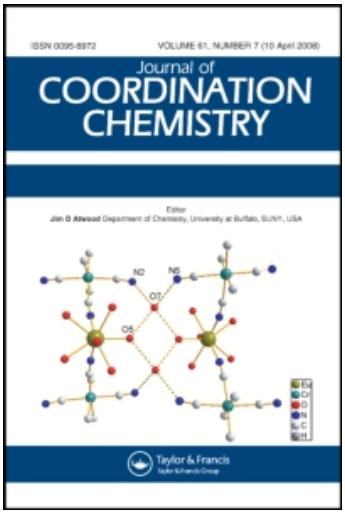


Journal of Coordination Chemistry
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713455674

Synthesis and antibacterial activities of metal(II) complexes with Schiff bases derived from 3,5-diiodosalicylaldehyde

Suo-Ping Xu[a]; Lei Shi[a]; Peng-Cheng Lv[a]; Rui-Qin Fang[a]; Hai-Liang Zhu[a]
[a] State Key Laboratory of Pharmaceutical Biotechnology, Nanjing University, Nanjing 210093, China; School of Chemistry and Chemical Engineering, Xuzhou Normal University, Xuzhou 221116, China

To cite this Article Xu, Suo-Ping , Shi, Lei , Lv, Peng-Cheng , Fang, Rui-Qin and Zhu, Hai-Liang(2009) 'Synthesis and antibacterial activities of metal(II) complexes with Schiff bases derived from 3,5-diiodosalicylaldehyde', Journal of Coordination Chemistry, 62: 12, 2048 — 2057
To link to this Article: DOI: 10.1080/00958970902741251
URL: http://dx.doi.org/10.1080/00958970902741251

Journal of Coordination Chemistry
Vol. 62, No. 12, 20 June 2009, 2048–2057

Synthesis and antibacterial activities of metal(II) complexes with Schiff bases derived from 3,5-diiodosalicylaldehyde

SUO-PING XU, LEI SHI, PENG-CHENG LV, RUI-QIN FANG and HAI-LIANG ZHU*

State Key Laboratory of Pharmaceutical Biotechnology, Nanjing University, Nanjing 210093, China; School of Chemistry and Chemical Engineering, Xuzhou Normal University, Xuzhou 221116, China

(*Received 7 July 2008; in final form 29 September 2008*)

Two new Schiff bases (2,4-diiodo-6-[(2-morpholin-4-yl-ethylimino)-methyl]-phenol and 2,4-diiodo-6-[(3-morpholin-4-yl-propylimino)-methyl]-phenol), condensed from 3,5-diiodosalicylaldehyde with 2-morpholinoethylamine and 3-morpholinopropylamine, have been designed and synthesized. Reaction of the Schiff bases with $Zn(OAc)_2 \cdot 2H_2O$, $Cu(OAc)_2 \cdot H_2O$, $Ni(OAc)_2 \cdot 4H_2O$, $Co(OAc)_2 \cdot 4H_2O$, $Cd(OAc)_2 \cdot 2H_2O$, $Mn(OAc)_2 \cdot 4H_2O$, $Fe(SO4)_2 \cdot 7H_2O$, and $Hg(OAc)_2$ led to the formation of 16 new mononuclear complexes. The complexes were characterized by UV, Infrared, ESI-MS, and elemental analyses, and 3,5-diiodosalicylalidene-2-morpholinoethylaminozinc(II) (**1**) and 3,5-diiodosalicylalidene-2-morpholinoethy laminocopper(II) (**2**) were characterized by single crystal X-ray diffraction. Based on crystal structural analysis of **1** and **2**, coupled with their spectral similarity with **3**–**16**, it can be concluded that **3**–**16** have structures similar to **1** and **2**. All the complexes were assayed for antibacterial activities against three Gram positive bacterial strains (*Bacillus subtilis*, *Staphylococcus aureus*, and *Streptococcus faecalis*) and three Gram negative bacterial strains (*Escherichia coli*, *Pseudomonas aeruginosa*, and *Enterobacter cloacae*) by 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl tetrazolium bromide method. Among the complexes tested, **8** and **16** showed the most favorable antibacterial activity with minimum inhibitory concentration of 0.781, 12.5, 6.25, 3.125, 3.125, 6.25 and 1.562, 6.25, 1.562, 3.125, 3.125, 1.562 $\mu g\,mL^{-1}$ against *B. subtilis*, *S. aureus*, *S. faecalis*, *P. aeruginosa*, *E. coli*, and *E. cloacae*, respectively.

