

**Review****A comprehensive review of filler, plasticizer, and ionic liquid as an additive in GPE for DSSCs****Mohd Fareezuan Abdul Aziz<sup>1,2</sup>, Nur Ezyanie Safie<sup>2</sup>, Mohd Asyadi Azam<sup>2,\*</sup>, Tunku Aidil Ilham Tunku Adaham<sup>2</sup>, Tan Jun Yu<sup>2</sup> and Akito Takasaki<sup>3</sup>**<sup>1</sup> School of Physics, Universiti Sains Malaysia, 11800 USM, Penang, Malaysia<sup>2</sup> Fakulti Kejuruteraan Pembuatan, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia<sup>3</sup> Department of Engineering Science and Mechanics, Shibaura Institute of Technology, 3-7-5 Toyosu, Koto-ku, Tokyo, 135–8548, Japan**\* Correspondence:** Email: [asyadi@utem.edu.my](mailto:asyadi@utem.edu.my); Tel: +60125048957.

**Abstract:** Low ionic conductivity in gel polymer electrolytes (GPEs) affects low dye-sensitized solar cells (DSSCs) performance is a crucial issue. Generally, the GPEs contain polymer (act as solvent holder), solvent, and salt (as ions provider). Usually, the GPE-based DSSCs are assembly with three necessary compartments: working electrode, GPE, and platinum electrode. The DSSCs parameters are included open-circuit voltage,  $V_{oc}$ ; short-circuit current density,  $J_{sc}$ ; fill factor,  $ff$  and efficiency, %. This review's main objective was to explore an additive such as plasticizer, filler, and ionic liquid effects on the ionic conductivity in GPEs by improving ions mobility and expanding the free volume of the GPE. The impact of additives in the GPE is also expected to enhance the DSSCs performance by increasing the  $J_{sc}$ ,  $V_{oc}$ ,  $ff$ , and efficiency. This comprehensive review discussed the latest progress of GPE utilizing the additive by listing the literature from the recent ten years.

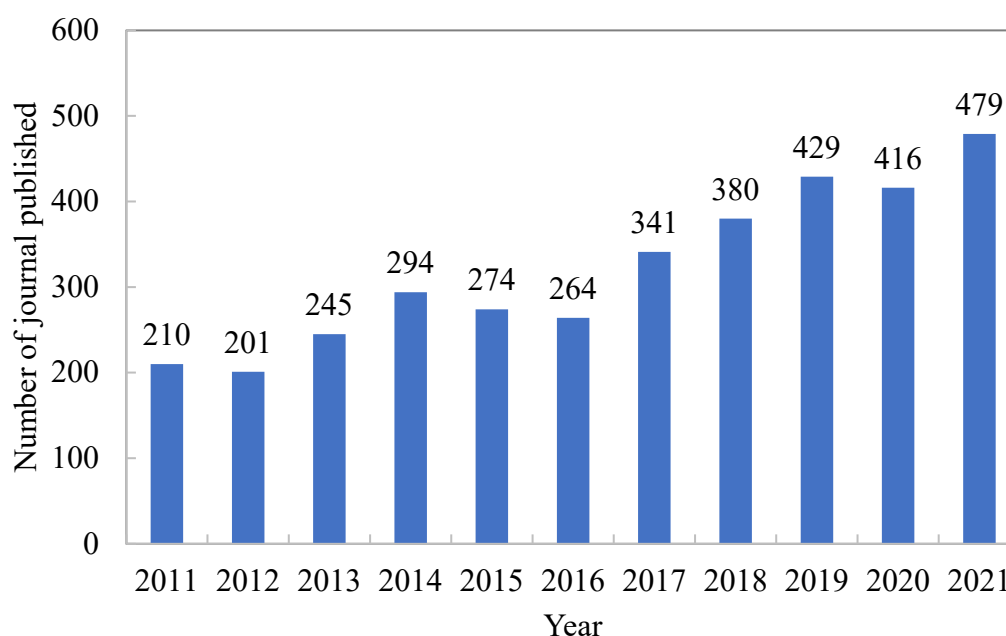
**Keywords:** ionic conductivity; gel polymer electrolytes; DSSCs efficiency; plasticizer; filler; ionic liquid

