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A Brief Updates on Nitrogen (N)-, Oxygen (O)-, and Sulfur (S)-Based Heterocycles Containing Advanced Materials in Next-Generation Energy Conversion and Storage Devices

Bhavesh S. Hirani[1,*], Sevak B. Gurubaxani[2]

Abstract

Subsequently, the market for wearable and portable electronics has expanded significantly due to the development of multifunctional energy generation and storage systems that can be folded, twisted, and reshaped in a variety of ways while maintaining their electrochemical efficiency. This progress has been fueled by the latest advancements in materials science and well-established techniques for constructing state-of-the-art organic semiconductors, heterocyclic solids, and flexible energy-generating and storage systems. Flexible photoelectrochemical cells and supercapacitors are now made possible by improving the optical, electrochemical, and physical characteristics of materials, opening the door to innovative energy solutions. As researchers gain deeper insights into the multifunctional properties of various materials, they have been able to tailor them for a wide range of flexible energy devices, making them more efficient and practical. These advancements extend to a variety of materials with diverse properties such as ionic conductivity, exceptional thermal stability, light harvesting capability, and non-toxicity. Notably, the design of these materials requires careful consideration of their scalability, sustainability, and overall performance in real-world applications. We will also describe some appealing examples that demonstrate the multifunctionality of these materials, showcasing their potential in flexible devices. Furthermore, we emphasize the importance of designing effective materials and their integration into next-generation flexible energy generation and storage systems. Looking ahead, flexible and wearable energy-conversion and storage technologies are expected to play a pivotal role in shaping future-oriented advancements, offering innovative solutions to the growing demand for portable, efficient, and sustainable energy sources.

Keywords: N/O/S based heterocycle, organic ionic conductors, energy-conversion and storage devices, non-toxicity

INTRODUCTION

Fossil fuel burning is the world's main energy source. Due to the depletion of fossil fuels, efficient, economical, ecologically friendly, clean and renewable energy generation, and storage solutions have been developed [1]. The development of sustainable energy-generating and storage systems for a range of uses, from grid-scale electricity to small portable devices, has advanced significantly during the past 20 years [2, 3]. It is important to create energy generation and storage systems that are also flexible and portable, so they can keep working well even when they are bent or stretched, to support flexible electronics [4]. High-performance electronics materials as flexible energy generation and storage devices available in the market are described in Figure 1, such as dye-sensitized photoelectrochemical cells, organic–

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inorganic hybrid perovskite solar cells, organic solar cells, fuel cells, supercapacitors, and metal–ion batteries, have thus been the subject of numerous attempts. Since they are inexpensive, lightweight, highly flexible, easy to assemble, and environmentally friendly, photoelectrochemical, and supercapacitor devices have been thoroughly studied for genuine, real-world applications [5]. Electrolytes are crucial in electrochemical devices, maintaining charge and facilitating ion or charge transfer between positive and negative cathodes. Efforts have been made to create effective electrode and electrolyte materials for high efficiency, flexibility, and lifespan [6]. This chapter provides information on identifying novel multifunctional electrolytes with superior performance for flexible energy-generating and storage systems. Despite advancements, challenges, such as volatilization, leakage, photodegradation, electrode corrosion, and insufficient device sealing for extended outdoor usage persist, especially when liquid electrolytes are used in high-performance systems [7].

Figure 1. High-performance electronics materials, flexibles energy generation, and storage devices available in the market.

Flexible energy-generating devices use various techniques, including gelling agents, organic or inorganic hole conductors, solvent-free polymer electrolytes, and others. However, these alternative electrolytes often declines in performance due to inadequate pore-filling and decreased charge mobility in the solid or quasi-solid media [8]. For example, DS-PECs with polymer gel electrolyte (PGE) often do not see a gain in photoconversion efficiency (PCE) of higher than 8%. Despite their superior sealing ability and increased durability, these DS-PECs can outperform liquid electrolyte-containing Gratzel photoelectrochemical cells even at low performance levels. Flexible energy storage devices made of polymer hydrogel have disadvantages such as a small operating voltage window, poor electrode pore penetration, reduced energy density, and poor electrode/electrolyte interface contact [9]. Strong mechanical strains can cause the electrolyte or electrode interface to break, making solid-state electrolytes limited in practical applications. In recent years, researchers have been exploring the possibility of improving electrochemical performance through the creation of innovative, multipurpose, and eco-friendly electrolytes. Solid-state ionic conductors, also known as solid electrolytes, are materials that conduct electricity through ion movement [10]. Li+ conductors are essential for lithium–ion batteries and are categorized based on the type of ion they conduct. H+ conductors are used in fuel cells and other electrochemical devices, and materials with flaws in their crystal structure are known as mixed ionic–electronic conductors [11].

Solid-state electrolytes, such as PGE, quasi-solid-state electrolyte (QSSE), and hole transport materials, have been extensively studied in the field of flexible energy generation and storage. Heterocyclic solid-state organic ionic conductors (SOICs) are a promising class of SOICs for creating high-efficiency ss-DSSCs. SOICs offer substitute materials for flexible energy devices, addressing electrolyte-related problems [12]. They have superior electrochemical characteristics over traditional electrolytes. Imidazole iodide (IMI) salts are the most promising class for creating high-efficiency ss-DSSCs. Researchers have improved the performance of IMI-based electrolytes by replacing alkyl chains or adding bulky moieties. For example, replacing an ester group on IMI improved performance [13]. The POZ/PTZ-substituted IMI-based SOICs have outstanding conductivity, hole mobility, and thermal stability (Figure 2 (1,2)). Substituting donor moieties improved short-circuit current density and electron-transport characteristics in 14 SS-DSSCs, achieving a PCE of 5.5%. Di-cationic SOICs-based on pyrrolidinium and IMI salts have shown over 6.0% efficiency, indicating that increasing ionic sites on the moiety enhances ion transport, leading to improved conductivity. This suggests that more research is needed to create more effective SOICs with efficient substituents in traditional champion moieties, offering improved light harvesting, hole mobility, and redox-mediating capabilities [14]. The SOICs were originally presented and connected by an octyl chain between donor moieties. The bulkier photosensitive substitution on BIMI enhanced photo-generation ability and conductivity through bulkier cations. These SOICs absorb the visible region spectrum, supporting the enhancement of photo-generation for champion metal-free sensitizer SK3 (Figure 2 (3)) [15].

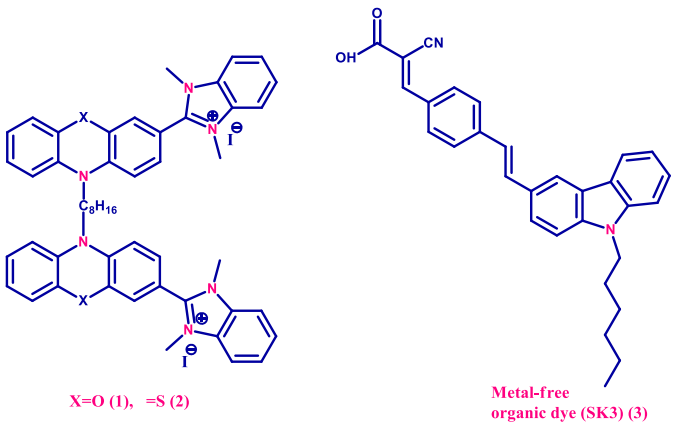


Figure 2. General structures of phenoxazine (POZ)/phenothiazines (PTZ)-substituted IMI-based (1,2) and SK3 (3) SOICs.

