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RESEARCH PAPER

pH Responsive and Nanoparticle Properties of Novel Azo Dye of Naphthol and Its Silver Complex

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ABSTRACT

Present work includes the synthesis of azo dye namely (E)-4-((4-isopropylphenyl)diazenyl)naphthalen-1-ol, derived from 1-naphthol and 4-isopropylaniline and its Nano silver(I) complex. The synthesized nano compounds were isolated in a high yield 95 and 80% and observed red and dark red color for azo dye ligand and its silver complex respectively. The synthesized compounds were characterized by physical and analytical methods including XRD, elemental analysis, mass, UV-Vis, H-NMR and FTIR spectroscopies. The azo dye showed significant changes in colors and UV-Vis which serves as pH indicator. The ligand displayed a notable color transition from yellow (acidic) to orange-red (neutral) to dark red (basic), reflecting enhanced conjugation and resonance extension under alkaline conditions. ^{1}H-NMR results supported the successful synthesis of the ligand and its silver complex, showing characteristic aromatic proton signals and confirming structural integrity. The silver complex exhibited slight chemical shifts in the proton environment, attributed to coordination with the silver ion. Collectively, the findings demonstrate the potential application of the synthesized azo dye as a pH-sensitive compound with distinctive optical properties. The synthesized compounds showed potential properties as crystalline nanomaterial.

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INTRODUCTION

Aromatic azo compounds azo have received large interesting in both basic and applied research fields which have been widely used in many practical applications such as colouring fibers, photoelectronics, printing systems, optical storage technology textile dyes as well as in many biological reactions and in analytical chemistry [1–3].

Plenty of azo dyes have been reported due to the growing interest on the coordination compounds of silver with various N-donor and O-donor ligands [4,5]. That due to their ability in the regions of chemistry and biology including antiviral, antitumor, bactericidal, fungicidal and nonlinear optical properties as well as their properties such as appearing characteristic structural flexibility, mimicking of protein active sites, ease of preparation, and stabilization of both oxidation states of the metal usual in biological systems [6–9].

Methyl red and methyl orange are azo dyes

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which act as indicators in the acid–base and complexometric titrations of analytical chemistry via changing the color due to the extent of electron delocalization [10–12]. Azo dyes possessing active functionals and the azo group lead to high conjugation and unique mechanism of changing colors in titration [13,14]. Therefore, we are interested in preparation new azo dye for 1-naphthol as pH-indicator and its silver complex.

MATERIALS AND METHODS

The 1 H-NMR spectra of synthesis compounds were recorded on a Bruker DMX-500 NMR spectrophotometer at a frequency of 400 MHz, using DMSO d6 as the solvent. Elemental analysis for C, N and H was performed on a PE 240C elemental analyser. Mass spectrometry was conducted with an Agilent instrument from the USA. The IR spectra using (KBr pellets) for synthesis compounds were recorded on a Broker FTIR spectrometer in the range of 4000-400 cm^{-1}. The UV-Vis spectra were recorded on a Shimadzu spectrometer in the range of 200-1000 nm. X-ray diffraction (XRD) data were carried out on Phillips, Holland PW 1710 X-ray diffractometer system, using a copper anode. The data obtained from this XRD were in the form of a chart of 2θ vs. intensity. The crystallite size (G) was calculated by using Scherrer equation.

The used materials were supplied commercially and used without further purification, 4-isopropylaniline and 1-naphthol (Sigma Aldrich), hydrochloric acid, sodium hydroxide and silver (I) nitrate were supplied from Merck. Solvents were purchased from Scharlau Company.

