

# **Synthesis and Characterization of Bioactive Schiff base ligand derived from 2-hydroxy-1-naphthaldehyde and 4-aminobenzonitrile and its Co(II), Ni(II) and Cu(II) Complexes**

## **ABSTRACT**

The Schiff base derived from 2-hydroxy-1-naphthaldehyde and 4-aminobenzonitrile ( $H_2L$ ) and its corresponding Co(II), Ni(II) and Cu(II) complexes were synthesized and characterized by means of FT-IR spectroscopy, thermal analysis, powder x-ray analysis (PXRD), conductivity measurement, magnetic susceptibility and elemental analysis. The mononuclear complexes have been obtained by liquid-assisted mechanochemical method. The Schiff base was synthesized in high yield > 90% with negligible waste. FT-IR study indicates a strong band in the spectra of the Schiff base at  $1577\text{ cm}^{-1}$  assigned to azomethine  $\nu(C=N)$  stretching vibration which shifted to new frequency regions in the spectra of complexes indicating the formation of the anticipated complexes. The Schiff base melted at  $197\text{ }^\circ\text{C}$  while decomposition temperatures of the complexes were in the range of  $265 - 300\text{ }^\circ\text{C}$ , signifying good thermal stability. Molar conductance values were in the range of  $4.39 - 1349\ \Omega^{-1}\text{cm}^2\text{mol}^{-1}$ , indicating non electrolytic nature of the synthesized complexes. Magnetic susceptibility measurement showed that Co(II), Ni(II) and Cu(II) complexes are paramagnetic and exhibit magnetic moment in the range of  $1.50 - 3.19\text{ BM}$ , the values correspond to the tetrahedral geometry. The relative percentage of the constituent elements (C, H, N) obtained from CHN analysis and calculated values using the proposed structures are similar and are in good agreement. The Schiff base and metal complexes were evaluated as agent for inhibiting growth of pathogenic bacteria using disc diffusion method. The results indicated that metal complexes are more active compared to Schiff base ligand against the test organisms.

*Keywords: Schiff base; azomethine; condensation reaction; complexes; mechanochemical synthesis; antimicrobial activity.*

## **1. INTRODUCTION**

"Schiff base are compounds with an azomethine group ( $-HC=N-$ ). Hugo Schiff described them for the first time in 1864. They are condensation products of aldehydes or ketones with primary amine" [1-4]. "The Schiff base formation is really a sequence of two types of reactions, i.e. addition followed by elimination. A large number of aldehydes and ketones have been condensed with various amines to give Schiff bases. The most common catalysts for the formation of Schiff bases are heat, acids or bases. The formation is generally driven to the completion by separation of the product or removal of water, or both. Many Schiff bases can be hydrolyzed back to their aldehydes or ketones and amines by aqueous acid or

base. Schiff bases are generally bidentate, tridentate, tetradentate or polydentate ligands capable of forming very stable complexes with transition metals via chelation" [5].

"Schiff bases are among the most common organic compounds. They are utilized as pigments and dyes, catalysts, organic synthesis intermediates, and polymer stabilizers. Schiff bases have also been demonstrated to have antifungal, antibacterial, antimalarial, antiproliferative, anti-inflammatory, antiviral, and antipyretic effects" [6,7]. "The presence of an imine group in such compounds has been proven to be critical to their biological actions" [8–12].

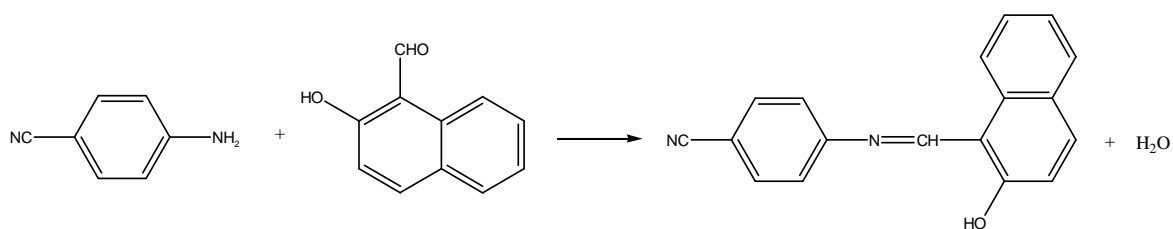
"Mechanochemistry can be as easy as grinding two reactants together in a pestle and mortar. Using mechanochemical synthesis can reduce environmental pollution and be more convenient than solvent-based synthesis" [13,14]. "Several groups have used this method to demonstrate mechanochemical synthesis of metal complexes" [15,16]. "Surprisingly, mechanochemical synthesis is often faster and more convenient than solvent-based techniques. As a society, we are becoming more conscious of the environmental effects of human activities and, as a result, of the need to produce cleaner, more energy-efficient technologies" [17]. In this paper, we will look at recent studies on the use of mechanochemistry (grinding) to generate Schiff base and metal complexes.

