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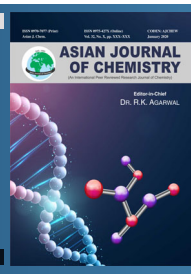

Spectroscopic, Thermal and Antimicrobial Study of Some Transition Metal(II) Complexes of β-Diketones

Rekha Rani[1], Umesh Kumar[1], Deepak Kumar[1], Siteshwar Pd. Yadav[2], Arjun Kr. Sinha[1] and S. Sharma[1,*]

[1]University Department of Chemistry, Magadh University, Bodh Gaya-824234, India
[2]Department of Chemistry, R.L. College, Madhawnagar-854205, India

*Corresponding author: E-mail: sharma.shivadhar@gmail.com

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A novel heterocyclic based β-diketone has been synthesized by the condensation of pyrazole-1-acetylchloride and sodium acetophenone. The product 1-phenyl-4-(pyrazole-1-yl)butane-1,3-dione has been used for complexation with Mn(II), Fe(II) and Co(II) metal ions. The ligand β-diketone is found to exist in keto-enol tautomeric forms. The comparision of infrared spectra of complexes clearly indicates the coordination of the ligand from its enolic form. The magnetic moment and electronic spectra of all the metal complexes leads to the distorted octahedral symmetry around the metal ion. The ligand as well as its metal complexes has been screened for antimicrobial activity and antifungal activities. It is observed that the antimicrobial and antifungal activities get enhanced in complexes in respect to the free ligand.

Keywords: β-Diketone, Pyrazole-1-acetylchloride, Sodium acetophenone, Transition metal(II) complexes, Distorted octahedral.

INTRODUCTION

The development of appropriate synthesis of mononuclear and multinuclear metal systems from building blocks of β-diketones possessing high spin ground state is a stimulating and promising field to be pursued by inorganic chemists. The use of wide range of radicals as nitrosyl nitroxide ligands which act as magnetic modulators for β-diketonate complexes and property of the resulting complexes which give rise to single chain magnets are better understood and clarified as well [1-5]. This has opened new perspectives in the field of β-diketone complexes. β-Diketone ligands have been found to coordinate in the equatorial plane of metal ions giving rise to quite flat complexes, which can contain coordinating solvents or monodentate ligands in the axial positions [6]. The growing interest in the transition metal deviation of β-diketones during last two decades is also due to their medicinal importance which have been revealed by several articles [7-10]. The research in this field has been stimulated by the versatility of metal complexes of β-diketones and their derivatives as NMR shift reagent, anticancer, antitumor, antioxidant agents [11-13].

Recently some reports the applications of transition metal complexes with β-diketones in chemical and photochemical catalysis [14-16]. Couto-Almeida *et al*. [17] have also reported the use of β-diketones and their complexes to design many lumine-scent compound and also to inhibit the growth of K562 cell. Particularly the β-diketone containing *N*-heterocyclic moiety *viz*. pyrazoles have been found to possess interesting pharma-cological activities like analgesic, antihypertensive, anti-inflammatory, *etc*. [18-23]. Pyrazole derivatives are also known to show a wide range of biological property including high affinity and selectivity towards Adenosine receptor antagonist [24]. To our best of knowledge, the literature revealed that no work on the metal complexes of β-diketones containing pyrazole moiety has been reported. It promoted us to pursue our work on metal complexes of pyrazole containing β-diketone, which is being reported herein.

EXPERIMENTAL

All the reagents used were of Anal.R grade. The ligand 1-phenyl-4-(pyrazole-1-yl)butan-1,3-dione was synthesized as reported method [24], which involves three steps. In first step, pyrazole-1-acetyl chloride was synthesized by reacting pyrazole and monochloroacetic acid in presence of sodium methoxide. In second step, pyrazole-1-acetic acid was treated with thionyl

chloride to form pyrazole-1-acetylchloride. Finally, in the third step, pyrazole-1-acetyl chloride was taken with sodium salt of acetophenone in toluene and the mixture was stirred using magnetic stirrer for 1 h and thereafter it was refluxed on water bath for 10-12 h. The reaction mixture was then kept in refrigerator for overnight and the excess toluene was removed. It was filtered on suction pump and recrystallized from ethanol. (**Scheme-I**).

