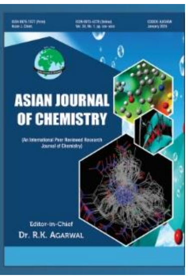
Asian Journal of Chemistry; Vol. 38, No. 2 (2026), 265-277

ASIAN JOURNAL OF CHEMISTRY

https://doi.org/10.14233/ajchem.2026.34266



REVIEW

Role of Schiff Bases and their Complexes in Optoelectronic Devices: A Review

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Received: 16 June 2025 *Accepted*: 24 December 2025 *Published online*: 31 January 2026 AJC-22246

Schiff bases, also known as imines, have gained considerable popularity for use in optoelectronic devices. Their ability to coordinate with different metal ions, unique electronic and charge transport properties and tunability *via* minor structural modifications make them suitable for these devices. The ease of modifying the optical energy band gap for desired absorption and emission characteristics, reduced toxicity, enhanced energy efficiency and material flexibility are attractive features promoting their use in optoelectronics. The present study compares important electrochemical, thermal and photophysical properties like optical band gaps, HOMO-LUMO energies, thermal stabilities and absorption/emission wavelengths of Schiff bases and their metal complexes. High fluorescence quantum and efficient charge transport are crucial for efficient light emission and enhanced performance of various optoelectronic devices. Schiff bases, their metal complexes and hybrid systems composed of Schiff base complexes with metal/semiconductor materials, which have been recently studied for use in organic light-emitting diode (OLED) and organic solar cells (OSC), have been reviewed in this article.

Keywords: Schiff bases, Metal complexes, Optoelectronic properties, Organic light-emitting diode, Organic solar cells.

INTRODUCTION

In the current era of smart technologies, optoelectronic devices have subtly transformed our lifestyles by enabling improved sensing, communication and data processing capabilities. It is the quantum mechanical nature of light and its interaction with the electronic materials that plays the key role in the functioning of optoelectronic devices [1-6]. The field of optoelectronics deals with the study and application of devices that emit, sense and control the properties of light, such as intensity and wavelength. Optoelectronic devices usually consist of semiconductor alloys embedded on substrates, like photodetectors, solar cells and LEDs [1,2]. The morphology of the functional materials influences the performance and properties of such materials for their diverse applications in chemical/ biological analysis, medicine, telecommunications, energy harvesting, *etc.* to name a few [1,2,7]. Usually, semiconducting inorganic compounds such as gallium arsenide phosphide ($GaAs_{1-x}P_x$) and indium phosphide (InP), have been used in the optoelectronic devices. However, they are associated with several disadvantages like rigidity, potential toxicity, trace existence, *etc.* making them cost ineffective [8-12]. On the other hand, functional metal-organic materials possess several advantages like stronger emission, large Stokes shift and photostability of transition-metal complexes, along with convenient synthesis and being cost effective [13-16]. Given these benefits, significant progress has been made in using organic materials in optoelectronics. Among these, Schiff bases have attracted considerable attention from researchers due to their simple synthesis protocols, stability and ability to form complexes with nearly all transition metal ions [17,18].

Schiff bases are formed by a condensation reaction between primary amines and aldehydes or ketones, as shown in Fig. 1. The first Schiff base was reported by Hugo Schiff (1834-1915) and was named after him [19,20]. Schiff bases are also recognised as imines or azomethines and function as ligands, coordinating to the metal ions by utilizing the nitrogen of the imine group, along with other available donor sites present in the molecule. This multidentate bonding results in the formation of chelates. The planar geometry exhibited by many Schiff base metal complexes enhances the quantum emission compared to free ligands [19-21]. Schiff bases are suitable for a wide range of applications due to their biodegradability, non-toxicity, desirable electrical properties and cost-effective production.

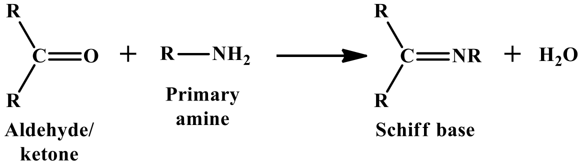


Fig. 1. General reaction for the formation of a Schiff base (SB)

Furthermore, the photochemical and photophysical features of Schiff base complexes can be easily tailored [21,22]. The properties of Schiff base can be customised by introducing, modifying or substituting appropriate functional groups in the ligands, using different solvent systems and forming metal complexes [23]. The Schiff bases have been utilised as high-efficiency selective sensing materials for ion sensing applications [24-26]. Krylova *et al.* [27] observed that incorporating a redox-active ligand in the coordination sphere of a complex modulated the energy of the frontier molecular orbitals, enhancing their availability and increasing the reactivity of the complex. This modified the redox properties of the metal centre, which affected the semiconductor and optoelectronic properties of the complex. Fig. 2 depicts structures of Schiff bases ligands (A and B) formed by condensation of 2-hydroxybenzaldehyde and hydrazinecarboxamide/thiosemicarbazone, Schiff base ligand-salophen (C) and Schiff base ligand D formed by condensation of 2,6-pyridinedicarboxaldehyde and 2-hydroxyphenylamines (D). Tin(IV) complexes of these Schiff bases show promising results when incorporated in optoelectronic devices such as organic light-emitting diodes (OLEDs) [28], solar cells and for fluorescence bioimaging [27]. The Sn-Schiff base complexes have coordination numbers five, six and seven, achieved by additional coordination to ethyl and 1,10-phenanthroline molecules. The gap between the highest occupied molecular orbitals (HOMOs) and lowest unoccupied molecular orbitals (LUMOs) of the tin derivatives obtained was similar to that found in organic semiconductors currently being used. Hyper-coordination and bond formation of Sn with N-donor were found to be a powerful method to tune the energy of the HOMO-LUMO levels [27].

A range of hybrid systems incorporating Schiff base complexes with metals or other semiconductor components has been extensively studied and reviewed for their remarkable optoelectronic properties. These materials demonstrate significant potential in advanced applications, which include converting light to energy in dye-sensitised solar cells [29-31], light activated catalysis, bio-labeling and photoactivated pro-drug delivery [32-34]. Novel hybrid materials comprising Schiff base metal complexes and metallic or semiconducting nanomaterials or polymer matrices exhibit an entirely new range of optical properties [32,33]. These hybrid systems modify and showcase the complex's novel catalytic and optical properties, which are not present in the individual components. Keeping this in mind, the present study focuses on the recent advancements in utilizing Schiff bases and their complexes/hybrids for OLEDs and OSCs (Fig. 3).

