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# Synthesis and Characterization Pd(II) Macrocyclic Complexes and Evaluation their Antibacterial Activity

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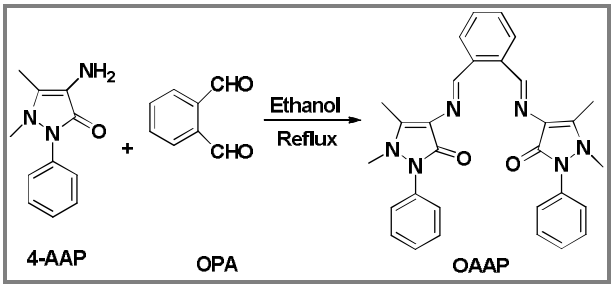
## ABSTRACT

*A series of Pd(II) complexes have been synthesized from macrocyclic Schiff base ligands. The ligands were prepared by condensation of 4-aminoantipyrine derivative with different diamines and hydrazides. Macrocyclic Schiff bases are tetradentate with N4 donor system around the metal ion. Octahedral geometry has been assigned for all complexes. All these compounds were characterized by different analytical methods such as elemental analysis, mass, FTIR, $^{1}$H-NMR, UV-Visible, magnetic susceptibility, molar conductance and thermal studies. All ligands and complexes were screened for their in vitro antimicrobial activity.*

### Graphical Abstract




Synthesis of OAAP.

**Keywords**: Antipyrine, Macrocyclic Schiff base, Pd complexes, Antibacterial activity.

## INTRODUCTION

1-Phenyl-2,3-dimethylpyrazol-5-one (antipyrine) is an important pyrazolone derivative and is widely used for a long time in medicine as antipyretic, analgesic and sedative[**1**]. The antipyrine derivatives possess a wide applications including biological, clinical and pharmacological area. Antipyrines also used as analytical reagents in the determination of metal ions. In addition they have also used in the purification of penicillin acylase, flow-injection and sequential injection spectrophotometric determination of salbutmol and ritodrine hydrochloride [**2**]. Furthermore, Schiff bases are important precursors for the synthesis of some bioactive compounds [**3**], which possess various biological activities such as antibacterial [**4**], antifungal [**5**], anti-HIV [**6**], anti-inflammatory [**7**], anticonvulsant

[8], antiviral [9] and anticancer [10] activates. Based on all of the above facts, we focused synthesis of various macrocylicpyrazolone derivatives and their palladium complexes.

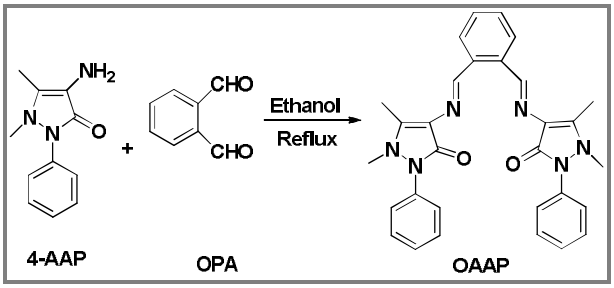
## MATERIALS AND METHODS

**Materials:** Palladium(II) chloride, 4-amino antipyrine, ortho-pthalaldehyde(OPA), 1,4-diamino butane, carbanohydrazide, 4H-1,2,4-traizole-3,5-diamine, 2-amino benzo hydrazide and naphthalene-1,8-diamine were purchased from Aldrich and all the organic solvents such as ethanol, methanol, dichloro methane, DMSO were analytical grade. These solvents distilled and preserved under molecular sieves. The purity of the compounds was confirmed by TLC using Merck 60F254 silica gel plates.

**Physical Measurements:** The percentages of carbon, hydrogen and nitrogen present in ligands and their complexes were determined by using PerkinElmer 2400 Series II CHNS/O analyzer. Buchi-510 melting point machine was used to find melting points. The Mettler-Toledo star system has been employed to perform thermogravimetric studies under an inert atmosphere of dry nitrogen <10 mg of sample masses utilized. The FTIR spectra of compounds have been recorded in the range of 4000–200 $cm^{-1}$ using Perkin Elmer-283 spectrophotometer. Brucker WH 300 (400 MHz) and Varian Gemini (100 MHz) spectrometers were used to record NMR spectra ($^{1}H$ and $^{13}C$ NMR). CEC-21-110B, Finningan Mat 1210 and MICROMASS7070 spectrometers operating at 70 eV using a direct inlet system and VG-Auto-Spec-M mass spectrometer were used for mass spectra. Electronic spectra were obtained with Shimadzu UV-160A, a UV-Visible double beam spectrophotometer with matched quartz cells of path length 1 cm. Conductance measurements were carried out on 10-3M solution of compounds in dichloromethane at 25°C on Dig sun Digital conductivity meter model DL-909. Gouy balance calibrated with $Hg[Co(NCS)_4]$ was employed for the estimation of magnetic susceptibilities of complexes at normal temperature.