This report analyzes the functions of heterocyclic-based SOICs in energy generation and storage systems. It discusses the impact of functional groups and additives on SOICs' electrochemical characteristics and flexible energy device functionality [16]. The report highlights the development of environmentally friendly, cost-effective, high-performing, and long-term stable flexible devices due to the single-component and multifunctional properties of SOICs, leading to their commercialization.

MECHANISMS OF CONDUCTION

Ionic conductivity in a material is dependent on the transport of ion types through the material. In ideal crystals, ions are arranged in a regular pattern and often stacked closely together, limiting their dispersion and vibration around the equilibrium position [17]. However, flaws can occur at any temperature, such as positional disorder, caused by deviation from optimal stacking. The severity of this condition can vary between materials and within the same material, depending on temperature or pressure. At zero temperature, the ion configuration in an ideal crystal is at its lowest potential energy. As the temperature rises, the contributions of entropy to free energy become more noticeable, leading to crystal defects due to the system's attempt to reduce free energy by raising entropy [18]. Schottky and Frenkel flaws are two significant categories of defects in crystals, which are considered "point defects." Schottky defects occur when two ions, one a cation and the other an anion, disappear, leaving their locations empty. Frenkel defects occur when a single ion wanders in interstitial spaces, which differ from normal sites in terms of their quantity or type. Although interstitial spaces are not energetically favorable for ions, entropy boosts drive their occupancy. An ion nearby can jump to empty sites created by Frenkel and Schottky defects, leaving gaps in the crystal. This frees up the ion's former location, which could now accommodate another ion. Conductivity may result from the movement of ions through the solid due to this action, known as "vacancy migration." Interstitial migration is another process, where an ion moves over great distances due to vacancy migration [19]. The interstitial mechanism is another mechanism that occurs when two or more ions move cooperatively. The locations of sites and interstitials in crystals where this process occurs are such that one or more neighboring ions must be pushed elsewhere for an ion to hop to a neighboring boring site or interstitial [20].

MOLECULAR ARCHITECTURE OF SOIC

SOICs are a new class of solid-state electrolytes that contain various cations and anions such as quaternary ammonium, phosphonium, imidazolium, pyridinium, and others [21]. These solid-state variants of ionic liquids have unique properties such as solidity, chemical durability, low toxicity, and electrochemical and thermodynamic stability. They tend to outperform ionic fluidity and exhibit superior thermal and electrochemical characteristics. SOICs can be created and altered to meet the needs of various electrochemical device types by substituting various functional groups. Some examples of SOICs include ester-functionalized imidazolium (IM) cations, alkyl chain length, non-aromatic pyrrolidinium dictations conjugated with various alkyl chains, carbazole groups, and hetero-anthracene derivatives [22]. These modifications can improve electrical, optical, and physical characteristics, conductivity, thermal stability, and selective wavelength absorption. SOICs are crucial for the development of effective, adaptable, and flexible energy-producing systems. However, very few SOICs are employed in the energy storage industry, and research on SOICs is still in its early stages [23]. SOICs can meet special requirements, like cation size, electron transfer rate, mechanical qualities, long-term cyclic stability, and ionic conductivity, by substituting tiny heterocyclic components and functional groups. The choice of electrolyte material for organic electrodes in energy storage devices typically does not restrict SOICs [24].

FLEXIBLE ENERGY GENERATION DEVICES

The future of cutting-edge technology, solar cells are a kind of renewable electricity-generating device with exceptional potential for large-scale solar power production [25]. Here, we examine four families of flexible third-generation solar cell technologies, DS-PECs, Bio-PECs, OIHPSCs, and OSCs, and talk about how well they work with different multifunctional SOICs [26].

Structural and Operating Mechanism of Flexible DS-PECs

DS-PECs have been extensively investigated as an alternative renewable energy source due to their advantages, such as low manufacturing costs, easy fabrication in environmental conditions, feedstock

availability, color and transparency, good performance in low light intensity, and a recyclable roll-to-roll coating process, for the TiO_2-photoanode. DS-PECs consist of four distinct parts: an n-type semiconductor (TiO_2), light-absorbing materials (metal and metal-free dye), an electrolyte, and a platinum back electrode [27]. The electrolyte significantly influences the longevity of DS-PECs and photovoltaic performance. The interaction between the electrolyte and photoanode surfaces significantly impacts photovoltaic parameters, including fill factor (FF), open-circuit voltage (Voc), and short-circuit current density (Jsc). An electrolyte with an iodide (I-)/triiodide (I3-) redox pair is necessary for DS-PECs, as it serves as a mediator, transfers charges from the anode to the cathode, and regenerates oxidized sensitizer at the TiO_2-photoanode/electrolyte interface. The greatest PCE of 12.8% has been reported using DS-PECs-based on a liquid (I-)/triiodide (I3-) redox pair and metal-free sensitizer. However, their long-term stability has been limited by counter electrode corrosion, light-absorbing material deterioration, and solvent leakage and volatilization. Extensive research has been conducted to develop substitute solid-state electrolytes [28, 29].

Role of SOICs in Flexible DS-PECs

DS-PECs have been developed using various changes and electrolyte types, resulting in classifications of all redox pair electrolytes into LSEs, QSSEs, and SSEs based on their physical states. Liquid state electrolytes, which contain iodide (I-) and iodine (I2) salts, have gained popularity due to their rapid dye regeneration, strong ionic conductivity, superior redox pair stability, ease of penetration into the TiO_2 matrix, and reduced light absorption. Other redox couple electrolytes include Co (II/III), Br-/Br3-, Fe/Fe+, SCN-/(SCN-3), Cu(I/II), and Ni (III/IV). DS-PECs were assembled using sensitizers based on porphyrins, with sensitizers working long with the cobalt redox shuttle having PCE values of 11% and 12%, respectively [30]. Organic solvents with higher dielectric constant and lower viscosity, such as N-methyl pyrrolidone (NMP), acetonitrile (ACN), Valero nitrile, 3-methoxy acetonitrile, ethylene carbonate, propylene carbonate (PC), propionitrile (PPN), and g-butyrolactone (GBL), are also present in these liquid redox couple electrolytes.

However, the presence of inorganic ionic salts and organic solvents in DS-PECs results in remarkable efficiency but also brings limitations, such as leakage, corrosiveness of iodine, dye degradation, when in constant contact with the liquid electrolyte, and the potential for triiodide ions to transform into polyiodide chains over time [31]. Constructing liquid electrolyte-based DS-PECs on a wide scale is challenging due to electrically connecting and chemically assembling each module in a single substrate. Water as an electrolyte could be less expensive and safer for the environment. Ionic conductors based on organic moieties are essential for solid-state DS-PECs due to their high ionic conductivity, improved thermal stability, and quick solidification [32]. Heterocyclic cations derived from the IM ring are the most used heterocyclic cations for solid-state DS-PECs. IM, a non-aromatic compound, offers exceptional electrochemical stability due to its high conductivity, non-toxicity, and low vapor pressure. Several imidazolium iodide-based ILs, such as DMPII, EMII, PMII, MPII, and BMIMI with various alkyl substituents, have been used in DS-PECs due to their high conductivity and low vapor pressure. However, due to their viscous nature, leakage is inevitable. To use flexible DS-PECs effectively, a solid-state electrolyte is required, as leakage occurs when using them for extended periods [33, 34].

