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Guanine Metal Complexes: Spectroscopic Studies, Dying Performance and as Indicator

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Abstract

Azo dyes are the most common and widely used dyes, accounting for more than half of each year's dyes. In this work, a complete description of a new innovative series of compounds with the elements [Ag (I), Zn (II)] generated from the guanine azo dye ligand (GAB) 8-[1-(3-carboxy) azo] guanine has been studied. The structural formula was studied using several physicochemical analyses and spectroscopic techniques (FT-IR spectra, UV-Vis). The FTIR spectrum of the ligand (GAB) was compared to the spectra of the metal ion complexes formed to determine its identity. Chelating caused some changes in the spectra of the complexes to appear to demonstrate that they could be linked to the ligand. The complexes have a tetrahedral geometry shape, the ligand functions as a bidentate ligand, and thermogravimetric analysis (TGA) is used to measure the thermal stability of compounds. The findings and equation presented by the analytical data seemed to be in good accord with the conclusions of the thermogravimetric investigation, which demonstrated that the disintegration of the ligand (GAB) and its complexes occur in multiple steps. The configuration that follows weakens thermal stability:

GAB(35.52%)>[Ag(GAB)(H_2O)$_2$]NO_3.2H_2O(34.6%)>[Zn(GAB)Cl_2].H_2O(31.54%), and the complexes have tetrahedral geometry shape. Furthermore, elemental analysis, mole ratio, and the mole ratio of each complex (1:1) (M:L). The ligand was effective as an acid-base indicator when the pH changed; they exhibited a striking color change, similar to how the ligand (GAB) and its complexes can be used to dye wool textiles due to their wide range of colors. It investigated how well the ligand (GAB) and its complex worked as a wool dye. The ligand GAB and its metal complexes were used to color most of the protein filaments in wool fiber, which have a complex structure with amino and carboxyl groups and colors ranging from orange to green.

Keywords: Azo dyes, Acid-base indicators, Dying performance, Guanine, Thermogravimetric analysis.

1. Introduction

Azo dyes account for most of the dye chemistry production volume, and their importance may grow. They are critical for the management of the dye and printing markets. These dyes are made using a simple diazotization and coupling technique. Many paths and changes are taken to achieve the dye's desired color characteristics, yield, and particle size for better dispersibility [1, 20].

Indicators include dyes or pigments that can be extracted from plants, fungi, and algae. Anthocyanins, also known as flavonoids, are a form of organic pigment that varies in color depending on the pH. It could be found in almost every red, blue, or violet flower. Anthocyanidins (anthocyanins) are essential plant pigments that cause the red, violet, and blue colors found in plant blossoms [2].

Previous research has revealed that commercially used markers come from natural sources or chemical synthesis. Acid-base indicators are structurally divided into three groups: phthaleins (e.g., phenolphthalein), sulphonaphthaleins (e.g., phenol red), and azo compounds (e.g., methyl orange) [2]. As their chemical forms change due to changes in their chemical environment, these compounds change color. Azo dyes are the most common synthetic colorants, and they are widely employed in the textile, printing, and paper industries [3,27,28].

Different colors can be produced by altering the functional groups integrated into the azo molecule. The main aim of this work is to prepare the [Ag (I), Zn (II)] complexes derived from the ligand (GAB) and identify them with some physicochemical methods.