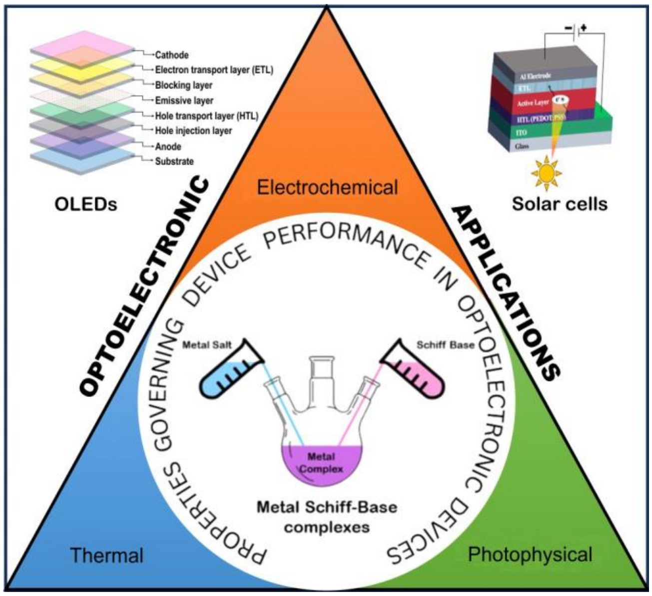


Fig. 3. Optoelectronics applications of Schiff bases and their complexes

Methodology: Articles related to the study were retrieved from 'Scopus', 'ScienceDirect' and 'Web of Science' databases with the focus on the work published between 2010 to 2024. The extraction process started by entering the key words like "Schiff base'', "complexes" "oled", "organic light emitting diode", "solar cell", "photovoltaic", "optoelectronics" and

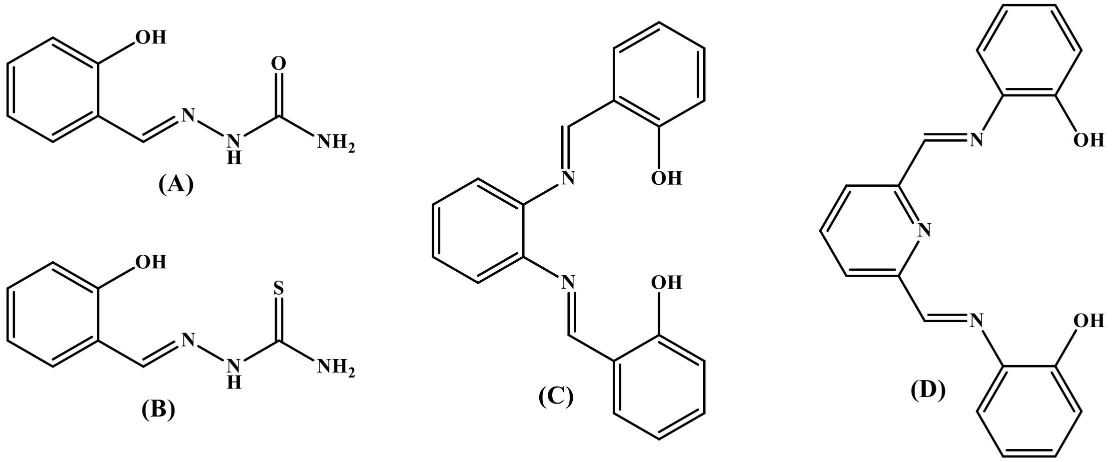


Fig. 2. Structure of some Schiff base ligands when coordinated to Sn(IV) exhibit useful optoelectronic properties [27]

their different combinations. The search excluded the single key words search due to an extremely large number of entries obtained. The following nine keyword combinations were used such as "Schiff bases, oled"; "Schiff base, organic light emitting diode"; "Schiff base complexes, oled"; "Schiff base, complexes, organic light emitting diode"; "Schiff base, solar cell"; "Schiff base, photovoltaic"; "Schiff base, complexes, solar cell"; "Schiff base, complexes, photovoltaic"; "Schiff base, optoelectronics" in each of the databases. These keyword combinations gave a total of 340, 667 and 1168 entries in the 'Scopus', 'ScienceDirect' and 'Web of Science' databases, respectively. The initial dataset thus contained 2175 entries. This data was uploaded in the 'Zotero' software (version 7.0. 30) and the duplicates (1395) and retracted papers (2) were removed to give 778 unique studies. These records were then screened to exclude the articles that involved purely synthesis of Schiff bases, experimental/computational studies or pure biological/medicinal studies or irrelevant reviews, books, reports, *etc*. Only the full-length relevant journal articles that focussed on Schiff bases and their complexes for their use in optoelectronics were included (189). A final refinement was further carried out to target 80 high quality journals that cover the synthesis strategies, key parameters for optoelectronic applications like optical band gaps, absorption and emission wavelengths of Schiff bases, their metal-coordination complexes and hybrid systems for OLEDs and OSC applications (Fig. 4). The review also focuses on the methods to enhance and improve the features of optoelectronic devices based on Schiff bases and its complexes. It is noteworthy to mention that other articles reviewed in the present study include historical information, future prospects and other general information.

Key properties of Schiff bases and their complexes for optoelectronic applications: The functionality of organic materials and hybrids used in optoelectronic devices mainly depends on their electrochemical, thermal and photophysical characteristics. For Schiff bases and their metal complexes, many of these key parameters fall within the required range, making them ideal candidates for use in optoelectronic devices.

Electrochemical properties: The energies of the HOMO and the LUMO are crucial parameters in determining the optoelectronic properties of Schiff base complexes. The electronic transitions, charge transport parameters and overall efficiency of devices such as OLEDs and photovoltaic cells depend on these energy levels and the optical energy band gap [35]. The optical energy band gap is the minimum energy required for photons to be absorbed by a material [33]. Electrochemical and spectral data are generally used to estimate the energy values of the HOMO and LUMO. Higher HOMO values are associated with the presence of electron-donating substituents in the same ligand scaffold. The effect of substitution has a greater impact on the values of LUMO levels compared to HOMO levels [36]. Complexes of Schiff bases with metals and variation or alteration of the ligand scaffold complexed to these metals are used to modify the optical energy band gap and tune their energies for desired absorption and emission characteristics [37-40].

Thermal properties: The heat resistance of materials plays a pivotal role in optoelectronic devices, as it directly impacts key attributes of efficiency and longevity. Gusev *et al.* [36] observed remarkable thermal characteristics of zinc complexes, enabling the fabrication of OLED devices using the vacuum thermal evaporation technique. It was proposed that

