**Dear Prof. Miloš Đuran**

Inorganic Chemistry Sub Editor of Journal of Serbian Chemical Society

We appreciate your assistance to improve our manuscript (entitled: Zinc, copper and nickel complexes of a macrocycle synthesized from pyridinedicarboxylic acid: A spectroscopic, thermal and theoretical studies). First of all, we sincerely thank for the heuristic comments put forward by the referees and the editorial office. We have performed our best to revise the manuscript. The electronic version of the manuscript with color fonts at the revised sections is also submitted. Thank you very much for your attention.

I am looking forward to receiving your confirmation.

Sincerely yours

Dr. Sayed Ali Naghi Taheri

**Reply to comments Referee 1**

In order to provide answer to **comments 1** and **2**, the procedure for preparation macrocycle ligand **1** and its complexes are shown in **Scheme** **1** and **2**, respectively. Thus representation was changed for **Scheme 1** and **2** compared to the previous.

Answer to **comment 3**: The possible structure for complexes is proposed on the basis of spectral evidence. In fact, much of the literature suggests complex structure without having crystallographic data. Such as the following articles:

[1] S. A. Elsayed, I. S. Butler, B. J. Claude, S. I. Mostafa, Synthesis, characterization and anticancer activity of 3-formylchromone benzoylhydrazone metal complexes. *Transition Met. Chem.* **2015**, 40, 179-187, doi:10.1007/s11243-014-9904-z

[2] O. A. El-Gammal, M. M. Bekheit, S. A. El-Brashy, Synthesis, characterization and in vitro antimicrobial studies of Co(II), Ni(II) and Cu(II) complexes derived from macrocyclic compartmental Ligand, *Spectrochim. Acta A* **2015**, 137, 207–219, doi:10.1016/j.saa.2014.08.016

[3] Samir Alghool, Mononuclear complexes based on reduced Schiff base derived from L-methionine, synthesis, characterization, thermal and in vitro antimicrobial studies, *J. Therm. Anal. Calorim.* **2015**, 121, 1309-1319, doi:10.1007/s10973-015-4610-4

[4] S. A. Amolegbe, S. Adewuyi, C. A. Akinremi, J. F. Adediji, A. Lawal, A. O. Atayese, J. A. Obaleye, Iron(III) and copper(II) complexes bearing 8-quinolinol with amino-acids mixed ligands: Synthesis, characterization and antibacterial investigation, *Arab. J. Chem.* **2015**, 8, 742–747, doi:10.1016/j.arabjc.2014.11.040

[5] H. A. R. Pramanik, P. C. Paul, P. Mondal, C. R. Bhattacharjee, Mixed ligand complexes of cobalt(III) and iron(III) containing N2O2-chelating Schiff base: Synthesis, characterisation, antimicrobial activity, antioxidant and DFT study, *J. Mol. Struct.* **2015**, 1100, 496-505, doi:10. 1016/j.molstruc.2015.07.076

[6] R. C. Maurya, D. Sutradhar, M. H. Martin, S. Roy, J. Chourasia, A. K. Sharma, P. Vishwakarma, Oxovanadium(IV) complexes of medicinal relevance: Synthesis, characterization, and 3D molecular modeling and analysis of some oxovanadium(IV) complexes in O,N-donor

coordination matrix of sulfa drug Schiff bases derived from a 2-pyrazolin-5-one derivative, *Arab. J. Chem.* **2015**, 8, 78–92, doi:10.1016/j.arabjc.2011.01.009

[7] N. K. Kar, M. K. Singh, R. A. Lal, Synthesis and spectral studies on monometallic ruthenium (III) complexes of N-(2-hydroxysalicyliden-1-yl)methylenebenzoylhydrazide, *Arab. J. Chem.* **2017**, 10, S76–S80doi:10.1016/j.arabjc.2012.05.007

[8] A. A. Kulkarni, S. B. Wankhede, N. D. Dhawale, P. B. Yadav, V. V. Deore, I. D. Gonjari, Synthesis, characterization and biological behavior of some Schiff’s and Mannich base derivatives of Lamotrigine, *Arab. J. Chem.* **2017**, 10, S184–S189, doi:10.1016/j.arabjc. 2012.07.020

[9] H. A. Hosseini, A. Nezhadali, M. Darroudi, Spectrophotometric study of complex formation between iodoquinol (IQ) and Co2+, Mn2+, Cd2+ , Pb2+, and Zn2+ in DMF/MeOH binary mixed solvents, *Arab. J. Chem.* **2017**, 10, S293–S296, doi:10.1016/j.arabjc.2012.07.036

[10] A. Ahmed, R. A. Lal, Synthesis, characterization and electrochemical studies of copper(II) complexes derived from succinoyl- and adipoyldihydrazones, *Arab. J. Chem.* **2017**, 10, S901–S908, doi:10.1016/j.arabjc.2012.12.026

[11] K. D. Patel, H. S. Patel, Synthesis, spectroscopic characterization and thermal studies of some divalent transition metal complexes of 8-hydroxyquinoline, *Arab. J. Chem.* **2017**, 10, S1328–S1335, doi:10.1016/j.arabjc.2013.03.019

And , etc.