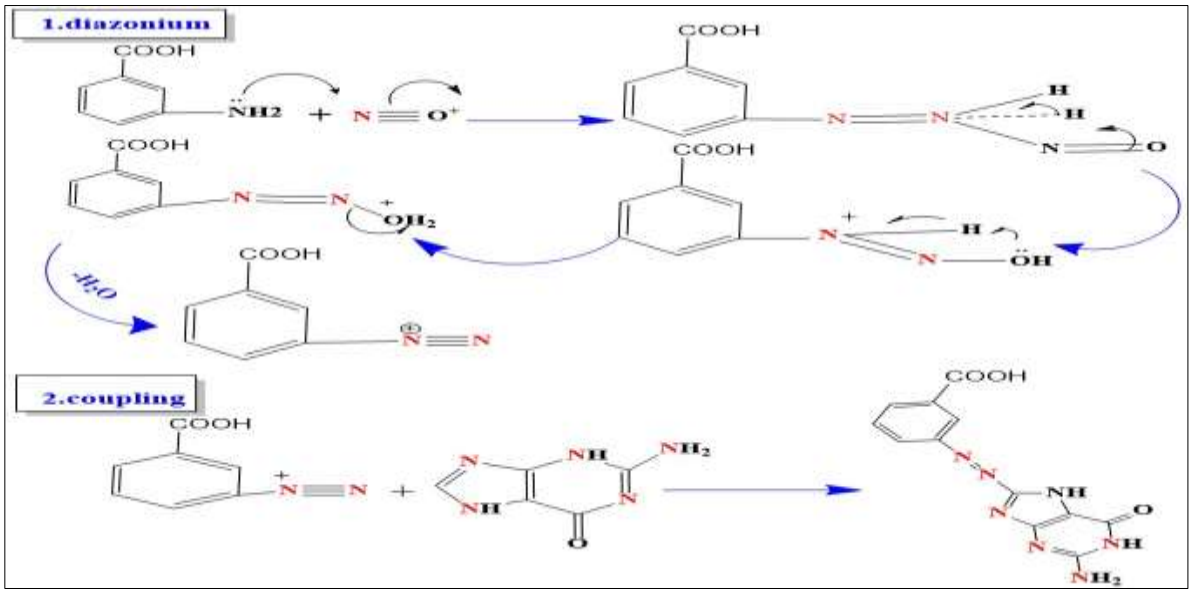
2. Materials and Methods

Solvents, materials, and tools were employed in each instance (C.H.N.) to determine the elemental analysis of the ligand (G.A.B.) and its complexes using an Eure EA 3000 Elemental Analyzer. When testing the pH of a sample, (HANNA equipment) was used. FT-IR spectra were acquired by using C.S.I. and a SHIMADZU 8400 spectrophotometer. The UV-Vis spectra of all the chemicals under investigation were examined using the (SHIMADZU 1800 UV-Vis spectrophotometer). Thermal gravimetric analysis (TGA) (S.D.T., Q600 V20.9 Build) was used to determine how much metal was in the synthesized ligands and complexes. Each chemical's melting points were determined using Gallenkamp's melting point equipment. The molar conductance of metal ion complexes was measured in deionized pure water at (10-3 M). The Mohr method was utilized to determine the chloride content in complexes.

2.1 Synthesis of the ligand 8-[1-(3-carboxy) azo] guanine (GAB)

A modified version of the procedure published in the literature [4, 21] was used to manufacture the ligand [GAB] (the diazonium reaction and coupling). The ligand [GAB] was synthesized by dissolving (0.01 mole; 1.371 g) m-aminobenzoic acid in (30 mL) ethyl alcohol, followed by a 4ml addition of concentrated acetic acid at (0-5°C). A cooled aqueous sodium nitrite solution (0.83 g in 25 mL water) was added to this solution with continuous stirring and held at approximately (0-5°C). The diazonium salt solution was slowly stirred into a cold solution (0.01 mole; 1.94 g) of guanine dissolved in [14 mL, 10% ethanolic NaOH solution]. The resulting liquid was neutralized to a pH of 5-6 by adding acetic acid or sodium hydroxide. The orange precipitate was filtered and

washed with a [distilled water: ethanol] [1:1] mixture before being gathered and desiccated [22] (**Scheme 1**).



Scheme 1. Synthesis of the ligand (GAB)

2.2 Synthesis of metal complexes

The synthesis of complexes was done in a mole ratio of [M:L] [1:1] by dissolving (1 mmol; 0.299 g) of the ligand GAB in a minimum quantity of deionized distilled water. The ligand solution was added gradually, with stirring, to the selected aqueous solution of metal salts [1 mmoL from each of $AgNO_3$ 0.1698 g or $ZnCl_2$ 0.1362 g], which were dissolved in the deionized distilled water. The mixture was refluxed for 3 hours, and the reaction was monitored using the TLC technique with the mixed solvent [0.8 mL methanol, 1.2 mL ammonia, and 0.4 mL butanol]. After filtering the colored solid, the residue is washed with a mixture of deionized distilled water and ethanol (1:1), then filtered and dried. The physicochemical properties of the ligand (GAB) and its complexes are tabulated in **Table 1** [5].

Table 1. Physical features and elemental properties

No.	Comp. (M.wt) (g/mol)	Color λmax (nm)	M:L	Λm ($S.mol^{-1}.cm^2$)	% Experimental % (Theoretical)				
					C	H	N	M	Cl
1	GAB(C_{12} H $_9$N $_7O_3$)	Yellow	---	---	48.142	3.007	32.086	---	---
	299.27	435.00			48.117	3.955	32.743		
2	[Ag(GAB)(H_2O)$_2$]NO_3.$2H_2O$	Yellowish orange	1:1	82.1	29.871	2.918	21.299	20.905	---
	516	476.00			30.232	2.519	21.705	20.502	
3	[Zn(GAB) Cl_2].H_2O	Reddish			31.729	2.285	21.602	14.412	3.751
	453.65	orange	1:1	25.7	31.472	2.424	21.607	14.951	3.022
		492.00							

3. Results and Discussion

3.1 Mole ratio

The most widely used technique for determining a complex's composition in solution is the mole ratio approach, which was applied in this case. **Figure 1** displays this process, the locating outcomes it produced, and **Table 2** presents the data. The results show that all the complexes built have a [1:1] ratio [M: L] mole ratio [6].