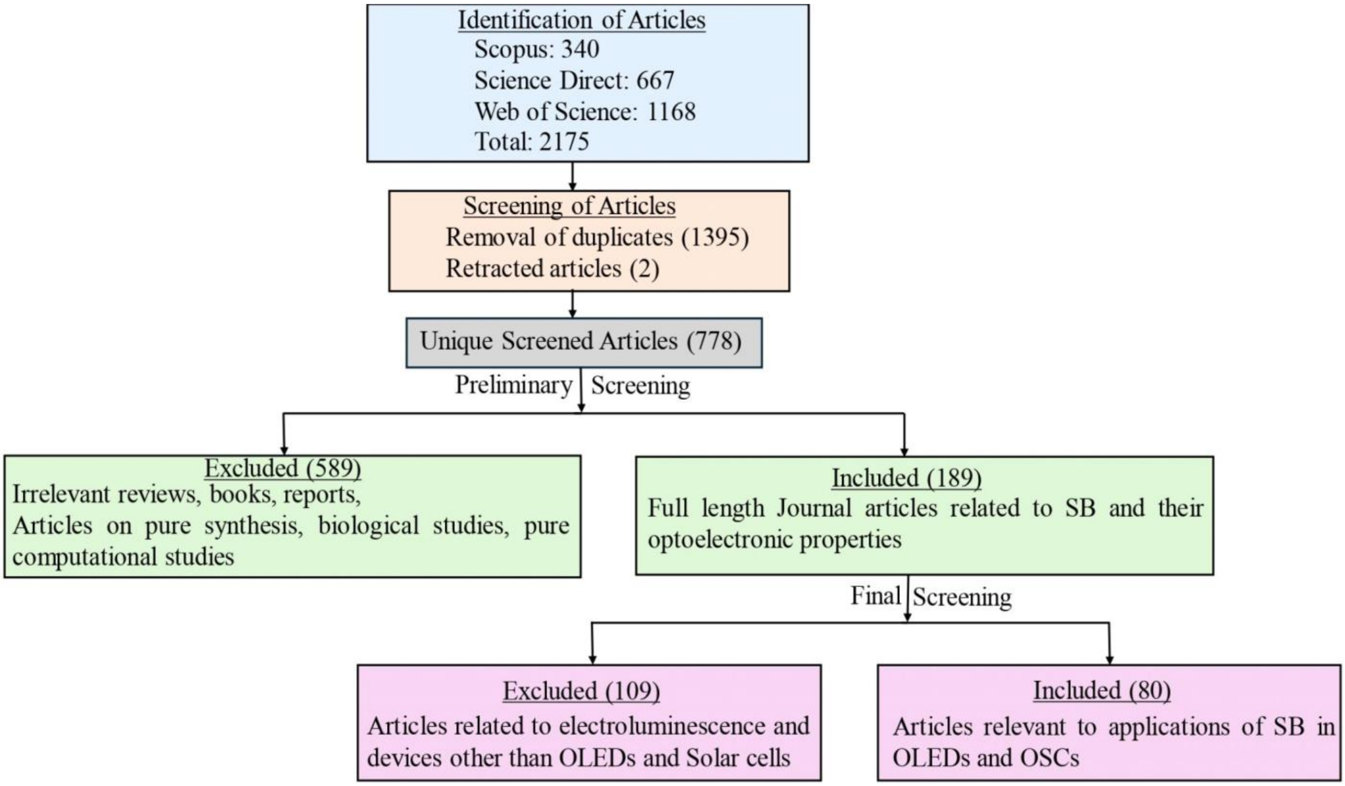


Fig. 4. Flow chart depicting the selection of literature (SB = Schiff base)

higher the degree of conjugation of ligands in a complex, the greater the ability to withstand high temperatures. Furthermore, these complexes exhibit higher glass transition temperatures and melting points, improving the durability and longevity of optoelectronic devices [23].

Photophysical properties: Absorbance, an important characteristic of solutions that tells us the percentage of light absorbed by a solution. Absorptivity is a measure of how well a material absorbs a photon. The Schiff base complexes often display intense absorption bands, while their emission spectra are influenced by their metal centres and surrounding ligands [41]. High fluorescence quantum and efficient charge transport are crucial for efficient light emission and enhanced performance of various optoelectronic devices [42,43].

Optoelectronic applications of Schiff bases, their complexes and hybrid materials

Organic light emitting diodes (OLEDs): Organic light emitting diodes (OLEDs) are solid-state devices consisting of an organic electroluminescent layer that emits light in response to an electric current. The OLED configuration (Fig. 5) incorporates a glass substrate layer and a transparent plastic foil that supports the device [44]. The electrons move from the cathode to the electron transport layer (ETL) and then to the emissive layer (EML). The ETL facilitates the movement of electrons but blocks the transport of holes. Similarly, holes reach the EML through the hole transport layer (HTL), which allows movement of holes and blocks electron movement. Together, ETL and HTL control the movement of charge carriers within the device and increase device efficiency [44].

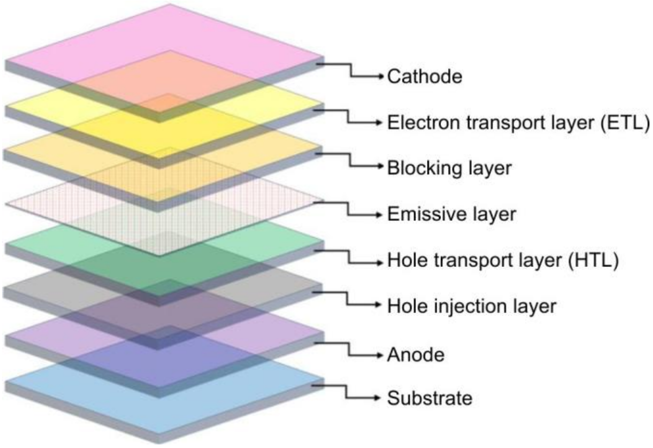


Fig. 5. Basic structure of an organic light emitting diode (OLED)

The OLEDs work on the principle of electroluminescence, *i.e.* light emitted by an organic material in response to the passage of electric current between the anode and the cathode [44]. The electrons and holes from the cathode and the anode enter the organic layer, where they recombine and release energy as light [44]. They are great for portable applications as they exhibit high light output, low power drain and are easy to read, even in direct sunlight. The electrochemical and photophysical properties of the Schiff base complexes explored in this review, with potential applications in OLEDs, are summarised in Table-1.

In 1987, Tang & Vanslyke [45] pioneered OLED research using tris(8-hydroxyquinolinato)aluminium (Alq_3) and demonstrated that metal complexes could generate colours in the visible range, for optoelectronic applications. Nishal *et al.* [46] synthesised and studied two novel soluble zinc Schiff base complexes with ligands synthesised by condensing *p*-aminodimethylaniline with 2-hydroxybenzaldehyde and 2-hydroxy-1-naphthaldehyde, respectively. The broad absorption maxima observed were attributed to π-π^* ligand transitions. Interestingly, spectacular results were obtained with these Schiff bases as the emitter, Alq_3 as the electron-transporting layer and (N,N′-di(1-naphthyl)-N,N′-diphenyl-(1,1′-biphenyl)-4,4′-diamine) (NPB) as the hole-transporting layer in OLEDs. The red OLEDs were obtained with 2-hydroxy-1-naphthaldehyde-based zinc Schiff base complex [Zn(SB-2)$_2$] (luminescence efficiency 0.28 cd A^{-1}), while yellow luminescence was observed with 2-hydroxybenzaldehyde-based zinc complex [Zn(SB-1)$_2$] (luminescence efficiency of 0.7cd A^{-1}) [23,45].

Nishal *et al.* [46] supported that substituents in the organic ligands complexed to the metals can be suitably modified to produce desired emission wavelengths. They studied the effect of elongating the bridging chain by increasing the number of alkyl groups. These salicylidiminato Schiff base ligand-based zinc complexes were found to be good blue light-emitting sources under UV radiation, which paved the way for the generation of white light for optoelectronic displays [46]. Thus, Schiff base zinc complexes are crucial light-emitting materials due to their good electron transport ability, light-emitting efficacy, ability to withstand high temperatures, redox stability and easily tunable electronic properties [46]. Several other complexes of zinc with salicylidiminato Schiff base ligands have been found to emit light in blue [47], green [48] and red [49] regions.