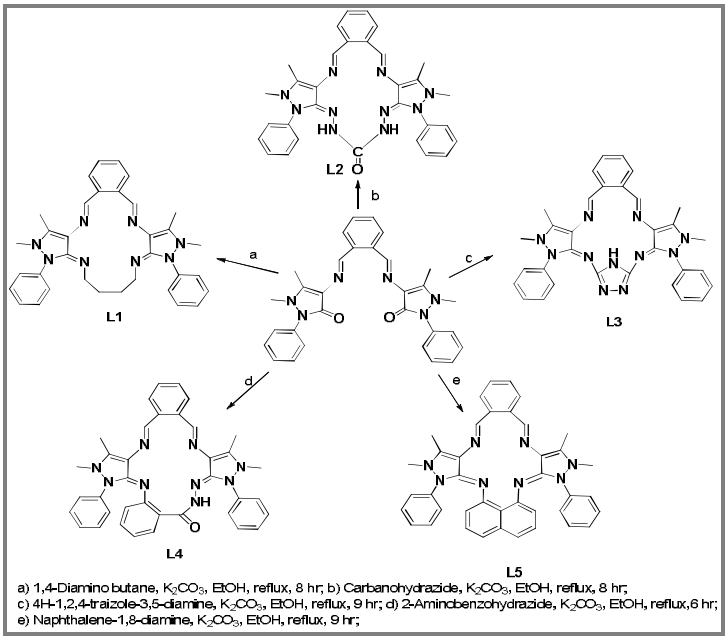
### General procedure for the synthesis of ligands

**Synthesis of OAAP:** A solution of 4-aminoantipyrine (2 mmol) and *o*-phthalaldehyde (1 mmol) in ethanol (25 mL) was refluxed for 4 h. After completion of the reaction (progress of the reaction monitored by TLC), the reaction solvent evaporated under reduced pressure and the crude kept aside at room temperature for two days. The yellow colored precipitate obtained has been filtered, rinsed with diethyl ether and recrystallized from ethanol to afford pure OAAP.



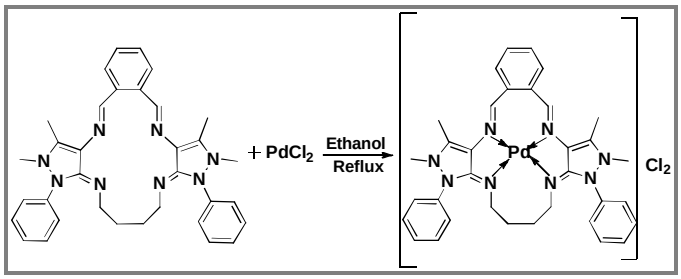

**Scheme 1.** Synthesis of OAAP.

**General synthesis of Schiff bases:** A ethanolic solution of OAAP (2 mmol) was refluxed with diamines (2 mmol) viz., 1,4-diamino butane(L1),and carnohydrazide (L2), 4H-1,2,4-traizole-3,5-diamine (L3), 2-amino benzo hydrazide (L4) and naphthalene-1,8-diamine (L5) correspondingly with the addition of 1 g of anhydrous $K_2CO_3$ for about 5-7 h, after completion of the reaction (progress of the reaction monitored by TLC). The solvent was reduced to one-third, treated with hot water and kept in refrigerator for 24 h. The solid product obtained was separated by filtration and recrystallized from ethanol.




**Scheme 2.** Synthesis of Schiff bases.

**General procedure for the synthesis of Schiff base Pd(II) complexes:** An ethanolic solution of newly prepared macrocyclic Schiff base ligands (2 mmol) and Palladium chloride(2 mmol) was refluxed until a solid formation, progress of the reaction monitored by TLC. After completion of the reaction, the reaction mixture concentrated to one-third of its volume. The solid product was filtered, washed with ethanol several times and dried in vacuum under vacuum over anhydrous calcium chloride (Yield: 65-78%).