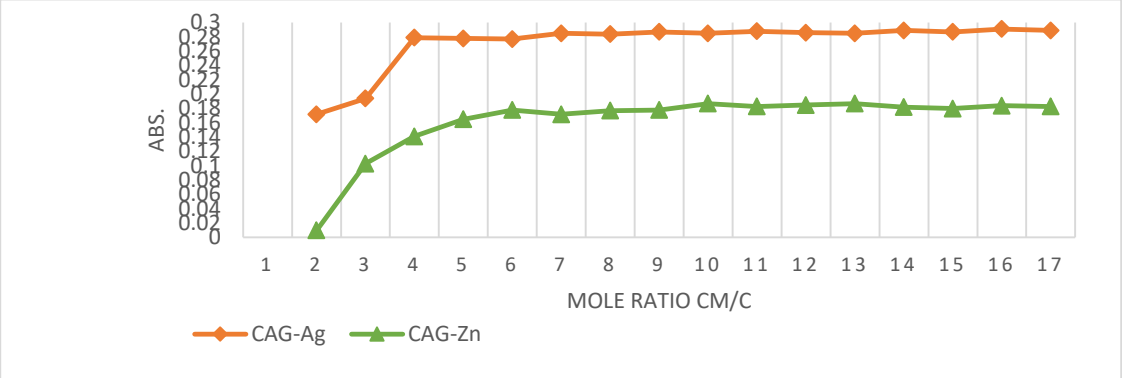


Figure 1. The mole ratio plot of complexes'

Table 2. Absorbance vs mole ratios for GAB-metal ion in solution

M:L	Absorbance (λmax nm)	
	GAB-Ag	GAB-Zn
	476	492
1:0.25	0.172	0.01
1:0.50	0.194	0.103
1:0.75	0.279	0.141
1:1	0.278	0.165
1:1.25	0.277	0.178
1:1.50	0.285	0.172
1:1.75	0.284	0.177
1:2	0.287	0.178
1:2.25	0.285	0.187
1:2.50	0.288	0.183
1:2.75	0.286	0.185
1:3	0.285	0.187
1:3.25	0.289	0.182
1:3.50	0.287	0.18
1:3.75	0.291	0.184
1:4	0.289	0.183

3.2 Thermogravimetric analysis (TGA)

The thermal behavior of the ligand (GAB) and its complexes during the ligand GAB degradation happens in four exothermic steps in the 50–800 °C range. The first stage of disintegration is between 25 and 50°C, resulting in a 2.11% weight loss; the second stage occurs between 50 and 290 °C, resulting in a 26.79% weight loss; and the third stage occurs between 290 and 600°C, resulting in a 25.61 weight loss. Between 600 and 800°C, the final stage of disintegration begins (9.356% weight loss). The suggestion is in **Figure 2A.** In the instance of $[Ag(GAB)(H_2O)_2]NO_3.2H_2O$, there are five steps, and the complex degrades **Figure 2B**. The first stage of

decomposition begins at 25 to 80°C, resulting in a weight loss of 2.86 kg of decomposed water [18]. The second breakdown stage was seen at 80–400°C (11.78% weight loss). Decomposition begins in the third stage at 400–500°C (12.80% weight loss). The fourth stage begins with a temperature range of 500–675°C (27.39% weight reduction). The last was at 675-800°C (a weight loss of 10.04%) [7]. Concerning [Zn(GAB)Cl_2]. The H_2O complex is divided into five steps **Figure 2 d**. The initial stage of decomposition begins at 25 to 55°C, resulting in a loss of 1.434 pounds. Between 55 and 84°C, the second stage of breakdown (17.63% wt loss) was observed. The third stage of decomposition begins between 84 and 560 (18.84% weight loss). Starting at 560–698°C, the fourth stage is 16.73% of its weight. The last stage is 698–800°C (13.83% weight loss) [8,9]. The findings and equation presented by the analytical data seemed to be in good accord with the findings of the thermogravimetric investigation, which demonstrated that the disintegration of the ligand (GAB) and its complexes occurs in multiple steps. The configuration that follows weakens thermal stability:

GAB(35.52%)>[Ag(GAB)(H_2O)$_2$]NO_3.2H_2O(34.6%)>[Zn(GAB)Cl_2].H_2O(31.54%)