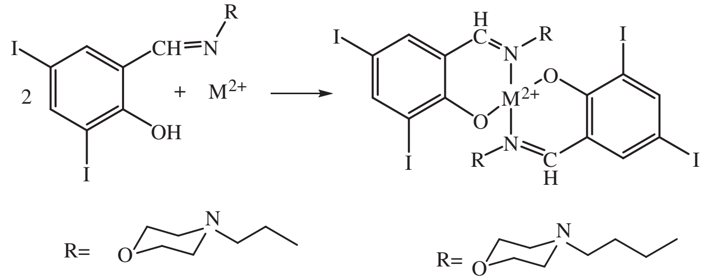
Keywords: 3,5-Diiodosalicylaldehyde; 2-Morpholinoethylamine; 3-Morpholinopropylamine; Metal(II) complexes; Antibacterial activity; Crystal structure

1. Introduction

Schiff bases derived from salicylaldehyde and their metal complexes show a wide spectrum of antimicrobial properties [1–4]. Researchers have studied the synthesis, characterization, and structure–activity relationship (SAR) of Schiff bases [5–8]; salicylaldehyde derivatives with one or more haloatoms in the aromatic ring showed antibacterial and antifungal activities [9]. In this article, we present two new Schiff

*Corresponding author. Email: zhuhl@nju.edu.cn

Journal of Coordination Chemistry
ISSN 0095-8972 print/ISSN 1029-0389 online
DOI: 10.1080/00958970902741251



1	M=Zn	**9**	M=Zn
2	M=Cu	**10**	M=Cu
3	M=Ni	**11**	M=Ni
4	M=Co	**12**	M=Co
5	M=Cd	**13**	M=Cd
6	M=Mn	**14**	M=Mn
7	M=Fe	**15**	M=Fe
8	M=Hg	**16**	M=Hg

Scheme 1. Syntheses of the complexes.

bases condensed from 3,5-diiodosalicylaldehyde with 2-morpholinoethylamine and 3-morpholinopropylamine. Reaction of the Schiff bases with different metal ions led to the formation of 16 new mononuclear complexes; all were assayed for antibacterial activities against Gram positive (*Bacillus subtilis*, *Staphylococcus aureus*, and *Streptococcus faecalis*) and Gram negative bacterial strains (*Escherichia coli*, *Pseudomonas aeruginosa*, and *Enterobacter cloacae*) by 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl tetrazolium bromide (MTT) method. Most complexes show potent antibacterial activity against the six bacterial strains. The results of this study may be useful to understand antimicrobial activity of metal(II) complexes with 3,5-diiodosalicylalidene Schiff bases.

2. Results and discussion

2.1. *Chemistry*

Two 3,5-diiodosalicylalidene Schiff bases have been synthesized by condensing 3,5-diiodosalicylaldehyde with 2-morpholinoethylamine and 3-morpholinopropylamine. Reaction of the Schiff bases with $Zn(OAc)_2 \cdot 2H_2O$, $Cu(OAc)_2 \cdot H_2O$, $Ni(OAc)_2 \cdot 4H_2O$, $Co(OAc)_2 \cdot 4H_2O$, $Cd(OAc)_2 \cdot 2H_2O$, $Mn(OAc)_2 \cdot 4H_2O$, $Fe(SO4)_2 \cdot 7H_2O$, and $Hg(OAc)_2$ led to the formation of 16 mononuclear complexes (scheme 1). All complexes gave satisfactory chemical analyses (±0.4%). UV, Infrared (IR) spectroscopy, and ESI-MS spectra were consistent with the assigned structures. Complexes **1** and **2** were characterized by X-ray crystallography (figures 1 and 2). Crystallographic and experimental data for **1** and **2** are provided in table 1. Selected bond lengths (Å) and angles (°) of **1** and **2** are illustrated in table 2. Their crystallographic data have been

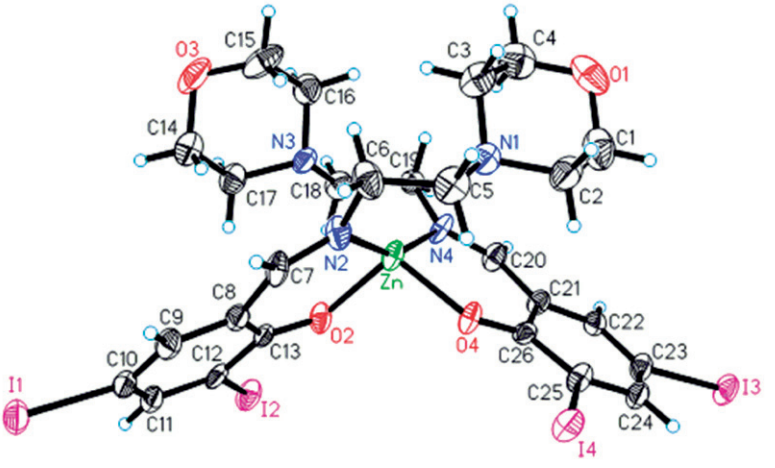


Figure 1. Crystal structure of **1** showing 30% probability displacement ellipsoids (arbitrary spheres for the H atoms.