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## 1. Introduction

Renewable energy applications such as solar cells in the current period have become an invention because of their advantage in transforming solar energy into electrical power. In initial generation solar cell fields including silicon layered based-solar cells [1–5] and thin film-based solar cells [6–10], there is a current generation of the solar cell, the dye-sensitized solar cell (DSSC), explored in 1991. This type of solar cell, invented by the Grätzel team, has good properties such as low-cost production, easy handling, and good energy conversion percentage [11]. The dye-sensitized solar cell also has more potential and advantages than initial generation solar cells [12]. The critical element in the DSSC assembly includes the electrolyte, photoanode, and counter electrode [13]. The poor interfacial electrolyte/electrode contacts in the DSSC brought the development of electrolytes and non-precious-metal counter electrodes in order to demonstrate an electrochemical stability and excellent catalytic activity towards triiodide reduction and [14,15]

The electrolyte is essential in building DSSCs that act as a medium for charge transport. Liquid types of electrolytes can make efficient DSSCs, but the properties such as corroding, leaking and flammability cause safety problem [16]. Gel polymer electrolytes (GPEs) are used as a replacement because of their properties, such as high ionic conductivity compared to solid polymer electrolytes and good stability. The performance of DSSCs also depends on the ionic conductivity factor. Ionic conductivity determines the efficient charge transfer from cathode to photoanode in DSSCs. Several factors determine a gel polymer electrolyte ionic conductivity. For instance, the lattice energy must be low when choosing a salt to ensure easy salt dissociation. The salts' cations and anions play essential roles in the DSSC process [17].



**Figure 1.** Number of published journals on GPEs through the years from 2011 to 2021. Source: Scopus database. Data are updated (January 2022).

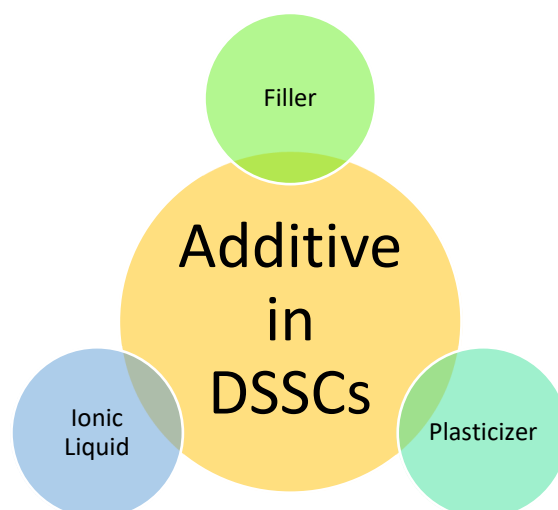
Currently, many GPEs have been proposed from the literature, which is made by mobilizing ions and plasticizing organic solvents in polymer matrices, and their potential use as application electrolytes is being actively pursued. The number of published journals on gel polymer electrolytes kept increasing through the years, confirming that the development of GPE for supercapacitors, Li-ion batteries and DSSC application is still significant (Figure 1). Even if the ionic conductivity of these gel polymer electrolytes is equivalent to that of liquid type, other aspects, including solvent holding, operation performance, and low-temperature conductivity, should be studied [18]. GPEs' ionic conductivity enhance because of the incorporation of organic solvents. Nevertheless, in these complex structures, solvent depletion due to evaporation or leakage is unavoidable [19]. Solvent absence leads to the collapse of the contact between electrode and electrolyte, as clearly as a reduction in conductivity.