## 2. MATERIALS AND METHODS

All the reagents were purchased from Sigma-Aldrich UK and are of analytical grade. They were not further purified before use.

### 2.1 Synthesis of Schiff base

"The solid reactants, 2-hydroxy-1-naphthaldehyde (1 mmol, 0.17218 g) and 4-aminobenzonitrile (1 mmol, 0.11814 g), were carefully weighed into a mortar. The reactant was ground for 45 minutes to yield a yellow orange powder. The powder was removed from the mortar and placed in a sample container" [18].



Scheme 1. Synthetic reaction of Schiff base

### 2.2 Synthesis of Complexes

"The reaction mixture of the metal(II) acetate (1 mmol) and Schiff base ligand (2 mmol) in a 1:2 mole ratio was ground in a mortar and pestle for 60 minutes to produce the coloured powder complexes. The products were air dried at room temperature (M = Ni or Cu)" [18].

### 2.3 Antibacterial and Antifungal activities

Antibacterial and antifungal activities were tested using the paper disk diffusion method. *Staphylococcus aureus*, *Escherichia coli*, and *Salmonella typhi* strains of bacteria as well as *Aspergillus fumigatus* and *Aspergillus flavus* strains of fungi were examined. Gram staining and biochemical tests were used to identify the microbial isolates, which were obtained from the Aminu Kano Teaching Hospital's Department of Chemical Pathology [19].

### 3. RESULTS AND DISCUSSION

#### 3.1 Physical properties, molar conductance and magnetic susceptibility

The Schiff base ligand and complexes that were synthesized were coloured. Electronic d-d transitions are what give complexes their colours. The percentage yield of the Schiff base and metal complexes synthesized by the mechanochemical method ranged from 84.5 to 92.2% (Table 1). A crystalline solid with a melting point of 197°C was found to be the Schiff base. Complexation is the cause of all complexes having a decomposition temperature higher than the melting point of the Schiff base. The complexes decomposed in the 265 to 292 °C range, demonstrating good thermal stability [20].

The molar conductivity values of the complexes in a 10<sup>-3</sup> M solution in DMF were measured. Within the bounds of their solubility, conductivity measurements have frequently been used to describe metal chelates. The higher molecular ions that a complex liberates in solution, the higher will be its molar conductivity, and vice versa. They offer a method of testing the degree of ionization of the complexes. The values of the molar conductance ( $\Lambda_m$ ) of the metal complexes are shown in Table 1. The findings revealed that the molar conductance values of the Co(II), Ni(II), and Cu(II) complexes were 13.49, 9.54, and 4.39  $\Omega^{-1}\text{cm}^2\text{mol}^{-1}$ , respectively. The conductivity data of the synthesized complexes make it abundantly clear that the complexes were discovered to be non-electrolytes and very low to account for any complex dissociation in DMF [21].

According to the effective magnetic moment values of the complexes (Table 1) derived from the Schiff base, Co(II), Ni(II), and Cu(II) complexes are paramagnetic in nature and have magnetic moments of 3.19, 2.67, and 1.50 BM, respectively, which are close to spin-only values and may be caused by orbital contribution and ligand effect. This value is less than the spin-only value reported by Kumari, 2017 (4.3–5.2 BM) for three unpaired electrons. Tetrahedral geometry matches the observed magnetic parameter [22].

