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Synthesis, Characterization and Biological Activities of Hydrazone Schiff Base and its Novel Metals Complexes

(Sintesis, Pencirian dan Aktiviti Biologi Hidrazon Pangkal Schiff serta Kompleks Baru Logamnya)

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ABSTRACT

Recently, researchers are focusing on the synthesis of Schiff base complexes due to their promising biological activities and frequent use in the pharmaceuticals. In the present study we synthesized Schiff base of salicyldehyde and 2, 4-dinitrophenylhydrazine by modified method and its novel metal complexes with Cu (II), Zn (II), Co (II), Ni (II), Mn (II), V (II) and AU *(IIII). For spectroscopic studies and characterization,* UV*-Vis and* FT-IR *spectroscopy were used. The Schiff base and its metals complexes were tested for antimicrobial, cytotoxic and antioxidant activities. Significant results are showed by the all synthesized complexes.*

Keywords: Biological activities; IR *spectroscopy; Schiff base;* UV*-visible*

ABSTRAK

Baru-baru ini penyelidik menumpukan perhatian kepada sintesis kompleks pangkal Schiff disebabkan oleh aktiviti biologi yang berpotensi dan kerap digunakan dalam bidang farmaseutis. Dalam kajian ini, Schiff pangkal salisildehid dan 2, 4-dinitrofenilhidrazin disintesis menggunakan kaedah ubah suai dan kompleks baru logamnya dengan Cu (II), Zn (II), Co (II), Ni (II), Mn (II), V (II) dan AU *(IIII). Untuk kajian spektroskopi dan pencirian,* UV*-Vis dan Spektroskopi* FT-IR *telah digunakan. Pangkal Schiff dan kompleks logam telah diuji untuk aktiviti antimikrob, sitotoksik dan antioksidan. Hasil ketara telah ditunjukkan oleh semua kompleks yang telah disintesis.*

Kata kunci: Aktiviti biologi; pangkal Schiff; spektroskopi IR*;* UV *nampak*

INTRODUCTION

Hydrazones are the derivatives of hydrazine and an essential group of drugs. Currently, they have been focused to, due to their wide range of pharmaceutical applications (Zahid et al. 1997). They have general formula R-NH-N=CRR”, synthesized through reduction reaction of hydrazine with carbonyl compounds (Hany et al. 2015). Hydrazones are more reactive than carbonyl groups (aldehyde or ketone) due to the presence of alpha active hydrogen atoms. They are used as precursor in organic reactions (Florian et al. 2006), particularly in the preparation of Schiff base ligand (Zeyrek et al. 2006) which formed coordination bond with transition metals *in vivo*, with promising biological properties (Savini et al. 2002), and also used as ligands in coordination chemistry. It is reported in the literature that hydrazones complexes employed in medicines also (Hajar et al. 2010; Ochiai 1977). Their complexes with different metals showed anti-tuberculosis (Katyal & Dutt 1975) herbicidal, insecticidal, and nematocidal activities (Amrata et al. 2014; Li-Fei et al. 2012; Rajarajan et al. 2016). It is also reported that hydrazones Schiff base and their metal complexes with various transition metals possess anti-analgesic, anti-inflammation, antibacterial, antifungal and anticancer activities (Hassan et al. 2007; Jin et al. 2006). On the basis of the cited valuable outcome and their vital role in medicine and wide applications in the pharmaceuticals, as therapeutic agents in the oxidation-reduction process in the chemical industries, in the oxidation-reduction reactions of different functional groups in organic chemists, used as insecticidal and agrochemicals in parts. The current initiative was therefore undertake, we synthesized salicyldehyde-2, 4-dinitrophenylhydrazone and its novel metals complexes Cu (II), Zn (II), Co (II), Ni (II), Mn (II), V (II) and AU (IIII).

EXPERIMENTAL DETAILS

PREPARATION OF SDNPH LIGAND ($C_{13}H_{10}N_4O_5$)

The Schiff base was synthesized through similar procedure cited in the literature (Shahawi et al. 2013; Silverstein & Bassler 1981) by the condensation reaction of salicyldehyde (0.414 mL, 3.9 mmol) with 2, 4-dinitrophenylhydrazine (0.594 g, 3 mmol) in 10 mL of ethanol (Scheme 1). The resultant mixture was stirred for 3 h at 25°C and monitored by TLC. It gives orange color precipitate, which was filtered, rinsed with EtOH and dried.