Alkyl-substituted Imidazole/Thiazole/Pyrrolidine-based SOICs

Electrochemical characteristics have the potential to provide rapid charge transfer and superior ionic conductivity. These SOICs, based on ester (–COOR) and functionalized IM cations of various alkyl chain lengths, offer several benefits, including rapid charge transfer, superior ionic conductivity, and sufficient ionic mobility [35]. The performance of solid-state DS-PECs is significantly influenced by electrolyte ionic conductivity. The short alkyl side chain-substituted COOR functionalized 3D ionic channel indicates good ionic mobility and conductivity without adding any chemicals. Conductivity significantly increases when supporting additives, such as iodide and lithium iodide, are added to (4). The Grotthuss exchange process allows charge transfer across an iodide chain as it is formed [36]. The solidification, conductivity, and thermal stability of SOICs depend heavily on the length of the alkyl chain. Extending the alkyl chain lowers the strong electrostatic interaction caused by small alkyl chain

substitutions in SOICs, while raising the melting point from (5) to (6) enhances the strong van der Waals force associated with long alkyl chain substitutions in SOICs. In the porous TiO_2-photoanode, the short alkyl chain of SOIC improves pore-filling, promoting the regeneration of oxidized dye molecules and producing a strong photocurrent response in DS-PECs [37]. The addition of the –COOR functional group to the IM ring greatly increased the efficiency of solid-state DS-PECs. Ionic crystals, made up of cations and anions with distinct electrochemical characteristics, have been used to demonstrate the presence of 3D ionic channels for I-. Heterocyclic moiety (IM)-based ILs (173) have shown remarkable performance in solid-state DS-PECs, with SOICs conjugated with thienyl and phenyl aromatic groups showing the highest PCE of 9.26% [38].

However, the study into the conductive process and how it works are limited because the majority of the electrolytes listed above are only compatible with IMI-based SOICs. This instance involves the synthesis of a number of distinct alkyl-substituted benzothiazole and thiazole dicationic SOICs (14, 18). Such a big size dications boost Voc by reducing electron recombination at the interface between the dye-sensitized TiO_2-photoanode and SOIC. Benzothiazole SOICs' ionic conductivity increases gradually when the alkyl chain is changed from propane to pentane (s = 0.215–0.444 mS cm^{-1}), but it quickly decreases when the hexane chain is changed [39]. The diffusion coefficient (Dapp) of the iodide–triiodide redox pair showed a similar pattern: it has the greatest triiodide Dapp, measuring 2.05×10^{-6} cm^2 S^{-1}. In contrast to the photovoltaic performance of various substituted dications thiazole-based SOICs, which translate to a PCE value of 7.90%. For solid state DS-PECs, another important metric for DS-PEC performance is Voc, which is likewise lower than that of DS-PECs-based on liquid electrolytes. Different alkyl chain substituted bis(pyrrolidinium) cations (alkyl chains such –C4H8–, –C5H10–, –C6H12–, and –C8H16–) and anions (SCN^-, $TFSI^-$, and Br^-)-based SOICs have been synthesized (19, 27) in order to improve the Voc and FF of solid-state DS-PECs. In comparison to DS-PECs reported with other pyrrolidinium-based SOICs, the photovoltaic performance of SOIC-24-based DS-PECs showed the greatest J_{sc} of 12.3 mA cm^{-2}, Voc of 810 mV, and FF of 0.60%, resulting in an overall PCE of 6.02% upon illumination of 100 mW cm^{-2}. Additionally, DS-PECs exhibit high ionic conductivity, melting point, and diffusion coefficient along with outstanding thermal stability [40]. Similarly, authors reported more dicationic SOICs (28, 29) based on pyrazolium cation and thiocyanate anion. Except for the nitrogen atoms' differing locations, the pyrazolium-based cation appears to share an aromatic heterocyclic skeleton with the IM cation. Excellent ionic conductivity and distinctive electrochemical properties that make them appealing for use as an electrolyte in DS-PECs are characteristics of derivatives based on pyrazole. The DS-PEC-based on bis-pyrazolium thiocyanate (Figure 3). produced an overall PCE of 7.30% with a J_{sc} of 11.7 mA cm^{-2} [41].

Pyridinium-Based SOICs

Conventional IMI-based SOICs and pyridinium-based SOICs can coexist. Compared to the IMI series of SOICs, they are less expensive and simpler to synthesize. Numerous pyridine derivatives, including 2-methylpyridine, 4-tert-pyridine, 4-tert-butyl pyridine, 2-methyl-4-propoxypyridine, and others, have been employed as electrolyte additives to raise the TiO_2 semiconductor's band edge (Fermi level) and investigate the cationic influence on the TiO_2-photoanode depending on the alkyl chain's length. In the sequence (30–36), the Voc and PCE steadily dropped when the $–CH_3$ group was substituted on a different location of the pyridinium SOIC (PCE of 7.50%) (37) [42]. Low toxicity, high ionic conductivity, a cationic effect, environmental sustainability, superior catalytic properties on a platinum electrode, and high stability in an aqueous medium are all demonstrated by the new electron-withdrawing chalcone functional group-substituted pyridinium cation with iodide and bromide anions-based SOICs (38). Figure 4 shows structure of pyridinium-based SOICs. Overall, PCE retention was 61.6% (PCE = 5.61% decreased to 3.46%) and 59.3% (PCE = 6.02% decreased to 3.57%) for the J–V characteristic of DS-PECs with water. Additionally, they used theoretical calculations to examine the impact of the chalcone group and solvent (acetonitrile and water) on pyridinium SOICs that include nitrogen cation atoms and redox potential [43] as well as the recombination rate. It is transparent throughout a wide spectrum of wavelengths and extremely oxidation-prone. It would be more beneficial to create inexpensive and extremely stable DS-PECs based on the aforementioned characteristics. Have

created several SOICs based on pyridinium that have alkyl (odd/even carbon chains) replaced. According to the conclusions of density functional theory (DFT) and time-dependent DFT, destabilization occurs when the cationic charge increases more in an acetonitrile solvent than in an aqueous environment. In comparison to pyridinium iodide (without a chalcone group), the redox potential of chalcone group-functionalized pyridinium SOICs rapidly regenerates oxidized dye and approaches the dye's ground-level state. Single-component SOICs that provided PCE of 0.9% and 0.7% made it possible for aqueous DS-PECs to function photovoltaically [44].