Preparation of azo dye ligand

The azo dye (IPDN) was produced via coupling of diazonium salt of 4-isopropylaniline with 1-naphthol in alkali medium as in Fig. 1. The diazonium salt was prepared by mixing 4-isopropylaniline (2.0 mmol, 0.27 g) dissolved in 15 mL water with 2 mL of hydrochloric acid under cooling then this solution was mixed with 5 mL of sodium nitrite (2.0 mmol, 0.14 g) dissolved in water under cooling and stirring for 30 minutes. The diazonium salt was reacted with 1-naphthol (2.0 mmol, 0.28 g) dissolving in 15 mL of ethanol in

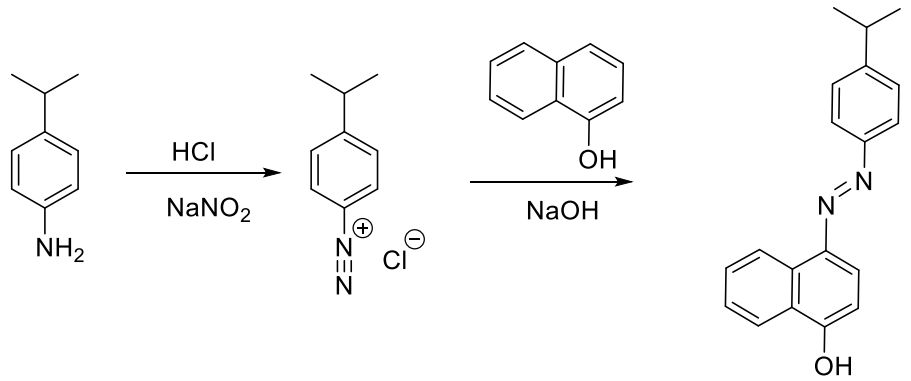

Fig. 1. Preparation steps of silver complex for IPDN.

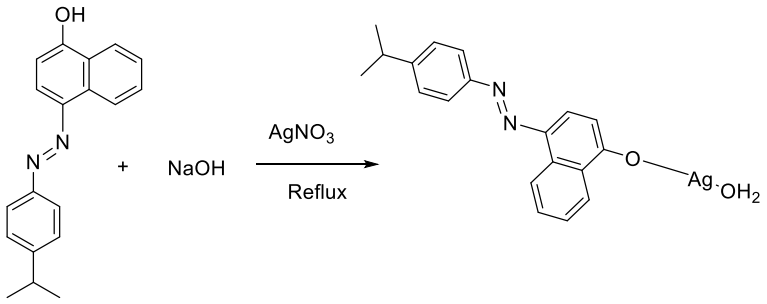

Fig. 2. Preparation steps of silver complex for IPDN.

the presence of 10 mL of sodium hydroxide (10%). The mixture solution was left under cooling and stirring for one hour. During the reaction, the color of the diazonium salt solution was changed from the red to the formed brown precipitate under reacting with 1-naphthol. This formed precipitate of azo dye was filtered of, washed several times with water then with cool ethanol, and afterward dried under a vacuum with yield equal to 95%.

Synthesis of the silver (I) azo dye complex

The azo dye complex of silver was synthesized via refluxing silver nitrate (2.0 mmol, 0.34 g) dissolved in 10 mL distilled water (DW) with 2.0 mmol, 0.58 g of the ligand dissolving in 20 mL of ethanol with 2.0 mmol, 0.08 g of sodium hydroxide for 2 hours at 75 °C. Aferwards the system was refluxed. During the reaction, the color of the solution was changed from reddish-brown to a