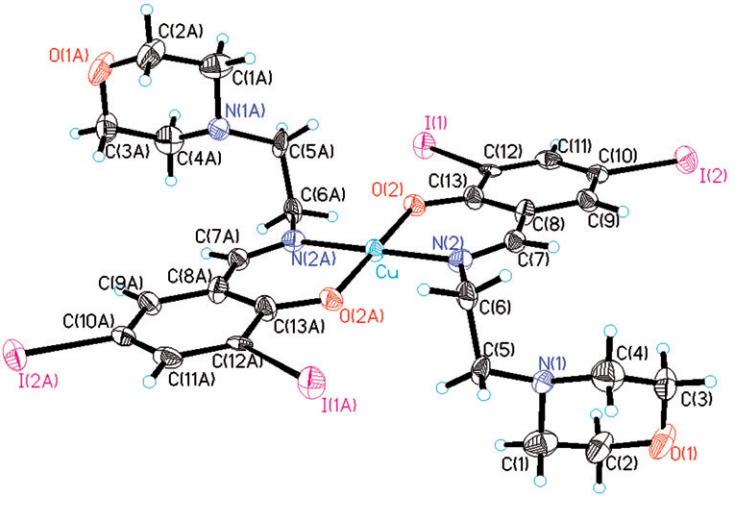


Figure 2. Crystal structure of **2** showing 30% probability displacement ellipsoids (arbitrary spheres for the H atoms.

deposited with the Cambridge Crystallographic Data Centre (CCDC 675135 for **1** and CCDC 675134 for **2**).

Complexes of formula $C_{26}H_{30}O_4N_4I_4M$ (M = Zn, Cu, Ni, Co, Cd, Mn, Fe, and Hg) were prepared from 2,4-diiodo-6-[(2-morpholin-4-yl-ethylimino)-methyl]-phenol ($C_{13}H_{16}O_2N_2I_2$), as described in the experimental section, in moderate yields (64–72%). IR spectra of the complexes (KBr pellets) display an intense absorption at ca 1610–1630 cm^{-1} attributable to ν(C=N), shifted ca 10–30 cm^{-1} to lower wavenumbers compared with 1640 cm^{-1} for $C_{13}H_{16}O_2N_2I_2$. UV spectra of the complexes display intense absorptions at 259–265 nm ($\pi \rightarrow \pi^*$) and 388–432 nm ($n \rightarrow \pi^*$).

Table 1. Crystallographic and experimental data for **1** and **2**.

Complex	**1**	**2**
Empirical formula	$(C_{13}H_{15}O_2N_2I_2)_2Zn$	$C_{13}H_{15}O_2N_2I_2Cu_{0.5}$
Formula weight	1035.51	516.84
Crystal system	Triclinic	Triclinic
Space group (Å, °)	$P\bar{1}$	$P\bar{1}$
a	9.938(2)	6.6090(13)
b	11.371(2)	10.377(2)
c	14.527(3)	12.550(3)
α	87.14(3)	113.29(3)
β	79.03(3)	93.84(3)
γ	76.20(3)	98.31(3)
V (Å^3)	1565.1(5)	774.9(3)
Z	2	2
T (K^{-1})	293(2)	293(2)
D_{Calcd} (g cm^{-3})	2.197	2.215
μ (mm^{-1})	4.767	4.726
F(000)	976	487
Maximum and minimum trans.	0.6471 and 0.3289	0.6493 and 0.4516
Data/restraints/parameters	6131/96/340	3028/0/172
θ range (°)	1.43 to 26.00	1.78 to 25.97
Index ranges (h, k, l)	$11 \le h \le 12$, $-13 \le k \le 14$, $0 \le l \le 17$	$-8 \le h \le 8$, $-12 \le k \le 11$, $0 \le l \le 15$
Reflections collected/unique	6509/6131	3308/3028
R_{int}	0.0401	0.0244
R ($I > 2$(I))	0.0548	0.0563
wR ($I > 2$(I))	0.1513	0.1475
($\Delta\rho$)max, ($\Delta\rho$)min (eÅ^3)	0.999 and −1.007	1.275 and −1.052

Note: $aR = \sum||F_o| - |F_c|/\sum|F_o|$, $bwR = [\sum[w(F_o^2 - F_c^2)^2]/\sum[w(F_o^2)^2]]^{1/2}$.