The synthesized ligand 1-phenyl-1(pyrazole-1-yl)butane-1,3-dione hence forth abbreviated as PPBD has been used for complexation with Mn(II), Fe(II) and Co(II) metal ions by usual method of refluxing the metal salts with ligand taken in 1:2 molar ratio in ethanolic medium for 3-4 h. Pyridine and β-picoline have been used as secondary ligands.

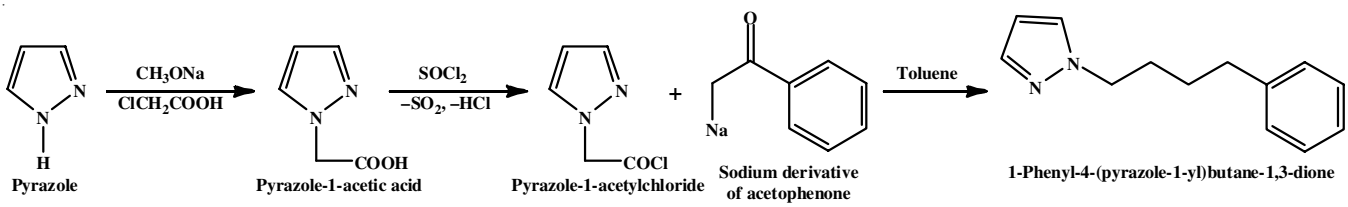
The elements *viz*. carbon, hydrogen and nitrogen were determined microanalytically on Carlo-Erva 1106 chemical analyzer, while the estimation of metals was carried out gravimetrically. The IR spectra of ligand and complexes were recorded on Perkin-Elmer FTIR spectrometer spectrum 2 using KBr disc method in the range of 4000-400 cm^{-1}. The molar conductance of complexes in DMF solution with 10^{-3} M concentration was determined using Elico Conductivity Bridge with dip type conductivity cell at room temperature. The magnetic moment of complexes was determined on Gouy balance at room temperature using $Hg[Co(CNS)_4]$ as calibrating agent and electronic spectra of complexes were recorded on Perkin-Elmer 137 spectrophotometer in DMSO solution.

Biological evaluation: The ligand 1-phenyl-4-(pyrazole-1-yl)butane-1,3-dione (PPBD) and its complexes with Mn(II), Fe(II) and Co(II) were screened for their *in vitro* antibacterial activity against bacteria *S. aureus* and *E. coli* by disc diffusion method using nutrient agar as medium and streptomycin as control. The *in vitro* antifungal activities of the ligand and its complexes were tested against the fungi *A. niger* and *C. albicans* by well diffusion method on potato dextrose agar as the medium and miconazole as control. The compounds were dissolved in DMSO and solution of different concentrations 25, 50 and 100 ppm were prepared separately. The well made on agar medium was inoculated with microorganism. The well was filled with the test solution using a micropipette and the plate was incubated 24 h for bacteria at 35 and 72 h for fungi at 30 ºC. The inhibition zone was developed at which the concentration was noted down. All the determinations were performed thrice.

RESULTS AND DISCUSSION

On the basis of microanalysis the percentage composition of the ligand as well as its complexes has been tabulated in Table-1. The low molar conductivity value (8.9-9.2 Ω^{-1} cm^{-1} mol^{-1}) in 10^{-3} M DMF solution clearly indicates their non-electrolytic nature [25,26]. On the basis of analytical data, the general formula of the complexes may be given as $[M(PPBD)_2X_2]$ where M = Mn(II), Fe(II) and Co(II), X = H_2O, Py and α-picoline.

