SYNTHESIS, CHARACTERIZATION AND ANTIBACTERIAL STUDIES OF AMINOBENZOTHIAZOLE DERIVATIVE AND ITS COPPER(II) COMPLEX

S. B. Rahardjo¹,², M. K. Widowati¹, Y. A. Rasyda¹, S.D. Marliyana¹,²

¹Department of Chemistry, Graduate Program, Faculty of Mathematics and Natural Sciences, Sebelas Maret University, Surakarta, Indonesia
²Department of Chemistry, Faculty of Mathematics and Natural Sciences, Sebelas Maret University, Surakarta, Indonesia

Corresponding Author: sentotbr@staff.uns.ac.id

ABSTRACT

This research aimed to synthesize and characterize Schiff base ligand (SB) derived from 2-aminobenzothiazole with o-anisaldehyde and its Cu(II) complex and investigate their antibacterial activities. The complex (Cu(II)-SB) was synthesized in a mole ratio of 1:2 of metal to the ligand. The complex’s physical measurements included UV-Vis, Atomic Absorption Spectrophotometer (AAS), Thermogravimetric Analysis/Differential Thermal Analysis (TG/DTA), FTIR, electrical conductivity, and magnetic susceptibility. The formation of the complex was indicated by a shift in the maximum wavelength of UV-Vis spectra toward smaller, from 823 nm for Cu(II) to 731 nm in the complex. The proposed complex formula was [Cu(SB)₂]SO₄·H₂O. The IR spectral data on the Cu(II)-SB showed that SB was coordinated through the C=N azomethine and C=N thiazole functional groups. The magnetic moment result µₑffective of 2.07 B.M, indicated that the complex was paramagnetic and was expected to have a square planar geometry. [Cu(SB)₂]SO₄·H₂O exhibited the highest antibacterial activity than the ligands and metals.

Keywords: Synthesis, Schiff Base, Copper, Complex, Aminobenzothiazole, Antibacterial.

INTRODUCTION

This research aimed to synthesize and characterize Schiff base ligand (SB) derived from 2-aminobenzothiazole with o-anisaldehyde and its Cu(II) complex and investigate their antibacterial activities. The complex (Cu(II)-SB) was synthesized in a mole ratio of 1:2 of metal to the ligand. The complex’s physical measurements included UV-Vis, Atomic Absorption Spectrophotometer (AAS), Thermogravimetric Analysis/Differential Thermal Analysis (TG/DTA), FTIR, electrical conductivity, and magnetic susceptibility. The formation of the complex was indicated by a shift in the maximum wavelength of UV-Vis spectra toward smaller, from 823 nm for Cu(II) to 731 nm in the complex. The proposed complex formula was [Cu(SB)₂]SO₄·H₂O. The IR spectral data on the Cu(II)-SB showed that SB was coordinated through the C=N azomethine and C=N thiazole functional groups. The magnetic moment result µₑffective of 2.07 B.M, indicated that the complex was paramagnetic and was expected to have a square planar geometry. [Cu(SB)₂]SO₄·H₂O exhibited the highest antibacterial activity than the ligands and metals.
compounds that can deal with bacterial resistance.\(^\text{15}\) Therefore, this research aims to synthesize copper complex compound with Schiff base ligand (SB) derived from aminobenzothiazole and observe its biological activity.

**EXPERIMENTAL**

**Material and Methods**

Chemicals in this research were used directly without purification. The analysis of SB was carried out using Infrared spectra, \(^1\)H-NMR, and \(^{13}\)C-NMR. Measurements of copper content were analyzed with Shimadzu AA-6650 Atomic Absorption Spectrophotometer. Electronic spectral data of Cu(II) solution and the complex solution were analyzed in the range of 400–800 nm using UV-Vis Lambda 25 Perkin Elmer spectrophotometer using methanol as solvent. Infrared spectra were recorded on an FTIR Prestige-21 Shimadzu spectrophotometer. Thermogram was recorded at a temperature in the range of 20 to 900°C with a heating rate of a 10 °C/min on a TG/DTA Linseis STA PT-1600. The electrical conductivity was calculated by Jenway CE 4071 conductivity meter. Standard solutions and copper complex solutions were made at the same concentration (0,001 M) in methanol.

**Synthesis of Schiff Base Ligand (SB)**

Synthesis of SB from 2-aminobenzothiazole and o-anisaldehyde was done by adding o-anisaldehyde solution (1 mmol; 0.136 grams; 3 mL methanol) dropwise on 2-aminobenzothiazole solution (1 mmol; 0.150 gram; 3 mL methanol). After stirring for 5 minutes, six drops of glacial acetic acid were added and stirred for an hour at room temperature. The reaction was monitored by TLC. After an hour, the solids appeared. The solids were filtered, then washed with methanol.