2.2. *Biological activity*

The biological activity of a particular substance depends on a complex sum of individual properties including complex structure, affinity for the target site, and survival in the medium of application, survival within the biological system, transport properties, and state of the target organism [10].

All the synthesized complexes were screened for antibacterial activity against three Gram (+) bacterial strains (*B. subtilis*, *S. aureus*, and *S. faecalis*) and three Gram (−) bacterial strains (*E. coli*, *P. aeruginosa*, and *E. cloacae*). The results are indicated in table 3. Generally, complexes with 2,4-diiodo-6-[(2-morpholin-4-yl-ethylimino)-methyl]-phenol were more active than with 2,4-diiodo-6-[(3-morpholin-4-yl-propylimino)-methyl]-phenol when the metal is the same.

Among the complexes, Hg complexes **8** and **16** showed most antibacterial activity with minimum inhibitory concentrations (MICs) of 0.781, 12.5, 6.25, 3.125, 3.125, 6.25 and 1.562, 6.25, 1.562, 3.125, 3.125, 1.562 μg mL^{-1}, against *B. subtilis*, *S. aureus*, *S. faecalis*, *P. aeruginosa*, *E. coli*, and *E. cloacae*, respectively.

3. Conclusions

In this article, two new Schiff bases derived from 3,5-diiodosalicylaldehyde with 2-morpholinoethylamine and 3-morpholinopropylamine have been synthesized. Reaction of the Schiff bases with different metals led to the formation of 16 new

Table 2. Selected bond lengths (Å) and angles (°) of **1** and **2**.

Bond	Distance	Bond	Distance	Bond	Distance
1					
Zn–O(4)	1.957(7)	Zn–O(2)	1.994(7)	Zn–N(2)	2.006(8)
Zn–N(4)	2.021(9)	I(1)–C(10)	2.083(10)	I(2)–C(12)	2.088(10)
I(3)–C(23)	2.109(10)	I(4)–C(25)	2.104(11)	O(1)–C(4)	1.397(18)
O(1)–C(1)	1.43(2)	N(1)–C(3)	1.481(16)	N(1)–C(5)	1.480(15)
N(1)–C(2)	1.483(16)	O(2)–C(13)	1.241(11)	N(2)–C(7)	1.307(13)
N(2)–C(6)	1.496(14)	O(3)–C(15)	1.347(19)	O(3)–C(14)	1.424(17)
O(4)–C(26)	1.271(11)	N(3)–C(16)	1.432(15)	N(3)–C(18)	1.462(15)
N(3)–C(17)	1.479(14)	N(4)–C(20)	1.264(12)	N(4)–C(19)	1.466(13)
2					
I(1)–C(12)	2.107(9)	I(2)–C(10)	2.093(11)	Cu–O(2)#1	1.910(7)
Cu–O(2)	1.910(7)	Cu–N(2)	1.981(9)	Cu–N(2)#1	1.981(9)
N(2)–C(7)	1.267(13)	N(2)–C(6)	1.490(14)	N(1)–C(1)	1.443(16)
N(1)–C(4)	1.466(17)	N(1)–C(5)	1.474(13)		
Angle	(°)	Angle	(°)	Angle	(°)
1					
O(4)–Zn–O(2)	105.0(3)	O(4)–Zn–N(2)	102.9(3)	O(2)–Zn–N(2)	94.4(3)
O(4)–Zn–N(4)	94.2(3)	O(2)–Zn–N(4)	101.6(3)	N(2)–Zn–N(4)	152.7(4)
C(4)–O(1)–C(1)	108.3(11)	C(3)–N(1)–C(5)	113.4(9)	C(3)–N(1)–C(2)	105.8(10)
C(5)–N(1)–C(2)	110.0(10)	C(13)–O(2)–Zn	127.3(7)	C(7)–N(2)–C(6)	116.0(9)
C(7)–N(2)–Zn	121.3(7)	C(6)–N(2)–Zn	122.1(7)	O(1)–C(1)–C(2)	110.1(12)
C(26)–O(4)–Zn	127.7(6)	C(20)–N(4)–Zn	120.6(7)	C(19)–N(4)–Zn	122.0(6)
2					
O(2)–Cu–N(2)	91.2(4)	O(2)#1–Cu–N(2)#1	91.2(4)	O(2)–Cu–N(2)#1	88.8(3)
N(2)–Cu–N(2)#1	180.0(6)	C(11)–C(12)–I(1)	116.1(7)	C(13)–C(12)–I(1)	120.2(7)
C(13)–O(2)–Cu	129.6(7)	C(9)–C(10)–I(2)	121.0(9)	C(11)–C(10)–I(2)	117.9(8)
C(7)–N(2)–C(6)	113.2(9)	C(7)–N(2)–Cu	125.6(8)	C(6)–N(2)–Cu	121.2(7)
N(2)–C(7)–C(8)	127.7(10)	C(1)–N(1)–C(4)	110.8(11)	C(1)–N(1)–C(5)	111.9(10)
C(4)–N(1)–C(5)	112.9(10)	N(1)–C(5)–C(6)	113.4(9)	C(3)–O(1)–C(2)	111.3(10)
N(2)–C(6)–C(5)	111.1(9)	N(1)–C(4)–C(3)	109.1(12)	O(1)–C(3)–C(4)	112.6(12)
O(1)–C(2)–C(1)	112.6(12)	N(1)–C(1)–C(2)	108.1(12)		