IR studies: The ligand consists of three parts: (i) pyrazole end, (ii) benzene end, and (iii) the groups present between pyrazole and benzene rings. A weak and sharp band observed at 3110 cm^{-1} is assigned to ν(C-H) stretching of five membered ring [27]. The bands at 1535 cm^{-1} (w), 1450 cm^{-1} (m) and 1390 cm^{-1}(m) are safely assigned to ring vibration of five membered pyrazole ring containing two heteroatoms [28,29]. These bands in the IR spectra of free ligand are good evidence in the favour of the presence of pyrazole moiety in the ligand. The weak band appeared at 3050 cm^{-1} is assigned to ν(C-H) stretching vibration of benzene ring [30]. The weak and medium bands at 1590 and 1500 cm^{-1} are the two characteristic bands for the skeletal vibration of benzene ring. The presence of benzene ring in the ligand is further confirmed by the appearance of a

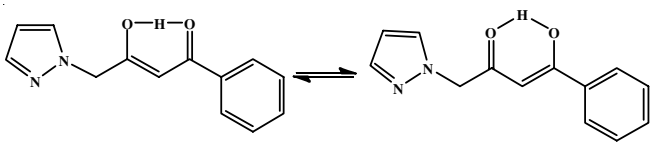


Scheme-I

TABLE-1
% COMPOSITION OF LIGAND AND COMPLEXES

Compounds	Elemental analysis (%): Found (calcd.)				Λ_m (Ohm^{-1} cm^{-1} mol^{-1})	μ_{eff} (BM)
	M	C	H	N		
PPBD ($C_{13}H_{12}N_2O_2$)	–	68.65 (68.42)	5.11 (5.26)	12.10 (12.28)	–	–
$[Mn(PPBD)_2(H_2O)_2]$	9.87 (10.09)	57.39 (57.25)	4.52 (4.77)	10.05 (10.275)	9.1	5.84
$[Mn(PPBD)_2(Py)_2]$	8.10 (8.24)	64.88 (64.76)	4.54 (4.79)	12.31 (12.59)	8.9	5.85
$[Mn(PPBD)_2(\alpha\text{-pico})_2]$	7.79 (7.91)	65.82 (65.61)	5.06 (5.17)	11.88 (12.08)	8.9	5.81
$[Fe(PPBD)_2(H_2O)_2]$	10.01 (10.26)	57.32 (57.14)	4.61 (4.76)	10.00 (10.25)	9.0	5.09
$[Fe(PPBD)_2(Py)_2]$	8.12 (8.38)	64.79 (64.67)	4.62 (4.79)	12.34 (12.57)	9.0	5.10
$[Fe(PPBD)_2(\alpha\text{-pico})_2]$	7.88 (8.045)	65.70 (65.52)	4.91 (5.17)	11.84 (12.068)	9.1	5.10
$[Co(PPBD)_2(H_2O)_2]$	10.52 (10.746)	57.00 (56.83)	4.43 (4.735)	9.92 (10.20)	9.2	4.80
$[Co(PPBD)_2(Py)_2]$	8.51 (8.79)	64.56 (64.38)	4.50 (4.77)	12.38 (12.52)	9.0	4.83
$[Co(PPBD)_2(\alpha\text{-pico})_2]$	8.39 (8.44)	65.42 (65.24)	4.93 (5.15)	11.88 (12.02)	8.9	4.81

weak and sharp band at 775 cm^{-1} due to out of plane bending vibration δ(C-H) of mono substituted benzene ring [31]. These IR bands provide strong evidence in the favour of the presence of the monosubstituted benzene ring in the ligand . The ligand is expected to absorb strongly in the region 1725-1700 cm^{-1} due to the presence of two carbonyl groups [32]. In the present study, however , the ligand doesn't display any band with great intensity in this range. It clearly indicates that the ligand doesn't exist in keto form. The IR spectrum of the ligand displays slightly a broad band at 1640 cm^{-1} with medium intensity which may be assigned to carbonyl vibration ν(C=O) of the ligand. In respect of the normal carbonyl vibration there is a greater reduction in its vibrational absorption frequency which may be attributed to the fact that the double bond character of C=O has undergone reduction by resonance.