For the exemplary dye-sensitized solar cell operation, the electrolyte should be in gel form, including various properties such as high conductivity, good stability, and no-leakage problem [20]. This show that the advancement of gel polymer electrolytes for DSSCs fabrication is still on challenging. In previous work, some various polymers has been used in the GPE for dye-sensitized solar cells, including poly(methyl methacrylate),  $(C_5O_2H_8)_n$  [21–28], poly(acrylonitrile),  $(C_3H_3N)_n$  [29–36], poly(vinylidene fluoride-co-hexafluoropropylene),  $(-CH_2CF_2-)_x[-CF_2CF(CF_3)-]_y$  [37–42], poly(vinylidene fluoride),  $(C_2H_2F_2)_n$  [43–45] and polyethylene glycol,  $C_{2n}H_{4n+2}O_{n+1}$  [46–50]. There are several works has been performed utilizing the GPEs including a salt such as  $K^+I^-$  [51],  $Li^+I^-$  [52],  $TBA^+I^-$  [53], and  $Na^+I^-$  [54], which are applied in DSSC fabrication. The idea of dual large-small cations in gel polymer electrolytes has been reported, such as  $TPA^+I^- / K^+I^-$  [55,56],  $TBA^+I^- / Li^+I^-$  [57],  $TBA^+I^- / K^+I^-$  [58–60],  $TPA^+I^- / Mg^{2+}I^-$  [61],  $THA^+I^- / K^+I^-$  [62] and  $THA^+I^- / Rb^+I^-$  [63]. The dye-sensitized solar cells' performance containing a double iodide system is expected to be high, but this combination of large-small cations shows low conductivity because of the sluggish mobility of large cations in the gel polymer electrolyte.

The standard GPE-based dye-sensitized solar cell fabrication is made by sandwiching the electrolyte with a photoanode (titanium dioxide layered with dye solution) and a counter electrode (layered platinum electrode). Prepare the photoanode by coating the titanium dioxide ( $TiO_2$ ) paste on the top of the active side fluorine tin oxide (FTO) glass. Usually, the preparation of  $TiO_2$  pastes by mixing the titanium dioxide powder with 2 mL of nitric acid (pH = 1). Then, spread the  $TiO_2$  paste using the “doctor blade” technique on the FTO glass. Then, the  $TiO_2$ -FTO will be heated in a closed furnace for 35 minutes at a temperature of 450 °C. Soak the  $TiO_2$ -FTO in Ruthenium N3 dye for 24 hours. This photoanode is ready to use after placing 24 hours in a dry desiccator. For the counter electrode preparation, the platinum solution is coated on the active side of the FTO glass and will be in the furnace for 35 minutes at 450 °C. Finally, the DSSC will be illuminated under sunlight conditions to measure efficiency and other parameters such as fill factor ( $ff$ ), short-circuit current density ( $J_{sc}$ ), and open-circuit voltage ( $V_{oc}$ ) [45,55,56].

The primary objective of this review is to inform and describe a summary of the different kinds of additives and their influence on the different types of gel polymer electrolytes employed for dye-sensitized solar cell fabrication. This review focused on developing additives in the GPEs for DSSCs in the past ten years. The effects of several additives in the GPEs have been divided into three types: plasticizer, filler, and ionic liquid, as in Figure 2. The fillers, such as silicon dioxide ( $SiO_2$ ), titanium dioxide ( $TiO_2$ ), zinc oxide ( $ZnO$ ), and aluminium oxide ( $Al_2O_3$ ), are listed in this review. Examples of a plasticizer such as ethylene carbonate (EC), propylene carbonate (PC), and diethyl carbonate (DEC) are discussed in this review. Several ionic liquids such as N-butyl-6-methylquinolinium bis

(trifluoromethylsulfonyl) imide [C<sub>4</sub>mquin] [NTf<sub>2</sub>], 1-methyl-3-propylimidazolium iodide (MPII), 1-butyl-3-methylimidazolium (BMII), 1-hexyl-3-methylimidazolium iodide (HMII), 1-butyl-3-methylimidazolium chloride (BMIC), 1-butyl-3-methylimidazolium tetrafluoroborate (BMITFB), and 1-glycidyl-3-methylimidazolium chloride (GMIC) is described. The influence of additives with several properties and identities on improving different GPEs in dye-sensitized solar cells is discussed. Hopefully, this review will help our current researcher who works on the GPEs to fabricate DSSCs.



