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Solvatochromic photoluminescence investigation of functional Schiff-bases: A systematic study of substituent effects

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ABSTRACT

In this paper a series of Schiff-bases, synthesized mechanochemically, were studied for their delicate changes in absorption and emission spectra by altering substituents and solvents at different concentrations. UV-vis and photoluminescence of aromatic salicylideneaniline Schiff-base (**ASASB**) derivatives with substituents ranging from electron donating to electron withdrawing ($-\text{CH}_3$, $-\text{Cl}$, $-\text{Br}$, $-\text{I}$, $-\text{C}(=\text{O})\text{CH}_3$, $-\text{C}(=\text{O})\text{OC}_2\text{H}_5$, $-\text{CN}$), and ring nitrogen containing derivatives were studied in solvents of different dielectric constant (*n*-hexane, toluene, ethyl acetate, THF, ethanol and methanol).

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1. Introduction

The basic processes in crystalline photochromic systems comprise hydrogen transfer, dimerisation, cyclization, ring opening, valence tautomerisation and isomerization. Crystalline Schiff-bases of salicylaldehyde (anils), aromatic Schiff-bases with *ortho* hydroxyl group, undergo photochromism or the related phenomenon of thermochromism initiated by an intramolecular hydrogen transfer from the *ortho*-hydroxy group to the nitrogen atom of the imine in the ground (thermochromism) or the excited (photochromism) electronic state, via a six-membered ring transition state, producing keto species with bathochromically shifted spectra [1–4]. Basically the intramolecular hydrogen transfer process at excited state can occur on molecules like salicylidene anilines [1], benzophenones [5], flavones [6], quinolones [7], quinoxalines [8], azoles [9], polymeric materials [10], etc.

Senier et al. [11] observed the reversible solid state photo-coloration of *N*-salicylideneanilines and noted that the various ring substituted compounds were polymorphic, of which only a few were photochromic. The observations revealed the effect of the structure on photochromism. Cohen, Hirshberg and Schmidt [11,12] showed that the presence of an *ortho*-hydroxy group is a

structural requirement for the photochromism of these compounds and proposed the H-transfer mechanism for the crystalline *N*-salicylideneanilines [13].

Schiff-bases are molecules that contain the azomethine group ($-\text{CH}=\text{N}-$) in their structure. They are generally synthesized by the condensation of primary amines and active carbonyl compounds [14,15]. This important class of organic molecules and their metal complexes are the focus in catalysis [16], chemosensing [17,18], bioimaging [17], organic-light emitting diodes (OLED) [19], dye-sensitized solar cell (DSSC) [15], medicine [20], etc. Along this line mainly salicylideneaniline Schiff-bases have also been the focus of non-linear optical (NLO) studies for potential applications such as data storage, information processing, telecommunication and optical switching. They are also considered as capable of competing with inorganic crystals for reversible photo-switching applications. And related studies have also been extended to other non-centrosymmetric photochromic crystals of salicylideneanilines in order to generate materials for data storage and optoelectronic switching [1], with wide applications it gives them an important place in diverse fields of chemistry, biochemistry and material science. Due to their photochromism and intramolecular hydrogen bonding, Schiff-bases with salicylideneaniline (SA) are a typical anil that shows photochromism and is one of the most important compounds in photochemistry [21–23]. The excited state intramolecular proton transfer (ESIPT) present in these compounds makes them also interesting objects for theoretical

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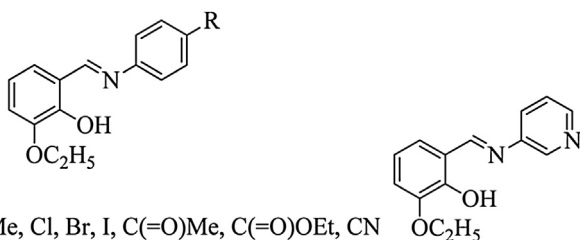


Fig. 1. The structure of ASASB used in this study.

studies of molecular dynamics in electronically excited states [2–4]. As a result, detailed knowledge of the photoactivity of the compounds and their structure is very important in order to achieve the above goals. Hence herein we are interested to present the photolytic behavior of salicylideneanilines derivatives (Fig. 2), particularly focusing on solvent and substituent effect.