The resonance effect is generally described as conjugate chelation to differentiate it from a normal hydrogen bonding effect [33]. This effect is further substantiated by the appearance of a broad band at 2705 cm^{-1} which is attributed to chelate -OH group [34-36]. It shows that the ligand exists in the enol form which is further substantiated by the appearance of a medium band at 1050 cm^{-1} and a weak band at 1510 cm^{-1} which are assigned to ν(C-O) stretching and ν(C=C) stretching vibrations, respectively [37-40]. A weak and sharp band appearing at 2850 cm^{-1} due to νCH_2 stretching vibration clearly indicates the presence of methylene group in the ligand, which is further confirmed by the appearance of a medium band at 1470 cm^{-1} due to δCH_2 group [41-43]. The IR spectra of the metal(II) complexes in comparison to the free ligand reveals the following facts about the coordination sites of the ligand. The band appearing at 2705 cm^{-1} in the IR spectra of the free ligand is absent in all of its complexes. It shows the deprotonation of -OH group and coordination through deprotonated oxygen [44-46]. The coordination through deprotonated enolic hydroxyl group is further supported by the slight increase in the absorption frequency of ν(C–O) stretching, which appears at 1055-1050 cm^{-1} in the IR spectra of complexes. Neither ν(N–N) stretching frequency of pyrazole ring (1010 cm^{-1}) nor ν(C=C-C=N) vibration (1605 cm^{-1}) of pyrazole ring undergoes appreciable change in the IR spectra of complexes. It is indicative of non-participation of endocyclic nitrogen atoms in complexation.

The other major change in the IR spectra of free ligand is observed in the stretching vibration of ν(C=O) group which undergoes red shift in the spectra of complexes and appears at around 1610 cm^{-1}. Obviously, it shows the coordination through carbonyl oxygen. The coordination through carbonyl oxygen is further confirmed by the appearance of new band 475-467 cm^{-1} due to ν(M–O) stretching vibration in the spectra of complexes [47,48]. Thus, it may be concluded that the ligand acts as bidentate monoanionic coordination through deprotonated enolic oxygen and carbonyl oxygen forming a six membered chelate with two conjugate π-bonds.

In addition, some new bands also observed in the IR spectra of complexes. A band appears at 960 cm^{-1} in the IR spectra of complexes which is the most diagnostic band for coordinated water in these complexes as suggested by Nakamoto [49]. The appearance of a weak band at 1000 cm^{-1} and a medium band at 745 cm^{-1} in the IR spectra of complexes III, VI and IX clearly indicates the presence of coordinated pyridine ring in these complexes. Similarly, complexes IV, VII and X display three new bands at 1675, 100 and 755 cm^{-1}, which may be taken as strong evidence in the favour of the presence of coordinated α-picoline in these complexes [50,51].

Magnetic moment and electronic spectra: The magnetic moment and electronic spectral bands of Mn(II) complexes have been displayed in Table-2. The magnetic moment values of Mn(II) complexes lie in between 5.81-5.85 B.M. which are very close to magnetic moment values due to five unpaired electrons. It shows that Mn(II) complexes are high spin octahedral magnetically dilute complexes [52]. The electronic spectra of Mn (II) complexes display three bands of very low intensity which may be assigned to the following spin forbidden transitions:

$$\nu_1 = {}^4T_{1g(4G)} \longleftarrow {}^6A_{1g}$$

$$\nu_2 = {}^4T_{2g(4G)} \longleftarrow {}^6A_{1g}$$

$$\nu_3 = {}^4E_{g(4D)} \longleftarrow {}^6A_{1g}$$

TABLE-2
ELECTRONIC SPECTRAL BAND OF
Mn(II) AND Fe(II) COMPLEXES

Compounds	ν_1 (cm^{-1})	ν_2 (cm^{-1})	ν_3 (cm^{-1})	μ_{eff} (BM)
$[Mn(PPBD)_2(H_2O)_2]$	20300	27140	30000	5.84
$[Mn(PPBD)_2(Py)_2]$	20050	24060	30080	5.85
$[Mn(PPBD)_2(\alpha\text{-pico})_2]$	19600	23590	29500	5.81
$[Fe(PPBD)_2(H_2O)_2]$	8800	18500	20180	5.09
$[Fe(PPBD)_2(Py)_2]$	8300	17400	20000	5.10
$[Fe(PPBD)_2(\alpha\text{-pico})_2]$	8700	18380	20100	5.10