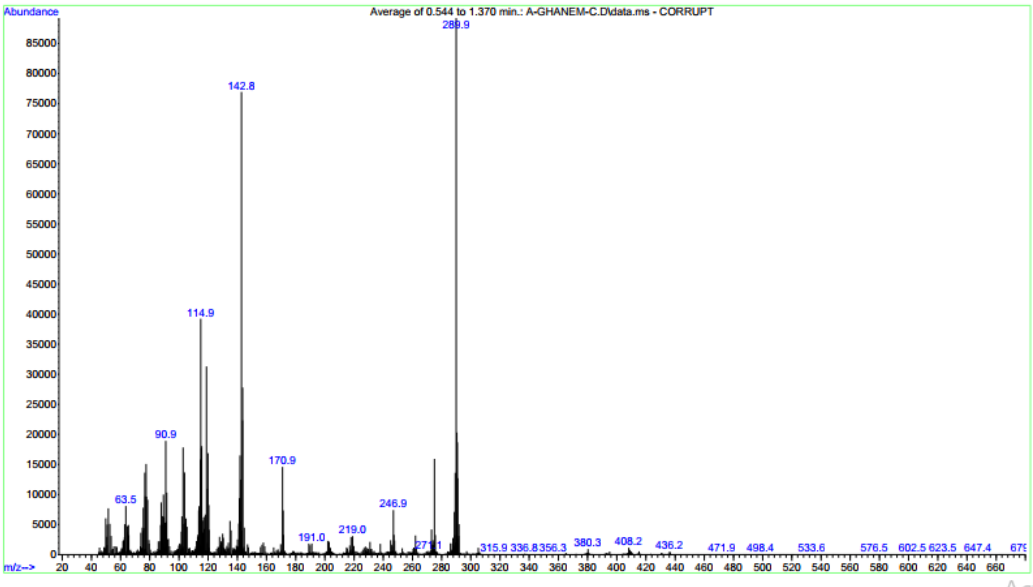

Fig. 3. Mass spectrum of IPDN ligand.

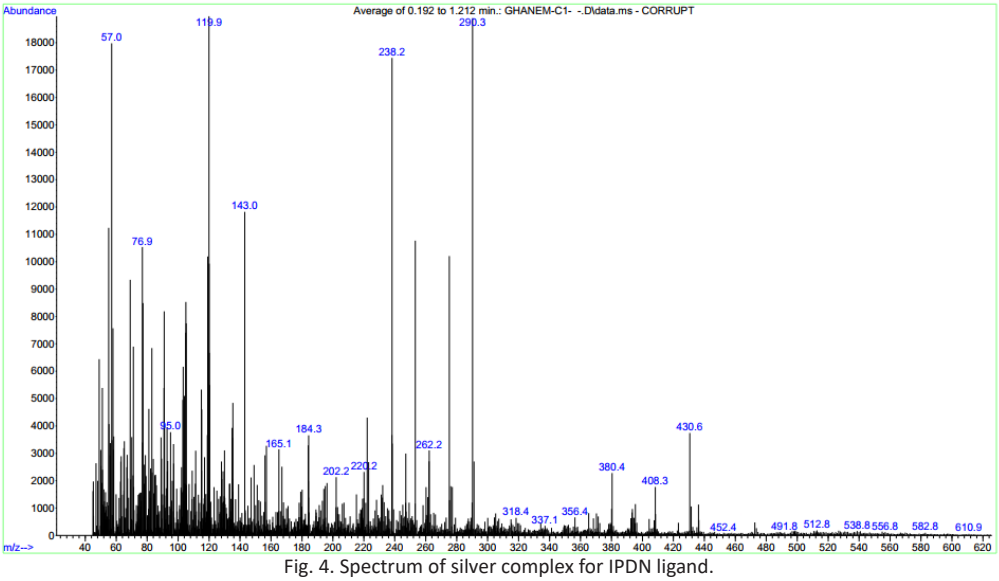

Fig. 4. Spectrum of silver complex for IPDN ligand.

dark brown precipitate. The precipitate was fltered of, washed several times with water/ ethanol, and finally dried under a vacuum with yield equal to 80%. The preparation steps of silver complex is depicted in Fig. 2.

RESULTS AND DISCUSSION

The results of elemental analyses of IPDN ligand, and silver complex are in agreement with that calculated for the suggested formula. Anal. Calc. (%): for ligand $C_{19}H_{18}N_2O$; (290.3 g/mol); C, 78.59; H, 6.25; N, 9.65; found C, 78.52; H, 6.21; N, 9.67. Whereas, Anal. Calc. (%): for $[Ag(C_{19}H_{17}N_2O)(H_2O)].H_2O$ complex; (433.26 g/mol); C, 52.67; H, 4.89; N, 6.47.; found C, 52.54; H, 4.75; N, 6.48. On the other hand, the expect mass results of mass spectra of IPDN ligand ($C_{19}H_{18}N_2O$) is m/z 290.37 in agreement with mother peak of experimental which is m/z 289.9. The mass of complex is m/z 430.6 in agreement with mother peak of expect molecular formula [M-2H] which is m/z 430.04.