**Scheme 3.** Synthesis of Schiff base Pd(II) complexes.

**Antibacterial activity:** The newly synthesized ligands and complexes have been screened to evaluate In-vitro antibacterial activity in opposition to microbes like *Escherichia coli, Klebsiella pneumonia, Basillus subtilis* and *Staphylococcus aureus*. The existing antibiotics drugs such as Streptomycin and Rifampicin were used as the standards for current study. This investigation was occurred with help of known reported procedure is so called cup plate method 23. Microorganisms were cultured in the combination of nutrient agar and nutrient broth (obtained from Hi-media, Mumbai). Sample compound was prepared 10 mg of compound in 10 mL of solvent water, methanol and DMSO. This sample is utilized to make a range of concentrations as 100, 50, 20, 15, 10, 5, 2, 1 μg $mL^{-1}$ by addition of solvent. Semi solid agar medium (25 mL) was poured on to uncontaminated Petri dishes and put aside for few hours to allow solidifies. Then well grown microorganism about 50 mL was added and distributed equally on to the above arranged agar medium dish with the help of fresh cotton swab. Fine borer was used to make 5 mm wide bores on the agar having cultured microorganism. These bores were further filled with sample solutions of compounds properly with micropipette. Similar

steps also used in the preparation of plates for antibiotics. In presence of free air conditions all plates were incubated at the temperature of 37°C for 24 h. 24 h later the zones of inhibition of growth for all compounds and antibiotics were measured and compared. The results were expressed in terms of active and inactive. Further minimum inhibitory concentrations of all active compounds were determined.

**Antifungal Activity:** The active macrocyclic metal complexes were tested for their in vitro growth inhibitory activity against the pathogenic fungus, namely *A. flavus* and *Fusarium* species cultured on sabour dextrose agar medium prepared by taking 11 mL of distilled water in a conical flask followed by the addition of following ingredients: mycological peptone, 10 g, dextrose, 30 g, agar, 12 g and the pH of the solution was adjusted to 5.7. Boiling was continued until complete dissolution. After that, the solution was sterilized by autoclaving at 15 lb pressure (120°C for 20 min) by the diffusion method and incubated at 28°C for 3 days. Several test solutions of different concentration (microgram per liter) were prepared in dimethyl sulphoxide solution. The percentage inhibition of fungal growth was determined on the growth in test plates compared to that of respective control plate, given by the Vincent equation.

## RESULTS AND DISCUSSION

Current work involved synthesis of new macrocyclic Schiff base ligands by condensation of derivative of 4-aminoantipyrine with different diamines, diamino hydrazides in presence of base conditions and their Pd(II) complexes. The carbon, hydrogen and nitrogen percentages in all newly prepared compounds were estimated by using CHN analyzer. The percentage of palladium in all complexes was found by known procedure. The tentative molecular Formulas for all compounds were proposed based on the physical and analytical data (Table 1).

**Table 1.** Physical data ofmacrocyclic Schiff base Pd(II) complexes

| Comp. No. | Pd(II) complex | Reaction time (h) | Decomp. temp. (°C) | Color | Yield (%) |
|---|---|---|---|---|---|
| 5.1. | $[Pd(L1)]Cl_2$ | 7 | 246 | Light Brown | 68 |
| 5.2. | $[Pd(L2)]Cl_2$ | 6 | 228 | Brown | 75 |
| 5.3. | $[Pd(L3)]Cl_2$ | 5 | 227 | Black | 72 |
| 5.4. | $[Pd(L4)]Cl_2$ | 8 | 241 | Ash | 65 |
| 5.5. | $[Pd(L5)]Cl_2$ | 5 | 236 | Light Yellow | 61 |

**IR spectral analysis:** The infrared spectra of the macrocyclic ligands were compared with their Pd(II) complexes to characterize the different binding modes of the macrocyclic ligands to Pd(I) ion. All the complexes exhibited two strong bands in the region of 1543-1586 $cm^{-1}$, which is assigned to the υ(C=N) stretching vibration. These bands were found in the lower region about 25-40 $cm^{-1}$ when compared to that of ligand spectra indicating the coordination bond formation between Schiff base

