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# Synthesis and Study of Some Hydrazone Metal Complexes

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Co(II), Ni(II) and Cu(II) metal complexes with 3-phenyl quinazoline-4 (3H)-one-2-carboxaldehyde hydrazone (PQCAH) is synthesized and characterized with the help of elemental analysis, molar conductance, magnetic suceptibility and electronic spectral data. On the basis of experimental data the complexes are found to be octahedral in geometry.

## INTRODUCTION

The interest in the study of hydrazones possessing potential donor sites has been growing as they are reported to be associated with anthelmintic, antibacterial, antifungal, antiinflammatory, antitubercular and antitumour activities.[1–7] Moreover, the biological activity of an active ligand is altered manyfold on coordinating with a suitable metal ion[8, 9]. Keeping the above facts in mind it is worth while to synthesize and characterize 3-phenyl quinazoline-4-(3H)-one-2-carboxaldehyde hydrazone (PQCAH) and its complexes with Co(II), Ni(II) and Cu(II).

## EXPERIMENTAL

All the chemicals used were of BDH quality except 3-phenyl quinazoline-4-(3H)-one carboxaldehyde which was prepared by earlier reported method[10].

### Preparation of the ligand PQCAH

The ligand PQCAH was prepared in good yield by condensing 3-phenyl quinazoline-4-(3H)-one carboxaldehyde with hydrazine hydrate in ethanol. The ligand, PQCAH is synthesized by refluxing the ethanolic solution of hydrazine hydrate with the solution of 3-phenyl quinazoline-4-(3H)-one carboxaldehyde dissolved in a minimum volume of tetrahydrofuran in 1 : 1 ratio. On cooling the solution so obtained, yellow coloured crystals were filtered, washed with cold ethanol followed by ether and dried in oven.

### Preparation of the complexes

Ethanolic solution of the ligand PQCAH was added to the solution of the respective metal halides dissolved in tetrahydrofuran in the molar ratio 2 : 1 with

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constant stirring. Resulting solution was refluxed over a water bath for 1 h, when a coloured compound separated out which was filtered and washed with ethanol followed by ether and dried in an oven.

The complexes were analysed for metal contents by standard procedures[11]. The carbon, hydrogen and nitrogen were determined by CE-440 elemental analyser. The UV spectra were recorded on a Cary-2390 spectrophotometer. The IR spectra were recorded on a Perkin-Elmer spectrophotometer. Magnetic moment was determined at room temperature by Gouy's method. The conductivity measurements were carried out in a systronics direct reading conductivity meter 303 using $10^{-3}$ M dimethyl formamide solution. Analytical data, colour, electronic spectral data, conductivity measurements and magnetic moments are recorded in Table-1.

TABLE-1

ANALYTICAL, MAGNETIC MOMENT, ELECTRONIC SPECTRA AND CONDUCTIVITY MEASUREMENTS FOR METAL COMPLEXES OF THE TYPE [M $(PQCAH)_2X_2$]

| Compound/Colour | % Analysis, Found (Calculated) | | | | $\lambda_{max}$ electronic ($cm^{-1}$) | $\mu_{eff}$ (B.M.) | $\Lambda_{max}$ ($ohm^{-1}$ $cm^{-1}$ $mol^{-1}$) |
|---|---|---|---|---|---|---|---|
| | M | C | H | N | | | |
| PQCAH<br>Light yellow | — | 63.97<br>(64.28) | 4.16<br>(4.28) | 19.83<br>(20.00) | — | — | — |
| [$Co(PQCAH)_2Cl_2$]<br>Orange | 8.80<br>(8.95) | 54.53<br>(54.71) | 3.52<br>(3.64) | 16.91<br>(17.02) | 8260<br>14800<br>18600 | 4.90 | 14.1 |
| [$Co(PQCAH)_2Br_2$]<br>Orange yellow | 7.69<br>(7.88) | 48.07<br>(48.21) | 3.09<br>(3.21) | 14.81<br>(14.99) | 8080<br>14600<br>19020 | 4.70 | 13.2 |
| [$Co(PQCAH)_2I_2$]<br>Yellow | 6.88<br>(7.00) | 42.61<br>(42.82) | 2.76<br>(2.85) | 13.19<br>(13.32) | 8360<br>15010<br>19210 | 5.10 | 16.8 |
| [$Ni(PQCAH)_2Cl_2$]<br>Green | 8.68<br>(8.92) | 54.61<br>(54.73) | 3.58<br>(3.64) | 16.91<br>(17.02) | 11020<br>17630<br>25710 | 3.24 | 14.6 |
| [$Ni(PQCAH)_2Br_2$]<br>Greenish yellow | 7.77<br>(7.86) | 48.09<br>(48.22) | 3.12<br>(3.21) | 14.86<br>(15.00) | 11700<br>17100<br>26010 | 3.11 | 17.4 |
| [$Ni(PQCAH)_2I_2$]<br>Brick red | 6.83<br>(6.98) | 42.67<br>(42.83) | 2.71<br>(2.85) | 13.10<br>(13.32) | 11900<br>17910<br>25100 | 3.18 | 18.3 |
| [$Cu(PQCAH)_2Cl_2$]<br>Dark green | 9.46<br>(9.59) | 54.18<br>(54.33) | 3.53<br>(3.62) | 16.76<br>(16.90) | 15100 | 1.90 | 14.7 |
| [$Cu(PQCAH)_2Br_2$]<br>Yellowish green | 8.39<br>(8.45) | 47.76<br>(47.91) | 3.08<br>(3.19) | 14.73<br>(14.90) | 15500 | 1.87 | 19.3 |

## RESULTS AND DISCUSSION

The elemental analysis shows that complexes have 1 : 2 stoichiometry of the type $[M(PQCAH)_2X_2]$, X = $Cl^-$, $Br^-$, $I^-$. The molar conductance values in DMF at the concentration $10^{-3}$ M are in the range of 13.2–19.3 $ohm^{-1}$ $cm^2$ $mol^{-1}$, indicating that all complexes are non-electrolytes.