UV-Visible absorption spectroscopy

UV-Vis spectra of IPDN ligand and its silver complex were recorded in DMSO solvent at room temperature as shown in Fig. 5. The IPDN ligand showed bands at 292 nm due to $\pi \rightarrow \pi^*$ and other band at 413 nm due to $n \rightarrow \pi^*$ [15–17]. The silver complex showed band at 301 nm for $\pi \rightarrow \pi^*$ transition and band at 401 nm for $n \rightarrow \pi^*$ [18–20]. On the other hand, the complex showed new band at 499 nm due to charge transfer from metal to the low energy level of π^* (antibonding) orbitals in ligand [21].

Infrared Spectra of the IPDN ligand and its silver complex

Infrared spectroscopy is a fundamental technique for identifying functional groups in organic compounds and metal complexes. Infrared spectra examine the bending and stretching vibrations of the synthesized dye ligand and its silver complex in the range of 400-4000 cm^{-1} as shown in Figs. 6 and 7. The IPDN ligand exhibited characteristic peaks attributed to important functional group such as CH aromatic group at 3047 cm^{-1}, the aliphatic group peak at 2955 cm^{-1}, the C=C group peak at 1624 cm^{-1}, 1571 cm^{-1}, and the C–H bending group at 1031 cm^{-1} and 814 cm^{-1}. It is worth noting that these peaks did not undergo a significant change after the ligand coordinated with the silver ions, indicating that these groups did not participate in the coordination process with the metals in the prepared complexes [22–24]. The IPDN ligand also showed a clear absorption of the azo group (N=N) at 1526 cm^{-1}, which remained constant in its position and intensity within the silver complex, confirming that this group did not participate in coordination with the metal ions.

In addition, a distinct peak for the hydroxyl group (O–H) appeared in the ligand spectrum at 3336 cm^{-1}, but it completely disappeared in the complex spectrum, indicating its participation in the coordination process. A peak for the C–O

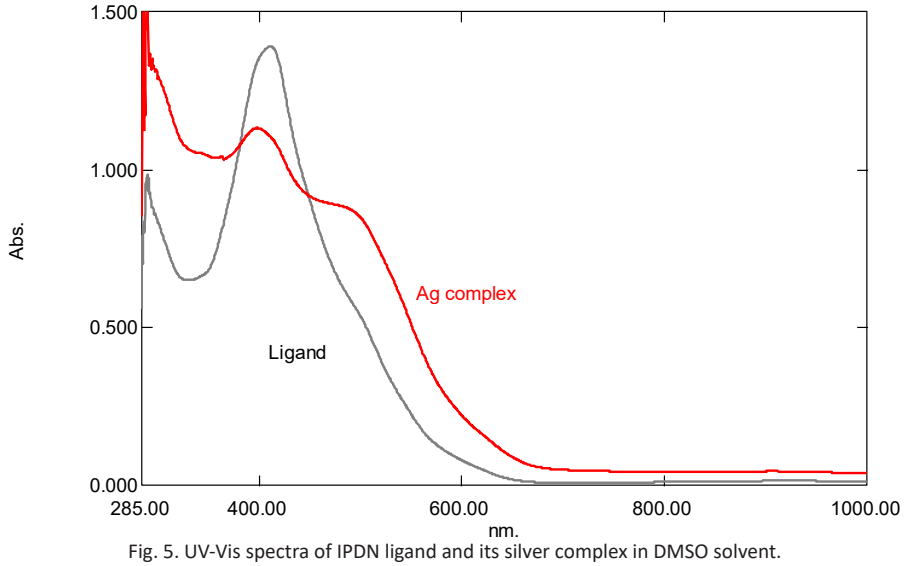

Fig. 5. UV-Vis spectra of IPDN ligand and its silver complex in DMSO solvent.

group was also recorded at 1228 cm^{-1} in the ligand spectrum, which shifted to 1247 cm^{-1} in the silver complex spectrum, further strengthening the evidence for its participation in coordination with the silver ion. Also, a new peak appeared in the silver spectrum at 434 cm^{-1}, attributed to

