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

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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 $[Cu(SB)_2]SO_4 \cdot H_2O$. 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 μ_{eff} of 2.07 B.M, indicated that the complex was paramagnetic and was expected to have square planar geometry. $[Cu(SB)_2]SO_4 \cdot H_2O$ exhibited the highest antibacterial activity than the ligands and metals.

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

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INTRODUCTION

The search for new compounds with biological activity has been very much done, one of which is a complex compound. The complexes of the Schiff base ligands have received much attention due to their good biological activity for antibacterial, antifungal, and antioxidant.¹⁻³ The Schiff base is a compound that has an azomethine (C=N) bond formed by the condensation reaction of carbonyl compounds with primary amines under certain conditions and is known for its strong coordination ability as a ligand.^{4,5} Previous studies have shown that metal ions which are bound to Schiff base ligands are capable of increased biological activity.⁶ Complex synthesis with Schiff base ligand has attracted many researchers because chelate from N and O donor atoms can bind to metal ions and exhibit good antibacterial activity to develop drug compounds.⁷⁻⁸

Research conducted by Kolate *et al.* (2020) showed that metal complex compounds have higher antimicrobial activity than free metals and ligands.⁹ Aminobenzothiazole ligands and their derivatives have good biological activity as antimicrobials, anticancer, and antioxidants.¹⁰⁻¹¹ Aminobenzothiazole is a benzothiazole derivative with a well-known heterocyclic group with good biological properties. Besides, benzothiazole shows increased activity when it is complexed with metals.¹² Some aldehydes and ketones have good antibacterial properties, so the metal complex consists of Schiff bases derived from aldehydes, or ketones will form new compounds with higher biological activity.¹¹

Various transition metals are biologically active compounds used for antibacterial compounds such as Cu, Ni, and Co.¹³ Among transition metals, copper is a widely studied metal because it is the third most abundant metal element in the human body, after iron and zinc.¹⁴

The progression of diseases is increasing due to the evolution of bacteria, which are increasingly resistant to drugs. The level of bacterial resistance that occurs can increase the risk of spreading infection, difficulty controlling infection, and can cause death. This concern underlies the search for new

compounds that can deal with bacterial resistance.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 s analysis of SB was carried out using Infrared spectra, ¹H-NMR, and ¹³C-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 CuSO₄·5H₂O (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.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 R_f value of this base Schiff ligand was 0.76. The reaction scheme of SB formation is shown in Fig.-1.

Fig.-1: Reaction Scheme, Formation of Schiff Base Ligand (SB)

From the ^1H -NMR proton data (Fig.-2), a chemical shift at 8.69 ppm shows an azomethine proton.¹⁷ 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.¹³ ^{13}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 $\text{C}_{\text{azomethine}}$ at the 165.6 ppm.¹⁸

Fig.-2: ^1H -NMR Spectrum of Schiff Base Ligand (SB)

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} .¹⁹ 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

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 spectra²⁰ 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 ($\text{CuCl}_2 \cdot \text{H}_2\text{O}$) to 775 nm in the complex.²¹

Fig.-5: Electronic Spectra of the Metal and the Complex

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 $\text{Cu}(\text{SB})_2(\text{SO}_4)(\text{H}_2\text{O})_n$ ($n = 0, 1, \text{ or } 2$).

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 $^{\circ}\text{C}$ accompanied by an endothermic peak at 160 $^{\circ}\text{C}$. The mass

reduction that occurs in the Cu(II)-SB complex is 3.15% equivalent to the release of one H₂O molecule (calc. 2.5%). H₂O 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 H₂O molecules that coordinate with the complex will begin to decompose at temperatures of 150-220 °C.²² 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 H₂O molecule as crystal water at a temperature of 30-130 °C.²³ From the thermogram results and the calculation of the release of H₂O molecules, the possible complex formula is Cu(SB)₂SO₄·H₂O.