The values of ν_2/ν_1 for these complexes are 1.34, 1.2 and 1.203, respectively. These values were fitted into Tanabe Sugano diagram to obtain D_q/B and ν/B values. The values of different crystal field parameters are given in Table-3. The C/B values derived in the present study for Mn(II) complexes are very close

TABLE-3
VALUES OF DIFFERENT CRYSTAL FIELD PARAMETER OF Mn(II) COMPLEXES

Compounds	ν_1/ν_2	D_q (cm^{-1})	B (cm^{-1})	C (cm^{-1})	β	C/B
$[Mn(PPBD)_2(H_2O)_2]$	1.34	1070.82	840.04	3025.68	0.1141	3.602
$[Mn(PPBD)_2(Py)_2]$	1.2	1034.47	862.06	3084.98	0.1136	3.579
$[Mn(PPBD)_2(\alpha\text{-pico})_2]$	1.203	1063.64	844.15	3029.89	0.137	3.589

to the theoretical value *i.e*. 3.8. The other crystal field parameters are in good agreement for the values reported for octahedral Mn(II) complexes [53].

The magnetic moment of Fe(II) complexes at room temperature are found 5.09-5.10 B.M (Table-1). These values are slightly higher than expected from four unpaired electrons (4.898 B.M.) in high spin octahedral complexes, which may be due to strong contribution from triply degenerated $^5T_{2g}$ ground state term [54]. The electronic spectrum of six coordinated Fe(II) complexes are expected to display only one band due to $^2E_g \rightarrow {}^2T_{2g}$ spin allowed transition but here three bands in their electronic spectra were observed. It shows the further splitting of $^5T_{2g}$ and ^5E_g levels due to Jahn-Teller distortion effect. Hence these three bands are assigned to the following spin allowed transitions.

$$\nu_1 = {}^5E_g \longleftarrow {}^5B_{2g}$$

$$\nu_2 = {}^5B_{1g} \longleftarrow {}^5B_{2g}$$

$$\nu_3 = {}^5A_{1g} \longleftarrow {}^5B_{2g}$$

ν_1 corresponds to $3D_s - 5D_t$ and ν_3 corresponds to $4D_s + 5\ D_t$; ν_2 is the measure of 10 D_q [55]. $\nu_1 + \nu_3$ will be $7D_s$ on the basis of these equations the values of different crystal field parametrers were calculated and the values are shown in Table-4. The values predicts the appreciable tetragonal distortion in octahedral symmetry of Fe(II) complexes.

TABLE-4
VALUES OF DIFFERENT CRYSTAL FIELD PARAMETER OF Fe(II) COMPLEXES

Complexes	$D_{q(xy)}$ (cm^{-1})	$D_{q(z)}$ (cm^{-1})	D_s (cm^{-1})	D_t (cm^{-1})
$[Fe(PPBD)_2(H_2O)_2]$	1850	583	4140	724.0
$[Fe(PPBD)_2(Py)_2]$	1740	400	4043	766.0
$[Fe(PPBD)_2(\alpha\text{-pico})_2]$	1838	558	4114	728.5

Magnetic moment of Co(II) complexes are found 4.80, 4.83 and 4.81 BM which is indicative of spin free octahedral complexes [56]. The values however, are greater than that corresponding to three unpaired electrons *i.e*. 3.87 BM (Table-3). This increase may be attributed to the strong orbital contribution from orbitally triply degenerate $^3T_{1g}$ ground state cubic field term. The electronic spectra of Co(II) complexes display three bands which may be assigned as below:

$$\nu_1 = {}^4T_{2g} \longleftarrow {}^4T_{1g} = 7200,\ 7300 \text{ and } 7250\ \ cm^{-1}$$

$$\nu_2 = {}^4A_{2g} \longleftarrow {}^4T_{1g} = 15250,\ 15360 \text{ and } 15375\ \ cm^{-1}$$

$$\nu_3 = {}^4T_{1g(p)} \longleftarrow {}^4T_{1g} = 19300,\ 19450 \text{ and } 19460\ \ cm^{-1}$$

The various crystal field parameters derived from these electronic bands are given in Table-5. The values of different crystal field parameters are in good agreement with octahedral Co(II) complexes with slight distortion [57].