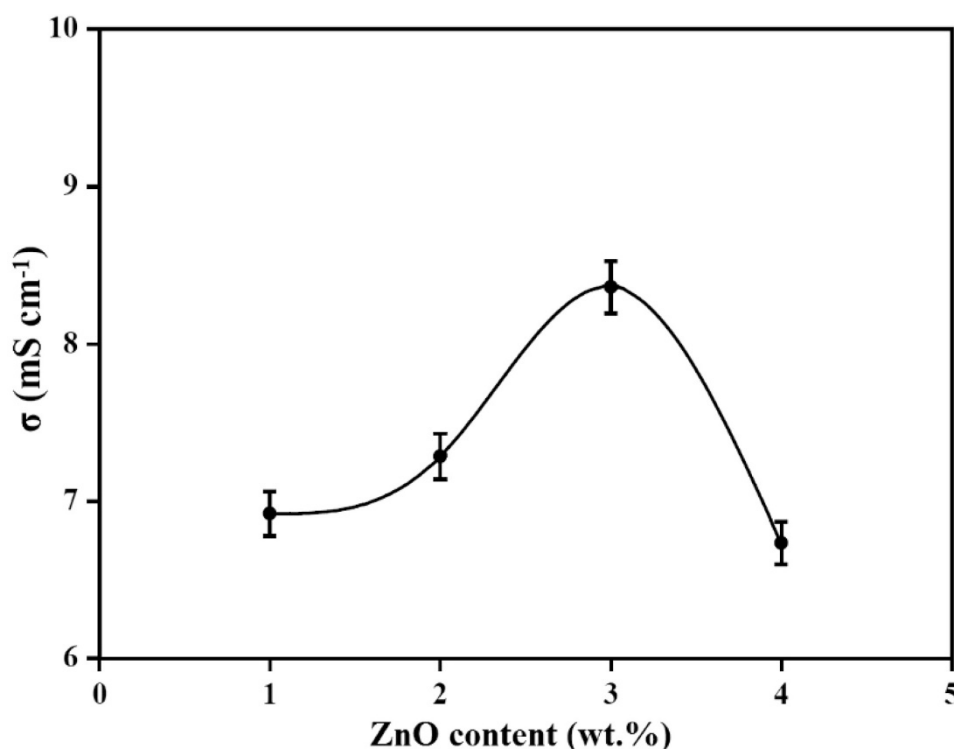
**Figure 2.** Classification of additives used in the dye-sensitized solar cells.

## 2. Filler as an additive in GPE

Based on the literature, gel polymer electrolytes containing nanoparticles as filler are expected to have higher ionic conductivity, and when such electrolytes are incorporated into dye-sensitized solar cells (DSSCs), performance is expected to improve. Tiautit *et al.* [64] incorporated silica (SiO<sub>2</sub>) and titanium oxide (TiO<sub>2</sub>) nanofillers into polymer gel electrolytes (PGE) and analyzed the effect on the dye-sensitized solar cell's performance. The poly (vinylidene fluoride-co-hexafluoro propylene) (PVdF-HFP) - polyvinyl alcohol (PVA) based PGE was produced by mixing the polymers in 1-Methyl-2 pyrrolidone (NMP) together with the SiO<sub>2</sub> and TiO<sub>2</sub> nanofillers. The performance test of the DSSC fabricated using this PGE revealed lower short-current density,  $J_{sc}$  and efficiency. The SiO<sub>2</sub>-TiO<sub>2</sub> nanofiller had enhanced the viscosity of the polymer gel and hindered the movement of ionic species in the PGE. This conclusion was supported by electrochemical impedance spectroscopy of the DSSC assembly, which measured a higher ion diffusion resistance ( $R_{diff}$ ) in the polymer electrolyte compared to the PGE without a nanofiller.