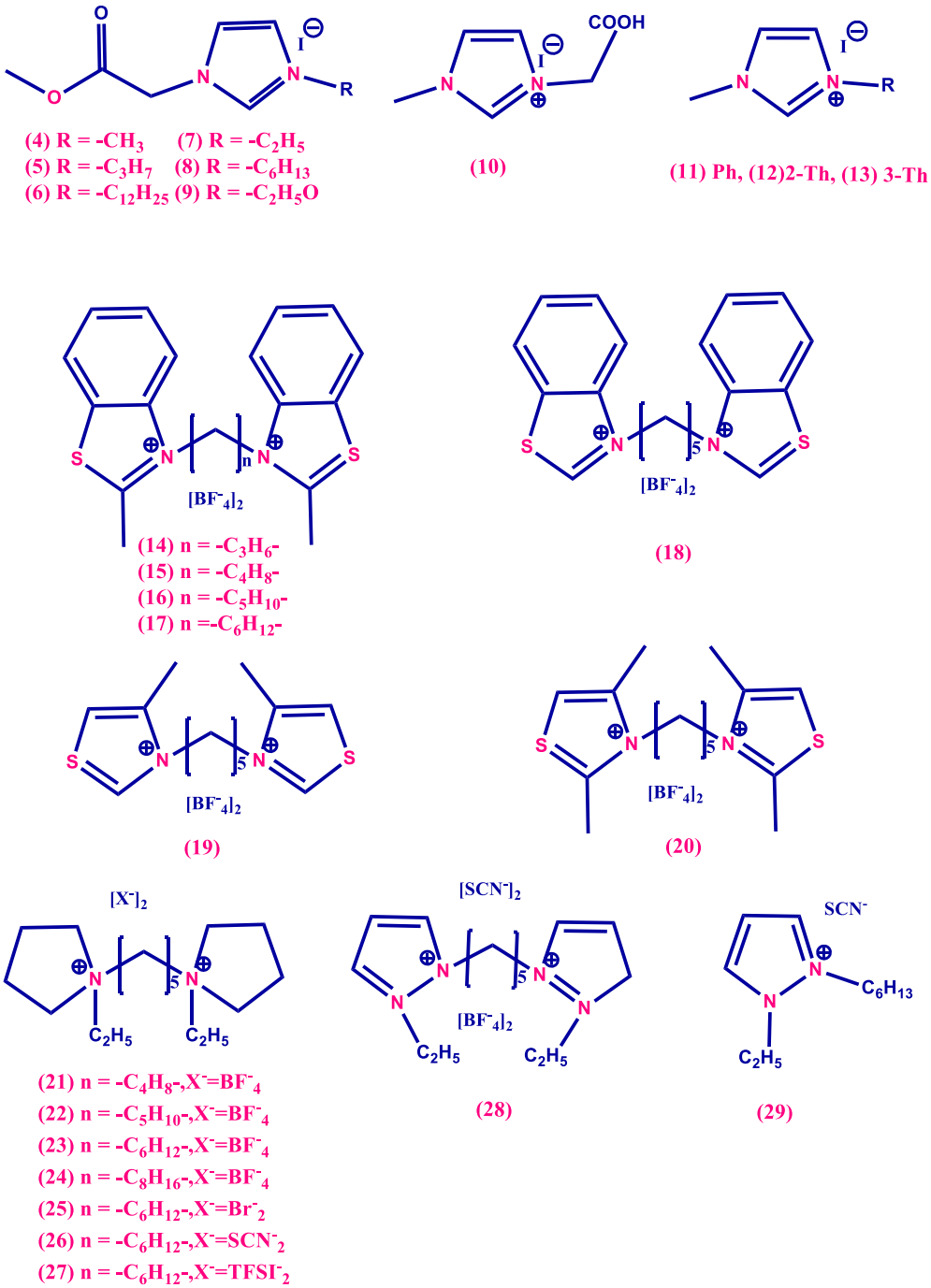
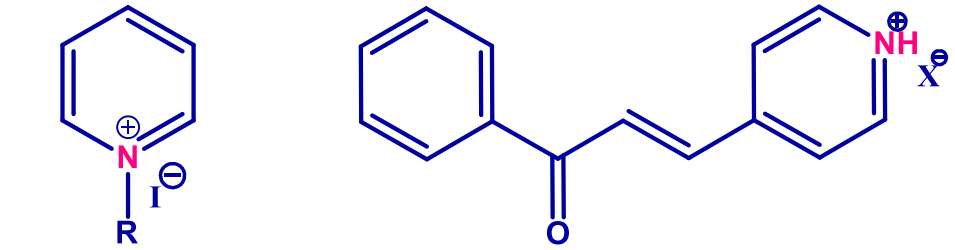


Figure 3. Structure of alkyl-substituted imidazole/thiazole/pyrrolidine-based SOICs.



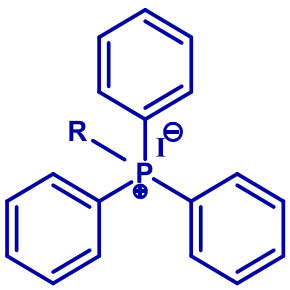
(30) R= C_2H_5
(31) R= C_4H_9
(32) R= C_5H_{11}
(33) R= C_6H_{13}
(34) R= C_7H_{15}
(35) R= C_8H_{17}
(36) R= C_9H_{19}

(37) X= I^-
(38) X= Br^-

Figure 4. Structure of pyridinium-based SOICs.

Phosphonium-Based SOICs

As previously mentioned, phosphonium-based conductors have received relatively little research focus, even though nitrogen-based SOICs, including benzothiazolium, pyrazolium, imidazolium, pyrrolidium, pyridinium, and piperidinium salts, have received a great deal of attention [45, 46]. Even though solid conductor-based phosphonium cations are intelligent and capable of becoming more durable and, going forward, having a longer life-span compared to ammonium cations, they have up until now been disregarded and given less attention in the application of energy-generating devices (particularly in DS-PECs). According to the paradigm, several ILs with minor alkyl chain substitutions on phosphonium cations and other anions produced electrochemical stability and staggered conductivity. Furthermore, ILs based on the phosphonium cation have superior ionic conductivity, low viscosity, and great thermal stability when compared to the quaternary ammonium counterpart. In 2016, authors created DS-PECs with phosphonium iodide based on ILs as the electrolyte. Under simulated light illumination, these devices show the highest PCE of 5.7% [47]. All of them, nevertheless, are still viscous liquids (or ionic liquids), and prolonged usage may cause leaks. Due to recent attempts to integrate a phosphonium-based solid-state electrolyte into DS-PECs, two phosphonium-based SOICs (39–40) have been reported. Figure 5 describes structure of pyridinium-based SOICs. LiI-doped solid-state DS-PECs have the following resulting J–V characteristics: PCE of 6.7%, and PCE of 2.4% [48, 49].

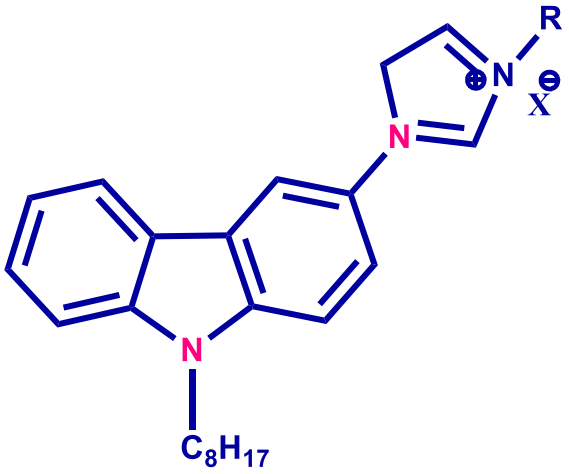


(39) R= H
(40) R= CH_3

Figure 5. Structure of pyridinium-based SOICs.

Carbazole-Based SOICs

Due to their superior charge mobility and straightforward fabrication process, organic hole conductors (OHCs) exhibit intriguing functionality and are viable substitutes for LSEs and ionic polymer electrolytes. Because of their better hole conductivity and purity, small OHCs based on tetraphenyl benzidines (TPB) and triphenylamines (TPA) have found widespread usage as a hole transport layer (HTL) [50]. The highly soluble and amorphous Spiro-OMeTAD is produced by substituting TPB with 2,20,7,70-tetrabromo-9,90-spirobifluorene. It is supplied at more than 6% PCE in solid state DS-PEC. The oxidized dye regenerates by utilizing the highest occupied molecular orbital (HOMO) electron of OHC and charge transport via a hopping process; however, Spiro-OMeTAD lacks any redox species (iodide–triiodide). Likewise, through the dual-functional channel, SOICs based on carbazole (CBZ)-IMI efficiently regenerate photo-excited dye molecules (41–43). Figure 6 describes structure of carbazole-based SOICs [51]. Since numerous polymeric CBZ derivatives have been employed as HTL in high-performance solid-state DS-PECs, CBZ was chosen as the OHC. Dual-functional channels for the hole and I_3^- transport are produced by the combination of the CBZ-IM cation and I^- anion. Under 100 mW cm^{-2} of illumination, the related solid-state DS-PEC had the greatest PCE of 2.85% [52].



