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Investigation of Spectrofluorimetric and Electrochemical Properties of Schiff Bases and Metal Complexes

Schiff Bazları ve Metal Komplekslerinin Spektroflorimetrik ve Elektrokimyasal Özelliklerinin İncelenmesi

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ABSTRACT

In this review article, the electrochemical and fluorescence properties of Schiff bases are discussed. Schif bases are structures that contain primary amines as a result of the condensation of aldehydes or ketones. They become radical with the excitation of electrons from the amine groups in their structures and show fluorescence properties. Likewise, radicalized electrons are an important container in electrochemical applications, especially in sensor applications, and Schiff bases such as ferrocene have shown significant sensitivity in suitable buffer solutions. This review article is informative for electrochemical and fluorescence applications with Schiff bases.

Keywords: Electrochemistry, Fluorescence, Metal Complexes, Schiff Base

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ABSTRACT

Bu derleme yazısında Schiff bazlarının elektrokimyasal ve floresans özellikleri tartışılmıştır. Schif bazları, aldehitlerin veya ketonların yoğunlaşması sonucu birincil aminler içeren yapılardır. Yapılarındaki amin gruplarından elektronların uyarılması ile radikalleşirler ve floresans özelliği gösterirler. Aynı şekilde, radikalleşmiş elektronlar, elektrokimyasal uygulamalarda, özellikle sensör uygulamalarında önemli bir kaptır ve ferrosen gibi Schiff bazları, uygun tampon çözeltilerde önemli bir hassasiyet göstermiştir. Bu derleme makalesi, Schiff bazları ile elektrokimyasal ve floresan uygulamaları için bilgilendiricidir.

Anahtar Kelimeler: Elektrokimya, Floresans, Metal Kompleksler, Schiff Bazı

1. Introduction

Compounds that are formed as a result of the condensation of aldehydes and ketones with primary amines under appropriate reaction conditions and that have C=N in their structures are called "Schiff bases" (Subasi & Subasi, 2022). Generally, compounds with C = N in their structure are called "azomethine or imine compounds" and the functional group consisting of C = N bonds is called the "Azomethine group" (figure 1) (Talukder, 2005). Aldehydes and ketones undergo addition and elimination (condensation) reactions with amines and Schiff base are synthesized by this reaction (Burinsky et al., 1984).

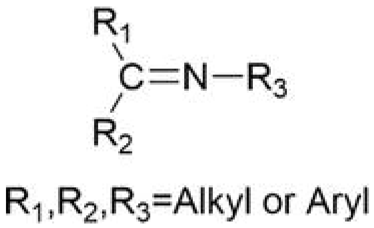

Figure 1: Skeleton of schiff bases (1)

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The reaction of aldehydes from Schiff bases is quite easy. But this process is not easy for ketones (Friedman, 2004; Layer, 1963). The reason for this is the selection of the appropriate pH range, the suitable catalyst for production of Schiff base form ketones, and the choice of solvent that will from an azeotrope mixture with the water to be formed in the reaction, making the reaction more specific (Gomez et al., n.d.). As a result of the reaction of primary amines with aldehydes, the carbon-nitrogen double bond in Schiff bases is called aldimine or azomethine, and the bond formed as a result of the reaction with ketone is called ketamine or imine (Subasi & Subasi, 2022). Reactions are very easy to synthesize using semicarbazide or thiosemicarbazide.

Semi carbazides and thiosemicarbazides are also widely used with carbonyl compounds (Dolman et al., 2006). Semicarbazones generally hydrolyze more readily than the correspnding oximes or hydrazones. The formation of semicarbazones from ketones and semicarbazides is catalyzed by aniline (More et al., 2019). Therefore, the mechanism differs from the normal general acid catalysis of semicarbazones. It is formed after the exchange of an aniline with a semicarbazide (1).

The reaction rate of Schiff bases and semicarbazides is faster than the reaction of semicarbazides with free carbonyl groups (Cordes & Jencks, 1962). This is most likely because it is more basic than the parent carbonyl groups from which the azomethine groups are derived. The reactions of aldehydes and ketones with 2,4-dinitrophenylhydrazine, semicarbazide, and hydroxylamine have generally been used to identify aldehydes and ketones (Feuell & Skellon, 1953). Their compounds, 2,4-dinitrophenylhydrazones, semicarbazones, and oximes, are solids with sharp characteristic melting points. The use of Schiff bases with metal complexes by researchers has become quite common.