Thermal studies: Thermogravimetric analysis (TGA) of the complexes were recorded in a static nitrogen atmosphere with heating rate of 10 ºC/min using Diamond TG/DTA analyzer. The thermograms (Fig. 1) revealed that all the synthesized metal(II) complexes decompose in four steps. The complexes I, IV and VII undergo first step decomposition with weight loss 6.3% (calcd. 6.58-6.60%) between 140-160 ºC corresponding to weight loss of coordinated water molecules. In rest of the complexes, the first decomposition occurs between 150-180 ºC with weight loss 23.4-26.5% (calcd. 23.6-26.76%), which could be due to loss of coordinated pyridine or α- picoline in these complexes. In almost all metal(II) complexes, the second decomposition takes place between 250-360 ºC with weight loss 41.2-52.6% (calcd. 41.4-52.84%) due to loss of $C_9H_8N_2$ moiety (phenyl and pyrazole moiety of the ligand). The third decomposition is observed in between 380-410 ºC corresponding to weight loss 24.0-30.6% (calcd. 24.17-30.82%) due to the loss of the remaining part of the ligand. Finally, the complexes are assumed to have converted into their metal oxides as the end product [58]. Thus, thermal studies revealed the fact that H_2O, pyridine and α-picoline are weakly coordinated to the metal(II) ions, which substantiates our proposition that complexes are tetragonally distorted octahedral with elongation along *z*-axis.

TABLE-5
CRYSTAL FIELD PARAMETERS OF Co(II) COMPLEXES

Complexes	ν_2/ν_1	10 Dq (cm^{-1})	B (cm^{-1})	β (%)
$[Co(PPBD)_2(H_2O)_2]$	2.118	10605	731.4	24.59
$[Co(PPBD)_2(Py)_2]$	2.104	11992	648.24	33.17
$[Co(PPBD)_2(\alpha\text{-pico})_2]$	2.12	10690	737.5	23.96

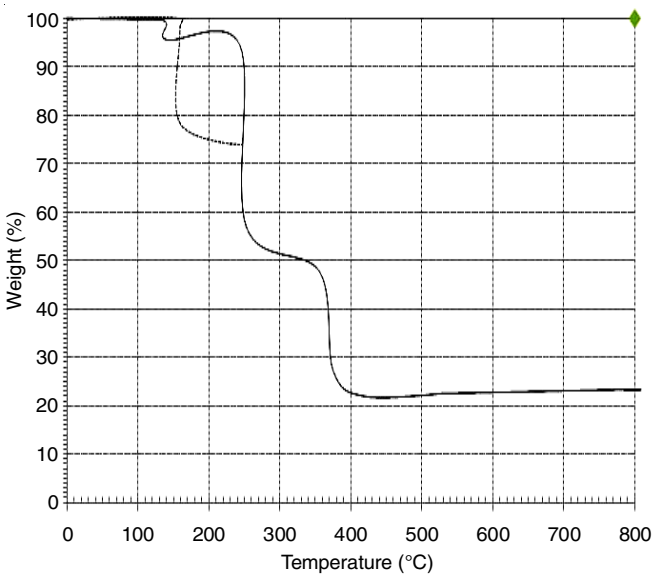


Fig. 1 Thermograms of synthesized metal(II) complexes

Biological evaluation: The synthesized ligand is active against both bacteria and fungi, however its metal(II) complexes are more active against both bacteria and fungi (Tables 6 and 7). The increase in antimicrobial activity of the complexes as compared to the ligand may be due to their chelation which reduces the polarity of the metal ion by partial sharing of positive charge with the donor group and also due to π-electron delocalization within the chelate ring [59,60].