(41) R = -C_6H_{13}, X^-= SCN^-

(42) R = -CH_3, X^-=I^-

(43) R = -C_6H_{13}, X^-=I^-

Figure 6. Structure of carbazole-based SOICs.

Hetero-Anthracene-Based SOICs

Previous literature published several hetero-anthracene donor moieties (i.e., phenothiazine (PTZ) and phenoxazine (POZ))-functionalized IMI and benzimidazolium iodide (BIMI)-based SOICs (44–47) in the context of the dual-functional (hole/I_3^-) SOICs investigation mentioned above. These SOICs have superior ionic conductivity, s = 2.94 mS cm^{-1}, s = 3.61 mS cm^{-1}, as well as improved thermal stability (4150°C) and melting temperatures (4100 °C). Compared to the previously mentioned alkyl-substituted IMI-based SOICs, the high ionic conductivity in SOICs allows charge transport from the densely packed polyiodides chain via the Grotthuss bond exchange mechanism, which is ten times larger. But because POZ has a better electron-donating capacity and a smaller interlayer distance, POZ-BIMI and POZ-IMI have better catalytic properties [53]. Hetero-anthracene moieties' highly conjugated and electron-donating characteristics also produce light-harvesting capabilities. The layer-by-layer thin film blend using POZ-substituted-SOIC materials and CBZ dye-sensitized TiO_2-photoanode showed an improvement in the absorbance spectrum of about 65 nm, resulting in increased efficiency in solid-state DS-PECs. Solid-state DS-PECs based on POZ-BIMI and POZ-IMI cations were carried out with a

maximum PCE of 5.3% and 5.7% in the presence of a single component. Additionally, they showed that under constant solar illumination, POZ-IM-based solid-state DS-PEC may remain stable for 41,000 hours [54]. Later, the same was applied to DS-PECs using di-anchoring carbazole-based dye. In liquid and solid-state electrolytes, it demonstrated overall PCEs of 5.20% and 4.02%. The SOICs based on hetero-anthracene functionalized BIMI cations enable photo-generation, harvesting capacity, and ionic conductivity. They are joined by a lengthy alkyl chain between two identical donor moieties (48–49). With their improved I^- diffusion/hole transport feature, the visible and ultraviolet spectrum absorbed by SOICs helps to improve dye photo-generation and serves a dual purpose in solid-state DS-PECs. More study is required to create possible SOICs with the appropriate substituent that might offer high PCE in solid-state DS-PECs, ionic conductivity, redox mediator activity, and remarkable light-harvesting capabilities considering the previously mentioned discoveries. Photo-generation, harvesting, and ionic conductivity are made possible by SOICs based on hetero-anthracene functionalized BIMI cations. They have two identical donor moieties connected by a long alkyl chain [55]. The visible and ultraviolet spectrum absorbed by SOICs contributes to enhanced dye photo-generation and serves a dual function in solid-state DS-PECs due to their enhanced I– diffusion/hole transport characteristic. The greatest PCE values of 7.9% and 6.8%. Figure 7 shows structure of hetero-anthracene-based SOICs.

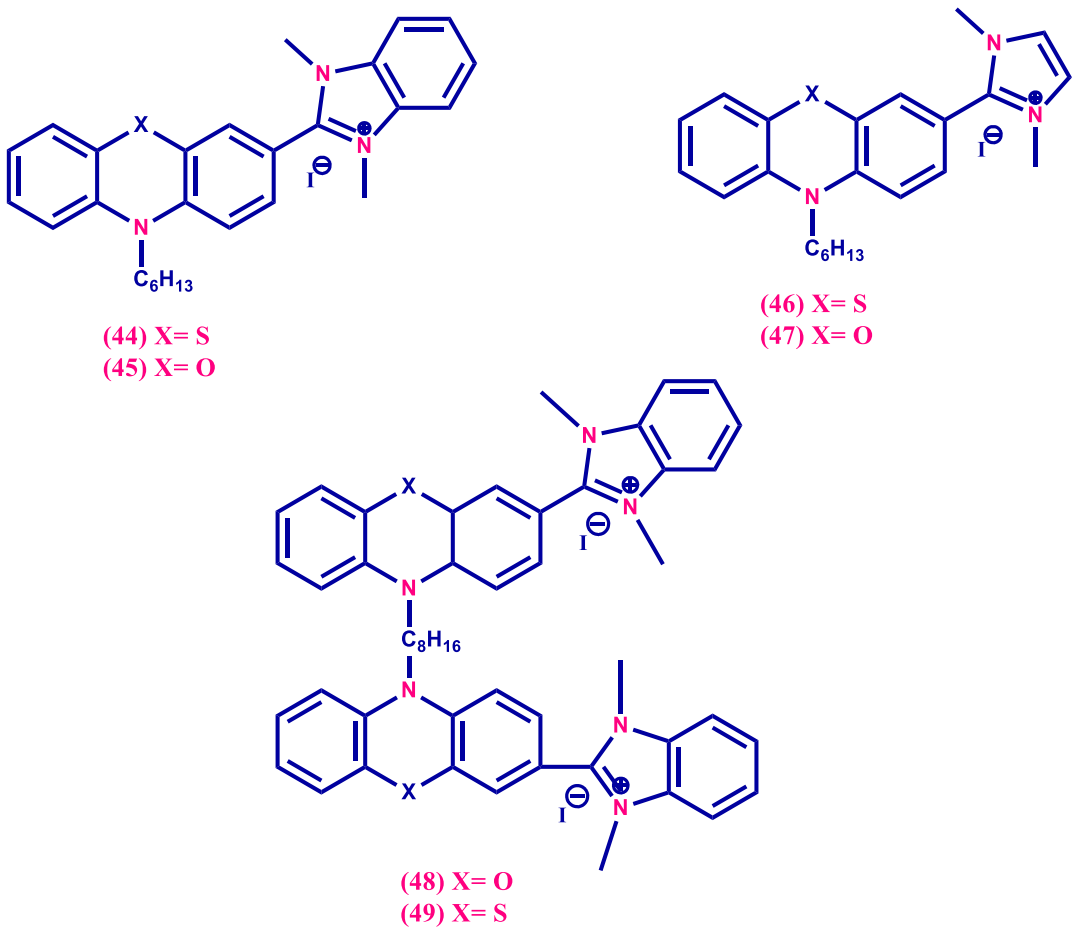


Figure 7. Structure of hetero-anthracene-based SOICs.

Two potential energy harvesting processes for solid-state DS-PECs were found by the researchers: (1) An electron from a photo-excited donor moiety moves straight to the TiO_2 conduction band, and (2) the FRET mechanism transfers the energy from the donor moiety to the dye molecule. In conclusion, solid-state DS-PECs can perform better overall and enhance their electron transfer kinetics and light-absorbing capacity by substituting SOIC for strong electron-donating moieties [56].