Metal complexes of Schiff bases have been known since the middle of the 19th century (Kaczmarek et al., 2018). N,N-ethylene bis (salicylideniminato) (Salen) is a prime example of this. Cationic, bridging, and dimeric complexes are seen in several Schiff bases (Gheller et al., 1981). The donor atoms in the molecule are taken into account when classifying metal complexes of Schiff bases (Raczuk et al., 2022). As a result, the most prevalent metal complexes are those with donor atom systems, such as N-O, O-N-O, O-N-S, N-N-O, O-N-N-O, and N-N-N-N (Radanović et al., 2022). Scientists have developed a wide range of complexes after becoming interested in schiff bases, which are used as ligands in the production of coordination compounds (Radanović et al., 2022). The metal complexes made from Schiff bases are employed as dyestuff in the dye business, particularly in the textile industry, since the groups in their structures give them color. The importance of Schiff base complexes in the medical world is increasing due to their anticancer activity and their use as a reagent in the fight against cancer is being investigated (Malik et al., 2018), (Liu et al., 2014). Some transition metals are known to have nucleolytic activities. Buredox active compounds break the phosphodiesteroskeleton of DNA molecules at physiological pH and temperature. Cu(II) complexes are very important in terms of biological activity and are known as antitumor, antiviral and anti-inflammatory agents (Liu et al., 2014; Subasi & Subasi, 2022). Especially Cu(II) complexes formed from Schiff base ligands have become important model compounds in the study of the physical and chemical behavior of biological copper systems. The Cu(II) complex of 1,10-Phenanthroline is the first synthetic transition metal complex to show effective nucleolytic activity. Copper complexes of some ligands such as bleomycin, pyrrole, thioether, oxime, peptide, and imidazole show DNA-clearing activity.

It is a known fact that Schiff bases show fluorescence properties. The color change of Schiff bases in fluorescence studies is important in terms of playing a decisive role in colorimetric sensor studies. It also affects the conductivity of the catalysts in the surface modification of the electrodes in electrochemical studies. Therefore, in this review article, the role of Schiff bases in the application areas, the application methods and applications in the electrochemical and fluorescence areas and how Schiff bases will advance in these areas in today's technology, this review has been created by scanning the literature reports in detail and citing these reports. The congress of this article was also presented at the ICON Data congress in 2021.

Fluorescence

The light emitted during the transition from an excited singlet system to a ground-state singlet system is called fluorescence (Beer & Longuet-Higgins, 2004). In the case of fluorescence and phosphorescence; an electron in a chromophore group of matter absorbs the short wavelength beam sent on the molecule in a very short time, like 10-14 seconds, and rises to an upper (sometimes two or three higher) energy level. From here, it returns to the basic state through various mechanisms (Westermayr & Marquetand, 2021). The state of the electron, when it goes up one or two levels without changing its spin, is called the excited singlet state (Hanson et al., 2007). A molecule in an excited singlet state is still diamagnetic as it is in its ground state, diamagnetic materials are also called electron-paired materials. A molecule changes from its basic electronic and vibrational state to its excited state by absorption (Myers et al., 1998). When the molecule is in its excited state, excess vibrational energy is dissipated by intermolecular collisions. The molecule then emits a beam to the ground energy level, creating fluorescence (Lichtman & Conchello, 2005).

Qualitative and quantitative analyzes of many organic and inorganic substances can be done by fluorescence spectroscopy of molecules. The most important feature of this method is that it is sensitive, that is, it can be analyzed in much smaller quantities. For organic molecules, the term fluorescence usually refers to the light energy emitted during a transition from the lowest excited singlet level to the fundamental singlet level (Y. Yu et al., 2021).

Stokes Shift

Transitions in molecular fluorescence bands always contain longer wavelengths or less energy than transitions in the absorbed radiation band that cause their excitation (Vincett et al., 2003). This change towards longer wavelengths is sometimes called the stokes shift (Y. Yu et al., 2021).