Achieving full white electroluminescence posed a serious challenge, as Gusev *et al.* [36] in 2021 observed that it was difficult to generate blue wavelengths in OLEDs. Available resources were restricted to organic aromatics, polymers and transition metal complexes of Ir(III), Os(II) and Pt(II), with restricted reserves of these metal ions [36]. However, zinc Schiff base complexes of benzoheterocycles successfully produce intense sky-blue fluorescence, made possible by the ease of tuning their properties *via* ligand functionalisation. In the same year, Kagatikar & Sunil [35] reviewed various organic light-emitting devices made from Schiff base and their complexes with metals, *viz.*, Zn, Pt, Mn, Fe and W. They observed that the wavelengths of the emissive material could be tuned by incorporating various substituents/metal atoms. Tuning influences the thermal stability of the emissive layer and its ability to emit blue, green, red and white colours. The work emphasised the need to improve operational stability and study the mechanism of intrinsic OLED degradation to develop better devices.

Nayak *et al.* [50] also synthesised two zinc Schiff base complexes, which were promising candidates for use as an emissive layer in OLEDs. Zn(SB-17) with Schiff base derived from 2-amino-5-methylphenol and 3-methoxy-4-phenoxybenzaldehyde emitted green light, while Zn(SB-18) where the Schiff base is derived from a condensation reaction between

TABLE-1
ELECTROCHEMICAL, THERMAL AND PHOTOPHYSICAL PROPERTIES OF SCHIFF BASE AND THEIR COMPLEXES USED IN ORGANIC LIGHT EMITTING DIODES (OLEDs)

SB/SB complex	E_{HOMO} (eV)	E_{LUMO} (eV)	Energy gap (eV)	T_m (ºC)	T_d (ºC)	Solution		Solid	Ref.
						Absorption Λ_{max} (nm)	Emission Λ_{max} (nm)	Emission Λ_{max} (nm)	
[Zn(SB-1)$_2$]	-5.00	-2.50	2.50	276	377	423	557		[23]
[Zn(SB-2)$_2$]	-4.90	-2.60	2.33	344	387	452	600		
[Zn(SB-3)]			3.14		~380				[46]
[Zn(SB-4)]			3.15		~380				
[Zn(SB-5)]			3.04		~380				
[Zn(SB-6)]			2.80		~380				
[Zn(SB-7)]			3.08		~380				
[Zn(SB-8)$_2$]	-5.33	-2.25	3.08	247	355	339	468	470	[36]
[Zn(SB-9)$_2$]	-5.21	-2.18	3.03	254	335	344	446, 459	445, 466	
[Zn(SB-10)$_2$]	-5.27	-1.87	3.40	267	330	307	436, 455	438, 458	
MacrocyclicSB11			3.45						[52]
MacrocyclicSB12			3.29						
[Sn(SB-13)(Et)$_2$]	-5.617	-1.514	4.103			386			[27]
[Sn(SB-13)(Et)$_2$(phen)]	-5.053	-2.074	2.979			385			
[Sn(SB-14)(Et)$_2$(phen)]	-5.350	-1.896	3.454			404			
[Sn(SB-15)(Et)$_2$]	-5.740	-2.156	3.584			463			
[Sn(SB-16)(Et)$_2$]	-5.644	-2.681	2.963			516			
[Zn(SB-17)]	-5.29	-2.87	2.42			265, 262			[50]
[Zn(SB-18)]	-5.30	-2.82	2.48			349, 337			
Yb(SB-19))(HSB-19)	-6.00	-4.80	1.20				~1000		[51]
Th-In (SB-20)	-5.70	-2.21	3.49			291	464	523	[53]
Th-Qn (SB-21)	-6.20	-2.81	3.40			295	374	523	
Th-TPA (SB-22)	-5.34	-2.36	2.99			436	530	578	

T_m = Melting point; T_d = Decomposition temperature; SB = Schiff base

2-aminophenol and 4-phenoxybenzaldehyde, emitted a yellowish-orange colour.

Kornikov *et al.* [51] synthesised and characterised two lanthanide complexes of ytterbium and gadolinium, Ln(SB-19)-(HSB-19), where H_2SB-19 ligand is 2-tosylamino-benzylidene-(2-benzo[*d*]thiazole)hydrazone. It was found that benzo[*d*]thiazole group in ligand contributed to the ytterbium complex exhibiting high electron mobility. It was successfully tested for near-IR emission in the 1000 nm range in host-free OLED owing to its high quantum yield (~ 0.9) and solubility.

Using *o*-phenylenediamine and 2-hydroxy-5-methylisophthalaldehyde/2-hydroxy-5-*tert*-butyl-1,3-benzenedicarboxaldehyde as the starting reagents, Barwiolek *et al.* [52] synthesised two macrocyclic Schiff bases. The fluorescence studies revealed that the compounds exhibited emission in the 454-516 nm and 452-417 nm ranges, respectively, on excitation at 295 nm. These materials form smooth films and show good semiconducting behaviour, making them good candidates for OLEDs [52]. Recently, Vite-Morales *et al.* [53] reported a green mechanosynthesis of three thiadiazole-derived Schiff bases (SB-20, 21, 22), without using any solvent. 1,3,4-Thiadiazole, when covalently bonded to indole, quinoline and triphenylamine groups (Th-In)/(Th-Qn)/(Th-TPA), exhibited suitable band gaps and photophysical properties suitable for optoelectronic and sensor applications.

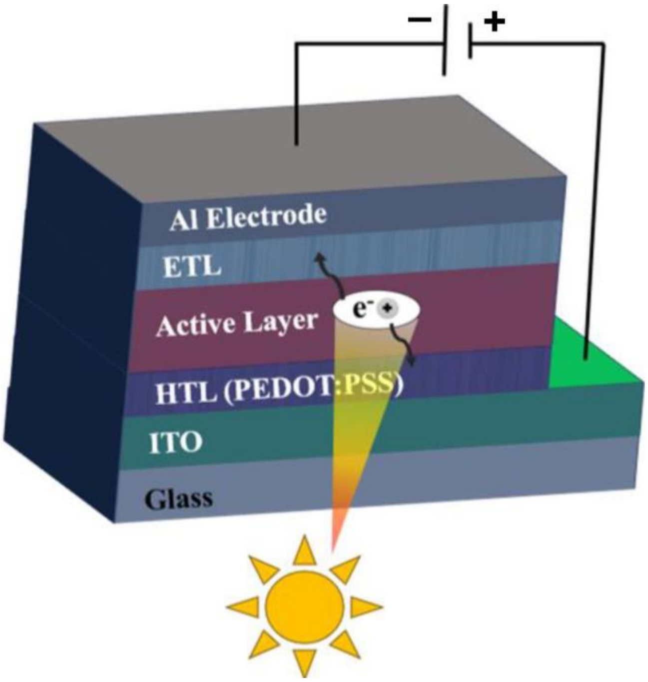


Fig. 6. Basic structure of an organic solar cell (OSC)

Organic solar cells (OSCs): Organic solar cells (Fig. 6) utilize organic semiconductor materials that, on exposure to sunlight, convert solar energy to electrical energy. This is achieved as solar exposure generates electron-hole pairs, which migrate to oppositely charged electrodes when subjected to an electric field, causing the current to flow [54,55]. As in OLEDs, ETL in solar cells facilitates electron travel, but now from the photoactive layer to the cathode, while inhibiting the movement of holes [56]. Meanwhile, the HTL allows the movement of holes from the photoactive layer to the anode, but does not allow electrons to move to the anode [54]. Additional layers in the device include the widely used poly(3,4-ethylenedioxythiophene):polystyrene sulfonate **(**PEDOT:PSS) conductive polymer as a hole transport layer and a transparent indium tin oxide (ITO) conductive layer, which functions as the anode while allowing light transmission to the photoactive layer [57].