PREPARATION OF METALS COMPLEXES

All the metals complexes were synthesized by the reaction of salicyldehyde 2, 4-dinitrophenylhydrazone (SDNPH) schiff base with metals salts (in the form of sulphate and chloride). SDNPH schiff base (0.4 m mol) was dissolved in 3 mL of ethanol and was added dropwise to the (0.2 m mol) metal solution (ethanol). The reactions mixture was stirred for 5-15 h at room temperature with a small amount of NaOH in ethanol. Different colors precipitates were obtained which was filtered, washed with EtOH and dried. The Proposed Structure of M (II) and M (III) metal complexes are shown in Scheme 2.

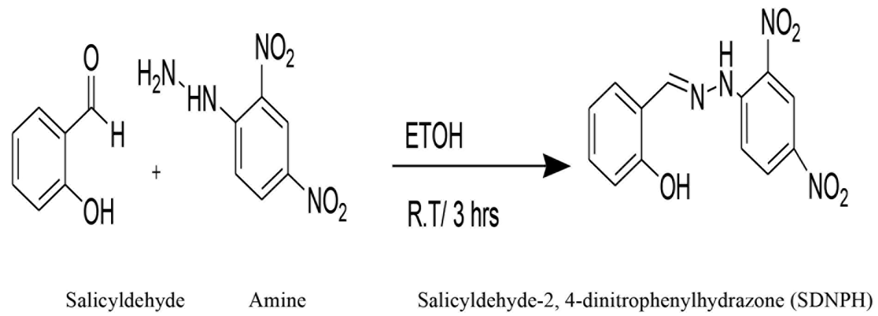
RESULTS AND DISCUSSION

All the synthesized compounds were of different color, stable and insoluble in polar solvents but readily soluble in DMF and DMSO. They do not have sharp melting point. The physicals properties are given in the Tables 1 and 2.

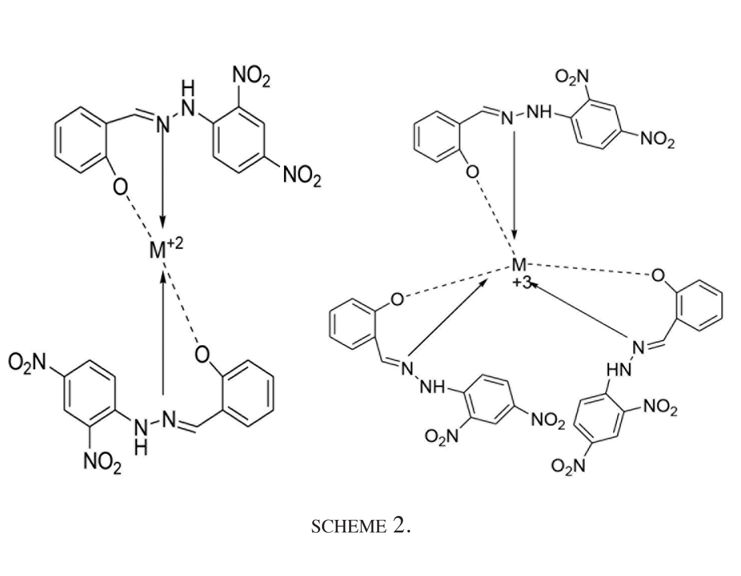
INFRARED SPECTROSCOPY

Infrared spectroscopy is a very useful method for the structure elucidation of compounds, that is ligands and their complexes to show the moiety bonded to the center metals atoms subsequently. The infrared records for the Schiff base ligand and its compounds are given in Table 3. Infrared bands of Schiff base and its complexes were weighed in the region of 4000-400 cm^{-1} to find out the coordination of ligand to metals ions. The difference between the ligand spectrum and its complexes showed coordination of Schiff base with metals ions. Salicyldehyde-2, 4-dinitrophenylhydrazone Schiff base show a characteristic band of ν(OH) of phenol group which reduced due to inter molecular H-bonding, is observed at 3300-3620 cm^{-1} in the spectrum of ligand. In addition, followed by 3273, 1616.79, 1513.41, 1334.55, 1272.00 and 1085.80 regions assigned to N-H, C=N, NO_2, Ar-O and N-N stretching vibration (Shahawi et al. 2013; Silverstein & Bassler 1981). The vanishing of the OH broad peak in all coordination compound confirmed M-O bond after complexation (Leniec et al. 2006). The absorption band at 1616.79 allocated to C=N of Schiff base, changing in frequency on complexation with metals ion representing the coordination of imine N to M formed M-N bond (Kaczmarek et al. 2009; Misbah et al. 2013).