In a previous study, zinc oxide (ZnO) was used as a nanofiller mixed in a blending of polyethylene oxide (PEO) and poly (vinylidene fluoride-co-hexafluoro propylene) (PVdF-HFP) [65]. The nanocomposite polymer (NCPE) was prepared by dissolving the PEO/ PVdF-HFP in the ethylene carbonate (EC) and propylene carbonate (PC) solvents. Then the sodium iodide (NaI), iodine crystal (I<sub>2</sub>) and ZnO nanofiller was added to the NCPE. From Figure 3, the highest conductivity of  $\sim 8.4 \times 10^{-3}$  S cm<sup>-1</sup> was obtained when the NCPE contained 3 wt. % of ZnO nanofiller. The lowest conductivity

of  $6.4 \times 10^{-3} \text{ S cm}^{-1}$  was obtained with the polymer electrolyte (0 wt. % of ZnO). The addition of ZnO nanofiller was expected to expand the amorphous region of NCPE. Thus, escalated the ionic conductivity of NCPEs by improving the free volume, which conducts the polymer chains to be expanded in the system. Thus, the redox couple ( $\text{I}^-/\text{I}_3^-$ ) mobility was increased while passing through free volumes gained in the NCPEs. The ZnO nanofiller also improved the electron recombination in the NCPEs by inducing the electron originating from the conduction of the titanium dioxide ( $\text{TiO}_2$ ) layer on the photoanode to the oxidation/reduction potential of  $\text{I}^-/\text{I}_3^-$ . DSSCs performance utilizing the NCPE based on PEO/ PVdF-HFP with ZnO nanofiller had a conversion efficiency of up to  $\sim 7.0\%$ . This phenomenon showed that adding ZnO nanofiller had improved the  $J_{sc}$ , resulting in a high-efficiency value. NCPEs containing ZnO nanofiller played an essential and influential role in enhancing ion transportation through transmittal channels, thus improving the DSSC performance.