Small azomethine molecules synthesised by Schiff base condensation chemistry have also been tested for developing organic photovoltaic (OPV) materials [58-62]. It is well-known that the azomethine groups in Schiff base function as π-conjugated organic materials with semiconducting and hole-transporting properties. The advantage of azomethines over vinylenes is that they can be prepared by inexpensive Schiff base condensation without the requirement of expensive transition metal catalysts, harsh chemicals or adverse reaction conditions and with water as a benign byproduct [58]. Small molecules are also beneficial due to their good solubility, well-defined structures and lower batch-to-batch variations. In azomethine compounds, the various optical and electro-optical properties can be tuned by protonating the free electron pair on nitrogen of the azomethine group [58]. The electrochemical and photophysical properties of the Schiff base complexes, used in OSCs, are also reviewed and are summarised in Table-2.

Petrus *et al.* [58] synthesised two small azomethine molecules, TPA-Th-TPA (SB-23) and TPA-TBT-TPA (SB-24), where TPA = 4-aminotriphenylamine; Th = thiophene; TBT = 4,7-*bis*(5-formylthiophen-2-yl)-2,1,3-benzothiadiazole. They found that when acidic PEDOT:PSS was replaced by neutral molybdenum oxide (MoO_x), the bulk heterojunction device output voltage increased from 0.45 V to 0.68 V, thereby increasing the efficiency from 0.35% to 0.42%. They observed that TPA-TBT-TPA (SB-24), with its small bandgap, exhibits the best overlap with the solar spectrum, resulting in higher current, open-circuit voltage and fill factor. These super qualities result in the best power conversion efficiency (PCE). The use of a reaction mixture enabled the direct fabrication of OPV devices, eliminating the hassles of product workup and resulted in an improved PCE of 0.6%. Thus, the use of azomethine chemistry for the synthesis of OPV devices is easy, clean, doesn't require any harsh conditions and is cost-effective [63,64].

Sek *et al.* [65] studied the condensation reaction of 2-naphthylamine with different aldehydes, each having a different number of phenyl rings. They obtained five unsymmetrical and one symmetrical Schiff bases, which were found to be thermally stable above 200 °C and absorbed light up to 420 nm. An examination of the HOMO and LUMO levels reveals that they lie in the same range (Table-2) (SB 25-30) and possess similar energy gaps, E_g. When these selected Schiff bases were tested as an acceptor component of the active layer in a bulk heterojunction solar cell, the highest PCE of 0.32% was achieved

TABLE-2
ELECTROCHEMICAL, THERMAL AND PHOTOPHYSICAL PROPERTIES OF SCHIFF BASE AND THEIR COMPLEXES USED IN ORGANIC SOLAR CELLS (OSCs)

SB/SB complex	E_{HOMO} (eV)	E_{LUMO} (eV)	Energy gap (eV)	T_m (ºC)	T_d (ºC)	Solution: Absorption Λ_{max} (nm)	Solution: Emission Λ_{max} (nm)	Solid: Emission Λ_{max} (nm)	Ref.
TPA-Th-TPA (SB-23)	-5.32	-3.13	2.19	200	>350				[58]
TPA-TBT-TPA (SB-24)	-5.26	-3.36	1.88	200	>350				
SB-25	-5.44	-3.86	1.58		267	267, 326 ($CHCl_3$) 268, 326 (NMP)	397, 433 ($CHCl_3$) 404 (NMP)	391	[65]
SB-26	-5.49	-3.90	1.59		423	291, 374 ($CHCl_3$) 292, 374 (NMP)	407 (NMP)	390	
SB-27	-5.48	-3.83	1.65			261, 405 ($CHCl_3$) 408 (NMP)	397, 420 ($CHCl_3$) 396 (NMP)	394	
SB-28	-5.45	-3.83	1.63		387	251, 335 ($CHCl_3$) 337 (NMP)	392, 414 ($CHCl_3$) 411 (NMP)	388	
SB-29	-5.47	-3.89	1.58		358	257, 286, 334 ($CHCl_3$) 285, 334 (NMP)	406 (NMP)	386	
SB-30	-5.46	-3.96	1.60		352	290, 333 ($CHCl_3$) 291, 336 (NMP)	407 (NMP)	388	
R_2SnO-SB-31 (R = nBu, Ph)	-5.2	-2.1	2.35			343, 479	564		[66]
R_2SnO-SB-32 (R = nBu, Ph)	-5.3	-2.5	2.31			351, 489	565		
R_2SnO-SB-33 (R = nBu, Ph)	-5.5	-2.5	2.33			354, 489	556		
R_2SnO-SB-34 (R = nBu, Ph)	-5.0	-2.1	2.25			336, 492	598		
SB-35 Dioxane	-5.490	-2.349	2.761			399	434		[67]
$CHCl_3$	-5.501	-2.266	2.675			402	443		
EA	-5.495	-2.308	2.720			404	440		
THF	-5.497	-2.297	2.718			405	441		
DCM	-5.495	-2.307	2.702			401	439		
SB-36 Dioxane	-5.730	-2.658	2.492			387	477		
$CHCl_3$	-5.738	-2.624	2.395			391	486		
EA	-5.733	-2.442	2.471			388	476		
THF	-5.734	-2.637	2.451			392	484		
DCM	-5.733	-2.641	2.426			391	487		
CuSB-37	-5.37	-3.14							[68]
PdSB-37	-5.40	-3.18							
PtSB-37	-5.36	-3.22							