Table 1. Physical properties, molar conductance and magnetic susceptibility of Schiff base (H<sub>2</sub>L) complexes

| Compound              | Molecular Formula  | Colour | Melting Point/Decomp.Temp (°C) | Yield (%) | Molar conductance ( $\Omega^{-1}\text{cm}^2\text{mol}^{-1}$ ) | $\mu_{\text{eff}}$ (BM) |
|-----------------------|--|--------|--------------------------------|-----------|---|-------------------------|
| (H <sub>2</sub> L)    | (C <sub>18</sub> H <sub>12</sub> N <sub>2</sub> O)                   | Orange | 197                            | 89.6      | -   | -                       |
| [Co(L) <sub>2</sub> ] | [Co(C <sub>18</sub> H <sub>11</sub> N <sub>2</sub> O) <sub>2</sub> ] | Red    | 292                            | 90.2      | 13.49   | 3.19                    |
| [Ni(L) <sub>2</sub> ] | [Ni(C <sub>18</sub> H <sub>11</sub> N <sub>2</sub> O) <sub>2</sub> ] | Yellow | 265                            | 84.5      | 9.54  | 2.67                    |
| [Cu(L) <sub>2</sub> ] | [Cu(C <sub>18</sub> H <sub>11</sub> N <sub>2</sub> O) <sub>2</sub> ] | Tan    | 280                            | 92.2      | 4.39  | 1.50                    |

#### 3.2 Powder x-ray diffraction

The powder x-ray diffraction analysis of 2-hydroxy-1-naphthaldehyde and 4-aminobenzonitrile revealed distinct and intense peaks that were missing from the Schiff base's powder x-ray diffraction (Fig 1). Instead, additional strong and sharp peaks were seen, indicating the formation of the target Schiff base [23]. Schiff base powder x-ray diffraction patterns show sharp reflections, indicating that the compound is crystalline [24,25]. The diffractogram of the complexes in relation to the reactants shows different reflection peaks as well.

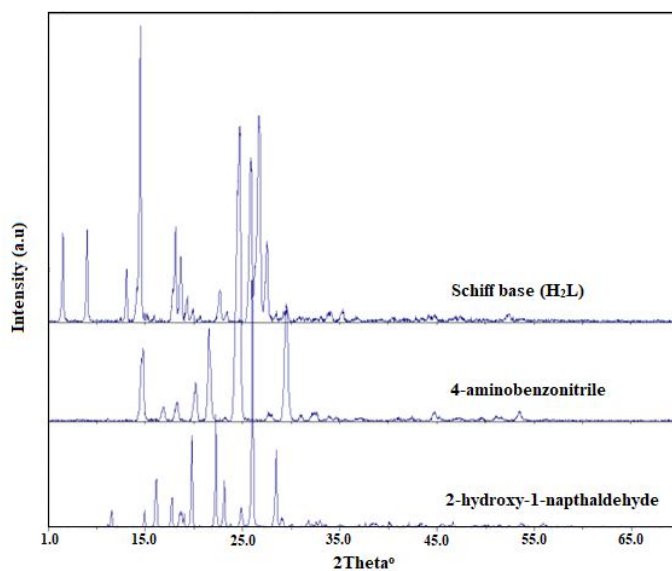


Figure 1. Comparison of PXRD patterns of 2-Hydroxy-1-naphthaldehyde, 4-aminobenzonitrile and Schiff base

### 3.3 Solubility test

The Schiff base is soluble in some aprotic solvents, such as DMSO, DMF, acetonitrile and insoluble in n-hexane. It was discovered that the metal complexes were soluble in aprotic solvents like DMF and DMSO, only slightly soluble in acetonitrile and acetone, and insoluble in n-hexane. This suggested that the produced compounds were soluble in most aprotic solvents. (Table 2).

Table 2. Solubility Test of Schiff base ( $H_2L$ ) and its metal complexes

| Compound                  | DMF | DMSO | Acetonitrile | Diethyl ether | Chloroform | Acetone | n-hexane |
|---------------------------|-----|------|--------------|---------------|------------|---------|----------|
| ( $H_2L$ )                | S   | S    | SS           | SS            | S          | SS      | IS       |
| [Co( $L$ ) <sub>2</sub> ] | S   | S    | SS           | SS            | SS         | SS      | IS       |
| [Ni( $L$ ) <sub>2</sub> ] | S   | S    | SS           | SS            | S          | SS      | IS       |
| [Cu( $L$ ) <sub>2</sub> ] | SS  | SS   | IS           | SS            | SS         | SS      | IS       |