Table 3. The TGA of ligand (GAB) and their complexes

Comp. & Molecular formula(molecular weight) g/mole	Step	TG. Range of the decomposition (^{0}C)	Suggested Assignment	% Mass loss	
				Cal. %	Found %
GAB $C_{12}H_9N_7O_3$	1	(25-50)	$H_3C_{0.25}$	2.00	2.117
	2	(50-290)	$H_2C_{6.5}$	26.73	26.79
	3	(290-600)	$H_4C_{5.25}N_{0.6}$	25.19	25.619
	4	(600-800)	N_2	9.356	9.967
	Residue	>800	$N_{4.4}O_3$	36.62	35.52
[Ag(GAB)(H_2O)$_2$]NO_3.2H_2O	1	25-80	H_2O	3.488	2.867
$AgC_{13}H_{13}N_8O_8$	2	80-400	$H_2O.C_3H_6$	11.62	11.78
	3	400-500	C_5H_3	12.2	12.80
	4	500-675	C_5N_5	27.90	27.39
	5	675-800	$N_2O_{1.5}$	10.07	10.04
	Residue	>800	$AgO_{4.5}$	34.6	35.09
[Zn(GAB) Cl_2].H_2O $ZnC_{12}H_{11}N_7O_4Cl_2$	1	25-55	$H_2O_{0.25}$	1.32	1.434
	2	55-84	Cl_2C	18.29	17.63
	3	84-560	C_7H_3	19.17	18.84
	4	560-698	$C_4O_{1.50}H_6$	17.19	16.73
	5	698-800	N_2O_2	13.22	13.83
	Residue	>800	$ZnN_5O_{0.75}$	32.40	31.54