**Table 2.** Analytical data of macrocyclic Schiff base Pd(II) complexes

| S. No. | Pd(II) Complexes | Molecular Formula | Found (Calculated)% | | | | MASS |
|---|---|---|---|---|---|---|---|
| | | | C | H | N | M | |
| 5.1 | $[Pd(L1)]Cl_2$ | $C_{34}H_{36}Cl_2N_8Pd$ | 55.60 (55.63) | 4.91 (4.94) | 15.22 (15.27) | 14.48 (14.50) | 734(M), 735($M^+$+1) |
| 5.2 | $[Pd(L2)]Cl_2$ | $C_{31}H_{30}Cl_2N_{10}OPd$ | 50.54 (50.59) | 4.08 (4.11) | 19.01 (19.03) | 14.44 (14.46) | 735.96(M), 737($M^+$+2) |
| 5.3 | $[Pd(L3)]Cl_2$ | $C_{32}H_{29}Cl_2N_{11}Pd$ | 51.53 (51.59) | 3.90 (3.92) | 20.62 (20.68) | 14.25 (14.29) | 744.97(M), 746($M^+$+2) |
| 5.4 | $[Pd(L4)]Cl_2$ | $C_{37}H_{33}Cl_2N_9OPd$ | 55.74 (55.76) | 4.15 (4.17) | 15.83 (15.82) | 13.32 (13.35) | 797(M), 798($M^+$+1) |
| 5.5 | $[Pd(L5)]Cl_2$ | $C_{40}H_{34}Cl_2N_8Pd$ | 59.71 (59.75) | 4.25 (4.26) | 13.91 (13.94) | 13.21 (13.24) | 804(M), 805($M^+$+1) |

nitrogen and metal ion. Further, it was confirmed by a non-ligand band with medium intensity around~500 $cm^{-1}$corresponding to metal nitrogen bond (M-N) stretching frequency (Table 2). The ligands L2 and L4 contain one C=O group and this band was appearing around ~1670 $cm^{-1}$ whereas this was shifted towards higher frequency region about to 20 $cm^{-1}$ revealing the fact that no involvement of C=O group in the complex formation. In complexes of ligands L2, L3 and L4 a positive shift observed for N-H frequency as compared to its position (3160-3144 $cm^{-1}$) for ligands indicating non-participation of nitrogen atoms in coordination. Stretching frequencies and wagging frequencies of aromatic rings have been found in the range of 1412-1365 and 3086-3045 $cm^{-1}$ for all ligands and important changes were not found in these frequencies in the case of respective complexes suggesting that ring atoms are not in coordination with the metal atom. The characteristic bands owing to the C=C, C=N (pyridine), N-N, =C-N (4-amino antipyrine ring) and aromatic rings in the spectra of respective macrocyclic Pd(II) complexes remain almost unchanged.

**Table 3.** Infrared spectral data of Macrocyclic Schiff base Pd (II) complexes

| Ligand/Complex | Selected bands($cm^{-1}$) | | | | |
|---|---|---|---|---|---|
| | $\upsilon_{N-H}$ | $\upsilon_{C=O}$ | $\upsilon_{C=N}$ | $\upsilon_{Pd-N}$ | $\upsilon_{Pd-Cl}$ |
| $[Pd(L1)]Cl_2$ | -- | -- | 1564, 1543 | 518 | -- |
| $[Pd(L2)]Cl_2$ | 3176 | 1691 | 1565, 1552 | 511 | -- |
| $[Pd(L3)]Cl_2$ | 3169 | - | 1586,1574,1558 | 517 | -- |
| $[Pd(L4)]Cl_2$ | 3174 | 1693 | 1556,1547 | 521 | -- |
| $[Pd(L5)]Cl_2$ | -- | -- | 1559,1546 | 523 | -- |

**Electronic spectral analysis:** The electronic spectra of the Pd(II) metal complexes (Table 4) were recorded in DMF as solvent. The Pd(II) metal complexes have been found to show a broad d-d transition band in the region of 440-478 nm due to $^1B_{1g} \leftarrow {}^1A_{1g}$ transition generally expected for the square planar geometry further, a relatively strong charge transfer band has been noticed in the region of 291-322 nm. These two transitions supporting the square planar geometry to all the Pd(II) metal complexes.