IR spectral shift of different compounds are comparable, representing like structures of the metals complexes. The new bands in range of complexes were observed for ν(M-O) and ν(M-N) stretching at region of 660-688 and 520-547 cm^{-1} (Guo et al. 2006) confirmed the coordination of Schiff base with metals ions. Spectral data shows that all the coordination compounds are



SCHEME 1.



SCHEME 2.

TABLE 1. Physicals properties of Schiff base and its complexes

Compounds	M. Mt	Color	M. Point	%Yield
$C_{13}H_{10}N_4O_5$	302	Orange	257	80.09
$Cu(C_{13}H_{10}N_4O_5)_2$	665.54	Light pink	268	63.05
$Ni(C_{13}H_{10}N_4O_5)_2$	660.69	Brown	>300	64.44
$Zn(C_{13}H_{10}N_4O_5)_2$	667.93	Yellowish	287	65.72
$Co(C_{13}H_{10}N_4O_5)_2$	660.93	Light orange	246	68.37
$Mn(C_{13}H_{10}N_4O_5)_2$	656.93	Dark pink	>300	69.30
$Au(C_{13}H_{10}N_4O_5)_3$	798.97	Brick red	265	62.98
$Cd(C_{13}H_{10}N_4O_5)_2$	714.41	Red	273	57.98
$V(C_{13}H_{10}N_4O_5)_2$	652.49	Reddish orange	260	55.79

TABLE 2. Solubility of Schiff base and its complexes

Solvents	Complexes								
	S.B	Cu	Ni	Co	Zn	Mn	Au	Cd	V
n-Hexane	N	N	N	N	N	N	N	N	N
Ethanol	P	P	P	P	P	N	P	CS	CS
Diethyl ether	P	P	P	P	P	P	P	P	CS
Chloroform	CS	P	P	CS	P	P	CS	CS	CS
THF	CS	CS	P	CS	CS	P	CS	CS	CS
Ethyl acetate	CS	CS	P	CS	P	P	CS	CS	CS
Methanol	CS	CS	P	CS	CS	P	CS	CS	CS
DMSO	CS	CS	CS	CS	CS	CS	CS	CS	CS
DMF	CS	CS	CS	CS	CS	CS	CS	CS	CS
Water	N	P	P	P	N	P	N	N	N

N = insoluble, P = partially soluble, CS = completely soluble, S.B = Schiff base

TABLE 3. IR data of Schiff base and its metals complexes

Compounds	ν OH	ν N-H	ν C=N	ν NO_2		ν Ar-O	ν N-N	ν M-N	ν M-O
				ν_1	ν_2				
$C_{13}H_{10}N_4O_5$	3300-3520	3273	1616.79	1513.41	1334.55	1272.00	1085.80	-----	-----
$Ni(C_{13}H_{10}N_4O_5)_2$	------	3275	1615.00	1513.19	1334.00	1272.66	1144.27	660	520
$Co(C_{13}H_{10}N_4O_5)_2$	------	3270	1616.48	1514.49	1335.07	1270.45	1144.66	663	533
$Cu(C_{13}H_{10}N_4O_5)_2$	-------	3273	1616.48	1510.10	1334.54	1260.73	1056.99	670	524
$Zn(C_{13}H_{10}N_4O_5)_2$	-------	3272	1616.33	1513.66	1334.45	1268.00	1135.23	688	545
$Mn(C_{13}H_{10}N_4O_5)_2$	-------	3265	1615.00	1513.66	1334.00	1272.66	1144.27	673	528
$Cd(C_{13}H_{10}N_4O_5)_2$	-------	3269	1615.21	1509.21	1332.75	1270.05	113368	677	543
$V(C_{13}H_{10}N_4O_5)_2$	-------	3267	1615.55	1511.69	1333.17	1272.13	1134.74	681	547
$Au(C_{13}H_{10}N_4O_5)_3$	-------	3260	1615.97	1513.22	1334.02	1272.64	1144.68	678	540

octahedral. The Schiff base is bidentate. Each metal ion coordinating to the two C=N azomethine moieties and two oxygen atoms from each hydroxyl group of phenol to completed the octahedral structure.