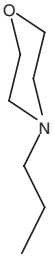
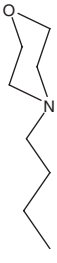
mononuclear complexes. All the complexes were assayed for antibacterial activities against three Gram positive (*B. subtilis*, *S. aureus*, and *S. faecalis*) and three Gram negative bacterial strains (*E. coli*, *P. aeruginosa*, and *E. cloacae*) by MTT method. Most complexes showed potent antibacterial activity against the six bacterial strains with **8** and **16** showing the most favorable antibacterial activity and, in general, 2,4-diiodo-6-[(2-morpholin-4-yl-ethylimino)-methyl]-phenol formed complexes that proved to be more active than those formed with 2,4-diiodo-6-[(3-morpholin-4-yl-propylimino)-methyl]-phenol.

4. Experimental

4.1. *Chemistry*

The 3,5-diiodosalicylalidene was synthesized with salicylaldehyde, KI, and KIO_3 [11]. The other chemicals (reagent grade) used were commercially available. UV spectra were

Table 3. MICs ($\mu g\,mL^{-1}$) of the complexes.

			Microorganisms' MICs ($\mu g\,mL^{-1}$)					
			Gram positive			Gram negative		
Complexes	R	M	*B. subtilis*	*S. aureus*	*S. faecalis*	*P. aeruginosa*	*E. coli*	*E. cloacae*
1		Zn	0.781	<50	1.562	3.125	0.781	12.5
2		Cu	12.5	25	1.562	0.781	>50	0.781
3		Ni	1.562	6.25	0.781	0.781	>50	1.56
4		Co	0.781	12.5	6.25	3.125	>50	12.5
5		Cd	0.781	12.5	12.5	0.781	0.781	1.562
6		Mn	0.781	25	1.562	>50	>50	6.25
7		Fe	0.781	12.5	3.125	0.781	>50	12.5
8		Hg	0.781	12.5	6.25	3.125	3.125	6.25
9		Zn	6.25	>50	1.562	6.25	1.562	12.5
10		Cu	25	>50	0.781	6.25	>50	6.25
11		Ni	0.781	12.5	1.562	0.781	>50	6.25
12		Co	>50	12.5	1.562	6.25	>50	0.781
13		Cd	3.125	12.5	12.5	6.25	3.125	6.25
14		Mn	3.125	>50	3.125	12.5	>50	6.25
15		Fe	0.781	6.25	1.562	6.25	12.5	6.25
16		Hg	1.562	6.25	1.562	3.125	3.125	1.562
Penicillin			1.562	1.562	1.562	6.25	6.25	3.125
Kanamycin			0.39	1.562	3.125	3.125	3.125	1.562

recorded on a U-3000 spectrophotometer. IR spectra were recorded on a Nexus 870 FT-IR. ESI-MS spectra were recorded on a Mariner System 5304 Mass spectrometer. Elemental analyses were performed on a CHN-O-Rapid instrument and were within ±0.4% of the theoretical values. Melting points were measured on a Boetius micro melting point apparatus.

4.1.1. General method of synthesis 1–16. Equimolar quantities (0.4 mmol) of 3,5-diiodosalicylaldehyde and 2-morpholinoethylamine or 3-morpholinopropylamine with 0.2 mmol of $Zn(OAc)_2 \cdot 2H_2O$, $Cu(OAc)_2 \cdot H_2O$, $Ni(OAc)_2 \cdot 4H_2O$, $Co(OAc)_2 \cdot 4H_2O$, $Cd(OAc)_2 \cdot 2H_2O$, $Mn(OAc)_2 \cdot 4H_2O$, $Fe(SO4)_2 \cdot 7H_2O$, or $Hg(OAc)_2$ were dissolved in methanol (10 mL) and stirred at room temperature for 15 min to give a clear solution. After standing for 3–5 days, the precipitates were separated by filtration, recrystallized from methanol, washed with methanol three times, and dried in a vacuum desiccator containing anhydrous $CaCl_2$.