Fluorescence the Effect of Molecular Structure

Fluorescence efficiency is typically increased by planarity, rotational inhibition, conjugation, and an increase in the number of rings (H. Yu & Ji, 2021). Compounds with aromatic rings that permit low energy * transitions in their structure emit the strongest fluorescence rays (Wachter et al., 1998). Additionally, aromatic rings with numerous conjugated double bonds in their structure, such as those found in aliphatic and alicyclic rings, exhibit fluorescence. However, their number is very low compared to the number of aromatic compounds. Apart from these, aliphatic and alicyclic compounds containing carbonyl groups and conjugated systems containing many double bonds also show fluorescence (H. Yu & Ji, 2021). These properties are further enhanced by the condensation of the aromatic rings. Heterocyclic rings such as pyridine, thiophene, pyrrole, and furan do not show fluorescence, substances excited as a result of the n → π* transition easily become triplet and show only phosphorescence (Wachter et al., 1998). Simple nitrogen-containing heterocyclic rings are characterized by n* low energy electronic transitions, which facilitate the transition between excited singlet and excited triplet states. Phosphorescence emission is used to transition from the triplet state to the ground state; in these situations, fluorescence is reduced or eliminated (Lower & El-Sayed, 1966).

Conjugation Effect on Fluorescence

In aromatic compounds, the electrons of the conjugated double bonds are delocalized. The greater the number of rings, the greater the delocalization. As a result, as the number of rings increases, the fluorescence intensity increases (Lower & El-Sayed, 1966; Wachter et al., 1998). In benzene, fluorescence occurs with high-energy photons, while in pentacene, it occurs with low-energy photons. While the fluorescence in benzene is in the UV region with higher energy, the fluorescence in pentacene is in the visible region (Lower & El-Sayed, 1966).

The Effect of The Rigidity and Planarity of the Molecule

The fluorescence property of a molecule is enhanced by the stiffness of its structure. A chelating complexing ligand's stiffness rises as it combines with a cation, which also raises fluorescence emission (26). Fluorescence with low energy transitions is exhibited in compounds with aromatic functional groups and is the strongest and most advantageous. A smaller number of compounds than aromatic systems can fluoresce, including those with a lot of conjugated double-bonded structures or aliphatic and alicyclic carbonyl groups (Lower & El-Sayed, 1966; Wachter et al., 1998).

Effect Of Substituents in the Molecule

A potential light transition between the excited singlet state and the ground state is typically increased by substituents in a luminous compound that can delocalize the substance's electrons (Wachter et al., 1998). Fluorescence also rises as a result of this. Ordinarily, substituents in the ortho-para position give the ring electrons, perhaps enhancing the fluorescence that will take place. The potential for fluorescence is decreased by the substituents at the meta-position, which take electrons away from the ring (Lower & El-Sayed, 1966).

Temperature and Solvent Effect

The fluorescence emission decreases as the temperature rises, increasing collision and facilitating conversion. Increasing the polarity of the solvent also affects the fluorescence [25]. Because polar solvents generally increase the energy difference in $n \rightarrow \pi^*$ transitions and decrease the fluorescence glow. In some cases, the energy of the $\pi \rightarrow \pi^*$ transition even falls below that of the $n \rightarrow \pi^*$ transition. Solvents containing heavy atoms, such as carbon tetrachloride and ethyliodide, reduce the intensity of fluorescence radiation (Lower & El-Sayed, 1966).

Viscosity Effect

Increasing the temperature and decreasing the viscosity of the solvent increase the probability of collision between the excited molecule and other molecules, and also inter-system transitions. In low temperature and high viscosity medium, the resting time is longer than the lifetime of the excited state and fluorescence increases (Lower & El-Sayed, 1966).

The Effect of Wavelength and Intensity of the Incident Beam

Luminescence efficiency may be reduced not only by physical events such as internal transformation, transition between systems and energy transfer, but also due to events such as bond breakage and product formation that may occur at stimulated levels. In addition, increasing the intensity of the incident beam increases the fluorescence. The lower limit of wavelengths that produce fluorescence is 250 nm (Talukder, 2005; Tuna Subasi, 2023). Therefore, fluorescences corresponding to the $\sigma^* \rightarrow \sigma$ transition are rarely encountered. Rays up to the aforementioned wavelength can only produce $\pi \rightarrow \pi^*$ and $\pi \rightarrow n$ transitions (Tuna Subasi, 2023).