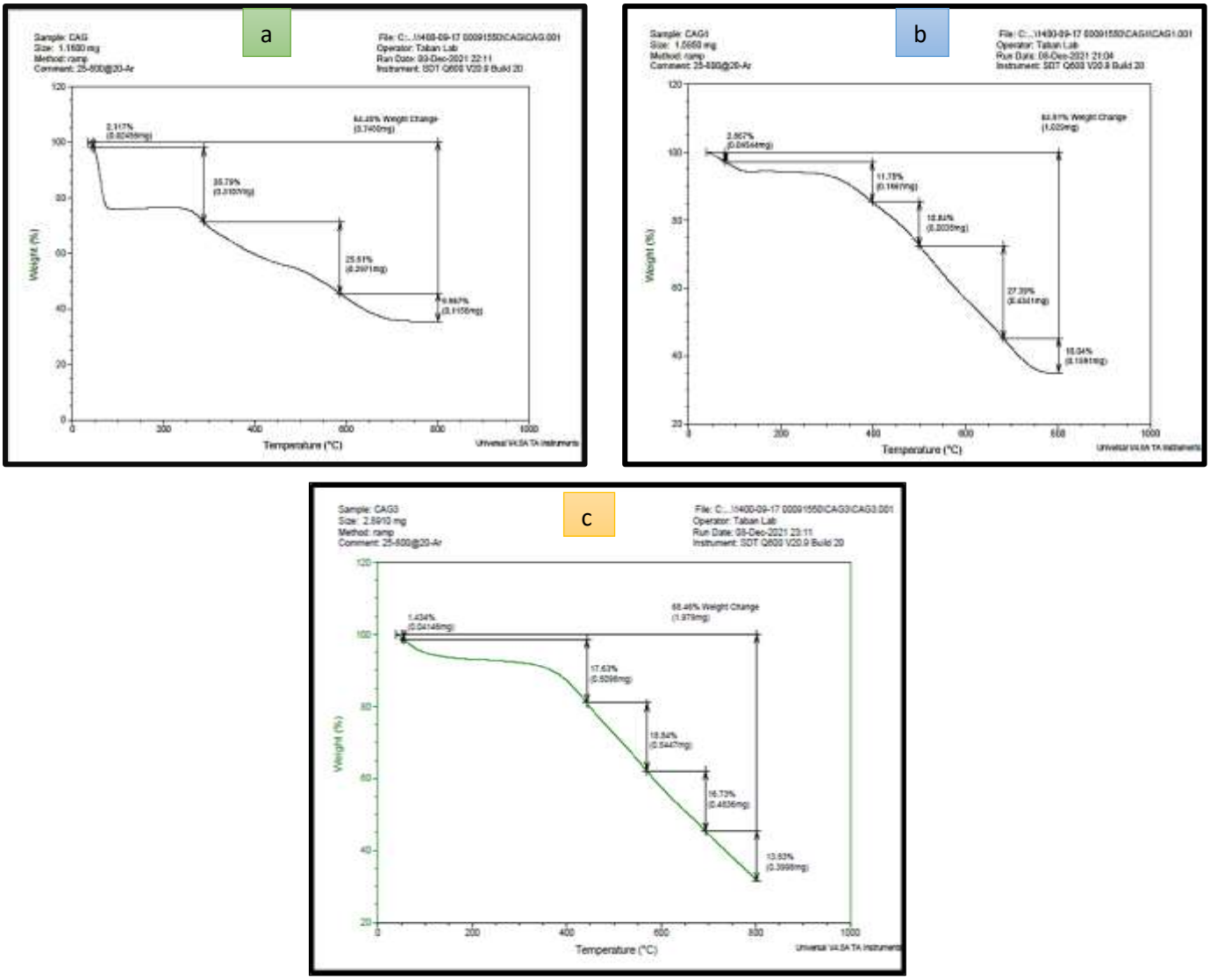


Figure 2. Thermogram for the (a) GAB ligand (b) $[Ag(GAB)(H_2O)_2]NO_3.2H_2O$ (c) $[Zn(GAB)Cl_2].H_2O$ complexes

3.3 The FTIR spectra for GAB ligand and its complexes

The FTIR spectra analysis can be utilized to determine how the ligand (GAB) will connect the metal ions to the formed complexes. The FTIR spectrum of the ligand (GAB) was compared to the spectra of the metal ion complexes formed to determine its identity. Chelating caused some changes in the spectra of the complexes to appear to demonstrate they could be linked to the ligand [29,30]. **Table 4** summarizes the principal bands for the ligand (GAB) and its metal ion complexes, whereas **Figure 3** shows the most moieties vibration was carried out in the range 400–4000 cm^{-1} using CSI disk [10, 11, 17].

Table 4. The FT-IR spectrum for the free ligand (GAB) and its complexes

Com.	υ (OH)	υ (NH_2)	υ $(C=O)_{py}$	υ $(C=N)_{imd}$	υ N=N	υ (-CN=N-C-)	υ $(M-N)_{imd}$	υ (M-N) azo	υ (M-Cl)	υ (MO) H_2O
GAB	3344 b	[3174 3112] d	[1691 1668] d	1573 Sh.	[1473 1419 1373] t	1259 W	---	---	---	---
$[Ag(GAB)(H_2O)_2]$ $NO_3.2H_2O$	3342 m	[3166 3114] d	[1693 1672] d	1560 w	[1415 1375]d	1261 W	603 w	503 w	---	459 vw
$[Zn(GAB)\ Cl_2].H_2O$	3342 m	[3172 3114] d	[1693 1672] d sh.	1564 w	[1415 1375] d	1263 w	605 w	503 vw	418 vw	---