4.1.1.1. *3,5-Diiodosalicylalidene-2-morpholinoethylaminozinc(II) (**1**).* Yellow crystals, yield 70%, m.p.: 249–250°C. UV (λ nm): 259; 397. Selected IR data

(cm^{-1}, KBr): 1630(s); 1446(s); 1218(s); 1113(s); 672(s). ESI-MS: 1036.11 ($C_{26}H_{31}O_4N_4I_4Zn^+$, $[M+H]^+$). Anal. Calcd for $C_{26}H_{30}O_4N_4I_4Zn$ (%): C, 30.64; H, 2.92; N, 5.43; Zn, 6.26. Found: C, 30.19; H, 2.90; N, 5.41; Zn, 6.31.

4.1.1.2. *3,5-Diiodosalicylalidene-2-morpholinoethylaminocopper(II)* (***2***). Green crystals, yield 72%, m.p.: 221–223°C. UV (λ nm): 260; 401. Selected IR data (cm^{-1}, KBr): 1628(s); 1434(s); 1220(s); 1112(s); 671(s). ESI-MS: 1034.26 ($C_{26}H_{31}O_4N_4I_4Cu^+$, $[M+H]^+$). Anal. Calcd for $C_{26}H_{30}O_4N_4I_4Cu$ (%): C, 31.04; H, 2.93; N, 5.55; Cu, 6.11. Found: C. 30.21; H, 2.90; N, 5.42; Cu, 6.15.

4.1.1.3. *3,5-Diiodosalicylalidene-2-morpholinoethylaminonickel(II)* (***3***). Green crystals, yield 66%, m.p.: 217–220°C. UV (λ nm): 263; 404. Selected IR data (cm^{-1}, KBr): 1621(s); 1445(s); 1210(s); 1113(s); 671(s). ESI-MS: 1029.32 ($C_{26}H_{31}O_4N_4I_4Ni^+$, $[M+H]^+$). Anal. Calcd for $C_{26}H_{30}O_4N_4I_4Ni$ (%): C, 30.44; H, 2.95; N, 5.51; Ni, 5.65. Found: C, 30.35; H, 2.92; N, 5.44; Ni, 5.71.

4.1.1.4. *3,5-Diiodosalicylalidene-2-morpholinoethylaminocobalt(II)* (***4***). Brown crystals, yield 64%, m.p.: 193–195°C. UV (λ nm): 265; 388. Selected IR data (cm^{-1}, KBr): 1617(s); 1435(s); 1223(s); 1116(s); 681(s). ESI-MS: 1029.36 ($C_{26}H_{31}O_4N_4I_4Co^+$, $[M+H]^+$). Anal. Calcd for $C_{26}H_{30}O_4N_4I_4Co$ (%): C, 31.06; H, 2.94; N, 5.52; Co, 5.67. Found: C, 30.34; H, 2.92; N, 5.43; Co, 5.72.

4.1.1.5. *3,5-Diiodosalicylalidene-2-morpholinoethylaminocadmium(II)* (***5***). Yellow crystals, yield 71%, m.p.: 248–250°C. UV (λ nm): 259; 399. Selected IR data (cm^{-1}, KBr): 1618(s); 1445(s); 1218(s); 1109(s); 669(s). ESI-MS: 1083.04 ($C_{26}H_{31}O_4N_4I_4Cd^+$, $[M+H]^+$). Anal. Calcd for $C_{26}H_{30}O_4N_4I_4Cd$ (%): C, 29.64; H, 2.82; N, 5.11; Cd, 10.32. Found: C, 28.85; H, 2.77; N, 5.17; Cd, 10.38.

4.1.1.6. *3,5-Diiodosalicylalidene-2-morpholinoethylaminomanganese(II)* (***6***). Black crystals, yield 65%, m.p.: 153–156°C. UV (λ nm): 262; 390. Selected IR data (cm^{-1}, KBr): 1610(s); 1423(s); 1220(s); 1113(s); 673(s). ESI-MS: 1025.47 ($C_{26}H_{31}O_4N_4I_4Mn^+$, $[M+H]^+$). Anal. Calcd for $C_{26}H_{30}O_4N_4I_4Mn$ (%): C, 31.16; H, 3.03; N, 5.48; Mn, 5.30. Found: C, 30.46; H, 2.93; N, 5.46; Mn, 5.36.