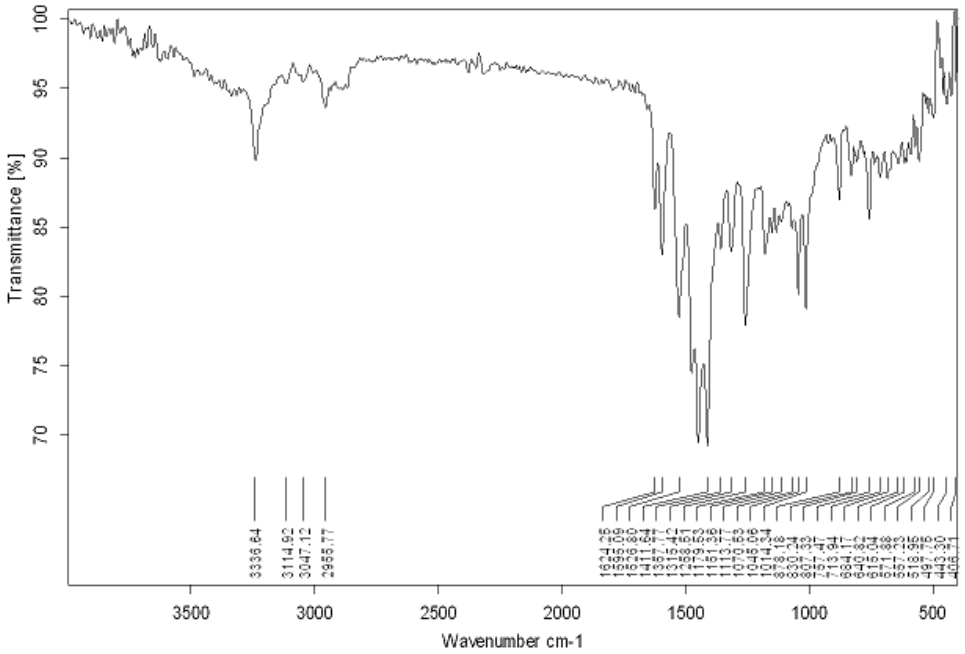

Fig. 6. Infrared spectrum of IPDN MPPD ligand in KBr pellet.

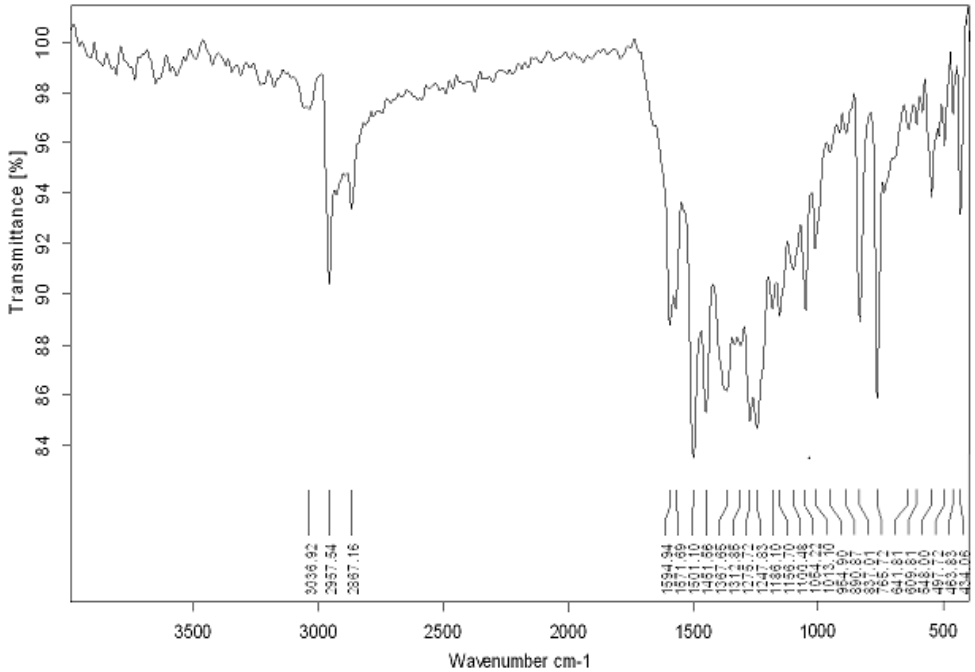

Fig. 7. Infrared spectrum of silver complex for IPDN ligand in KBr pellet.