**Synthesis of Cu(II) Complex (Cu(II)-SB)**

Synthesis of Cu(II) complex with SB was carried out by adding CuSO\(_4\)·5H\(_2\)O (0.124 g, 1 mmol) solution in methanol as solvent (5 mL) dropwise to SB solution (0.266 g, 2 mmol) in methanol (5 mL) as solvent. The solution was then stirred without heating for 3 hours until blue solids were observed. The obtained solids were filtered and dried with a vacuum desiccator.

**Antibacterial Study**

Antibacterial activity tests of complex compounds were carried out by agar diffusion method by measuring bacterial inhibition.\(^\text{16}\) Four ATCC bacteria used in testing were *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Escherichia coli*, and *Pseudomonas aeruginosa*. The antibacterial test instruments were first sterilized by heating using an autoclave at 121°C at a pressure of 15 psi (per square inch) for 15 minutes. Agar Muller Hinton's media was dissolved in water, and the mixture was heated on a water bath while stirring with a magnetic stirrer. The media suspension was inserted into the petri dish until it becomes solid. The bacterial suspension was made by dissolving a bacterial loop in sterile 9% NaCl. The compound solution was diluted using DMSO solvent with various concentrations. A compound solution with each concentration was dropped on 10 μL paper disks using a micropipette. The bacterial suspension was etched evenly on the surface of the media suspension. Chloramphenicol was placed on media as a positive control and DMSO as a negative control. The paper disk's location was spaced with one another, then incubated for 24 hours at 37°C. The inhibition of bacterial growth was known by measuring the clear zone's diameter around the test sample.

**RESULTS AND DISCUSSION**

**Synthesis of Schiff Base Ligand (SB)**

Synthesis of Schiff base compounds was carried out by reacting o-anisaldehyde with 2-aminobenzothiazole in a mole ratio of 1: 1 and by adding glacial acetic acid catalyst at room temperature for 3 hours. TLC test was performed to determine whether SB had formed. The eluents used were N-hexane: ethyl acetate = 8: 2. TLC analysis was carried out to ensure no other compounds were formed so that the Schiff base ligand produced a single spot-on TLC. The Rf value of this base Schiff ligand was 0.76. The reaction scheme of SB formation is shown in Fig.-1.
From the $^1$H-NMR proton data (Fig.-2), a chemical shift at 8.69 ppm shows an azomethine proton. At a chemical shift of 6-8 ppm, the multiplet signal indicates the aromatic ring proton. The absence of a chemical shift with a broad peak at 1-2 ppm, a shift from the proton amine, shows that the Schiff base ligand has formed through the carbonyl and the amine group. C-NMR spectrum (Fig.-3) shows the number of carbon shifts corresponding to the number of carbon atoms in the Schiff base ligand and also shows a signal of C$_{\text{azomethine}}$ at the 165.6 ppm.
Analysis of SB was also performed using infrared spectroscopy. Infrared spectra of SB, 2-aminobenzothiazole, and o-anisaldehyde can be seen in Fig.-4. There is no absorption of N-H belonging to aminobenzothiazole at 3394 and 3271 cm\(^{-1}\). Also, there is a loss of C=O absorption corresponding to anisaldehyde at 1823 cm\(^{-1}\). These two groups disappear because aminobenzothiazole and o-anisaldehyde were reacted to form SB with the azomethine (C=N) group. This reaction is confirmed by the appearance of azomethine (C=N) absorption at 1604 cm\(^{-1}\). The infrared spectra results can strengthen the analysis that SB has been successfully formed.

![Fig.-4: FTIR Spectra of 2-aminobenzothiazole, o-anisaldehyde, and SB](image)

**Synthesis of Cu(II)-SB Complex**

Synthesis of the complex was successfully carried out. The formed complex was indicated by a shift in the maximum wavelength towards the shorter one in the UV-Vis spectra from 823 nm to 778 nm as shown in Fig.-5. The same thing happened in Cu(II)-(6-methyl-pyridin-2-ylamino)acetic acid. The maximum wavelength shifted from 850 nm (CuCl\(_2\)·H\(_2\)O) to 775 nm in the complex.

![Fig.-5: Electronic Spectra of the Metal and the Complex](image)

**Determination of Empirical Formula of the Complex**

AAS result showed that Cu(II)-SB contains 9.05% of copper. Theoretical calculations in various complex formulas of Cu(II)-SB were compared with the AAS result. (Table-1). Thus, the complex formula can be estimated to be Cu(SB)\(_2\)(SO\(_4\))(H\(_2\)O)\(_n\) (n = 0, 1, or 2).