4.1.1.7. *3,5-Diiodosalicylalidene-2-morpholinoethylaminoiron(II)* (***7***). Black crystals, yield 65%, m.p.: >300°C. UV (λ nm): 260; 427. Selected IR data (cm^{-1}, KBr): 1630(s); 1437(s); 1218(s); 1113(s); 674(s). ESI-MS: 1026.25 ($C_{26}H_{31}O_4N_4I_4Fe^+$, $[M+H]^+$). Anal. Calcd for $C_{26}H_{30}O_4N_4I_4Fe$ (%): C, 31.17; H, 2.95; N, 5.40; Fe, 5.37. Found: C, 30.44; H, 2.92; N, 5.46; Fe, 5.44.

4.1.1.8. *3,5-Diiodosalicylalidene-2-morpholinoethylaminomercury(II)* (***8***). Brilliant yellow crystals, yield 72%, m.p.: 192–194°C. UV (λ nm): 260; 432. Selected IR data (cm^{-1}, KBr): 1603(s), 1443(s), 1215(s), 1118(s), 667(s). ESI-MS: 1171.12 ($C_{26}H_{31}O_4N_4I_4Hg^+$, $[M+H]^+$). Anal. Calcd for $C_{26}H_{30}O_4N_4I_4Hg$ (%): C, 27.20; H, 2.62; N, 4.71; Hg, 17.15. Found: C, 26.67; H, 2.56; N, 4.78; Hg, 17.13.

4.1.1.9. *3,5-Diiodosalicylalidene-3-morpholinopropylaminozinc(II)* (***9***). Yellow crystals, yield 62%, m.p.: 165–167°C. UV (λ nm): 260; 381. Selected IR data

(cm^{-1}, KBr): 1622(s), 1448(s), 1221(s), 1118(s), 669(s). ESI-MS: 1064.02 ($C_{28}H_{35}O_4N_4I_4Zn^+$, $[M+H]^+$). Anal. Calcd for $C_{28}H_{34}O_4N_4I_4Zn$ (%): C, 32.35; H, 3.24; N, 5.22; Zn, 6.21. Found: C, 31.67; H, 3.20; N, 5.27; Zn, 6.15.

4.1.1.10. *3,5-Diiodosalicylalidene-3-morpholinopropylaminocopper(II)* (***10***). Green crystals, yield 67%, m.p.: 170–172°C. UV (λ nm): 263; 379. Selected IR data (cm^{-1}, KBr): 1621(s), 1443(s), 1222(s), 1109(s), 672(s). ESI-MS: 1062.16 ($C_{28}H_{35}O_4N_4I_4Cu^+$, $[M+H]^+$). Anal. Calcd for $C_{28}H_{34}O_4N_4I_4Cu$ (%): C, 32.30; H, 3.26; N, 5.20; Cu, 5.92. Found: C, 31.67; H, 3.20; N, 5.27; Cu, 5.99.

4.1.1.11. *3,5-Diiodosalicylalidene-3-morpholinopropylaminonickel(II)* (***11***). Green crystals, yield 66%, m.p.: 251–253°C. UV (λ nm): 261; 406. Selected IR data (cm^{-1}, KBr): 1618(s), 1450(s), 1224(s), 1118(s), 671(s). ESI-MS: 1057.12 ($C_{28}H_{35}O_4N_4I_4Ni^+$, $[M+H]^+$). Anal. Calcd for $C_{28}H_{34}O_4N_4I_4Ni$ (%): C, 32.64; H, 3.30; N, 5.23; Ni, 5.50. Found: C, 31.82; H, 3.22; N, 5.30; Ni, 5.55.

4.1.1.12. *3,5-Diiodosalicylalidene-3-morpholinopropylaminocobalt(II)* (***12***). Brown crystals, yield 60%, m.p.: 230–232°C. UV (λ nm): 262; 404. Selected IR data (cm^{-1}, KBr): 1616(s), 1445(s), 1223(s), 1118(s), 669(s). ESI-MS: 1057.31 ($C_{28}H_{35}O_4N_4I_4Co^+$, $[M+H]^+$). Anal. Calcd for $C_{28}H_{34}O_4N_4I_4Co$ (%): C, 32.54; H, 3.24; N, 5.25; Co, 5.64. Found: C, 31.81; H, 3.22; N, 5.29; Co, 5.58.

4.1.1.13. *3,5-Diiodosalicylalidene-3-morpholinopropylaminocadmium(II)* (***13***). Yellow crystals, yield 70%, m.p.: 254–256°C. UV (λ nm): 260; 431. Selected IR data (cm^{-1}, KBr): 1620(s), 1448(s), 1218(s), 1120(s), 667(s). ESI-MS: 1110.75 ($C_{28}H_{35}O_4N_4I_4Cd^+$, $[M+H]^+$). Anal. Calcd for $C_{28}H_{34}O_4N_4I_4Cd$ (%): C, 31.24; H, 3.12; N, 5.02; Cd, 10.07. Found: C, 30.28; H, 3.06; N, 5.04; Cd, 10.12.