2. Investigation of Fluorescence Properties of Some Sciff Bases and Metal Complexes

2.1. Study of the silver(I) ion Complex

At wavelengths where ligands fluoresced brightly following complex formation, a change was observed. The emission wavelength of 460 nm to 470 nm has been moved from the excitation wavelength of 385 nm of 375 nm (Talukder, 2005b; Tuna Subasi, 2023). In contrast to other silver compounds developed in this investigation, complex has comparatively modest fluorescence intensity (Wachter et al., 1998). The rigid conformational shape of the ligand upon complexation or the strong coordination with the Ag(I) metal center are likely the causes of the rise in fluorescence intensity of complexes. The silver electron density was found to reduce the fluorescence band's intensity in the complexes (ex = 320 nm , em = 395 nm em = 469 and, ex = 406 nm) (Lower & El-Sayed, 1966).

2.2. Examination of Cu/Mn Complexes

The four schiff bases [Cu(L1)$_2$]n (1), [Mn(L1)$_2$]n (2), [Cu(L2)$_2$]n (3), [Mn(L2)$_2$]n (4, HL1 = 2-(((4H-1,2,4-triazol-4-yl)imino)methyl)-4,6-dichlorophenol; HL2 = 2-(((4H-1,2,4-triazol-4 yl)imino)methyl)-4,6-dibromophenol) was synthesized.

The luminescence compounds' properties free and 1-4 HL1 by using DMF [24]. A wavelength of 438 nm was used to stimulate the luminescence of the HL1, 1 and 2 series. Upon photoexcitation at 438 nm, a luminescence band of free HL1 ligand was observed at 470 nm with maximal weighted π–π* transition fluorescence (25). At the same 438 nm photoexcitation, and also observed emission bands and lower fluorescence intensity at 527 and 514 nm, redshifting the wavelength to 57 nm and 44 nm, respectively, compared to the HL1 ligand. This could be caused by the paramagnetic action of the corresponding d9 and d5 ions, Cu(II) or Mn(II) (1).

2.3. Investigation of Sc^{3+}, Pd^{2+}, Sr^{2+}, Bi^{3+} , Ag^{+}, Co^{2+}, Cd^{2+}, Ni^{2+}, Cr^{3+}, Zn^{2+}, K^{+}, Ca^{2+}, Fe^{3+}, Li^{+}, Be^{2+} , Mn^{2+}, Cu (II), Mg^{2+} , Na^{+} and Al^{3+} Complexes

It was created using 4-(1-(4-hydroxy-3-methoxybenzyl)-1H-benzo[d]imidazol-2-yl)-2-methoxyphenol (L). Spectra of the ligand acetonitrile:water (1:1) in terms of emission and absorption We looked at the ligand's fluorescence spectrum in the medium. The maximum fluorescence spectra showed emission at 360 nm when the ligand was stimulated with 300 nm (1). Investigated are the ligand's fluorescence spectra in acetonitrile water (1:1) with 10 equivalents of excess metal ions. Under the effect of ions such as these title ions, the ligand's fluorescence spectra did not significantly alter. However, the Cu^{2+} ion effectively quenches fluorescent light (Carlson, 2015). At 360 nm, it displayed the degree of fluorescence of the unbound ligand and the ligand solution with metal ions. Cu^{2+} ion is preferred by the ligand over other metal ions. This outcome is in line with findings in the literature (1), (Lower & El-Sayed, 1966; Wachter et al., 1998).