ELECTRONIC SPECTROSCOPY

The ultraviolet absorption spectrums of the prepared compounds took in DMSO at 25°C. The data of the maximum absorption wavelength (λ max) are given in

Table 4. The characteristic bands of SDNPH Schiff base assigned at 275, 296 and 405 nm, respectively. The higher energy band represent electron transition π→π* in benzene sphere and imine or azomethene moiety (Felico et al. 2001; Guo et al. 2006). The lower energy transition represents the n→π* transition of N atom p-orbital lone pair in imine and benzene sphere (Youssef et al. 2008). The ultraviolet absorption spectrums of all the coordination compounds Ln (II) and Ln (III) Schiff base metals complexes show resemblance which represents alike structures are given in the Table 4. The difference between the spectrums of the complexes and ligand after complexation confirmed coordination bond. Further changing in the wavelength also indicated that the coordination of the schiff base occurred to the metals ions.

ANTIBACTERIAL STUDY

The antibacterial activity was evaluated by *in vitro* method against four bacteria strains, gram positive *B. subtilis* and *S. aureus* and gram negative *E. coli* and *S. typhi* bacteria. The two concentrations (1 and 2 mg/mL) of each sample were used against tested microorganism through agar well diffused assay. In well diffused method, diffusion of microorganism occurred on nutrient agar medium plates and incubated for 24 h at 37°C. The inhibitory zone of microorganism growth around the well was measured in millimeter. The data of the antibacterial activity of all the compounds are given in Table 5. All the metals complexes showed high activity than Schiff base, it is due to the coordination of Schiff base with metals which enhance their activities significantly (Chohan et al. 2004). Among the metals complexes Copper complex showed high activity against *E. coli* and *S. typhi*. Zinc complex showed activity against *B. subtilis.* The tetracycline was used as standard for *E. coli, S. typhi* and cephradine used for *B. subtilis* and *S. aureus*.

ANTIFUNGAL ACTIVITY

Antifungal study was evaluated against four fungus species, *C. albicans, R. stolonifer, T. viride* and *A. nigar*, in their *in vitro* method through agar well diffused standard protocol. For this purpose, potato dextrose agar was used as medium. In this method, each species of fungus was streaked on potato dextrose agar medium plates and incubated for 48 h at 37°C. Two different concentrations of 1 and 2 mg/mL were applied. The activity was determined by the calculation of zones of inhibition in (mm) are tabulated in the Table 6. Schiff base showed less antifungal activity, while the metals complexes showed high activities. The experimental data was evaluated with standard antifungal medicine fluconazole under similar condition followed in tested compounds.

Copper complex showed high activity among the metals complexes against *C. albicans* followed by *R. stolonifer, T. viride* and *A. nigar*. Nickel complex also showed high activity toward *C. albicans* and *R. stolonifer* than other meal complexes.

The results showed that some metals complexes show high activities as compared of Schiff base, however, it is also known that Schiff base containing N and O donor atoms increased its activity through complexation with metals ions (Arulpriya et al. 2010; Chohan et al. 2002).