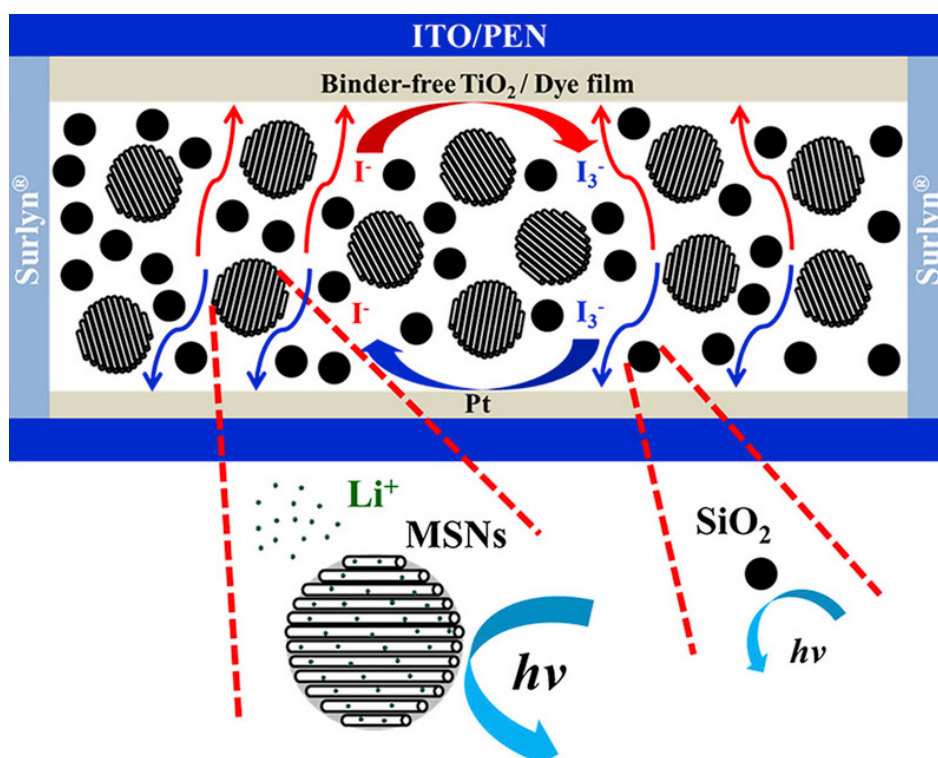


**Figure 3.** Conductivity vs weight percentage (wt. %) of ZnO amount. Reproduced with permission from Elsevier [65].

In this report [66], the authors suggested that adding  $\text{SiO}_2$  to the gel polymer electrolytes would improve the polymer chain flexibility, possibly enhancing the ions' mobility. In this study, GPEs consisting of polymer blends of poly (vinylidene fluoride)-co-hexafluoropropylene (PVdF-HFP) and Poly (ethylene oxide) (PEO) with ethylene carbonate (EC), propylene carbonate (PC), sodium iodide (NaI), and iodine ( $\text{I}_2$ ) was incorporated with  $\text{SiO}_2$  nanofiller. The GPEs without  $\text{SiO}_2$  (0 wt. %) nanofiller only showed ionic conductivity of  $6.40 \times 10^{-3} \text{ S cm}^{-1}$ . When the  $\text{SiO}_2$  nanofiller was added to the GPEs system, ionic conductivity increased to  $\sim 9.0 \times 10^{-3} \text{ S cm}^{-1}$  (13 wt. % of  $\text{SiO}_2$ ). The  $\text{SiO}_2$  nanofiller could improve the local diffusion of ions by creating pathways and reducing the crystalline structure of the GPEs, resulting in the creation of chain mobility for ions movement. This work also revealed that the performance of the DSSCs was raised by improving the short-circuit current density,  $J_{sc}$ ,

from  $14.0 \text{ mA cm}^{-2}$  (0 wt. % of  $\text{SiO}_2$ ) to  $27.0 \text{ mA cm}^{-2}$  (13 wt. % of  $\text{SiO}_2$ ). Thus, the GPE containing 13 wt. % of  $\text{SiO}_2$  nanofiller showed the highest DSSC efficiency of  $\sim 9.4\%$ .

Another researcher used mesoporous silica nanoparticles (MSNs) in a gel electrolyte (GE) which was applied in the fabrication of plastic-based dye-sensitized solar cells (DSSCs) [67]. The MSNs based gel-electrolyte was prepared by mixing lithium iodide (LiI), tetrabutylammonium iodide, iodine ( $\text{I}_2$ ), silica ( $\text{SiO}_2$ ) and MSNs at various weight percent. From the DSSCs study, the gel-electrolyte containing 10 wt. % MSNs showed the lowest series resistance,  $R_s$ , compared with the other samples.  $R_s$  was identified as the charge transfer resistance at the platinum electrode/GE interlayer and the photoanode/GE interlayer. Another series resistance,  $R_s$ , is the Warburg element ( $Z_w$ ) which is the resistance of the ionic distribution ( $\text{I}^-/\text{I}_3^-$ ) in the GE. MSNs (in nm size pores) assisted in the redox couple ( $\text{I}^-/\text{I}_3^-$ ) ion distribution, resulting in higher ionic conductivity. The DSSCs parameter showed that the gel-electrolyte without MSNs (0 wt. %) had low short-circuit current density,  $J_{sc}$  of  $\sim 9.90 \times 10^{-3} \text{ A cm}^{-2}$  and efficiency of  $\sim 4.6\%$ . However, gel-electrolyte containing 10 wt. % of MSNs showed high  $J_{sc}$  and efficiency of 5.45%. From the literature [67], the MSNs enhance the ionic conductivity of the gel-electrolyte by forming more conductive pathway in the gel-electrolytes. This phenomenon also enhanced the  $V_{oc}$  because the  $\text{Li}^+$  (from the electrolyte) adsorbed into the mesopores of MSNs in the electrolyte. As shown in Figure 4, this is presumably because the addition of MSNs in the gel-electrolyte exhibited a better  $J_{sc}$ ,  $V_{oc}$  and higher efficiency.