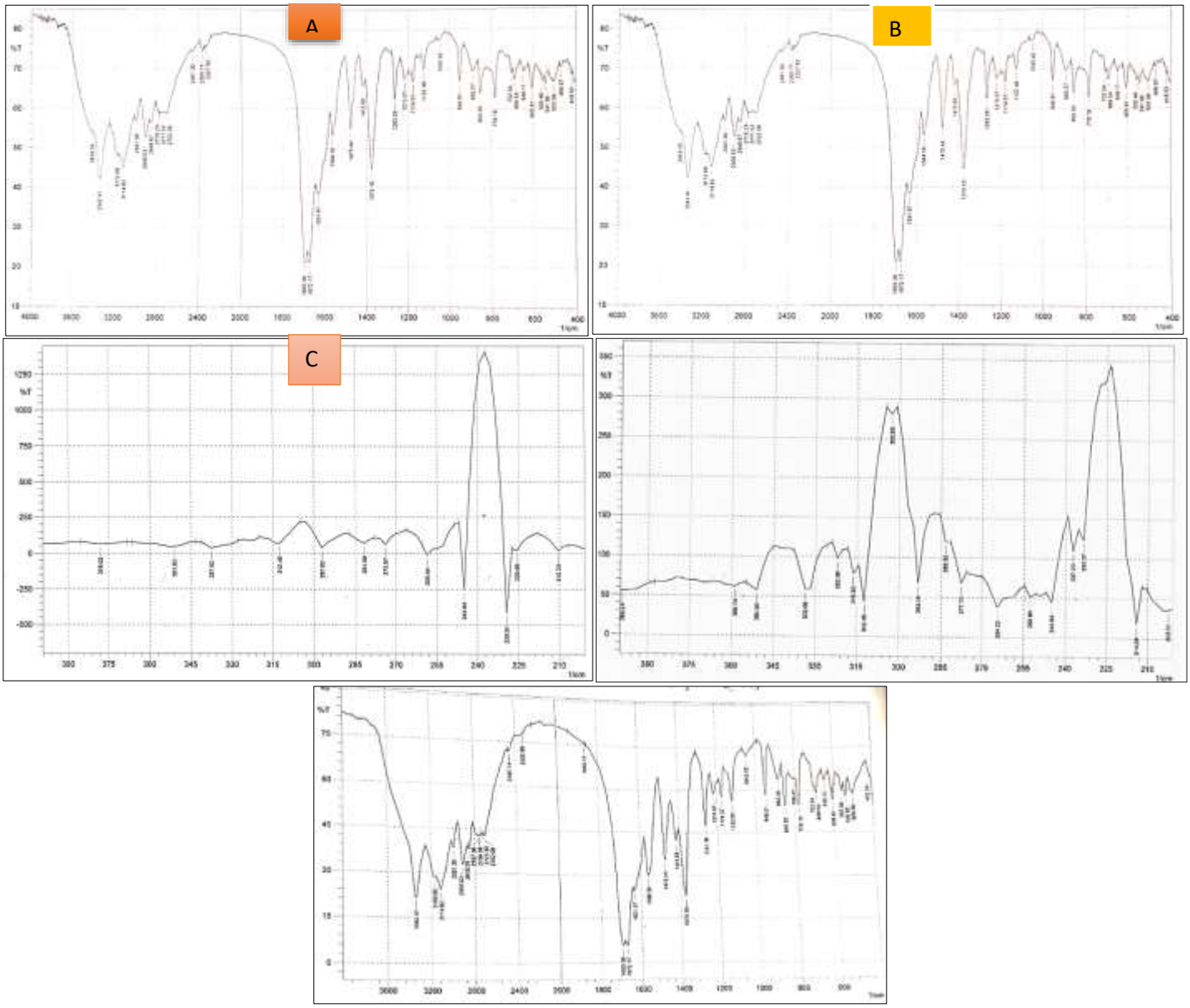


Figure 3. The FTIR assignment for the (A) GAB ligand (B) [Ag (GAB) $(H_2O)_2]NO_3.2H_2O$ (C)$[Zn(GAB)Cl_2].H_2O$ complexes

3.4 Electronic spectrum of the (GAB) ligand and its complexes

The electronic spectra of the ligand (GAB) and its complexes were dissolved in water [10^{-4} M] with the range (280-1100) nm. The data of the ligand and its complexes are shown in **Table 5** and **Figure 5** [12,13,19].

Table 5. The data of spectrum of UV-Vis for (GAB) ligand and their complexes

Compound	ƛ(nm)	Wavenumber (cm^{-1})	Assignment	Hybridization	Geometry
GAB	291	34364	$\pi \rightarrow \pi *$	---	---
	409	24449	$n \rightarrow \pi^*$		
$[Ag(GAB)(H_2O)_2]NO_3.2H_2O$	476	21008	CT	Sp^3	Tetrahedral
$[Zn(GAB)\ Cl_2].H_2O$	492	2032	CT	Sp^3	Tetrahedral