ANTI-OXIDANT STUDY

The DPPH free radical protocol was employed to study the standard free radical potentials. The antioxidant activity of Schiff base salicylalde-2, 4-dinitrophenylhydrazone and its metals complexes were analyzed by *in-vitro* method. Different concentrations (10, 20, 30, 40 & 50 ppm) of tested compounds were used for the evaluation of antioxidant activity. Ascorbic acid was used as a standard which show 100% activity. The scavenging reaction between (DPPH) free radical and an antioxidant (H-D) can be written as:

$$\underset{\text{(Purple)}}{\text{(DPPH)}} + \text{(H-D)} \rightarrow \underset{\text{(Yellow)}}{\text{DPPH-H}} + \text{(D)}$$

The results of free radical scavenging activities of Cu, Ni, Cd, and Au Complexes and V, Mn, Co, Zn are given in the Figures 1 and 2, respectively. Ligand showed less activity while on complexation with metals increased the

TABLE 4. UV-visible spectral data wavelength (nm) for the Schiff base and its complexes

S. No	Compounds	Wavelength(nm)	Band assignment
1	$C_{13}H_{10}N_4O_5$	275, 296, 405	π→ π*, n→ π*
2	$Ni(C_{13}H_{10}N_4O_5)_2$	339, 350, 470	π→ π*, n→ π*
3	$Co(C_{13}H_{10}N_4O_5)_2$	297, 315, 460	π→ π*, n→ π*
4	$Cu(C_{13}H_{10}N_4O_5)_2$	312, 319, 465	π→ π*, n→ π*
5	$Zn(C_{13}H_{10}N_4O_5)_2$	325, 339, 474	π→ π*, n→ π*
6	$Mn(C_{13}H_{10}N_4O_5)_2$	343, 360, 500	π→ π*, n→ π*
7	$Cd(C_{13}H_{10}N_4O_5)_2$	300, 360, 484	π→ π*, n→ π*
8	$V(C_{13}H_{10}N_4O_5)_2$	285, 304, 495	π→ π*, n→ π*
9	$Au(C_{13}H_{10}N_4O_5)_3$	291, 331, 447	π→ π*, n→ π*

TABLE 5. Zone of inhibition for antibacterial study

Compounds	Gram positive				Gram negative			
	B. subtilis		*S. aureus*		*E. coli*		*S. thypi*	
	1 mg/mL	2 mg/mL	1 mg/mL	2 mg/mL	1 mg/mL	2 mg/mL	1 mg/mL	2 mg/mL
$C_{13}H_{10}N_4O_5$	07	08	00	00	00	00	00	00
$Cu(C_{13}H_{10}N_4O_5)_2$	12	16	11	15	8	12	08	10
$Ni(C_{13}H_{10}N_4O_5)_2$	08	10	10	12	07	09	06	09
$Zn(C_{13}H_{10}N_4O_5)_2$	09	11	11	13	10	12	07	09
$Co(C_{13}H_{10}N_4O_5)_2$	00	06	07	10	00	00	00	00
$Mn(C_{13}H_{10}N_4O_5)_2$	00	06	08	10	06	07	05	06
$Au(C_{13}H_{10}N_4O_5)_3$	06	09	07	09	00	00	*06*	00
$Cd(C_{13}H_{10}N_4O_5)_2$	06	08	07	09	00	00	00	00
$V(C_{13}H_{10}N_4O_5)_2$	00	06	00	07	00	00	00	00
Tetracycline	20	22	20	22	----	----	----	---
Cephradine	-----	-----	-----	-----	16	18	15	17

TABLE 6. Zone of inhibition for anti-fungus study

Compounds	*C. albicans*		*R. stolonifer*		*T. viride*		*A. nigar*	
	1 mg/mL	2 mg/mL	1 mg/mL	2 mg/mL	1 mg/mL	2 mg/mL	1 mg/mL	2 mg/mL
$C_{13}H_{10}N_4O_5$	00	00	00	00	00	00	00	00
$Cu(C_{13}H_{10}N_4O_5)_2$	14	16	12	14	11	12	10	11
$Zn(C_{13}H_{10}N_4O_5)_2$	08	10	07	19	08	09	06	08
$Ni(C_{13}H_{10}N_4O_5)_2$	12	13	10	12	07	09	06	08
$Co(C_{13}H_{10}N_4O_5)_2$	08	09	07	09	06	08	00	00
$Mn(C_{13}H_{10}N_4O_5)_2$	08	10	06	08	07	09	06	07
$Au(C_{13}H_{10}N_4O_5)_2$	06	08	06	09	06	07	05	06
$Cd(C_{13}H_{10}N_4O_5)_2$	06	07	05	06	00	06	00	05
$V(C_{13}H_{10}N_4O_5)_2$	05	06	00	06	06	07	06	06
Fluconazole	19	22	17	20	16	20	15	20

activity significantly. All the metal complexes showed comparable or less activity than that of standard (Ascorbic acid). The high antioxidant potential of these complexes can be recognized to the electron retreating consequences of the Cu(II), Ni(II), Zin(II) and Cd(II) ions which assisted the liberate of hydrogen to squeeze the DPPH free radical (Tetteh et al. 2014).