S-Soluble, SS- Slightly soluble, IS- Insoluble

### 3.4 Infrared spectroscopy

The Schiff base's IR spectra reveal the formation of a new peak ( $1577\text{ cm}^{-1}$ ) caused by the azomethine group (Table 3). The O-H absorption bond in the Schiff base was also discovered to be present at  $3692\text{ cm}^{-1}$ . The shift of the azomethine peak in the complex spectra from to  $1599\text{ cm}^{-1}$  indicates that the coordination activity of the azomethine nitrogen. The emergence of the  $434\text{--}475\text{ cm}^{-1}$  and  $542\text{--}545\text{ cm}^{-1}$  bands in the low-frequency region for the respective M–O and M–N ions provided additional evidence in favor of the coordination of the Schiff base to the metal ion. In the spectra of the ligand, these bands were absent. These two novel bands (M–O and M–N) have metal ions that coordinate through phenolic oxygen and azomethine nitrogen. higher wavenumber of M–N than M–O in

Infrared spectroscopy was due to the difference in electronegativity between the two elements (N and O) [26,27].

Table 3. Infrared spectra result of Schiff base (H<sub>2</sub>L) and its complexes

| Compound              | v(OH) cm <sup>-1</sup> | v(C=N) cm <sup>-1</sup> | v(M-O) cm <sup>-1</sup> | v(M-N) cm <sup>-1</sup> |
|-----------------------|------------------------|-------------------------|-------------------------|-------------------------|
| (H <sub>2</sub> L)    | 3692                   | 1577                    | -                       | -                       |
| [Co(L) <sub>2</sub> ] | -                      | 1581                    | 475                     | 545                     |
| [Ni(L) <sub>2</sub> ] | -                      | 1599                    | 445                     | 542                     |
| [Cu(L) <sub>2</sub> ] | -                      | 1581                    | 434                     | 542                     |

### 3.5 Elemental analysis

The calculated values using the suggested structures are similar to and in good agreement with the relative percentage of the constituent elements (C, H, and N) obtained from CHN analysis (Table 4). This is in support of the complexes' proposed structures. Small variations in some of the values that are within the allowable range are explained by [28].

Table 4. Elemental Analysis of Schiff base complexes

| Compound              | C (%)             | H (%)             | N (%)             |
|-----------------------|-------------------|-------------------|-------------------|
|                       | Found(Calculated) | Found(Calculated) | Found(Calculated) |
| [Co(L) <sub>2</sub> ] | 70.60 (71.88)     | 3.22 (3.69)       | 8.72 (9.31)       |
| [Ni(L) <sub>2</sub> ] | 70.50 (71.91)     | 3.30 (3.69)       | 8.60 (9.32)       |
| [Cu(L) <sub>2</sub> ] | 70.02 (71.34)     | 3.20 (4.46)       | 8.60 (9.32)       |

### 3.6 Antimicrobial activity

The antibacterial activity of the Schiff base was reasonable against bacterial isolates, with a maximum zone of inhibition of 13 mm against *E. coli* (Table 5). The complexes derived from the Schiff base exhibit significant activity, particularly at higher concentrations against the organism tested with inhibition zone range of 08 - 16 mm. At varying concentrations of the tested fungal isolates, Schiff base and its complexes also exhibit moderate antifungal activity with inhibition zones ranging from 07 to 16 mm (Table 6).

It has been established that the synthesized compounds' antimicrobial properties are due to the azomethine group (5,8–10). The chelation of the Schiff base with metal ions may be responsible for the increased activity of the metal(II) complexes compared to the free ligand. The polarity of the metal ion will be reduced to a greater extent due to the overlap of the ligand orbitals during the chelation process and the partial sharing of the metal ion's positive charge with donor groups, this will increase the permeability of the compounds through the lipid layer of cell membranes [29].