Recently, solvent and substituent effects on absorption and emission spectrum have been reported in literatures. Pearson et al. in their earlier study showed substituent effects in the keto-enol tautomerism of fused 1,4-naphthalenediols [24]. Zakerhamidi et al. have also mentioned substituent and solvent effects on the spectroscopic properties and dipole moments of hydroxyl benzaldehyde azo dye and related Schiff-bases [25]. Wanko, et al. lately have studied the optical absorption properties of *p*-, *m*-, and *o*-nitrophenolate in vacuo and in bulk solution [26] and Kaya et al. also declared the effect of substituent position on optical, electrical, electrochemical, and fluorescence properties of

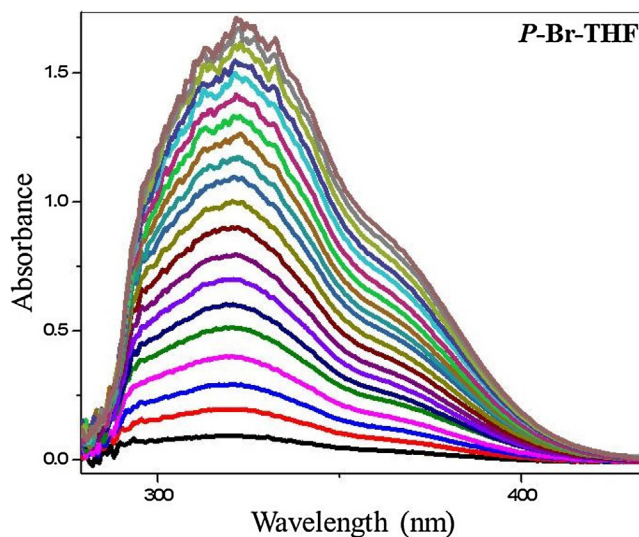


Fig. 3. Absorption spectra of *p*-Br ASASB in THF at twenty increments of concentrations.

polyphenol species derived from methyl substituted aminopyridine based Schiff-bases [27]. Particularly, Kim et al. [28], Chuang et al. [29], Hanson et al. [30], Mutai et al. [31] and Sakai et al. [32], have presented substituents effect on ESIPT molecules, which resulted in change of the ESIPT emissions due to different power of electron donating (or withdrawing) effect.

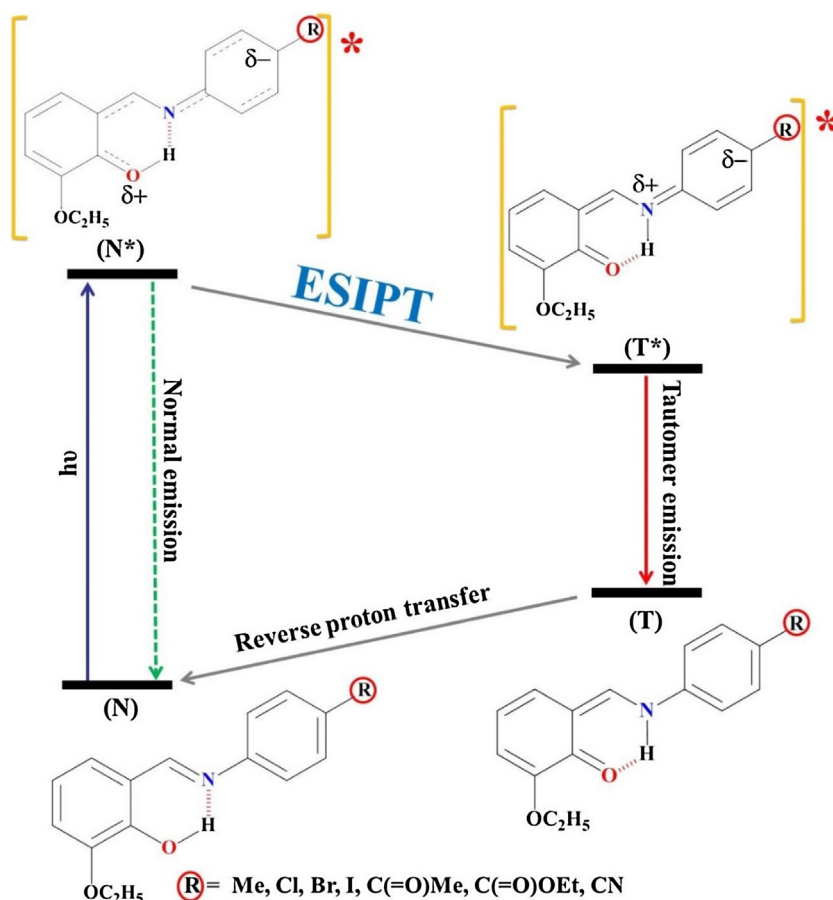


Fig. 2. Principal photophysics on mechanism of ESIPT, illustrated by ASASB derivatives.

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