[Ni- (SB-38)$_2$]	-5.54	-2.10	3.44					[69]
[Ni- (SB-38)$_2$]COOH	-5.72	-2.41	3.31					
[Ni- (SB-38)$_2$]CN	-6.00	-2.70	3.30					
[Ni- (SB-38)$_2$]NO$_2$	-6.04	-3.13	2.91					
[Cu- (SB-38)$_2$]	-5.79	-1.86	3.92					
[Cu- (SB-38)$_2$]-COOH	-5.96	-2.38	3.58					
[Cu- (SB-38)$_2$]-CN	-6.24	-2.65	3.59					
[Cu- (SB-38)$_2$]-NO$_2$	-6.28	-3.12	3.16					
[Zn(SB-38)$_2$]	-5.81	-1.95	3.86					
[Zn(SB-38)$_2$]-COOH	-6.02	-2.44	3.58					
[Zn(SB-38)$_2$]-CN	-6.27	-2.70	3.57					
[Zn(SB-38)$_2$]-NO$_2$	-6.33	-3.12	3.21					
I-8 (SB-39)			1.09, 3.35					[82]
I-6 (SB-40)			1.07, 2.79					
SB-41	-5.65	-2.68	2.97		Gas	512, 316		[83]
					Ethanol	477, 334		
					Acetone	478, 334		
					DMF	477, 335		
					DMSO	477, 335		
SB-42	-5.62	-2.64	2.98		Gas	510, 312		
					Ethanol	474, 325		
					Acetone	474, 325		
					DMF	474, 326		
					DMSO	473, 326		
SB-43	-5.88	-3.81	2.07		Gas	410, 502		
					Ethanol	334, 478		
					Acetone	330, 240		
					DMF	332, 240		
					DMSO	335, 467		
SB-44	-5.55	-2.55	3.00		Gas	308, 230		
					Ethanol	330, 240		
					Acetone	330, 240		
					DMF	332, 240		
					DMSO	331, 240		
SB-46	-4.43/-5.00	-3.81						[85]
SB-47	-4.45/-4.98	-3.82						
Sn(IV)-SB-49a			3.53					[87]
Sn(IV)-SB-49b			3.60					
Sn(IV)-SB-49c			3.53					
Sn(IV)-SB-49d			3.63					

T$_m$ = Melting point; T$_d$ = Decomposition temperature; NMP = N-Methyl-2-pyrrolidone; SB = Schiff base

under illumination of 1.3 mW/cm^2. In 2018, Cantón-Díaz *et al.* [66] prepared four organotin compounds bearing Schiff bases *via* a multicomponent microwave-assisted reaction. They used 2-hydroxy-1-naphthaldehyde, 2-aminophenol derivatives and the corresponding diorganotin oxide and recorded their optoelectronic properties. Due to the electron-withdrawing nature of the azomethine C=N group, complexation with the metal ions leads to fascinating ‘‘push-pull’’ molecules, with interesting and useful nonlinear optical properties. These could be utilised to make photoinduced charge transfer possible and they can be applied to OPV devices like heterojunction solar cells. Another important feature of these complexes is their absorption (480-490 nm) in the visible region, near the maximum solar emission, along with an optical band gap of around 2.3 eV [R$_2$SnO-SB-31-34, (R = nBu, Ph)], affording them as potential electron acceptor materials in the synthesis of OPV materials [66].

It is also well known that two basic parameters, the energy band gap and refractive index, control the optical and electronic behaviour of semiconductors. In this regard, the two low molar mass Schiff base hydrazone derivatives, 1-(4-nitrophenyl)-2-(4-phenoxybenzylidene)hydrazone and 1-(2,4-dinitrophenyl)-2-(4-phenoxybenzylidene)hydrazone, were synthesised and characterised (Table-2, SB 35 and SB 36) by Sıdır *et al.* [67]. The energy band gap of a semiconductor depends on its light absorption threshold, whereas the refractive index depends on the degree of transparency of the material to incident light. Both optoelectronic parameters govern the semiconductor band structures, but the relationship between the two remains debatable.

Wang *et al.* [68] synthesised several Schiff base metal complexes (MSB-37, where M = Pt, Pd and Cu and SB-37 are phthalocyanine and porphyrin-based Schiff base) to test their performance as hole transport materials (HTMs) in perovskite solar cells. It was observed that variations in the central metal atom allow for better alignment of energy levels with the perovskite absorber. The Cu-based device was found to have the highest PCE of 8.07% among the analysed MSB-37 devices. This could be attributed to well-matched energy levels, low surface roughness and the high crystalline nature of the thin film. The PCE of these HTMs was as follows: CuSB-37 = 6.82 ± 0.88 (8.07)%; PdSB-37 = 4.90 ± 0.53 (6.25)%; PtSB-37 = 6.28 ± 0.61 (7.35)%.

A study by Irfan *et al.* [69] investigated the influence of metal and electron-withdrawing groups on the charge transport and optoelectronic properties of *bis*(dithiocarbazate)-based SB-38 [H-(Ap-sadtc)] complexes containing Ni(II), Cu(II) and Zn(II), which were synthesised by Nanjundan *et al.* [70]. They studied the effects of –COOH, –CN and $–NO_2$ groups on the frontier molecular orbitals, energy gap, reorganisation energy, ionisation potential and electron affinity of $[Ni\text{-}(SB\text{-}38)_2]$, $[Cu\text{-}(SB\text{-}38)_2]$ and $[Zn\text{-}(SB\text{-}38)_2]$. They concluded that the complexes and their derivatives are promising candidates for optoelectronic devices [69]. Recent research has indicated that phenothiazine (PTZ)-based materials have potential as electron donors/acceptors in organic solar cells or as HTMs in OLEDs and perovskite solar cells. These applications are attributed to several desirable properties, including a low energy band gap, tunable energy levels and reversible redox properties. In this context, Revoju *et al.* [71] reviewed the optoelectronic properties of numerous phenothiazine (PTZ)-based materials for use in various devices, including organic solar cells (OSCs), dye-sensitised solar cells (DSSCs) [29-31,72-78], perovskite solar cells (PSCs) [63,79], OLEDs and batteries [80]. It was observed that the molecular properties and, consequently, device performance could be easily adjusted through minor modifications of the PTZ core or its conjugate backbone. Later, Huang *et al.* [81] reviewed multiple studies on modifying the optoelectronic properties of nitrogen-based conjugated molecules by adding boron-based Lewis acids, creating $B \leftarrow N$-bridged conjugated units and using them to develop OPV materials. Although these synthesised units may possess outstanding optoelectronic properties, they face challenges due to the Lewis acidity of boron, which makes them sensitive to moisture and limits their application in OPVs. Therefore, further research into the stability of these materials and devices involving B-N-embedded systems is necessary.

Alamro *et al.* [82] synthesised a series of liquid crystalline materials, focusing on the Schiff base derivatives with laterally positioned methoxy groups and terminal alkoxy chains of varying lengths (6-12 carbon atoms), specifically, (*E*)-3-methoxy-4-[(*p*-tolylimino)methyl]phenyl 4-alkoxybenzoates. It was observed that on illumination with white light, compound SB-39, with 8 carbon atoms in the alkyl chain, exhibited 5 times higher electrical conductivity *vis-à-vis* dark conditions. Further investigations revealed two direct optical band energy gaps, one in the UV and one in the visible light range. Moreover, compound SB-40, with a 6-carbon alkyl chain, has band energy gaps of 1.07 eV and 2.79 eV, which are desirable for various solar energy applications. The effect of electron donor groups (methyl) and electron acceptor groups (nitro), along with different solvent polarities, on the absorption spectra of Schiff bases synthesised from substituted thiophene carboxaldehyde/furaldehyde and 2-aminobenzamide (SB 41-44) was studied by Irfan *et al.* [83]. It was observed that when the $\text{-}NO_2$ group replaced the furan at the 5th position in compound SB-43, it resulted in a red shift. These results helped tailor Schiff base compounds for various organic electronic device applications, serving as n-type (compound SB-43), p-type (compound SB-44) and balanced hole and electron transport materials (compounds SB-41/SB-42) (Fig. 7).