The thermogram of Cu(II)-SB is presented in Fig.-6. The thermogram of the complex showed a decrease in mass at a temperature of 30-160 °C accompanied by an endothermic peak at 160 °C. The mass
reduction that occurs in the Cu(II)-SB complex is 3.15% equivalent to the release of one H$_2$O molecule (calc. 2.5%). H$_2$O molecules that were uncoordinated to the complex and have a position as crystal water will begin to decompose at a temperature of 10-170°C, while H$_2$O molecules that coordinate with the complex will begin to decompose at temperatures of 150-220 °C. This also occurs in other complexes, such as the Cu(II) complex with the nicotinohydrazide derivative Schiff base ligand, which experiences the release of one H$_2$O molecule as crystal water at a temperature of 30-130 °C. From the thermogram results and the calculation of the release of H$_2$O molecules, the possible complex formula is Cu(SB)$_2$SO$_4$·H$_2$O.

Table-1: AAS Result and the Possible Complex Formulas

| Empirical Formula | Molecular Weight (g/mol) | Theoretical Calculation (%) | AAS Result (%) |
|-------------------|--------------------------|-----------------------------|----------------|
| Cu(SB)$_2$(SO$_4$) | 696.16                   | 9.12                        | 9.05           |
| Cu(SB)$_2$(SO$_4$)(H$_2$O)$_2$ | 732.19               | 8.67                        |                |

![Fig.- 6: TG/DTA Data of the Complex](image)

The results of electrical conductivity measurements of standard and complex compounds in methanol are shown in Table-2. The number of ions of the complex can be determined by comparing the molar conductivity of a complex solution with a standard solution's molar conductivity. The molar conductivity value of Cu(II)-SB complex approaches the electrical conductivity value of CuSO$_4$·5H$_2$O solution with the ratio of cation: anion = 1:1, so the estimated complex formula formed is [Cu(SB)$_2$]SO$_4$·H$_2$O.

Table-2: Molar Conductivity of 1.10$^{-3}$ M Standard Solutions and the Complex in Methanol

| Compounds      | Molar Conductance $\Lambda^\infty$ (S.cm$^2$/mol) | Cation: Anion Ratio | Number of Ions |
|----------------|-----------------------------------------------|---------------------|----------------|
| Methanol       | -                                             | -                   | 0              |
| CuSO$_4$·5H$_2$O | 8                                             | 1 : 1               | 2              |
| FeSO$_4$·7H$_2$O | 16                                            | 1 : 1               | 2              |
| Cu(NO$_3$)$_2$·5H$_2$O | 100                                          | 2 : 1               | 3              |
| Co(NO$_3$)$_2$·6H$_2$O | 130                                          | 2 : 1               | 3              |
| Cr(NO$_3$)$_3$·6H$_2$O | 160                                          | 3 : 1               | 4              |
| Cu(II)-SB      | 6                                             | 1 : 1               | 1              |

**Infrared Analysis**

From the results of infrared data, the complex undergoes a shift in functional group absorption. The infrared spectra are shown in Fig.-7. IR spectrum of [Cu(SB)$_2$]SO$_4$·H$_2$O showed a shift in C=N azomethine and C=N thiazole groups' absorption. There was a shift of C=N azomethine and C=N thiazole absorption from 1601 and 1558 cm$^{-1}$ in SB to 1648 and 1546 cm$^{-1}$ in the complex, respectively. This shift indicates the coordination...
of the bidentate Schiff base ligands through the $C=N_{azomethine}$ and $C=N_{thiazole}$ functional groups. The appearance of Cu-N absorption also indicates that the ligand was coordinated to the metal via the N donor atom. The presence of O-H absorption at 3299 cm$^{-1}$ also indicates the presence of H$_2$O in the complex.

Electronic Spectra
Complex [Cu(SB)$_2$]SO$_4$·H$_2$O has one peak absorption at 778 nm with a relatively small value of molar absorptivity ($\varepsilon$) (<1000 L/mol·cm) ($\varepsilon = 48$ L/mol·cm; $\nu = 12,853.47$ cm$^{-1}$). This absorption can be indicated as a d-d transition, namely the $^2B_{1g} \rightarrow ^2A_{1g}$ transition, which has a smaller wavelength shift than the CuSO$_4$·5H$_2$O metal absorption (823 nm; $\varepsilon = 32$ L/mol·cm; $\nu = 12,150.66$ cm$^{-1}$). This shift indicates that SB has a greater ligand strength than H$_2$O. Cu(II) complex with the basic ligand Schiff salicylaldehyde derivative has an absorption peak of 622 nm with a molar absorptivity value ($\varepsilon$) 80 L/mol·cm which is indicated as a d-d transition. Square planar complexes of Cu(II)-sulfisoxazole and Cu(II)-sulfanilamide have one peak absorption at 793 nm, respectively, with low molar absorptivity. [Cu(SB)$_2$]SO$_4$·H$_2$O is proposed to form square planar geometry.