M–O bond vibrations, confirming the association of oxygen with the silver ion within the complex [25–27].

Proton nuclear magnetic resonance (^{1}H-NMR) spectroscopy of the IPDN ligand and its silver complex

The structure of the IPDN ligand was characterized using proton nuclear magnetic resonance (^{1}H-NMR) spectroscopy, as shown in Fig. 8. The spectrum showed eight distinct signals in the aromatic region, attributed to the protons of the phenyl ring and naphthalene rings associated by azo group in the ligand structure.

A signal was observed at a chemical shift of 8.90 ppm, attributed to the proton at position (10) on the naphthalene ring. The high activity of this proton is attributed to the electronic influence of the hydroxyl group.

Two triplet signals were also observed, corresponding to the protons of the naphthalene ring, as well as two doublet signals, each representing two protons, corresponding to the protons of the phenyl ring. In the aliphatic region, a doublet signal was recorded at 1.18 ppm, resulting from the methyl groups (CH_3) with a total proton

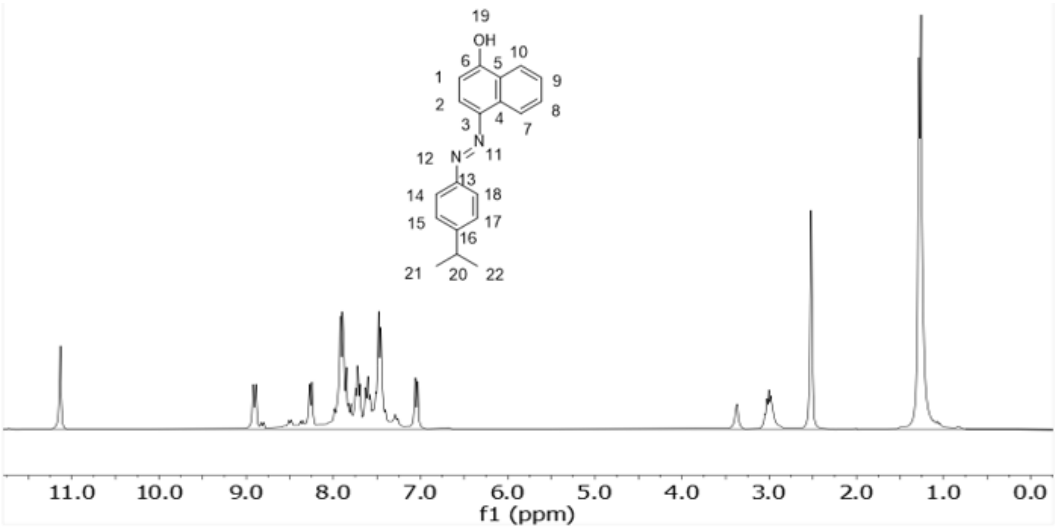

Fig. 8. ^{1}H-NMR spectrum of IPDN ligand in DMSO d6.

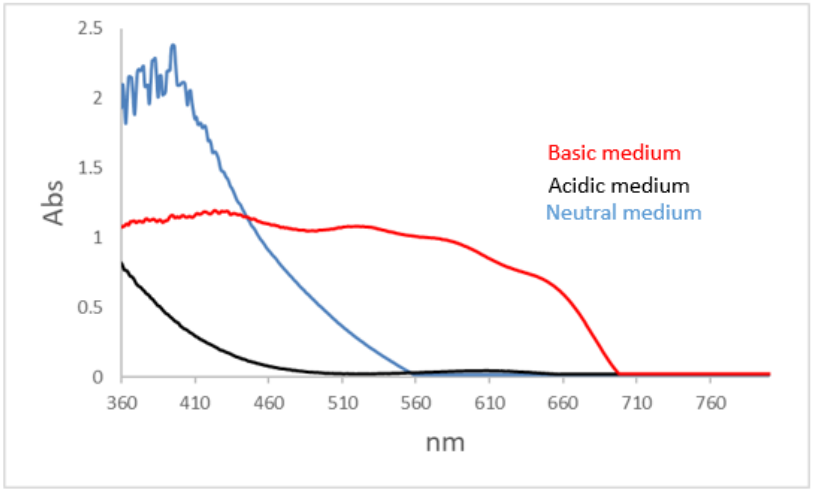

Fig. 9. pH effects on UV-Vis spectra of IPDN ligand in aqueous solution.

count of six. On the other hand, a triplet signal at 2.62 ppm is due to CH of isopropyl. The aromatic signals of the ligand were distributed within the range of 8.90–7.05 ppm.