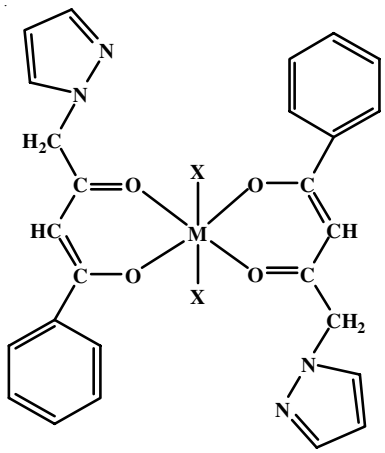
On the basis of elemental analysis, molar conductivity, magnetic moment, spectral and thermal studies, the tentative structure of complexes may be given as below:

TABLE-6
ANTIBACTERIAL SCREENING OF LIGAND AS WELL AS ITS COMPLEXES

Compound	Diameter of inhibition zone (mm); *S. aureus*			Activity index (%)			Diameter of inhibition zone (mm); *E. coli*			Activity index (%)		
Concentration	25	50	100	25	50	100	25	50	100	25	50	100
PPBD ($C_{13}H_{12}N_2O_2$)	15	16	17	71	70	61	13	15	16	72	65	64
$[Mn(PPBD)_2(H_2O)_2]$	17	18	20	81	78	71	14	16	17	78	70	68
$[Mn(PPBD)_2(Py)_2]$	18	18	22	86	78	79	15	17	18	83	74	72
$[Mn(PPBD)_2(\alpha\text{-pico})_2]$	18	19	22	86	83	79	15	18	19	83	78	76
$[Fe(PPBD)_2(H_2O)_2]$	19	20	23	90	87	82	16	20	21	89	87	84
$[Fe(PPBD)_2(Py)_2]$	20	21	23	95	91	82	17	21	22	94	91	88
$[Fe(PPBD)_2(\alpha\text{-pico})_2]$	20	21	24	95	91	86	17	21	22	94	91	88
$[Co(PPBD)_2(H_2O)_2]$	18	20	21	86	87	75	16	18	20	89	78	80
$[Co(PPBD)_2(Py)_2]$	19	20	22	90	87	79	16	19	20	89	83	80
$[Co(PPBD)_2(\alpha\text{-pico})_2]$	19	20	22	90	87	79	15	18	20	83	78	80
Streptomycin	21	23	28	100	100	100	18	23	25	100	100	100

TABLE-7
ANTIFUNGAL SCREENING OF LIGAND AS WELL AS ITS COMPLEXES

Compound	Diameter of inhibition zone (mm); *A niger*			Activity index (%)			Diameter of inhibition zone (mm); *C. abicans*			Activity index (%)		
Concentration	25	50	100	25	50	100	25	50	100	25	50	100
PPBD ($C_{13}H_{12}N_2O_2$)	14	15	20	64	60	67	16	17	18	67	71	64
$[Mn(PPBD)_2(H_2O)_2]$	16	18	21	73	72	70	17	19	19	71	79	68
$[Mn(PPBD)_2(Py)_2]$	16	19	21	73	76	70	17	18	20	71	75	71
$[Mn(PPBD)_2(\alpha\text{-pico})_2]$	17	19	22	77	76	73	18	19	20	75	79	71
$[Fe(PPBD)_2(H_2O)_2]$	19	22	26	86	88	87	20	21	23	83	88	82
$[Fe(PPBD)_2(Py)_2]$	20	22	26	91	88	87	21	22	24	88	92	86
$[Fe(PPBD)_2(\alpha\text{-pico})_2]$	20	23	27	91	92	90	20	22	25	83	92	89
$[Co(PPBD)_2(H_2O)_2]$	18	20	23	82	80	77	19	20	21	79	83	75
$[Co(PPBD)_2(Py)_2]$	17	20	23	77	80	77	19	19	21	79	79	75
$[Co(PPBD)_2(\alpha\text{-pico})_2]$	18	21	24	82	84	80	18	20	20	75	83	71
Miconazole	22	25	30	100	100	100	24	24	28	100	100	100



where M = Mn(II), Fe(II), Co(II)

Conclusion

In this work, synthesis of new heterocyclic based β-diketone ligand and its few transtion(II) has been synthesized and characterized. The ligand is prepared by the condensation of pyrazole-1-acetylchloride and sodium acetophenone. Thermal analysis shows that all the metal(II) complexes are appreciably stable and there is elongation along *z*-axis in their distorted octahedral geometry. All the complexes are active against both bacteria and fungi. The ligand itself is also biologically active and its activity gets enhanced after complexation with metal ions.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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