Table 5. Antibacterial activity of Schiff base and complexes showing the inhibition zone (mm)

| Compound              | <i>S. aureus</i> |    |    | <i>E. Coli</i> |    |    | <i>S typhi</i> |    |    |    |
|-----------------------|------------------|----|----|----------------|----|----|----------------|----|----|----|
|                       | Conc(mg/ml)      | 60 | 30 | 15             | 60 | 30 | 15             | 60 | 30 | 15 |
| Gentamycin (standard) | --               | 32 | -- | --             | 30 | -- | --             | 35 | -- | -- |
| DMSO(Control)         | 6                | 6  | 6  | 6              | 6  | 6  | 6              | 6  | 6  | 6  |
| (H <sub>2</sub> L)    | 12               | 10 | 08 | 13             | 09 | 08 | 12             | -- | -- | 11 |
| [Co(L) <sub>2</sub> ] | 16               | 12 | 09 | 14             | 12 | 11 | 14             | 13 | 12 | 12 |
| [Ni(L) <sub>2</sub> ] | 15               | 13 | 11 | 15             | 13 | 09 | 12             | 11 | -- | -- |
| [Cu(L) <sub>2</sub> ] | 13               | 11 | 09 | 14             | 10 | 09 | 15             | 12 | 10 | 10 |

Table 6. Antifungal activity of Schiff base and complexes showing the inhibition zone (mm)

| Compound                            | <i>C. albicum</i> |    |    | <i>A. flavus</i> |    |    | <i>A. fumigatus</i> |    |    |
|-------------------------------------|-------------------|----|----|------------------|----|----|---------------------|----|----|
| Conc(mg/ml)                         | 60                | 30 | 15 | 60               | 30 | 15 | 60                  | 30 | 15 |
| Nystatin<br>(standard)              | --                | 32 | -- | --               | 30 | -- | --                  | 35 | -- |
| DMSO(Control)<br>(H <sub>2</sub> L) | 6                 | 6  | 6  | 6                | 6  | 6  | 6                   | 6  | 6  |
| [Co(L) <sub>2</sub> ]               | 10                | 08 | 07 | 12               | 11 | 09 | 11                  | 09 | -- |
| [Ni(L) <sub>2</sub> ]               | 15                | 11 | -- | 15               | 13 | 10 | 16                  | 10 | 09 |
| [Cu(L) <sub>2</sub> ]               | 14                | -- | 16 | 09               | 13 | 14 | 12                  | 10 | 07 |
| [Cu(L) <sub>2</sub> ]               | 14                | 10 | -- | 14               | 12 | 10 | 10                  | 16 | 09 |

#### 4. CONCLUSION

This paper describes the synthesis and characterization of Co(II), Ni(II) and Cu(II) complexes containing N, O donor bidentate Schiff base ligand. The Schiff base was synthesized by mechanochemical reaction of 2-hydroxy-1-naphthaldehyde and 4-aminobenzonitrile. The synthesized compounds were characterized by elemental analyses, FT-IR, PXRD, magnetic susceptibility, and conductivity measurement. The Schiff base and complexes were found to be coloured, non-electrolyte and thermally stable. Based on FT-IR spectra, the complexes were found to be coordinated by the Schiff base's azomethine nitrogen and phenolic oxygen. Elemental analysis revealed M:L ratios of 1:2. A proposed structure is well supported by the complexes' spectroscopic data (Figure 2). The antimicrobial screening results of the test compounds show that they have mild to moderate antimicrobial activities when compared to standard drugs. The nonpolluting nature of the mechanochemical approach qualifies it as a green method for the synthesis of Schiff bases and complexes.

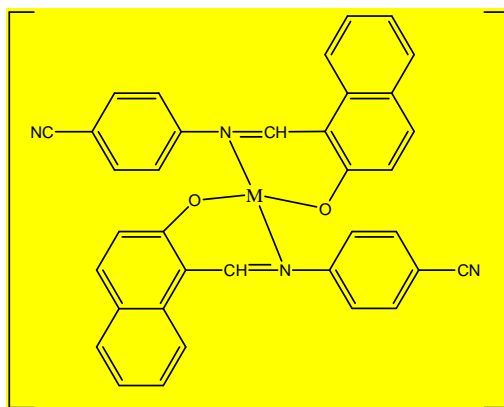


Figure 2. Proposed Structure of Complexes  
Where M = Co(II), Ni(II), or Cu(II)

#### REFERENCES

- Hernandez-Molina R, Mederos A i. Acyclic and macrocyclic Schiff base ligands. ChemInform. 2004;35(48): 411-46. DOI: 10.1002/chin.200448246
- Selim EAB, Al-Douh MH, Al-Nohey DS, Al-Haik WM, Sonbol KS. Synthesis, Characterization, and Antimicrobial Activity Studies of New Schiff Base Tetradentate