Proton nuclear magnetic resonance (^{1}H-NMR) spectrum of the silver complex preparing in the same solvent (DMSO-d*6*) was measured. The spectrum showed a pattern similar to that of the free ligand in terms of the number of signals, with some shifts in the chemical positions observed as a result of the complex formation with the silver ion. The aromatic signals of the complex were distributed within the range of 8.36–7.64 ppm. This shift is attributed to the coordination effect between the ligand and the silver ion, which alters the electronic environment surrounding the

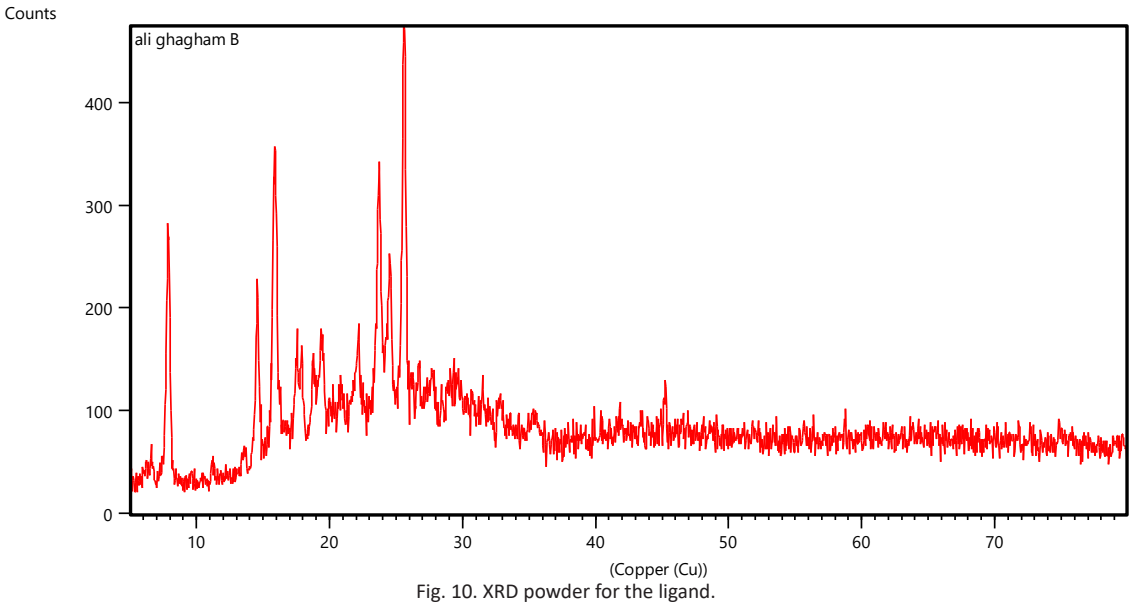

Fig. 10. XRD powder for the ligand.