Magnetic Moment
[Cu(SB)$_2$]SO$_4$·H$_2$O has an effective magnetic moment value of 2.07 B.M. The complexes with the effective magnetic moment of 1.73-2.12 B.M. are paramagnetic with 1 unpaired electron. These results also indicate that there is no interaction or bonding between Cu metals. The effective magnetic moment value smaller than 1.73 will produce a complex with diamagnetic properties. Cu(II)-sulfisoxazole complex has an effective magnetic moment value of 2.12-215 B.M and square planar geometry. Therefore, [Cu(SB)$_2$]SO$_4$·H$_2$O is paramagnetic and estimated to have a square planar geometry. From the characterization above, the proposed structure of the complex is shown in Fig.-8.

![Fig.-7: FTIR Spectra of SB and Cu(II)-SB](image)

![Fig.-8: Purposed Structure of Cu(II)-SB Complex](image)
Antibacterial Test
The antibacterial activity test was carried out by using the disc diffusion method or disc paper diffusion. DMSO was used as a negative control because it did not provide an inhibitory zone and could dissolve the sample, while for positive control, several antibiotics were used. The results of the bacterial inhibition zone are shown in Table-3.

Table-3: Average Inhibitory Zone

| Compound | Concentration (µg/disk) | Control | DMSO | Antibiotic (+)* |
|----------|-------------------------|---------|------|-----------------|
|          | 15  | 30  | 45  | 60  | 75  | (-) | (+) |
| CuSO₄·5H₂O | -  | -   | -   | -   | -   | -   | 24.40 |
| SB       | 6.45 | 6.51 | 6.67 | 6.78 | 6.92 | -   | 24.40 |
| Cu(II)-SB | 6.95 | 7.4  | 7.56 | 7.70 | 8.25 | -   | 24.40 |

For *Staphylococcus aureus* bacteria, it was found that at the highest concentration of 75 µg/disk, the ligand inhibition zone was very low and tended not to inhibit bacterial growth. While at the same concentration, the complex inhibition zone was higher (8.25 mm). In the *Staphylococcus epidermidis* bacteria, the inhibition zone of bacteria on CuSO₄ metal was not visible, and only on the SB showed the inhibition zone at the highest concentration 75 µg/disk. Whereas in the complex, there was an inhibition zone, 7.12 mm, with quite low values at the highest concentrations. In *Escherichia coli*, SB's inhibition zone began to appear at a concentration of 45 µg/disk and continued to increase at the highest concentration. However, this ligand inhibition zone's results were still low compared to the complex inhibition zone, which had begun to appear to inhibit at a concentration of 15 µg/disk and continued to increase with increasing concentrations, 7.33 mm at a concentration of 75 µg/disk. In *Pseudomonas aeruginosa* bacteria, the inhibition zone on CuSO₄ metal and SB was not visible, while in the complex, the inhibition zone was seen at the lowest concentration to the highest concentration with the inhibition zone at a concentration of 75 µg/disk of 8.84 mm.

The results in Table-3 show that SB and CuSO₄·5H₂O have a low bacterial inhibition zone. Meanwhile, the complex showed a significant increase in antibacterial activity. This improvement may be due to copper complexing with C=N, which causes a decrease in polarity and an increase in copper ions' lipophilicity so that the Cu(II)-SB can easily enter the bacterial cell membrane. It can damage bacterial metabolism. Another thing that may also influence is the nature of Cu metal, which can kill bacterial cells at various concentrations due to the electron affinity for Cu metal ions.
CONCLUSION

SB was successfully synthesized by reacting o-anisaldehyde and 2-aminobenzothiazole in methanol at a 1: 1 mole ratio with glacial acetic acid as catalyst. The ligand analyzed with $^1$H-NMR showed the loss of chemical shift from proton amines and the appearance of the chemical shift of azomethine protons. Based on $^{13}$C-NMR data, the number of C atoms also corresponded to the expected ligand. The copper(II) complex with SB was successfully synthesized with the maximum wavelength in the UV-Vis spectrum of 778 nm. A possible complex formula was [Cu(SB)$_2$]$\text{SO}_4\cdot\text{H}_2\text{O}$. Cu(II)-SB complex was electrolyte, paramagnetic, and was estimated to have a square planar geometry via C=N azomethine and C=N thiazole groups of SB. Cu(II)-SB complex has slightly higher antibacterial activity than copper(II) and SB.

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