- Ligands and Their Manganese (II) Complexes. *Asian J Chem Sci.* 2020;7(4):35–46. DOI: 10.9734/AJOCS/2020/v7i419030
- Abdalahem Bin Selim E, Hadi Al-Douh M, Hadi Abdullah H, Salim Al-Nohey D. Electronic Structure of Two New Bis-Schiff Base Ligands using DFT Method. *Asian j Appl Chem R* 2021;10(2):10–5. DOI: 10.9734/AJACR/2021/v10i230231
  - Farajzadeh S, Moghadam PN, Khalafy J. Synthesis of Schiff-base pendant groups on PGMA-MAn for removal of colored pollutants. *J Iran Chem Soc.* 2020;1–10
  - Sani S, Kurawa MA, Siraj IT, Koki IB. Mechanochemical synthesis: A suitable method to the synthesis of some diamines Schiff bases. *Bayero J Pure Appl Sci.* 2019;12(1):90–6. DOI: 10.4314/bajopas.v12i1.12
  - Przybylski P, Huczynski A, Pyta K, Brzezinski B, Bartl F. Biological properties of Schiff bases and azo derivatives of phenols. *Curr Org Chem.* 2009;13(2):124–48. DOI: 10.2174/138527209787193774
  - Alibrahim KA, Alsuhaibani AM, Refat MS. Synthesis and spectroscopic characterizations of manganese (II), iron (III), copper (II) and zinc (II) hydrazine complexes as catalytic activity agents. *Bull Chem Soc Ethiop.* 2022;36(1):33–44. DOI: 10.4314/bcse.v36i1.4
  - Bringmann G, Dreyer M, Faber JH, Dalsgaard PW, Staerk D, Jaroszewski JW, et al. Ancistrotanine C and Related 5, 1'-and 7, 3'-Coupled Naphthylisoquinoline Alkaloids from *Ancistrocladus tanzaniensis*. *J Nat Prod.* 2004;67(5):743–8. DOI: 10.1021/np0340549
  - Guo Z, Xing R, Liu S, Zhong Z, Ji X, Wang L, et al. Antifungal properties of Schiff bases of chitosan, N-substituted chitosan and quaternized chitosan. *Carbohydr Res.* 2007;342(10):1329–32. DOI: 10.1016/j.carres.2007.04.006
  - Souza AO de, Galetti F, Silva CL, Bicalho B, Parma MM, Fonseca SF, et al. Antimycobacterial and cytotoxicity activity of synthetic and natural compounds. *Quím Nova.* 2007;30(7):1563–6. DOI: 10.1590/S0100-40422007000700012
  - Sani S, Siraj IT. Copper (II) and Zinc (II) Complexes Synthesized by Green Mechanochemical Method and their Antimicrobial Studies. *Commun Phys Sci.* 2021;7(2):67–75.
  - Haggam RA, Soylem EA, Assy MG, Arastiedy MF. Synthesis and antimicrobial evaluation of new series of quinazolin-5-one derivatives. *J Iran Chem Soc.* 2020;1–9. DOI: 10.1007/s13738-020-01896-0
  - Braga D, Maini L, Grepioni F. Mechanochemical preparation of co-crystals. *Chem Soc Rev.* 2013;42(18):7638–48. DOI: 10.1039/C3CS60014A
  - Luo M, Li HM. One-pot synthesis and biological and catalytic applications of organometallic complexes involving oxazolines and (R)/(S)- $\alpha$ -phenylethylamine. *J Iran Chem Soc.* 2020;17(4):963–71. DOI: 10.1007/s13738-019-01830-z