Non-Aromatic-Based SOICs

Imidazolium, pyrazolium, benzothiazolium, pyrrolidium, and thiazolium-based SOICs (50–52) are viable options for solid-state DS-PEC applications, as was previously mentioned. Such SOICs often have reduced ionic conductivity, which makes solid-state DS-PECs based on them unfavorable and incapable of operating well in the absence of additives. However, adding I_2 and LiI to SOICs improves both the efficiency of solid-state DS-PECs and their ionic conductivity. Additives or multi-component SOICs will raise uncertainty, which is undesirable for production scaling up. High-performance OIHPSCs based on organic hole transport materials (OHTMs) also require additives. To improve OIHPSC performance, for example, additives, such as $LiN(CF_3SO_2)_2$, Li-TFSI, $N(PhBr)_3$-$SbCl_6$, and Co complex (FK102, Co (III)-TFSI), were added to OHTMs to increase conductivity. A photovoltaic device's stability may be impacted by unknown side reactions, and the inclusion of multiple components adds complexity and raises the device's cost, among other disadvantages [57]. High-performance, reliable solid-state DS-PECs heavily depend on the design of highly conductive single-component SOICs. Thus, for the use of solid-state DS-PECs, the authors produced pendant propargyl-functionalized piperidinium and IM-based SOICs (50–52). This SOIC uses the attraction of hydrogen (H) bonds and Coulomb forces to distribute the 1D lamellar packing of anions (iodide) and cations (piperidinium or IM). Because piperidinium-based SOIC had greater crystallinity and a smaller interlayer (7.6 Å) distance than IM-based SOIC (9.2 Å), its ionic conductivity was higher for (s = 40 mS cm^{-1}) than for (s = 9 mS cm^{-1}). Additionally, it was noted that the IPCE for piperidinium-based SOIC exceeded 65–70% in the 380–450 nm region and reached 77% at 500 nm [58]. According to the researchers, iodine and a substituent (propargyl group) are essential for creating high-performance SOICs with superior ionic conductivity in long-range one-dimensional (1D) lamellar crystallites. Additionally, for use in solid-state DS-PECs, a non-aromatic cyclic piperidine iodide-based SOIC in Figure 8, devoid of any substituted functional groups, is created. To produce a 1D lamellar phase with a small interlayer distance and superior ionic conductivity, a substituted piperidinium SOIC was incorporated. To encourage a self-constructed 1D lamellar structure using H-bonding and Coloumb forces, the researchers looked at the SOIC with a small amount of I_2. Single-component SOICs' reported ionic conductivity of 4.20 mS cm^{-2} is higher than that of commonly used conventional and IL-based electrolytes. Some authors announced a new non-aromatic SOIC based on pyrrolidium iodide (53–54). Iodide, succinonitrile were combined in different molar ratios to create this solid-state electrolyte. Solid-state DS-PEC photovoltaic characteristics using the electrolyte demonstrated an overall PCE of 6.7% [59].

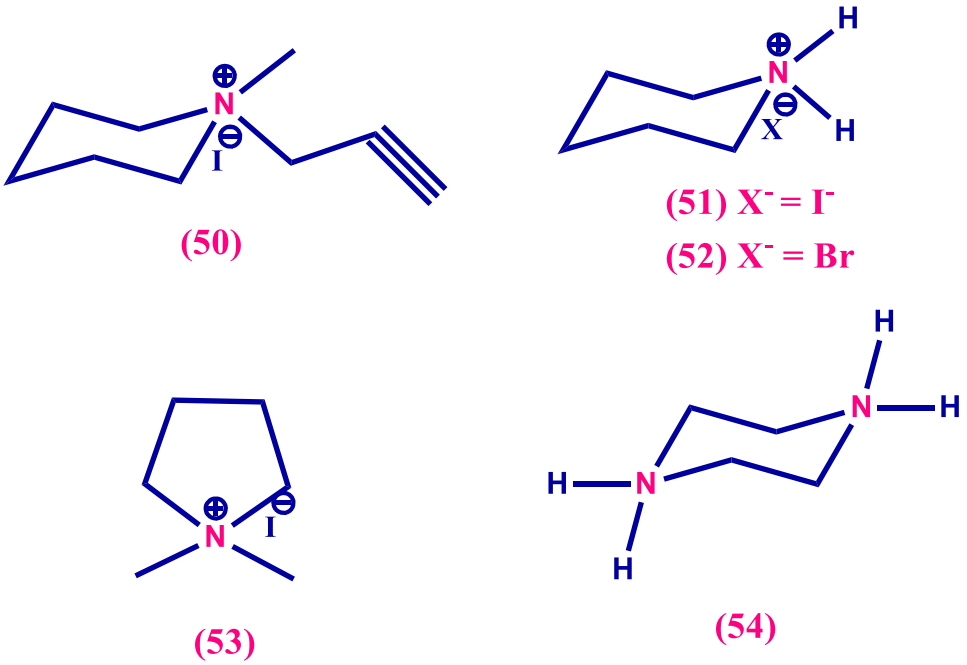


Figure 8. Structure of non-aromatic-based SOICs.

Flexible Bio-Photoelectrochemical Cells (Bio-PECs)

The application of photosynthetic pigment–protein complexes for non-renewable resources and sustainable technologies, like adaptable Bio-PECs, is gaining attention. Light-sensitive biomolecules, such as the reaction center complex (iron or sulfur) of pigments, proteins containing chlorophylls, and other cofactors, perform light absorption and charge separation in flexible bio-systems [60]. Reaction centers (RCs) or RC/light-harvesting antennas (RC-LH1) from the bacteria Rhodobacter (Rba.) sphaeroides are present in most protein complexes used to form protein solar cells. Numerous investigations into Rba. Sphaeroides' RC has confirmed its possible use in flexible Bio-PECs. With a 100% quantum efficiency, RC is the most efficient light energy conversion system found in nature [61].

Role of SOICs in Flexible Bio-Photoelectrochemical Cells

In flexible Bio-PECs, the design and synthesis of multifunctional SOICs can get beyond those restrictions from a materials standpoint. According to the aforementioned, photosynthetic protein and dicationic SOICs have been combined to create flexible Bio-PEC-type photodetectors that can detect extremely low UV light intensity and turn sunlight into energy. Following that, they built Bio-PECs on a flexible substrate for use in wearable technology and tested a tiny electronic device in the presence of UV light, establishing it as a promising coating option for sunglasses with UV protection illustration in Figure 9. The creation of different SOICs for improved performance and practical applications is made possible by the strategy of integrating RC-LH1 into SOICs in dual-function electrical generation and sensing devices [62].

Figure 9. Role of SOICs in sunglasses for eye protection from UV light.

Flexible OIHPSCs and OSCs

There has never been a solar cell, like OIHPSCs in the history of third-generation solar cells, which in just seven years have attained an astounding PCE of 25.2%. Because of the exceptional electrical, chemical, and optical characteristics of perovskite materials, OIHPSCs achieve performance that surpasses the PCEs of CdTe polycrystalline thin film (B21.20%), CIGS (B22.80%), thin film crystalline Si (B21.20%), and multicrystalline Si (B22.30%) solar cells. More than 12,000 scientific papers have been published in recent years, and several commercial companies, including Dyesol, Hunt Perovskite Technologies, Oxford PV, Microquanta Semiconductor, Frontier Energy Solutions, Saule Technologies, and others, are establishing their own technologies for the commercialization of OIHPSC panels. Because of their inexpensive cost, mechanical stability, and R2R production capability, OIHPSCs are suitable for creating flexible solar cells. Although flexible OIHPSC manufacturing has successfully employed scalable R2R deposition techniques, the measured PCE is still as low as 5% [63].