CYTOTOXICITY

Cytotoxicity was evaluated through brine shrimp lethality assay. Different concentrations (1000, 250 & 100 µg/mL) of each sample were tested against 10 naupuli in vial. Blank solution of DMSO was used as negative control and doxorubicin 4 mg/mL was used as positive control. After the completion of incubation period, dead naupli were counted in each vial. The results found were promising given in the Table 7. Metals complexes showed high percent mortality as compared to ligand, respectively (Tarafder et al. 2000). These finding matched with the previous studied showed that the metals, complexes of Copper, Nickel, Zinc and Gold showed 100% mortality at all concentrations (Afrasiabi et al. 2005).

CONCLUSION

The results showed that the synthesized compounds showed significant antimicrobial (antibacterial and antifungal), antioxidant and cytotoxic activities. Schiff base showed less biologicals activities than metals complexes. Among the metals complexes Cu (II), Zinc (II), Co (II) and Ni (II) were found to show high activities than other metals complexes.

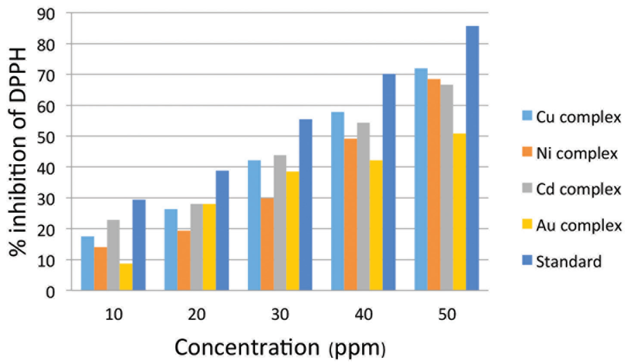


FIGURE 1. Antioxidant potential of complexes of Cu, Ni, Cd, Au and standard

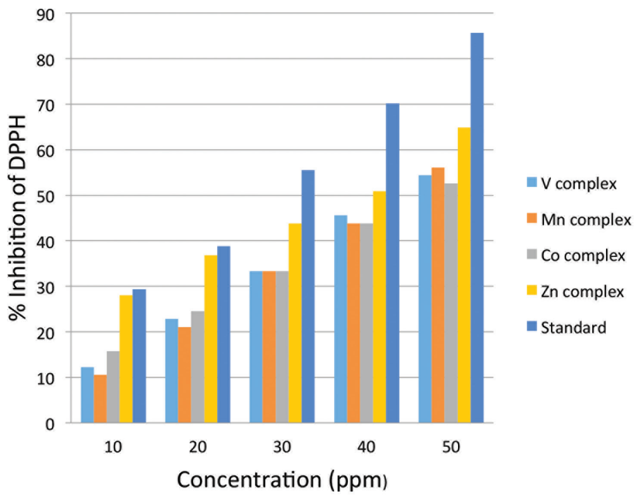


FIGURE 2. Antioxidant potential of complexes of V, Mn, Co, Zn and standard

TABLE 7. % Mortality of Schiff base and its complexes

Compounds	% Mortality		
	100 μg/mL	250 μg/mL	1000 μg/mL
$C_{13}H_{10}N_4O_5$	50	50	60
$Cu(C_{13}H_{10}N_4O_5)_2$	100	100	100
$Ni(C_{13}H_{10}N_4O_5)_2$	100	100	100
$Zn(C_{13}H_{10}N_4O_5)_2$	100	100	100
$Co(C_{13}H_{10}N_4O_5)_2$	70	70	80
$Mn(C_{13}H_{10}N_4O_5)_2$	70	70	80
$Au(C_{13}H_{10}N_4O_5)_3$	100	100	100
$Cd(C_{13}H_{10}N_4O_5)_2$	60	60	80
$V(C_{13}H_{10}N_4O_5)_2$	40	40	60

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