB1**, **SB2** and their Zn(II) complexes

The ^{1}H-NMR spectra of the **SB1** and **SB2** showed peaks at 13.49 and 12.91 ppm, respectively, for the OH protons (Figures S5 and S6). However, these peaks disappeared in the spectra of the Zn(II) complexes (Figures S7 and S8) indicating deprotonation followed by complexation. The spectra of the zinc(II) complexes showed a singlet at 8.18 or 8.20 ppm of the azomethine proton (-C*H* = N-) of **ZnSB1** and **ZnSB2** (Figures S7 and S8), respectively which was observed at 8.36 or 8.42 ppm in the free Schiff bases spectra (Figures S5 and S6). These chemical shift values for the free ligands and the zinc(II) complexes were in agreement with those reported previously for similar compounds [22–26].

The ^{13}C-NMR spectra of the **SB1** and **SB2** (Figures S9 and S10) showed a sharp signal at 159.8 or 159.5 ppm which may be assigned to azomethine carbon while the peaks at 161.2 or 161.1 ppm may be assigned to the carbon atoms bonded to the hydroxyl group [32]. These characteristic azomethine or hydroxyl carbon signals underwent a downfield shift and appeared at 167.4 and 170.2 ppm for **ZnSB1** (Figure S11) and at 167.5 and 169.2 ppm for **ZnSB2** (Figure S12) invoking its coordination through azomethine nitrogen and hydroxyl groups. A similar shift in the ^{13}C-NMR shifts for the azomethine carbon is reported previously [32]. Moreover, aromatic signals have undergone slight shift upon coordination while almost no changes are observed for the methyl signal which appeared in the range of 21.0-21.2 ppm.

Electronic absorption spectra

The UV-Vis absorption spectra of the Schiff bases and their Zn(II) complexes were measured in DMSO using 8×10^{-5} M solutions. The spectra of the **SB1** and **SB2** (Figures S13 and S14) showed an absorption band at 365 and 374 nm, respectively which may attribute to ligand-ligand charge transfer; $\pi \rightarrow \pi^*$ or $n \rightarrow \pi$, of the conjugated system and the azomethine group. This band is red shifted in the spectra of the complexes (**ZnSB1**: 402 nm, **ZnSB2**: 407 nm) (Figures S15 and S16). Similar shift has been reported for analogous systems [22–26].

Crystal structures of SB1 and SB2

Crystals suitable for X-ray structure determination of **SB1** and **SB2** were grown from saturated MeOH solution. Their structures with atomic numbering schemes are depicted in Fig. 1. The biphenyl moieties in these compounds are almost planar. The imine N = C bond distances (C8-N1 = 1.281(4) Å (**SB1**), 1.270(4) Å (**SB2**)) are similar to those reported for similar Schiff bases (1.265–1.285 Å) [24–26]. These

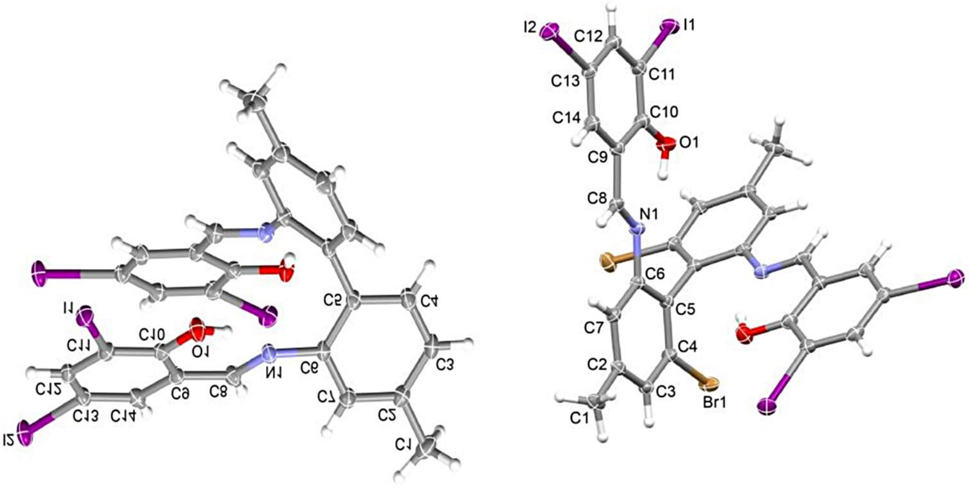


Fig. 1 Molecular structures and atom labeling schemes of **SB1** (left) and **SB2** (right) determined by X-ray diffraction. The ellipsoids represent a probability of 50%, and only the crystallographically independent atoms are labeled

Table 2 Selected bond lengths (Å) and angles (°) for **SB1** and **SB2**.