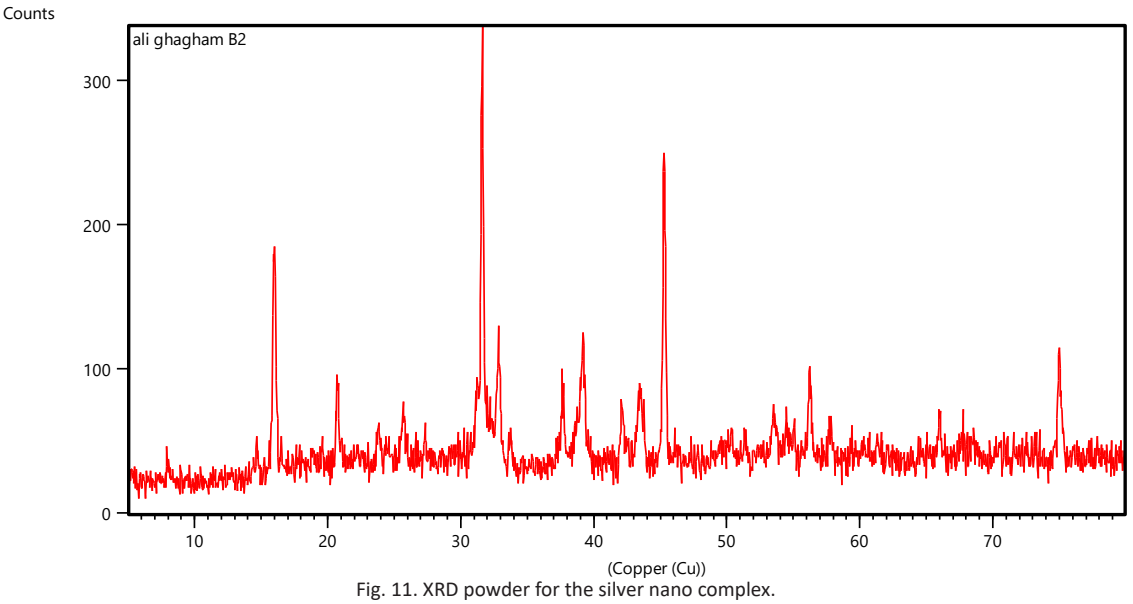

Fig. 11. XRD powder for the silver nano complex.

aromatic protons.

pH effect

The color and absorption behavior of the IPDN ligand was studied using UV-visible spectrophotometer under acidic, neutral, and basic conditions, as shown in Fig. 9. From the spectra, it was observed that the ligand in neutral medium exhibited an orange-red color in aqueous solution, with a maximum absorption at 502 nm. In acidic conditions, clear changes occurred, represented by yellow color, accompanied by a significant blue shift in the absorption spectrum to 439 nm due to protonation of the ligand's active sites, which reduced the effectiveness of the electron resonance system[28].

In basic conditions, the color of the solution turned dark red, with a significant red shift (bathochromic shift), with the maximum absorption at 522 nm. This change indicates an enhanced extension of the resonance system in the ligand's molecular structure and absorption at higher wavelengths resulting from the deprotonation of the hydroxyl group. Fig. 9, showed significance of the blue and red shifts resulting from changes in the nature of the medium surrounding the ligand, reflecting a clear response to the surrounding chemical environment and its direct impact on the optical and absorption properties of the compound.

XRD characterization

XRD spectrum of ligand and its complex are presented in Figs. 10 and 11. The ligand showed the XRD peaks at 2θ equals to 7.890º, 14.617º, 15.921º, 23.747º, and 25.669º. The complex showed the XRD peaks at 2θ equals to 15.852º, 20.726º, 31.778º, 45.301º, 56.216º, and 75.025º. The ligand and its complex showed high intensity and sharp peaks which may be attributed to long range order of the molecules and high degree of the crystallite nature. The crystallite size was calculated using Scherrer formula where the ligand showed a crystallite size 11.9 nm and the complex showed crystallite size (6.34 nm) in the range of nanoparticle materials.

CONCLUSION

In this study, a novel pH-responsive azo dye incorporating aromatic rings was successfully synthesized through the diazotization of 4-Isopropylaniline followed by coupling with 1-naphthol. Furthermore, its silver(I) complex was prepared with a yield of 80%. The structural, optical, and physicochemical properties of the synthesized compounds were comprehensively characterized using elemental analysis, mass spectrometry, FTIR, UV-Vis, ^{1}H-NMR, techniques. The FTIR spectra confirmed the preservation of the azo (–N=N–) functionality in both the ligand and the silver complex, while clear evidence indicated the participation of the hydroxyl group in metal coordination. The UV-Vis analysis revealed significant bathochromic and hypsochromic shifts in the absorption spectra under varying pH conditions, confirming the dye's pH-responsiveness.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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