Two polymorphs of anthracene SB-45 (N-(anthracen-9-ylmethylene)2,5-dichloroaniline) were obtained using the same solvent, hexane [84]. It was observed that Form I was long needle-shaped and elastically bendable and Form II was block-shaped and brittle, as depicted in Fig. 8. The difference in the mechanical properties was attributed to the nature of crystal packing. However, when their optical properties were studied, it was revealed that both forms have the same wavelength maxima (405 nm) and similar photoluminescence (PL) and photo-luminescence excitation (PLE) values. These results suggested that the optical properties were independent of the structure [84].

Upendranath *et al.* [85] synthesised and characterised two new SB 6-nitro-benzimidazole derivatives (SB-46 and SB-47). Good absorption properties, emission in the blue region and suitable chemical reactivity make them potential candidates for use in OLEDs, sensors and photosensitizers in third-gene-

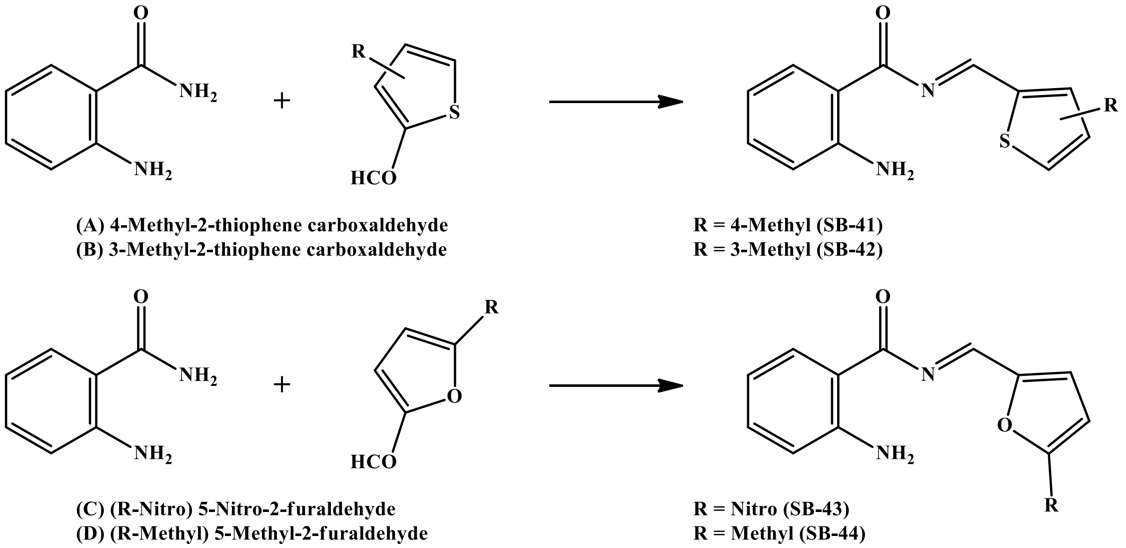


Fig. 7. Synthetic route of substituted thiophene carboxaldehyde/furaldehyde and 2-aminobenzamide-based Schiff bases (SBs 41-44) [83]

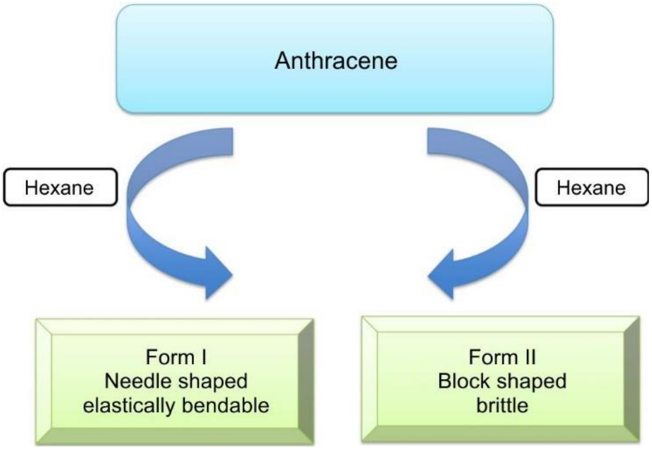
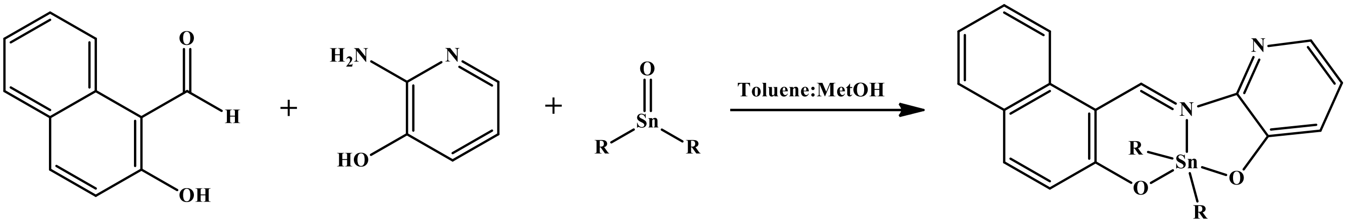


Fig. 8. Dimorphic forms of anthracene SB-45 crystals [84]

ration dye sensitised solar cells (DSSCs). They also exhibit good intramolecular charge transport properties and superior light stability [85]. Similarly, Tahmasbi *et al.* [86] reported a new series of Cu(II) complexes of the SB-48 ligand N,N′-(1,2-diphenyl ethane-1,2-diylidene)*bis*(3-nitrobenzohydrazide) (synthesised by the reaction between 3-nitrobenzohydrazide and benzil). Their absorption spectra displayed two electronic transitions at 355 nm and 432 nm, which are likely n→π* or π→π* transitions. The band gap energies of 2.67 eV and 2.89 eV were estimated for the SB-48 ligand and Cu(II) complex, respectively. These results corroborate the potential use of such Schiff base compounds for various nonlinear photonic, optical and optoelectronic applications, such as optical wavelength converters [86]. Furthermore, Sánchez-Vergara *et al.* [87] synthesised four penta-coordinated organotin(IV) complexes *via* one-pot synthesis using 2-hydroxy-1-naphthaldehyde, 2-amino-3-hydroxypyridine and organotin oxides. Hybrid films fabricated using organotin(IV) complexes with PEDOT: PSS and graphene exhibited increased charge mobility. Analysis of their mechanical and optical properties (Sn(IV)-SB 49a-d), revealed dependence on the nature of the additional ligands present in these complexes (Fig. 9).