Role of SOICs in Flexible OIHPSCs and OSCs

Due to their remarkable optoelectronic characteristics, which include long diffusion lengths, intense photo-harvesting, low exciton binding energies, high charge transport mobility, and long lifetime, organic–inorganic halide perovskites ($CH_3NH_3PbX_3$, where X = I^-, Cl^-, or Br^-) have made significant improvements in PCE possible over the last ten years. The organic hole transport material (OHTM), a vital part of OIHPSCs, needs to be tuned for high PCE. To attain greater PCEs (25.2% reported in

2019), a number of OHTM types (spiro-OMeTAD), PEDOT:PSS (Poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate)), PTAA (Poly(bis(4-phenyl)(2,4,6-trimethylphenyl)amine), and P3HT (Poly(3-hexylthiophene-2,5-diyl)) have been studied. Organic or inorganic dopants such Cu (III) complex, tBP, CuSN, Li-based salts (Li-TFSI, NaTFSI, and AgTFSI), and Cu (II) pyridine are necessary to improve OIHPSC performance. OIHPSCs can attain exceptional PCEs by employing the additive technique initially employed in solid-state DS-PECs38. A trace quantity of supporting chemicals used to increase the overall PCE of OIHPSCs is present in the majority of reported OHTMs [64]. Nevertheless, the hygroscopic nature of Li-TFSI additions also makes it easier for perovskite and OHTM materials to degrade (or change their electrical characteristics). Many attempts have been made to create dopant-free OHTMs for OIHPSCs to overcome device instability, and these devices have demonstrated over 18% PCE. Based on the description above, a series of dopant-free A(acceptor)–D(donor)–A(acceptor)-type SOICs (55–57) for OSC and IOHPSC device applications employing pyridinium dication and bromide or bis(trifluoroethane)sulfonamide (TFSI_) anions (Figure 10) [65]. Numerous advantageous characteristics, including improved hole mobility, appropriate energy levels, superior ionic conductivity, and higher light absorption capability, are present in these highly conjugated pyridinium cation-based SOICs [66]. Additionally, the conductivity, hole mobility, and device performance of the SOICs were significantly impacted by the pyridinium cation with different anions (Br_ or TFSI_). In terms of photovoltaic performance, OIHPSCs and OSCs made with single component showed the highest PCE values of 17.4% and 6.89%, respectively. These values are very comparable to those of conventional Spiro-OMeTAD (with additive)-based OIHPSC (PCE = 17.9%). Additionally, it was claimed that two ionic OHTMs were created using a microwave reactor (58–59) and POZ substitution BIMI cation with distinct anions (I^- and Br^-). These SOICs' superior ionic conductivity, hole mobility, and heat stability (4200 °C) have led to their development as a replacement for dopant OHTMs. When exposed to one sunlight intensity, the corresponding photovoltaic performance of the single component based OIHPSC shows a higher PCE value of 15.0% than PCE = 12.0%. By including Li-TFSI, the researchers saw an improvement in photovoltaic performance. J–V characteristics revealed that + Li-TFSI had an overall PCE value of 18.1%, the PCE of the maintained almost 90% of its initial value, indicating exceptional long-term stability of the OIHPSC [67].

FLEXIBLE ENERGY STORAGE DEVICES

SOICs in Flexible Supercapacitors

Because of their superior interaction with porous electrodes, liquid electrolytes, such as aqueous, organic, and ionic liquid electrolytes, have been utilized as energy storage components (supercapacitors and batteries) for many years. Although organic and ionic liquid electrolytes have a lower ionic conductivity, they may nevertheless function at higher potential ranges. Aqueous electrolytes have a limited operating voltage range while having outstanding ionic conductivity and capacity for performance. There was a chance that the liquid electrolyte-based SC may leak, which could lead to several safety issues. Solid-state polymer electrolytes present encouraging chances to stop leaks and reduce the safety risk in this situation. Because of their delayed penetration into porous electrodes, weak ionic conductivity, and poor surface contact, the described solid electrolytes often perform worse [68]. Furthermore, these solid-state electrolyte-based SCs had a short lifespan in real-world use. PGEs, which combine the advantages of solid and liquid electrolytes, have gained interest due to their dual functionality as an electrolyte and separator. Poly-(methyl methacrylate) (PMMA), poly(vinyl alcohol) (PVA), poly(ethyl oxide) (PEO), poly(acrylic acid) (PAA), and numerous other polymer components are present in the reported PGEs.

Different kinds of additives (inorganic salts) are added to the gel electrolyte matrix to enhance the ionic conductivity and electrochemical performance of PGEs. For example, adding $LiNO_3$, LiOH, $CuCl_2$, LiCl, and so on enhances the performance of PVA/H_2SO_4-based hydrogels. However, virgin PGEs have extremely low ionic conductivity due to the absence of inorganic salts. Furthermore, adding more than one additive raises the price of flexible SCs, adds complexity, and may have an impact on stability over the long run. By substituting SOICs for hazardous inorganic salts and low-conductive ILs,

the aforementioned problems can be eliminated [69]. Additionally, the development of effective and stable flexible SCs may result from the combination of SOIC with pluronic polymers, which may enhance ionic conductivity, interface contact between electrodes and electrolyte, and electrode pore-filling. Pluronic polymers show promise as hydrogel candidates for a variety of solid-state devices, including DS-PECs, zinc ion batteries, supercapacitors, and Few of them, meanwhile, have been investigated for SOIC application in flexible energy storage systems. According to recent reports, flexible SCs with thermos-responsive Pluronic hydrogels generated by SOICs have a broad operating temperature range. To improve the hydrogel's electrochemical characteristics, two SOICs piperidinium iodide and piperidinium bromide, were added. Because of their superior mechanical endurance and decreased volatility, SOIC-induced hydrogels lessen the problem of device leakage. It was discovered that the flexible SCs using SOICs as the ionic conductors outperformed those made with traditional LiBF4 salt in the electrolyte [70].

The flexible SCs not only function exceptionally well at both low and high temperatures, but they also have a thermoresponsive self-healing feature. Intense mechanical stresses, including bending, twisting, stretching, etc., are a part of the practical use of flexible SCs. These stresses can lead to delamination and cracks in the solid electrolyte and electrolyte/electrode interface of the flexible SC, respectively. High conductivity was demonstrated by 30 w/w% SOIC hydrogel (s = 2.04 mS cm^{-1} at 293 K). This is due to the 3D intrinsically connected nanochannels found in hydrogel, which enable quicker ion transport inside the polymeric matrix.

They put together Ti3C2 NS FSCs in three distinct configurations – sandwich, twisted fiber, and interdigitated Ti_3C_2 NS FSCs to examine the advantages of SOIC-induced hydrogel in FSC, the performance and long-term stability of FSCs are usually enhanced by these setups. This led to the creation of a sandwich structure of Ti_3C_2 NS-based flexible SCs with 30 w/w% SOIC hydrogel, which demonstrated long life cycles and outstanding electrochemical characteristics. In addition to FSCs' high capacitance, flexibility, and lightweight are essential properties for scaling up FSCs for wearable and portable applications. Therefore, they created the interdigitated FSC structure, which has numerous benefits over the traditional sandwich FSC configuration. The primary benefit of this interdigital design is the short distance between electrodes, which eliminates the necessity for the membrane (or separator) that is typically used in traditional FSC High performance is achieved due to the lateral ion transport in layered Ti_3C_2 NS side by side and the numerous accessible edges that the interdigitated Ti_3C_2 NS provides. Reduce the resistance of ion transport, further reduce the distance between the Ti_3C_2 NS electrodes, and work to improve power density and cycle stability at high current densities. Additionally, they created twisted fabric Ti_3C_2 NS FSC on a polyester cotton blend fabric substrate to assess the electrochemical performance of Ti_3C_2 NS FSC as a flexible energy source for wearable and smart electronics applications. Numerous fiber FSCs that have been reported are installed in a twisted configuration, which enables them to be used in conjunction with other fiber-shaped flexible electronic devices to achieve low-cost and multipurpose technology [71].