**Table 4.** Electronic spectral, Magnetic moment and Molar conductance data of Schiff base Pd(II) complexes.

| Comp. No. | Pd(II) complex | $\lambda_{max}$ nm($cm^{-1}$) | $\Lambda_M$ ($\Omega^{-1}cm^2mol^{-1}$) |
|---|---|---|---|
| 5.1. | $[Pd(L1)]Cl_2$ | 448, 305 | 57.4 |
| 5.2. | $[Pd(L2)]Cl_2$ | 440, 322 | 51.2 |
| 5.3. | $[Pd(L3)]Cl_2$ | 453, 318 | 54.3 |
| 5.4. | $[Pd(L4)]Cl_2$ | 465, 291 | 62.5 |
| 5.5. | $[Pd(L5)]Cl_2$ | 478, 298 | 56.8 |

**Conductance measurements:** The molar conductance values (Table 4) for all the Schiff base Pd(II) metal complexes were measured in dichloromethane at $10^{-3}$ M concentration. The molar conductance values of all the complexes found in the range of 51.2-62.5 $mho^1$ $mole^{-1}$ $cm^{-2}$ giving the evidence for 1:2 electrolytic in nature attributable to two chlorides present in ionic sphere of the complexes. Further silver nitrate test was carried out for these complexes and gave positive results.

**Magnetic properties:** The magnetic susceptibility studies have been conducted for all macro cyclic Schiff base Pd(II) complexes and these compounds were found to be diamagnetic in nature and hence Pd(II) ion is in the low spin configuration. The diamagnetic nature of complexes was further confirmed by the sharp, well defined signals in the $^1H$ NMR spectra (Table 4).

**Thermal analysis:** The thermogravimetric study results of Pd(II) complexes were demonstrating that they are stable up to ~230°C indicates that all are in the anhydrous state. The thermo grams of all the Pd(II) complexes contain only two main decomposition stages which are loss of organic ligand

moiety and oxidation of metal ion. The all Pd(II) complexes are not shown any peak in the range of 70-230$^{o}$C is supporting the non existence of a lattice and coordinated water, which later also confirmed by the absence of endothermic peak of DTA in this temperature range. The First sharp weight loss was found in the range of 250-300°C representing that the decomposition curve due to loss of organic ligand moiety was further supported by DTA curve in this same temperature range and curves above 500°C resulting from the final products of decomposition corresponds to oxidation of oxide in all the complexes.

**$^{1}$H NMR spectral analysis:** In $^{1}$H NMR spectra all the ligands signal corresponding to CH=N protons were observed in the range of 8.05-8.44 δ while, in the spectra of Schiff base Pd(II) complexes, these signals are appeared in the higher region of 9.12-8.74 δ confirming the coordination of ligand to Pd(II) ion. There is no appreciable change was observed in the peak positions of remaining all protons (Table 5).

**Table 5.** $^{1}$H NMR spectral data of Pd(II) complexes

| Comp. No. | Pd(II) complex | Peaks (ppm) |
|---|---|---|
| 5.1. | $[Pd(L1)]Cl_2$ | 8.74(2H, s, CH=N), 7.50-7.68(4H, m, Ar-H), 6.71-6.96(10H, m, Ar-H), 3.74(4H, t, $CH_2$-N=C), 2.72(6H, s, N-$CH_3$), 2.21(6H, s, C-$CH_3$), 1.91(4H, t, $CH_2$-C) |
| 5.2. | $[Pd(L2)]Cl_2$ | 8.93(2H, s, CH=N), 7.55-7.69(4H, m, Ar-H), 6.62-6.86(10H, m, Ar-H), 6.13(2H, s, N-H), 3.67(6H, s, N-$CH_3$), 2.27(6H, s, C-$CH_3$). |
| 5.3. | $[Pd(L3)]Cl_2$ | 8.89(2H, s, CH=N), 7.59-7.72(4H, m, Ar-H), 6.68-6.88(10H, m, Ar-H), 4.74(1H, s, N-H), 2.75(6H, s, N-$CH_3$), 2.29(6H, s, C-$CH_3$). |
| 5.4. | $[Pd(L4)]Cl_2$ | 8.98(2H, s, CH=N), 7.53-7.95(8H, m, Ar-H), 6.69-7.22(10H, m, Ar-H), 6.64(1H, s, N-H), 2.82(6H, s, N-$CH_3$), 2.28(6H, s, C-$CH_3$). |
| 5.5. | $[Pd(L5)]Cl_2$ | 9.12(2H, s, CH=N), 7.52-7.73(10H, m, naphthalene+Ar-H), 6.59-6.71(10H, m, Ar-H), 2.74(6H, s, N-$CH_3$), 2.33(6H, s, C-$CH_3$). |