2.4. Investigation of Zn(II)/Ln(III) Complexes

2,3,Dihydroxynaphthalene-1,4-dicarbaldehyde and 2-[O-(1-ethyloxyamide)] are Schiff bases. As well as [Zn_2(L)Ho(m2-OAc)2(OAc)(MeOH)] and oxime-6-methoxyphenol. Complexes of [Zn_2(L) Er (m2-OAc)$_2$] and [CH_2Cl_2] OAc were created. Recently, it has been discovered that several lanthanide compounds of the Salamo type show exceptional fluorescence characteristics. Components of ZnII were used as sensitizers for lanthanide fluorescent lighting (Tuna Subasi, 2023). In-depth measurements were made of the emission spectra of H4L in CH_3OH:$CHCl_3$ (v/v=1:1) solution and the corresponding metallic complexes 1 and 2 in methanol solution. H4L has a wavelength of 442 nm and a 350 nm intra-ligand $^{-}$* transition. Complexes 1 and 2 displayed rather significant emission peaks at 447 and 448 nm, respectively, at a wavelength of roughly 350 nanometers. These bathochromically shifted, comparatively strong emission peaks can be attributed to ligand-to-metal charge transfer (LMCT) transitions (Štěpnička et al., 1999).

3. Electrochemical Methods

Scientists like Galvani, Volta, and Cavendish recognized that electricity had intriguing and significant interactions with mammalian tissues as early as the experimental sciences era. For instance, electrical demands cause the muscles to stretch and contract. Physiological electricity is produced by some creatures, like the torpedo, which is considerably more problematic (Figure 2). Over 50 billion nerve endings on flat, wing-like structures on the left and right of the torpedo abruptly exude acetylcholin compound to the membranes on the underside of the wings (Gary D & Purnendu K, 2013). Acetylcholine induces the sodium ions (Na^+) membranes to suddenly travel through membranes, perhaps differentiating this from a quick load separation (Altuner, Ozalp, Yilmaz, Sudagidan, et al., 2022; Dunant & Israel, 1985). The sea water surrounding the torpedo experiences a current of several amps as a result of this potential differential.

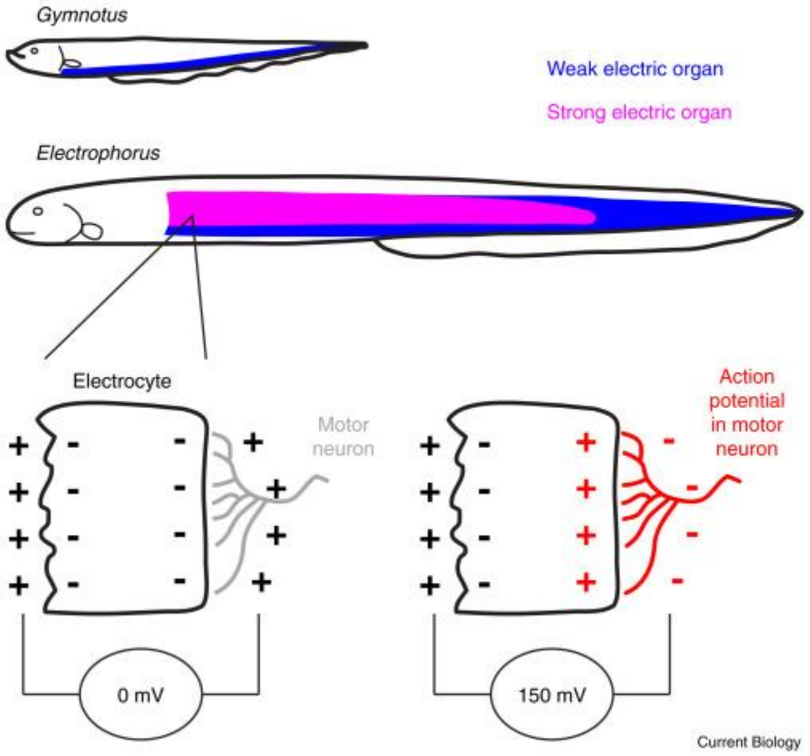

Figure 2.: Potential distinction of the torpedo fish on the right and left wings (Carlson, 2015).

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3.1. Comparison of Redox Reactions with Acid-Base Reactions

The notion of Bronsted-Lowry is used to compare redox reactions to acid-base reactions. Both involve the transfer of one or more laden particles from the transmitter to the recipient (Altuner, Ozalp, Yilmaz, Sudagidan, et al., 2022). In general, the Bronted-Lowry reaction occurs as in equation 1 (Eq.1):

Acid (I) + Base (II) $\leftrightarrow$ Acid (II) + Base (I) (Eq.1) (30).