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	714.17	8.89	
Cu(SB) ₂ (SO ₄)(H ₂ O) ₂	732.19	8.67

Fig.- 6: TG/DTA Data of the Complex

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 CuSO₄·5H₂O solution with the ratio of cation: anion = 1:1, so the estimated complex formula formed is [Cu(SB)₂]SO₄·H₂O.

Table-2: Molar Conductivity of 1.10⁻³ M Standard Solutions and the Complex in Methanol

Compounds	Molar Conductance Λ_m^* (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]SO₄·H₂O 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 $C=N_{\text{azomethine}}$ and $C=N_{\text{thiazole}}$ 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_2O in the complex.

Fig.-7: FTIR Spectra of SB and Cu(II)-SB

Electronic Spectra

Complex $[Cu(SB)_2]SO_4 \cdot H_2O$ has one peak absorption at 778 nm with a relatively small value of molar absorptivity (ϵ) ($<1000\text{ L/mol}\cdot\text{cm}$) ($\epsilon = 48\text{ L/mol}\cdot\text{cm}$; $\nu = 12,853.47\text{ cm}^{-1}$). 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 $CuSO_4 \cdot 5H_2O$ metal absorption (823 nm; $\epsilon = 32\text{ L/mol}\cdot\text{cm}$; $\nu = 12,150.66\text{ cm}^{-1}$). This shift indicates that SB has a greater ligand strength than H_2O . Cu(II) complex with the basic ligand Schiff salicylaldehyde derivative has an absorption peak of 622 nm with a molar absorptivity value (ϵ) $80\text{ L/mol}\cdot\text{cm}$ which is indicated as a d-d transition.²⁴ Square planar complexes of Cu(II)-sulfisoxazole and Cu(II)-sulfanilamide have one peak absorption at 793 nm, respectively, with low molar absorptivity.²⁵ $[Cu(SB)_2]SO_4 \cdot H_2O$ is proposed to form square planar geometry.

Magnetic Moment

$[Cu(SB)_2]SO_4 \cdot H_2O$ 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 electron²⁶. 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.²⁷ Cu(II)-sulfisoxazole complex has an effective magnetic moment value of 2.12-2.15 B.M and square planar geometry.²⁵ Therefore, $[Cu(SB)_2]SO_4 \cdot H_2O$ is paramagnetic and estimated to have a square planar geometry.²⁸⁻²⁹ From the characterization above, the proposed structure of the complex is shown in Fig.-8.

Fig.-8: Purposed Structure of Cu(II)-SB Complex

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

<i>Staphylococcus aureus</i> Inhibition Zone Diameter (mm)							
Compound	Concentration (µg/disk)					Control	
	15	30	45	60	75	DMSO (-)	Antibiotic (+)*
CuSO ₄ ·5H ₂ O	-	7.32	7.41	7.60	7.80	-	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
<i>Staphylococcus epidermidis</i> Inhibition zone diameter (mm)							
CuSO ₄ ·5H ₂ O	-	-	-	-	-	-	24.40
SB	-	-	-	6.67	6.68	-	24.40
Cu(II)-SB	6.36	6.47	6.72	6.94	7.12	-	24.40
<i>Escherichia coli</i> Inhibition zone diameter (mm)							
CuSO ₄ ·5H ₂ O	6.12	6.45	6.58	6.83	6.87	-	28.25
SB	-	6.46	6.57	6.64	6.73	-	28.25
Cu(II)-SB	6.82	6.92	7.12	7.23	7.33	-	28.25
<i>Pseudomonas aeruginosa</i> Inhibition zone diameter (mm)							
CuSO ₄ ·5H ₂ O	-	-	-	-	-	-	19.93
SB	-	-	-	-	-	-	19.93
Cu(II)-SB	8.18	8.35	8.63	8.64	8.84	-	19.93

* Antibiotics for *St. aureus*: vancomycin; *St. epidermidis*: vancomycin; *P. aeruginosa*: gentamycin; *E. coli*: chloramphenicol;

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.³⁰ 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.³¹

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 ^1H -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 $[\text{Cu}(\text{SB})_2]\text{SO}_4 \cdot \text{H}_2\text{O}$. 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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