Answer to **comment 4**: Solvents such as methanol, ethanol and acetonitrile were used to produce single crystal of complexes. The studied temperature was 40 to 65 ºC. Unfortunately, our attempts to obtain single crystals of all the complexes have not been successful.

The X-ray diffraction (XRD) pattern is not useful for new compounds (macrocycle ligand **1** and three of its complex **2**, **3** and **4**). Because X-ray diffraction pattern to identify and confirm for known compounds.

Answer to **comment 5**:

The caption of **Scheme 1** is changed as follows: Reaction for preparation of macrocycle **1**.

The caption of **Scheme 2** is changed as follows: Reaction for preparation of the complexes [M(L)Cl2] and its formula structure.

Answer to **comment 6**: Corresponding number for compounds were corrected in Tables 1 and 2 in the text of manuscript.

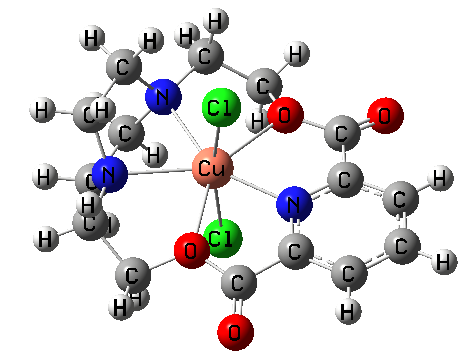
Answer to **comment 7**: The language of this manuscript was corrected by a native English speaker.

**Reply to comments Referee 2**

In the previous work, there was no evidence to show the pyridine nitrogen connection to the central ions in complexes. Therefore, the most likely structure for these complexes with coordination number 6 was octahedral geometry. But in this work, IR spectra show that the pyridine nitrogen was coordinated to metal ions. Therefore, the most likely structure proposed for these complexes with coordination number 7 can be pentagonal bipyramidal geometry. On the other hand, the optimized structure of these complexes is compatible with pentagonal bipyramidal geometry (bond angles of 70.77-73.71 º in plane, and 176.38-178.90 º on axis).

In our previous work, there were no theoretical calculations to predict the structure of complexes.

Now, bipyramidal geometry structure is proposed for those complexes by using theoretical calculations. Molecular structure and geometric parameters are as follows:



|  |  |  |  |
| --- | --- | --- | --- |
| **Table X** Selected optimized geometry parameters of **Cu** complexes (bond length in Angstroms and bong angle in degrees) from **pervious paper**. | | | |
| Parameters |  |  |  |
| Bond length |  | Bond length |  |
| N1-C2 | 1.35 | C14-N15 | 1.48 |
| N1-C6 | 1.35 | C19-N16 | 1.48 |
| C2-C8 | 1.50 | C17-N15 | 1.49 |
| C6-C7 | 1.50 | C18-N16 | 1.49 |
| C7-O9 | 1.24 | C17-C18 | 1.56 |
| C8-O10 | 1.24 | N1-M | 2.11 |
| C7-O11 | 1.36 | O11-M | 2.31 |
| C8-O12 | 1.36 | O12-M | 2.31 |
| C13-O11 | 1.47 | N15-M | 2.33 |
| C20-O12 | 1.47 | N16-M | 2.33 |
| C13-C14 | 1.53 | Cl37-M | 2.39 |
| C19-C20 | 1.53 | Cl38-M | 2.46 |
| Bond angle |  |  |  |
| N1-M-O11 | 73.00 |  |  |
| N1-M-O12 | 73.00 |  |  |
| N15-M-O11 | 74.20 |  |  |
| N16-M-O12 | 74.16 |  |  |
| N15-M-N16 | 65.51 |  |  |
| Cl37-M-Cl38 | 168.45 |  |  |

Lines 372 - 373 related to energy of ligand and complexes were excluded.

Table 4 shows energy bonds of compounds (macrocycle ligand 1 and three of its complexes) and Table 6 shows AIM charges of compounds which were added to the text.

Other proposed notes were carried out in the revised text.

With best regards

Dr. Sayed Ali Naghi Taheri