**$^{13}$C NMR spectral analysis:** The $^{13}$C NMR spectra of all the ligands exhibited characteristic signals in the range of 153.1-165.8 δ owing to the azomethine carbon is doubly bonded to nitrogen where as in the spectra of Schiff base Pd(II) complexes appeared in the region of 141.6-150.2 ppm, is a down field shift. It gives evidence for the coordination of ligands to the metal ion via nitrogen donor atom. The peaks due to carbons adjacent to donor nitrogen atom were shifted to a little downfield in case of respective complexes. Noticeable changes in signal positions were not observed in case of remaining all carbon atoms in complexes (Table 6).

**Table 6.** $^{13}$C NMR spectral data of Pd(II) complexes

| Comp. No. | Pd(II) Complexes | $^{13}$C NMR peaks( δ ppm) |
|---|---|---|
| 5.1. | $[Pd(L1)]Cl_2$ | 9.9(2C, C-$\underline{C}H_3$), 29.2(2C, C-$\underline{C}H_2$-C), 36.4(2C, N-$CH_3$), 47.2(2C, C-$\underline{C}H_2$-N=), 109.5(2C, =C-N-), 123.4, 124.1, 130.6, 130.9, 132.3, 134.5, 136.6 (18C, Ar-C), 145.6(2C, =C-N-), 142.3(2C, C=N), 150.2(2C, CH=N). |
| 5.2. | $[Pd(L2)]Cl_2$ | 9.5(2C, C-$\underline{C}H_3$), 37.4(2C, N-$CH_3$), 110.3(2C,=C-N-), 122.4, 123.7, 129.5, 130.1, 132.2, 134.1, 136.5 (18C, Ar-C),146.3(2C,=C-N-), 141.6(2C, C=N), 155.6(1C, C=O), 149.7 (2C, CH=N). |
| 5.3. | $[Pd(L3)]Cl_2$ | 9.3(2C, C-$\underline{C}H_3$), 34.2(2C, N-$CH_3$), 111.2(2C, =C-N-), 121.1, 124.2, 130.3, 131.5, 132.3, 134.1, 137.0 (18C, Ar-C), 143.2(2C,=C-N-), 142.6(2C, -C=N) 144.5(2C, -C=N), 148.2 (2C, CH=N). |
| 5.4. | $[Pd(L4)]Cl_2$ | 8.5(2C, C-$\underline{C}H_3$), 33.8(2C, N-$CH_3$), 107.1-111.2(2C,=C-N), 121.0, 123.2, 124.1, 126.5, 126.8, 127.2, 130.1, 131.2, 132.1, 134.2, 135.3, 135.5, 146.1(24C, Ar-C), 146.5-147.1(2C,=C-N), 142.8(2C, C=N) 147.5 (2C, CH=N), 167.2(1C, C=O). |
| 5.5. | $[Pd(L5)]Cl_2$ | 9.0(2C, C-$\underline{C}H_3$), 38.1(2C, N-$CH_3$), 112.1(2C, =C-N), 122.8, 124.1, 128.8, 131.5, 132.1, 135.2, 137.1 (18C, Ar-C), 145.1(2C,=C-N-),143.4(2C, C=N), 148.9 (2C, CH=N), 116.1, 121.1, 126.8, 128.2,137.2,148.1(10C, naphthalene ring). |

## APPLICATION

**Antibacterial activity:** Based on preliminary investigation, it is observed that Schiff-base Pd(II) complexes have shown activity against four different strains of bacteria in the order of: 5.2>5.5>5.4>5.1>5.3 for *Bacillus subtilis*, 5.3>5.1.>5.4>5.2>5.5 for *Staphylococcus aureus*, 5.1>5.5>5.4>5.3>5.2 for *Escherichia coli* and 5.2>5.4>5.3>5.5>5.1 for *Klebsiella pneumonia* (Table 7).