In addition to the electrode design and configurations, SOIC-induced hydrogel also plays a pivotal role in obtaining outstanding stability and performance of FSCs. This hydrogel's self-assembly microcrystalline phase in an aqueous environment, which improves ionic conductivity and promotes ion mobility within the device, is its greatest benefit. Long polymeric chains found in hydrogel can function as a micro-insulator to prevent titanium (Ti_3C_2 NS) oxidation and the flow of tiny discharge currents. Hydrogel's high hydrophilicity improves the electrolyte electrode interface and electrode wettability. Additionally, it was discovered that SOIC hydrogel outperformed traditional electrolytes such as 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIM-BF4). All the research above suggests that in-plane interdigitated Ti_3C_2 NS FSC performed exceptionally well under multiple cycling, different bending angles, and high current densities, while SOIC-induced hydrogel aids in preserving electrode–electrolyte interfacial contact under severe mechanical bending [72].

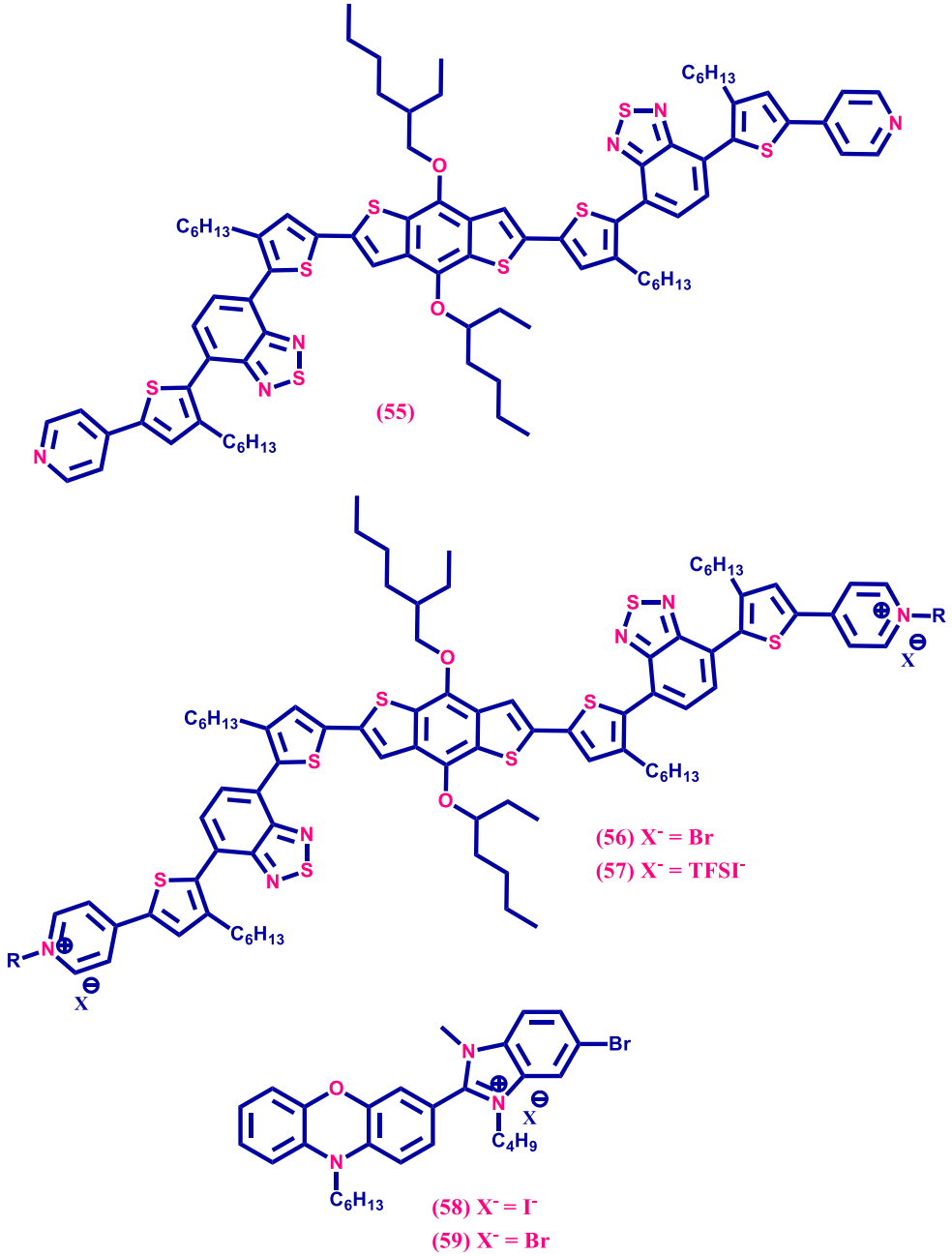


Figure 10. Structures of heterocyclic SOICs used in flexible OIHPSCs and OSCs.

CONCLUSIONS AND FUTURE PROSPECTS

This chapter examined the significant challenges associated with the stability and performance of traditional electrolytes in flexible energy generation and storage devices, as well as the creation of novel solutions, with a focus on alternative SOICs. Regardless of the internal architecture of the cell, we have talked about the most recent literature reports on different types of SOICs and their application in various photo, electrochemical cell, and supercapacitor devices as an essential part for high-performance cells. Nevertheless, the published SOICs for OIHPSC and flexible DS PECs have not yet been released. Over the past several years, we have emphasized the advancement and gains made in the structural design and application of SOICs, with as a focus on flexible energy-generating and storage devices. Compared to traditional electrolytes, the overall PCEs of all solid-state photoelectrochemical devices based on innovative single-component SOICs are greater. In particular, the maximum PCEs were recorded at over 8.0% in solid state DS PECs and OIHPSC devices, and 18.0% when employing

innovative SOICs with an additive. Innovative notions necessitate the identification of SOICs molecular architecture and its work functions, which are distinct from standard electrolytes utilized in DS PECs used to achieve high performance, multifunctionality, and stability. Among the many advantageous characteristics of the highly conjugated SOICs molecules are their easily processed, diverse electrical and optical characteristics. Strong backbone interactions between heterocyclic compounds enhanced close molecule-to-molecule contact and made electron/hole charge carrier transit possible.

Most notably, SOICs with only one component, that is, no additives, showed excellent conductivity and charge mobility. Even though solid-state DS PECs are much more durable than liquid electrolyte devices, issues with stability, scaling up production on flexible substrates, and additional PCE improvements still exist. With dye-sensitized photoanode and counter electrode, it is possible to address out existing issues with the electrolyte interfaces and improve the performance and longevity of reported flexible DS PECs. Furthermore, new SOICs for solid state DS PECs can be created using as stable organic radical (such as 2 azaadamantane Nsoxyl (AZA) sor 2,2,6,6,tetramethyl,1piperidinyloxy (TEPO)) as a solid state electrolyte rather than an unstable iodide radical because the organic radical is a potential redox mediator with dual channel for the charges (electron/hole) transport and improved DS PECs, performance and stability. Eliminating iodide components from solid-state DSPECs or the SOIC's skeleton will greatly extend device life and prevent photo corrosion from affecting the counter electrode. Nonetheless, there is still a great deal of space for advancement in terms of stability, efficiency, and adaptable, flexible energy gadgets via the logical development of new SOICs.

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