The IR spectrum of the ligand showing a medium intensity broad band observed around 3200 $cm^{-1}$ is assigned to ν(N—H). This band remains unperturbed in the complexes which indicates that primary amino group is not involved in the bond formation with the metal ions. The ligand shows a sharp and broad band at 1700–1620 $cm^{-1}$ attributed to ν(C=N) stretching shifts towards lower frequency region (1660–1610 $cm^{-1}$) inferring thereby the participation of azomethine N of quinazoline ring ($N^1$) as well as hydrazone group in complexation[13].

In the light of previous assignments,[14, 15] the bands in the far IR region 470–400 $cm^{-1}$ to ν(M—N) coupled with a ligand vibration and the band in the region 310–260 $cm^{-1}$ may be attributed to ν(M—X). The infrared spectral data are recorded in Table-2.

TABLE-2

INFRARED SPECTRAL DATA ($cm^{-1}$) OF PQCAH AND ITS METAL COMPLEXES

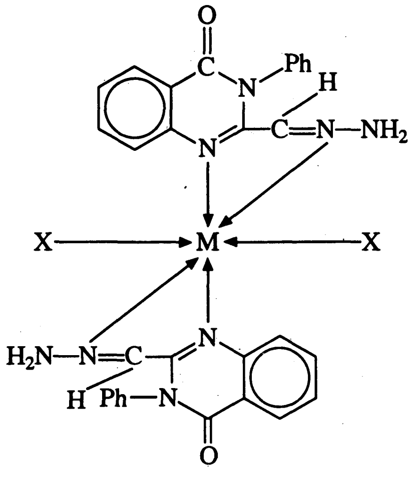
| Compounds | ν(N—H) | ν(C=N) (Hydrazone) | ν(C=N) (Azomethine) | ν(M—N) | ν(M—X) |
|---|---|---|---|---|---|
| PQCAH | 3170 m,b | 1635 s,b | 1700 s,b | – | – |
| $[Co(PQCAH)_2Cl_2]$ | 3165 m,b | 1610 s,b | 1675 s,b | 465 m | 250 m |
| $[Co(PQCAH)_2Br_2]$ | 3170 m,b | 1605 s,b | 1670 s,b | 445 m | 260 m |
| $[Co(PQCAH)_2I_2]$ | 3165 m,b | 1620 s,b | 1660 s,b | 450 m | 270 m |
| $[Ni(PQCAH)_2Cl_2]$ | 3170 m,b | 1610 s,b | 1650 s,b | 460 m | 290 m |
| $[Ni(PQCAH)_2Br_2]$ | 3165 m,b | 1615 s,b | 1655 s,b | 445 m | 300 m |
| $[Ni(PQCAH)_2I_2]$ | 3170 m,b | 1600 s,b | 1650 s,b | 455 m | 310 m |
| $[Cu(PQCAH)_2Cl_2]$ | 3170 m,b | 1605 s,b | 1640 s,b | 440 m | 280 m |
| $[Cu(PQCAH)_2Br_2]$ | 3165 m,b | 1610 s,b | 1660 s,b | 460 m | 285 m |

s = strong, b = broad, m = medium

### Electronic spectra and magnetic moments of the complexes

Electronic spectrum of the complex $[Co(PQCAH)_2X_2]$ displays three bands in the range 8360–8100, 15050–14600 and 19210–18060 $cm^{-1}$ which could be assigned to ${}^4T_{1g} \rightarrow {}^4T_{2g}(F)$, ${}^2A_{2g}(F)$ and ${}^4T_{1g}(P)$ transitions respectively. The second transition appears as a shoulder. $\mu_{eff}$ values are 4.7–5.1 B.M. Thus an octahedral geometry is assigned[16] for Co(II) complexes. The octahedral and six coordinated Ni(II) complexes display simple spectra consisting of three transitions from $^3A_{2g}(F) \rightarrow {}^3T_{2g}$, $^3T_{1g}(F)$ and $^3T_{1g}(P)$ levels. These are reported to occur in the following regions: 13000–10000, 18000–15000 and 27000–24000 $cm^{-1}$ respectively.[17] $\mu_{eff}$ values are 3.11–3.24 B.M., which are well within the range of reported values of hexa-coordinated Ni(II) complexes (2.9–3.4 B.M.)[18]. For octahedral Cu(II) complexes[19] the expected $^2E_g \rightarrow {}^2T_{2g}$ band is around 15500 $cm^{-1}$ and $\mu_{eff}$ values are 1.80–1.90 B.M. These values agree well with the expected spin-only values reported for the Cu(II) complexes (1.75–2.20 B.M.)[20].

Hence on the basis of above discussion the complexes of the Co(II), Ni(II) and Cu(II) of the type $[M(PQCAH)_2X_2]$ can be presumed to have octahedral geometry as shown in Fig. 1.



$[M(PQCAH)_2X_2]$
M = Co(II), Ni(II) and Cu(II)
$X = Cl^-, Br^-$ or $I^-$

Fig. 1.

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