SB1		SB2	
C11-I1	2.093(3)	C11-I1	2.088(3)
C13-I2	2.097(3)	C13-I2	2.098(3)
C10-O1	1.339(4)	C10-O1	1.339(4)
C8-N1	1.281(4)	C8-N1	1.270(4)
C6-N1	1.419(4)	C6-N1	1.422(4)
		Br1-C4	1.899(3)
C8-N1-C6	120.8(2)	C8-N1-C6	123.9(3)
C7-C6-N1	121.5(3)	C7-C6-N1	124.1(3)
C5-C6-N1	117.6(3)	C5-C6-N1	115.2(3)
C9-C8-N1	120.4(3)	C9-C8-N1	120.8(3)

bond lengths are clearly shorter that the N-C single bonds of 1.419(4) and 1.422(4) Å for **SB1** and **SB2** respectively, within the same compounds. The carbon-oxygen bond distances of the hydroxyl group (C10-O1 = 1.339(4) Å for both **SB1** and **SB2**) are slightly shorter than the corresponding distance of the 3-nitro- (1.354(2) Å) or 3-methoxy- (1.353(7) Å) substituted Schiff bases. The C-I bond lengths are within the normal C(sp^2)-I distances. Moreover, the C-Br bond distance of **SB2** (1.899(3) Å) is comparable to those found in similar systems [24–26]. The imine moieties of **SB1** and **SB2** are almost planar with sp^2 hybridization of atoms as indicated by the bond angles which are close to 120º (Table 2). The imine functionality adopted a torsion angle of 177.3(3)º and 180.0(3)º relative to the benzene plane for **SB1** and **SB2**, respectively. However, a noticeable difference in the dihedral angle between the iodophenyl rings between that of **SB1** (8.34º) and **SB2** (88.04º) is observed.

Crystal structures of ZnSB1 and ZnSB2

The molecular structures of complexes **ZnSB1** and **ZnSB2** are presented in Fig. 2. The Schiff bases are coordinated to the central zinc atom in a tetradentate fashion through two nitrogen atoms of the imine moieties in addition to two oxygen atoms of the deprotonated hydroxyl groups. The geometry around the Zn atom in **ZnSB1** is tetrahedron as indicated by the O-Zn-O, O-Zn-N and N-Zn-N angles (Table 3). Similar

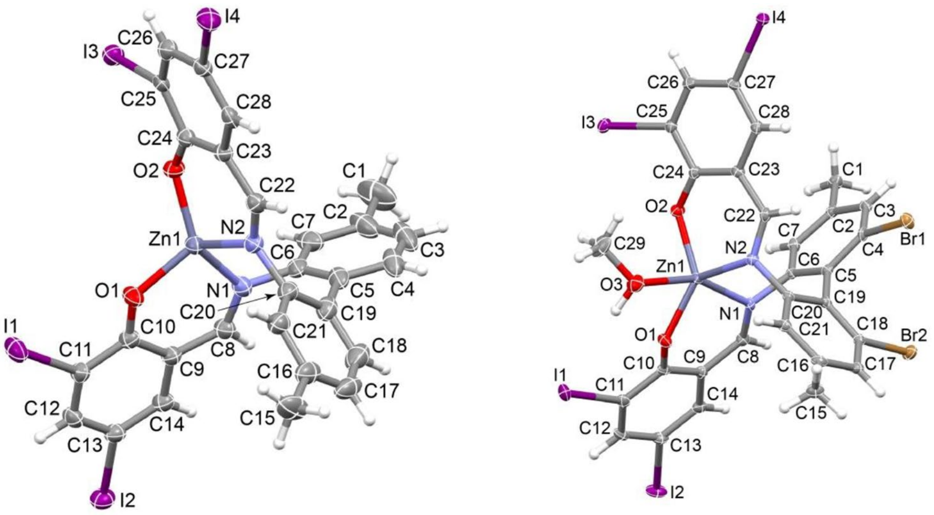


Fig. 2 Molecular structures and atom labeling schemes of **ZnSB1** (left) and **ZnSB2** (right) determined by X-ray diffraction. The ellipsoids represent a probability of 50%, and only the crystallographically independent atoms are labeled