A conjugated acid-base pair is reacting, as seen by the reaction of Eq.1. Electrons in redox processes and protons that neutralize are examples of loaded particles. An acid becomes a conjugate base that can accept protons when it gives a proton. Similar to this, when a base electron offers, it transforms into an electron-accepting amplifier (Altuner, Ozalp, Yilmaz, Sudagidan, et al., 2022).

3.2. Electrochemical Cells

Two conductors, referred to as "electrodes," each submerged in electrolyte solution, make up an electrochemical cell. A salt bridge is inserted into the solutions to stop mixing. Thus, the migration of potassium ions and chloride ions in one direction from the salt bridge is what transmits the electrical solution from one electrolyte solution to the other electrolyte solution. The direct interaction of copper metal and silver ions is nevertheless avoided (Altuner, Ozalp, Yilmaz, Sudagidan, et al., 2022). Electrochemical cathode and anode reaction samples resemble equality 2 (Eq.2) and equality 3 (Eq.4), respectively.

$$Ag^{+} + e^{-} \leftrightarrow Ag(k)$$
$$Fe^{3+} + e^{-} \leftrightarrow Fe^{2+}$$
$$NO_3^{-} + 10H^{+} + 8e^{-} \leftrightarrow NH_4^{+} + 3\,H_2O \text{ (Eq.2) (30)}$$

By applying the proper voltage to an inert electrode, such as platinum, these reactions are created.

$$Cu(k) \leftrightarrow Cu^{2+} + 2\,e^{-}$$
$$2\,Cl^{-} \leftrightarrow Cl_2(k) + 2e^{-}$$
$$Fe^{2+} \leftrightarrow Fe^{3+} + e^{-} \text{ [Eq.3] (30).}$$

The cathode irtiary environment is also used to solve the problems in Eq 3. The literature is replete with accounts of electrochemical research (Alizadeh et al., 2022; Altuner, Ozalp, Yilmaz, Bekmezci, et al., 2022; Altuner, Ozalp, Yilmaz, Sudagidan, et al., 2022).

3.3. Electrochemical applications for Schiff Bases

Ferrocene and its derivatives have been extensively researched in organometallic chemistry ever since their discovery (Scottwell et al., 2014; Ye et al., 2005). Particularly intriguing is the iron core redox chemistry (Woo Rhee et al., 2000) and structure (Frantz et al., 2004; Wong et al., 2001). Ferrocene/ferrocenium (Fc/Fc^{+}) is a one-electron redox pair that is strictly reversible in nature. However, substituents on the ferrocene moiety would affect the redox behavior by altering the HOMO's energy level (Szarka et al., 2001), which may drastically slow down reversibility (Štěpnička et al., 1999). Arylamines and carbonyl compounds condense to form schiff bases, which are extremely stable and used as adaptable intermediates in the synthesis of a number of important molecules. Ferrocene moiety incorporation into Schiff bases provides the chemical and physical characteristics that are missing or hardly noticeable in the parent material.

Conclusion

The investigations and significance of Schiff bases in electrochemical and fluorescence studies, particularly sensors, are discussed in this review article. Aldehydes or ketones condense with primary amines to form Schiff bases. Since Schiff bases form solid complexes with many metals, particularly transition metals, they are preferred as a suitable ligand in the identification of these metals. The chemical characteristics of Schiff bases and metal complexes, which are used in many different fields, must be determined. When molecules' fluorescence characteristics are looked at, flatness, inhibition of rotation, conjugation, and an increase in the number of rings all tend to boost fluorescence effectiveness. Compounds with aromatic rings are given by the strongest fluorescence rays, enabling the transition. Therefore, the color change of Schiff bases under fluorescence rays is important for use in colorimetric fluorospectrometric sensors. This colorimetric color change is also used in electrochemistry, affecting the rate of electron transition in electrochemical sensors. Electron transition of catalysts in the system coated with Schiff bases is related to the condensation of aromatic rings, and therefore, Schiff bases form complexes with various metals, giving an insight to new studies.

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