**Table 7.** Zone of inhibition of Macrocyclic Schiff base Pd(II) metal complexes

| Comp. No. | Schiff base Pd(II) metal complexes (1000 µg mL$^{-1}$) | Zone of inhibition (mm) | | | |
|---|---|---|---|---|---|
| | | gram(+) | | gram(-) | |
| | | *B. subtilis* | *B. subtilis* | *B. subtilis* | *B. subtilis* |
| 5.1 | $[Pd(L1)]Cl_2$ | 25 | 31 | 38 | 07 |
| 5.2 | $[Pd(L2)]Cl_2$ | 36 | 24 | 11 | 39 |
| 5.3 | $[Pd(L3)]Cl_2$ | 16 | 39 | 23 | 23 |
| 5.4 | $[Pd(L4)]Cl_2$ | 27 | 26 | 28 | 26 |
| 5.5 | $[Pd(L5)]Cl_2$ | 28 | 11 | 29 | 17 |
| Drug-1 | Streptomycin | 30 | 16 | 15 | 11 |
| Drug-2 | Ampicillin | 16 | 05 | 18 | 06 |
| Drug-3 | Rifampicin | 44 | 41 | 44 | 38 |

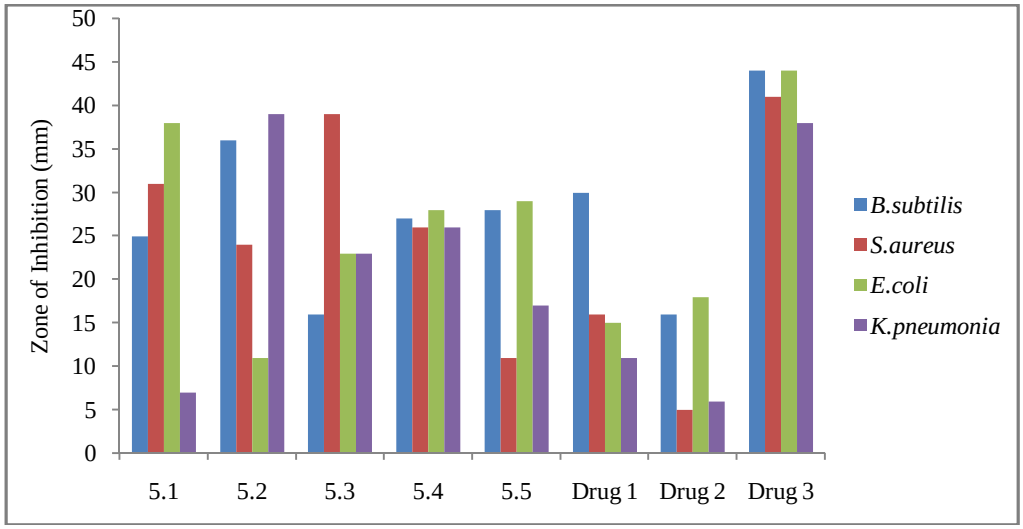



**Figure 1.** Zones of inhibition of Schiff-base Pd(II) metal complexes

**Antifungal activity:** All the complexes were showed good antifungal activity against both fungal organisms compared to their corresponding free ligand (Table 8).

**Table 8.** Zone of inhibition of Macrocyclic Schiff base Pd(II) metal complexes

| Comp. No. | Schiff base Pd(II) metal complexes | *A.flavus*(µg mL$^{-1}$) | | *Fusarium* (µg mL$^{-1}$) | |
|---|---|---|---|---|---|
| | | 500 | 1000 | 500 | 1000 |
| 5.1 | $[Pd(L1)]Cl_2$ | 18 | 26 | 34 | 27 |
| 5.2 | $[Pd(L2)]Cl_2$ | 32 | 35 | 32 | 41 |
| 5.3 | $[Pd(L3)]Cl_2$ | 34 | 37 | 33 | 47 |
| 5.4 | $[Pd(L4)]Cl_2$ | 36 | 32 | 37 | 45 |
| 5.5 | $[Pd(L5)]Cl_2$ | 28 | 40 | 32 | 49 |
| Drug-1 | Amphotericin | 36 | 61 | 42 | 61 |
| Drug-2 | Itrazole | 47 | 63 | 53 | 64 |
| Drug-3 | Bavistin | 41 | 66 | 45 | 67 |

## CONCLUSION

We have successfully synthesized and characterized novel 4-aminoantipyrine based macrocyclic Schiff base Pd(II) complexes. All the complexes are non-electrolytic and para in nature. According to the results of elemental and spectral studies octahedral structures were assigned tentatively for all the complexes. The screening result of antimicrobial activity resulted that the complexes were shown medium to good activity.

## ACKNOWLEDGEMENTS

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