4.1.1.14. *3,5-Diiodosalicylalidene-3-morpholinopropylaminomanganese(II)* (***14***). Black crystals, yield 64%, m.p.: 145–148°C. UV (λ nm): 260; 430. Selected IR data (cm^{-1}, KBr): 1618(s), 1438(s), 1223(s), 1101(s), 678(s). ESI-MS: 1053.26 ($C_{28}H_{35}O_4N_4I_4Mn^+$, $[M+H]^+$). Anal. Calcd for $C_{28}H_{34}O_4N_4I_4Mn$ (%): C, 32.64; H, 3.26; N, 5.27; Mn, 5.16. Found: C, 31.93; H, 3.23; N, 5.32; Mn, 5.22.

4.1.1.15. *3,5-Diiodosalicylalidene-3-morpholinopropylaminoiron(II)* (***15***). Brown crystals, yield 62%, m.p.: 198–201°C. UV (λ nm): 262; 429. Selected IR data (cm^{-1}, KBr): 1609(s), 1431(s), 1219(s), 1114(s), 678(s). ESI-MS: 1054.22 ($C_{28}H_{35}O_4N_4I_4Fe^+$, $[M+H]^+$). Anal. Calcd for $C_{28}H_{34}O_4N_4I_4Fe$ (%): C, 32.56; H, 3.27; N, 5.28; Fe, 5.22. Found: C, 31.91; H, 3.23; N, 5.31; Fe, 5.30.

4.1.1.16. *3,5-Diiodosalicylalidene-3-morpholinopropylaminomercury(II)* (***16***). Brilliant yellow crystals, yield 72%, m.p.: 183–185°C. UV (λ nm): 262; 422. Selected IR data (cm^{-1}, KBr): 1607(s), 1455(s), 1215(s), 1120(s), 664(m). ESI-MS: 1198.01 ($C_{28}H_{35}O_4N_4I_4Hg^+$, $[M+H]^+$). Anal. Calcd for $C_{28}H_{34}O_4N_4I_4Hg$ (%): C, 28.62; H, 2.80; N, 4.60; Hg, 16.78. Found: C, 28.05; H, 2.84; N, 4.67; Hg, 16.73.

4.2. *Antibacterial activity*

The antibacterial activity of the synthesized complexes was tested against *B. subtilis*, *S. aureus*, *S. faecalis*, *P. aeruginosa*, *E. coli*, and *E. cloacae* using MTT medium. The MICs of the test complexes were determined by a colorimetric method using the dye MTT [12]. A stock solution of the synthesized complex (50 μg mL^{-1}) in DMSO was prepared and the test complexes were incorporated in specified quantity of sterilized liquid medium. A specified quantity of the medium containing the complex was poured into microtitration plates. Suspension of the microorganism was prepared to contain $\sim 10^5$ cfu mL^{-1} and applied to microtitration plates with serially diluted complexes in DMSO to be tested and incubated at 37°C for 24 h for bacterial. After the MICs were visually determined on each of the microtitration plates, 50 μL of PBS containing 2 mg mL^{-1} of MTT was added to each well. Incubation was continued at room temperature for 4–5 h. The content of each well was removed and 100 μL of isopropanol containing 5% 1 mol L^{-1} HCl was added to extract the dye. After 12 h of incubation at room temperature, the optical density (OD) was measured with a microplate reader at 570 nm. The observed MICs are presented in table 3.

4.3. *Crystal structure determination*

Crystal structure determination of **1** and **2** were carried out on a Nonius CAD4 diffractometer equipped with graphite-monochromated Mo-Kα ($\lambda = 0.71073$ Å) radiation. The structure was solved by direct methods and refined on F^2 by full-matrix least-squares using SHELX-97 [13]. All nonhydrogen atoms were refined anisotropically. Hydrogens were placed in calculated positions and assigned fixed isotropic thermal parameters at 1.2 times the equivalent isotropic U of the atoms to which they are attached and allowed to ride on their respective parent atoms. Contributions of these hydrogens were included in the structure-factor calculations. Crystallographic and experimental data for **1** and **2** are given in table 1. Selected bond lengths (Å) and angles (°) are illustrated in table 2. Crystallographic data have been deposited with the Cambridge Crystallographic Data Centre (CCDC 675135 for **1** and CCDC 675134 for **2**).

Acknowledgment

The work was financed by a grant (Project 30772627) from National Natural Science Foundation of China.

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