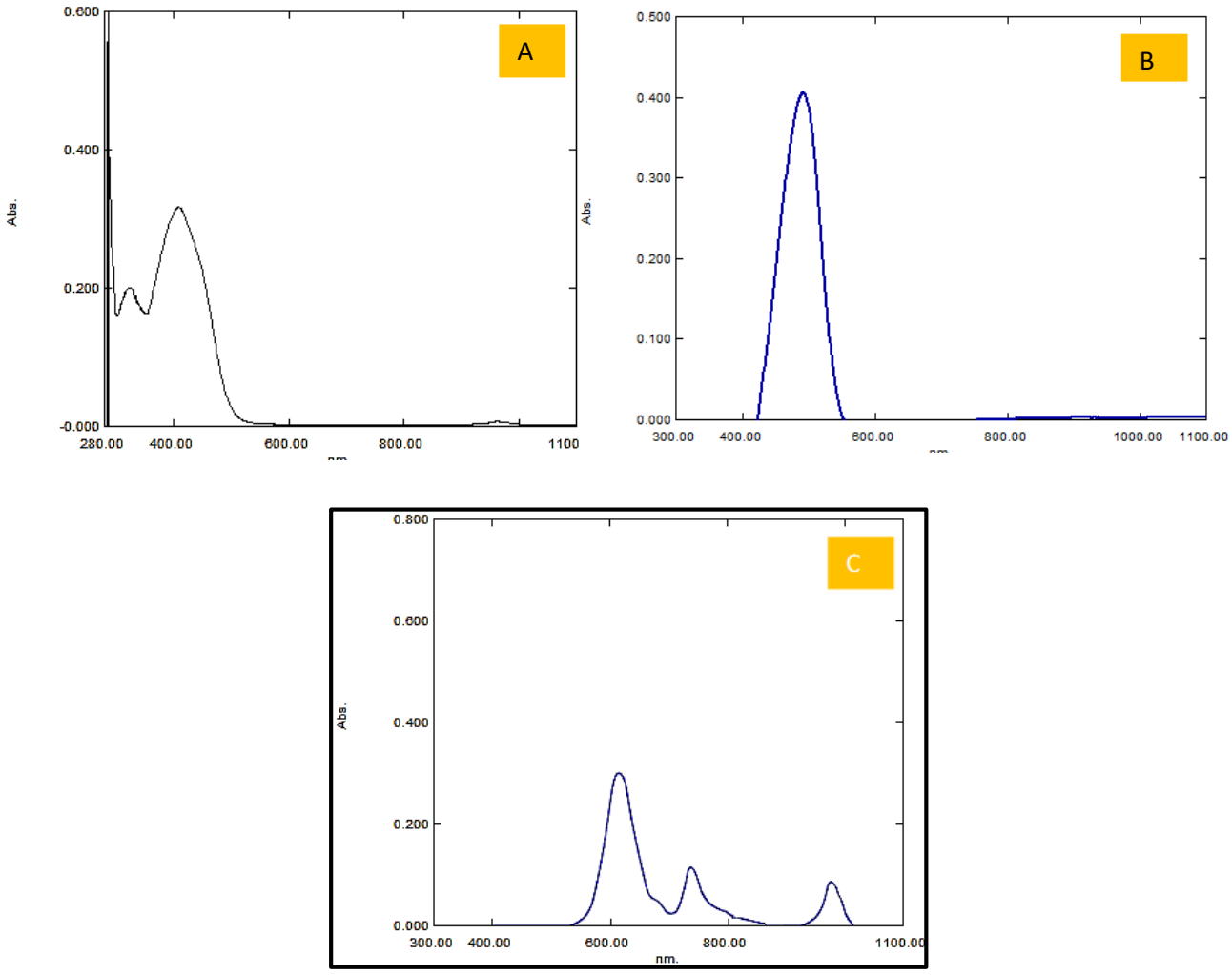


Figure 4. The electronic spectra for (A) GAB ligand (B) $[Ag(GAB)(H_2O)_2]NO_3.2H_2O$ (C) $[Zn(GAB)Cl_2].H_2O$

3.5 Dying performance

The effectiveness of the ligand (GAB) and its complex as a wool dye were examined. Most of the protein filaments in wool fiber, keratin, have a complex structure with amino and carboxyl groups [14]. Keratin is also the majority of the protein filaments in wool fiber. Early theories of wool dyeing were developed on the wool's ability to absorb acids. Interactions between the negatively charged dye anions and the positively charged amino groups on the fibers could explain the entire process of dying wool in an acidic environment. Polar groups like -NH, -SH, and -OH are among

the numerous polar groups discovered in wool fiber. In polyamide fiber, a terminal amino group and a sizable number of NH groups are conceivable. A specific pH level is required for these functional groups to interact covalently with reactive dyes such as vinyl sulphone and chlorotriazine. It is important to emphasize that this is happening in an acidic environment. Numerous variables, such as the type of metal and its valence state, the concentration of the solution, pH, time, temperature, and others, affect the amount and rate of absorption. The free carboxyl groups of acidic amino acids are the most likely binding sites because they can offer negatively charged binding sites over a wide pH range. Particularly at alkaline pH values, the nitrogen atoms of amin and amide groups can form coordination linkages [15]. The wool fabric in **Figure 5**, which has colors ranging from orange to green, was colored using the ligand GAB and its metal complexes [25,26].

Table 6. Data on coloring and different fastness characteristics of GAB ligands and complexes on wool textile

Compounds	Color	Color fastness	Staining Dye	Notes
GAB	Yellow	4	4	Acceptable on grayscale
$[Ag(GAB)(H2O)_2]NO_3.2H_2O$	Yellowish orange	3	3	Acceptable on grayscale
$[Zn(GAB)\ Cl_2].H_2O$	Reddish orange	4	4	Acceptable on grayscale

A **B** **C**

Figure 5: The dying of ligand (GAB) and some of their complexes A=GAB ligand ,**B**=$[Ag(GAB)(H2O)_2]NO_3.2H_2O$,**C**=$[Zn(GAB)\ Cl_2].H_2O$.

3.6 Azo dyes as acid-base indicators

Organic dyes with distinct colors in solutions with different pHs are known as acid-base indicators. They are frequently used in acid-base titrations to establish the equivalency point. When the pH changes, they exhibit a striking color change. Due to their capacity to alter color in response to pH, azo dyes are the most widely used chemical molecules as acid-base indicators [16,23,24]. **Table 7** contains the data from acid-base titrations, which is used to assess the indicator properties of azo dyes. **Figure 6** displays the azo dye solutions in both acidic and basic environments.