15. Bowmaker GA, Chaichit N, Pakawatchai C, Skelton BW, White AH. Solvent-assisted mechanochemical synthesis of metal complexes. *Dalton Trans.* 2008;(22):2926–8. DOI: [10.1039/B804229M](https://doi.org/10.1039/B804229M)
16. Sani S. Green Synthesis, Characterization and Antimicrobial Studies of Ni (II) and Cu (II) Schiff Base Complexes. *Int J Green Chem.* 2022;8(1):9–18.
17. Garay AL, Pichon A, James SL. Solvent-free synthesis of metal complexes. *Chem Soc Rev.* 2007;36(6):846–55. DOI: [10.1039/B600363J](https://doi.org/10.1039/B600363J)
18. Stilinović V, Cinčić D, Zbačnik M, Kaitner B. Controlling solvate formation of a Schiff base by combining mechano-chemistry with solution synthesis. *Croat Chem Acta.* 2012;85(4):485–93. DOI: [10.5562/cca2111](https://doi.org/10.5562/cca2111)
19. Iniyama GE, Nfor EN, Okon ED, Iorkpilig IT. Antimicrobial activities of synthesized Zinc (II) mixed ligand complexes derived from 2-acetylpyridine-4-phenylsemicarbazone and nitrogen-sulphurmonodentate ligands. *Int J Sci Techn Res.* 2014;3(11):73–7.
20. Nejo AA, Kolawole GA, Nejo AO. Synthesis, characterization, antibacterial, and thermal studies of unsymmetrical Schiff-base complexes of cobalt (II). *J Coord Chem.* 2010;63(24):4398–410. DOI: [10.1080/00958972.2010.532871](https://doi.org/10.1080/00958972.2010.532871)
21. Shaker SA, Aziz YFA, Salleh AA. Synthesis and Characterization of Mixed Ligand Complexes of 8-Hydroxyquinoline and o-hydroxybenzylidene-1-phenyl-2, 3-dimethyl-4-amino-3-pyrazolin-5-on with Fe (II), Co (II), Ni (II) and Cu (II) ions. *Eur J Sci Res.* 2009;33(4):702–9.
22. Carabineiro SA, Silva LC, Gomes PT, Pereira LC, Veiros LF, Pascu SI, et al. Synthesis and characterization of tetrahedral and square planar bis (iminopyrrolyl) complexes of cobalt (II). *Inorg Chem.* 2007;46(17):6880–90. DOI: [10.1021/ic062125w](https://doi.org/10.1021/ic062125w)
23. Zhong W, Zhong GQ, Zhang Y, Zhong Q. Solvent-Free Synthesis and Characterization of the Zn (II) Complexes with Amino Acid Schiff Base. *Advanced Materials Research. Trans Tech Publ;* 2012. p. 740–5. DOI: [10.4028/www.scientific.net/AMR.455-456.740](https://doi.org/10.4028/www.scientific.net/AMR.455-456.740)
24. Cinčić D, Kaitner B. Schiff base derived from 2-hydroxy-1-naphthaldehyde and liquid-assisted mechanochemical synthesis of its isostructural Cu (II) and Co (II) complexes. *CrystEngComm.* 2011;13(13):4351–7. DOI: [10.1039/C0CE00421A](https://doi.org/10.1039/C0CE00421A)
25. Cindrić M, Uzelac M, Cinčić D, Halasz I, Pavlović G, Hrenar T, et al. Three routes to nickel (II) salicylaldehyde 4-phenyl and 4-methylthiosemicarbazonato complexes: mechanochemical, electrochemical and conventional approach. *CrystEngComm.* 2012;14(9):3039–45. DOI: [10.1039/C2CE06611D](https://doi.org/10.1039/C2CE06611D)
26. Joseyphus RS, Nair MS. Synthesis, characterization and biological studies of some Co (II), Ni (II) and Cu (II) complexes derived from indole-3-carboxaldehyde and glycylglycine as Schiff base ligand. *Arab J Chem.* 2010;3(4):195–204.
27. Zaky R, Fekri A. Solvent-free mechanochemical synthesis of Zn (II), Cd (II), and Cu (II) complexes with 1-(4-methoxyphenyl)-4-(2-(1-(pyridin-2-yl) ethylidene) hydrazinyl)-1H-pyrrole-3-carbonitrile. *Green Process Synth.* 2018;7(6):515–23. DOI: [10.1515/gps-2017-0057](https://doi.org/10.1515/gps-2017-0057)

28. Tigineh GT, Liu LK. Studies on Mechanochemistry: Solid Coordination Compounds from Primary Aromatic Amines and Cobalt (II) Chloride Hexahydrate. *J Chin Chem Soc.* 2014;61(11):1180–7.
29. Kumar D, Chadda S, Sharma J, Surain P. Syntheses, spectral characterization, and antimicrobial studies on the coordination compounds of metal ions with Schiff base containing both aliphatic and aromatic hydrazide moieties. *Bioinorg Chem Appl.* 2013;2013. DOI: [10.1155/2013/981764](https://doi.org/10.1155/2013/981764)