Recently, inverted perovskite solar cells with a p-i-n configuration have been widely explored due to their simple configuration, minimal hysteresis, enhanced operational stability and low-temperature manufacturing process. However, their PCE needs further improvement compared to classical n-i-p perovskite solar cells. This can be achieved through inserting appropriate charge transport and buffer interlayers between the primary ETL and the top metal electrode [88]. In this context, Akbulatov *et al.* [88] examined how the long-term stability of these devices could be enhanced by using redox-active ligands such as Schiff base (salicylimine) or 2,3-dihydroxynaphthalene with tin complex and 2,3-dihydroxyphenazine with germanium complex. This improvement is attributed to surface recombination, which aligns the material energy levels across the interface and reduces interfacial losses. It was observed that redox-active ligands, when coordinated with tin, significantly lower the energy of LUMO levels while increasing the energy of the HUMO levels, resulting in a HOMO-LUMO gap of approximately 2 eV, which imparts semiconductor properties and fluorescence in the visible range. Chen *et al.* [89] noted that when the SB-50 (chloromethylene) dimethyliminium chloride (CDCl) was introduced between the TiO_2 electron transport layer and the methyl ammonium lead iodide ($MAPbI_3$) light absorber layer, the performance of printable mesoscopic perovskite solar cells (p-MPSCs) could be greatly enhanced. It was suggested that CDCl binds to the layers through coordination and hydrogen bonds, which helps improve crystallinity and effective defect passivation, thus suppressing non-radiative recombination. This also optimised the interfacial energy level alignment and promoted effective charge transfer. This combination resulted in a 19.74% PCE, the highest PCE reported so far for the printable carbon-based hole-conductor-free PSCs. The effect of a layer of isonicotinohydrazide and pyrene-based SB-51 (PyMIs) on the main electronic parameters of a Schottky diode, metal-semiconductor junction diode, was studied in the fabricated Al/PyMIs/*p*-Si/Al diode by Tezcan *et al.* [90]. The presence of an organic layer between the metal and semiconductor has been found to enhance the electrical and photoelectric characteristics. The results obtained for diode parameters, *viz.* saturation current (I_0), ideality factor (n) and barrier height (ϕ_b) for Al/PyMIs/*p*-Si/Al diode indicate its ability for diverse photoelectrical device applications.

To summarize, the Schiff base ligands and their metal complexes possess remarkable potential as key components for the manufacture of optoelectronic devices. The nature of the SB scaffold influences the band gap and thermal stability of OLEDs. The substituents on the Schiff base scaffold and the metal center to which they are coordinated also influence the band gap and thermal degradation of devices. For a given ligand scaffold, LUMO values change to a greater extent on



Sn(IV)-SB-49a: R = Bu
Sn(IV)-SB-49b: R = Cy
Sn(IV)-SB-49c: R = Ph
Sn(IV)-SB-49d: R = $CH_2Si(CH_3)_3$

Fig. 9. Organotin(IV) complexes of SB-49 with additional ligands (a-d) [63]

substitution. However, the same ligand scaffold in the presence of electron-donating substituents increases the HOMO value. A higher degree of conjugation enhances the thermal stability, which is apparent from the higher glass transition and melting temperatures and also resulted in emissions at higher wavelengths. Based on these findings, efforts are being made to decrease the conjugation. Increasing the alkyl chain length in zinc-complexed Schiff base ligands is an effective strategy for achieving blue emission, which is essential for realizing full white electroluminescence. The band gaps for the different metal-Schiff base complexes studied were found to be in the range of 2.5-3.5 eV for Zn-Schiff base, 3.0-4.0 eV for Sn-Schiff base and around 1.2 eV for Ln-Schiff base complexes.

Enhancement of PCE has also been the priority for OPV devices. Several strategies and combinations of metal- Schiff base complexes have been explored to improve the PCE and the device stability. Phthalocyanine and porphyrin-based Schiff base complexes of Cu had achieved a PCE of 8.07% due to well-matched energy levels, low surface roughness and highly crystalline nature. The effect of substituents on the energy gap has also been studied. These studies highlight that electron-withdrawing groups can decrease the energy gap between frontier molecular orbitals to overlap with the solar spectrum. The use of a suitable organic layer between the electron transport layer and light absorber layer in PSCs was found to optimize the interfacial energy level alignment and charge transfer. This resulted in the highest PCE of 19.74% achieved so far for printable carbon-based hole-conductor-free PSCs.

Most literature surveyed on metal-Schiff base complexes has primarily focused on the band energy gap. However, another fundamental parameter, the refractive index of these complexes, has received little attention. The transparency of a semiconducting material depends on the refractive index. The optical and electronic response of these materials is influenced by transparency. Data on thermal stabilities and melting points of most Schiff base complexes are missing.

Challenges and prospects: Over the years, there has been tremendous progress in the development of Schiff base based optoelectronic devices. Some challenges that have prevented the widespread application of Schiff bases in optoelectronics need to be addressed. One such challenge is the fabrication of monochromatic blue luminophores as active materials in optoelectronics. This is due to a broad electronic band gap in blue light-emitting materials. A broad electronic band gap leads to poor alignment of energy levels with adjacent layers in the device. Although a high operating voltage may be required, excessive voltage can induce material degradation, thereby reducing device lifetime and operational stability [91,92]. The development of red-emitting devices remains challenging, primarily due to the small band gap required for red emission [93,94]. Furthermore, the purity of the organic material critically influences the efficiency and overall performance of the device. Despite this, hardly any studies have demonstrated the influence of impurities and moisture [95,96]. Furthermore, the dissociation of Schiff base materials may yield products which might result in a voltage rise along with diminished device efficiency [97], however, some external factors, such as temperature, deteriorate the device's lifespan. An increase in temperature accelerates the degradation of optoelectronic devices and beyond a critical threshold, it can trigger device failure [98,99]. Despite extensive studies, a well-defined mechanism governing OLED degradation is still not fully understood [100,101]. Therefore, further systematic investigations are required to optimize the performance of Schiff base complexes for the optoelectronic applications.

With continuous efforts in this field, we anticipate more impact-driven innovations in optoelectronics. The emergence of flexible optoelectronic smart devices is already in its nascent phase. The mechanical resilience of the Schiff base-based elastic organic crystals makes them a desirable contender for application in flexible optoelectronic devices. Thus, their use in such devices is gaining widespread scientific attention. This makes us strongly believe that the future holds great potential for flexible organic micro/nano-crystals in the development of smart optoelectronic devices [102,103].

Apart from this, Schiff bases and its metal complexes are also finding applications in the emerging interdisciplinary field of 'bioelectronics' [104]. Bioelectronics involves designing of biocompatible electronic devices. This involves the use of biologically derived materials in electronics/optics. In this regard, several research studies have stipulated the antimicrobial and electronic functionality of Schiff base-based metal complexes [105,106]. However, their biocompatibility remains poorly understood and limited information is available on their mechanical properties. These aspects are crucial for their successful application in implantable systems [107]. Thus, comprehensive research integrating the electrical properties, biological activity, and mechanical reliability of Schiff base complexes is urgently required. In parallel, the incorporation of Schiff base complexes into biocompatible polymer matrices may significantly advance the development of hybrid, flexible and stimuli-responsive platforms compatible with biological tissues. Such progress could facilitate the realization of wearable diagnostic systems, implantable sensors and smart therapeutic devices, ultimately enabling the fabrication of more efficient and environmentally sustainable optoelectronic devices.

ACKNOWLEDGEMENTS

The authors are sincerely grateful to The Principal, St. Stephen's College, Delhi, India, for support of this work.

CONFLICT OF INTEREST

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

DECLARATION OF AI-ASSISTED TECHNOLOGIES

During the preparation of this manuscript, the authors used an AI-assisted tool(s) to improve the language. The authors reviewed and edited the content and take full responsibility for the published work.

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