Table 7. Titration of acid (0.1M) against NaOH (0.1M) for (GAB)

NO.	Volume of acid	Volume of NaOH	volume of $NaHCO_3$ (mL)	Color in acid	Color in base
1	(HCl) 5 mL	5.3 mL	---	Dark yellow	Light yellow
2	(ACOH) 5 mL	6 mL	---	Yellow	Color less
Titration of acid (0.1M) against NaHCO3 (0.1M)for (GAB)					
3	(HCl) 5 mL	---	7.3 mL	Yellow	Color less
4	(ACOH) 5 mL	---	6 mL	Light yellow	Yellow

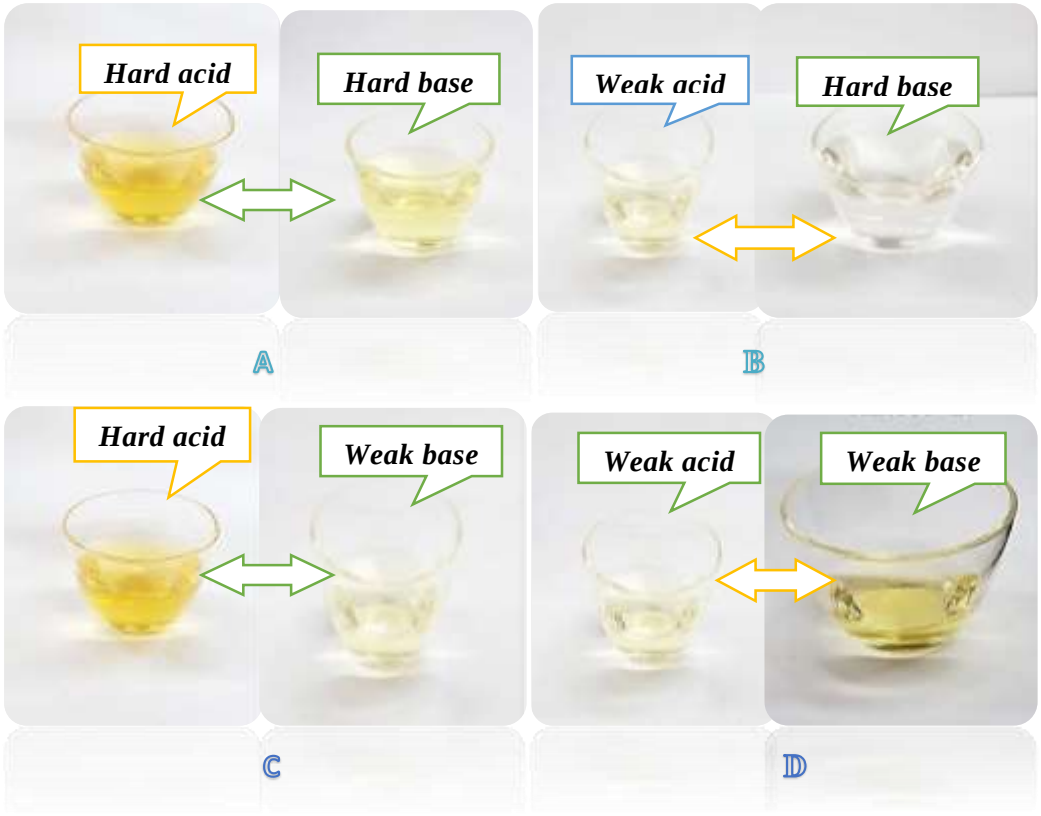


Figure 7. Change in color of Azo ligand (GAB): A= HCl with NaOH, B= ACOH with NaOH, C=HCl with $NaHCO_3$, D= ACOH with $NaHCO_3$

4. Conclusion

The Ag (I) and Zn (II) complexes were from the azo ligand 8-[1-(3-carboxy)azo]. The mole ratio at which guanine (GAB) is produced is [M:L][1:1]—and studied using various spectroscopic methods. The complexes have tetrahedral geometry; the ligand functions as a bidentate ligand. They can be used as a suitable indicator for acid-base titrimetric assays in solid acid/strong base and weak acid/strong base titrimetric analyses. The ligand and its complexes have a variety of colors that have been confirmed to be used for dying wool fabrics, and the result of the ultraviolet protection factor shows a good UV-absorbing ability. The primary goal is to assist with various analytical tasks in research and teaching labs.

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Conflict of Interest

The authors declare that they do not have any competing interests.

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Ethical Clearance

This work has been approved by the Scientific Committee at the University of Baghdad/ College of Science.

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