Table 3 Selected bond lengths (Å) and angles (°) for **ZnSB1** and **ZnSB2**

ZnSB1		ZnSB2	
Zn1-O1	1.932(4)	Zn1-O1	1.937(2)
Zn1-O2	1.901(4)	Zn1-O2	1.950(2)
Zn1-N1	2.010(5)	Zn1-N1	2.026(2)
Zn1-N2	2.017(5)	Zn1-N2	2.184(2)
C11-I1	2.087(6)	C11-I1	2.093(3)
C13-I2	2.088(6)	C13-I2	2.102(3)
C25-I3	2.100(5)	C25-I3	2.099(3)
C27-I4	2.091(6)	C27-I4	2.101(3)
		C4-Br1	1.902(3)
		C18-Br2	1.902(3)
C6-N1	1.419(4)	C6-N1	1.433(3)
C8-N1	1.277(8)	C8-N1	1.284(4)
C20-N2	1.435(7)	C20-N2	1.433(4)
C22-N2	1.294(7)	C22-N2	1.293(4)
C24-O2	1.292(7)	C24-O2	1.300(3)
O1-Zn1-O2	115.20(17)	O1-Zn1-O2	134.49(9)
N1-Zn1-N2	94.54(19)	N1-Zn1-N2	88.65(9)
O1-Zn-N1	93.91(18)	O1-Zn-N1	85.61(8)
O1-Zn1-N2	126.27(19)	O1-Zn1-N2	130.43(9)
O2-Zn1-N1	134.86(19)	O2-Zn1-N1	112.48(9)
O2-Zn-N2	94.56(18)	O2-Zn1-N2	92.81(9)
		O1-Zn1-O3	73.05(8)
		O2-Zn1-O3	88.00(9)
		N1-Zn1-O3	157.57(8)
		N2-Zn1-O3	99.81(9)
C5-C6-N1	120.0(6)	C5-C6-N1	119.0(2)
C7-C6-N1	119.5(6)	C7-C6-N1	119.1(2)
C8-N1-C6	118.0(5)	C8-N1-C6	117.7(2)
C9-C8-N1	126.5(5)	C9-C8-N1	125.9(3)
C22-N2-C20	118.0(5)	C22-N2-C20	116.8(2)

tetrahedron geometries for 4-bromosalicylideneamino-4,4’-diethylcarboxylato-1,1’-biphenylzinc complex with comparable bond angles (N-Zn-N: 96.373(13), N-Zn-O: 94.137(13), 140.589(16), 140.295(15), 94.630(14), O-Zn-O: 101.090(14)º) and bis(3-methyl-1-phenyl-4-[(2,4,6-trimethylphenyl-imino)-methyl]-1 H-pyrazol-5-olate)zinc were reported [33, 34]. The Zn-N (2.010(4), 2.017(4) Å) and the Zn-O (1.932(4), 1.901(4) Å) bond lengths of **ZnSB1** are similar to the corresponding lengths of 4-bromosalicylideneamino-4,4’-diethylcarboxylato-1,1’-biphenylzinc and bis(3-methyl-1-phenyl-4-[(2,4,6-trimethylphenyl-imino)-methyl]-1 H-pyrazol-5-olate)zinc [33, 34]. **ZnSB2** showed similar coordination mode of the Schiff base, in which two imine nitrogen atoms and two phenoxo oxygen atoms in addition to a methanol molecule is coordinated to the Zn center. The resulted geometry of the Zn atom is between that of square pyramid and of trigonal bipyramid as observed for similar complexes [24, 34]. The Zn-O and Zn-N bond lengths (Table 3) are not exceptional and similar to those of **ZnSB1** and (2,2-dimethylpropane-1,3-diimine-3-ethoxysalicylaldehyde)zinc [34]. Moreover, the C-Br and C-I bond distances of the complexes are very comparable to those of the parent Schiff bases as these bonds are away from the bonding sites [24, 35].

Conclusion

Two new Schiff bases based on biphenyl and iodo-substituted salicylaldehyde were reported and reacted with zinc acetate to generate the corresponding zinc(II) complexes in which the Schiff bases are tetracoordinate to the zinc(II) metal. The molecular structures of the two Schiff bases are determined and show a planar geometry of the imine fragments. The structures of the zinc(II) complexes are also determined and found to have a distorted tetrahedral geometry for **ZnSB1** and a distorted square planar one for **ZnSB2**.

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Author contributions TMJ and TMS conceptualization, supervision, experimental work, data collection, writing experimental part, review. ME writing the manuscript, collecting ideas, supervisor, HM X-ray structure measurements, writing experimental X-ray part. YY proof reading and x-ray discussion. HK supervision and revision.

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Data availability No datasets were generated or analysed during the current study.

Declarations

Conflict of interest The authors declare that they have no competing interest.

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