**Figure 4.** Structure of DSSC utilizing GPEs with the addition of MSNs. Reproduced with permission from Elsevier [67].

The other example of incorporating  $\text{SiO}_2$  as filler to boost the ionic conductivity and DSSCs performance was performed by this researcher [68]. This researcher discussed that the influence of filler in the hybrid polymer electrolyte (HPE) matrix of poly (ethylene oxide) (PEO) and poly (vinylidene

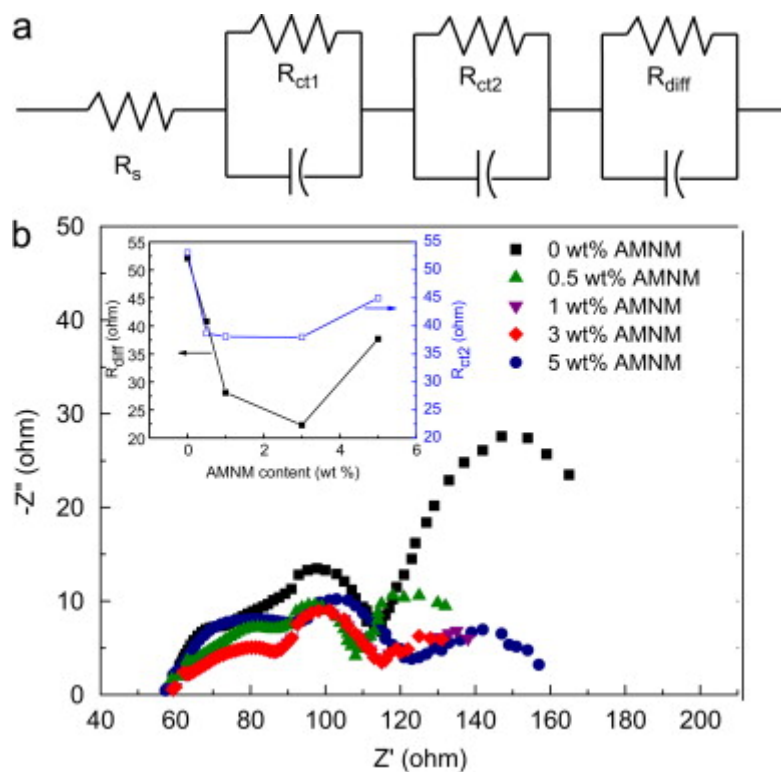
fluoride)-hexafluoropropylene (PVDF-HFP) expanded the amorphous phase of HPE, thus assisting in the formation of more ion conduction pathways. This ionic conductivity improvement was demonstrated by the HPE consisting of 20 mg of SiO<sub>2</sub>, which attained a high value of  $\sim 1.4 \times 10^{-3} \text{ S cm}^{-1}$  at the normal temperature of 27 °C. In contrast, the activation energy ( $E_a$ ) of this HPE (with 20 mg of SiO<sub>2</sub>) was at the lowest value of 0.65 eV, indicating that the ions were more mobile in the HPE with added SiO<sub>2</sub> filler, which may be due to the increased free volume in the HPE, resulting in high ion mobility. In the DSSCs fabrication tests, the HPE with added SiO<sub>2</sub> filler showed improved  $J_{sc}$  and efficiency,  $\eta$ . This HPE (10 mg SiO<sub>2</sub>) had its  $J_{sc}$  values increased from  $11.27 \times 10^{-3} \text{ A cm}^{-2}$  to  $13.63 \times 10^{-3} \text{ A cm}^{-2}$  and its efficiency improved from 3.91 % to 4.85 %. The SiO<sub>2</sub> filler could improve the contact between the photoanode and the HPE, thereby improving charge transportation and decreasing electron recombination. However, the HPE containing 20 mg of SiO<sub>2</sub> had its  $J_{sc}$  decrease due to excessive SiO<sub>2</sub>, resulting in aggregation and blocking of the charge carrier transport in the HPE, leading to a higher charge recombination rate and decreased the DSSCs efficiency.

Jeon and Kim briefly discussed utilizing nano-sized alumina (Al<sub>2</sub>O<sub>3</sub>) nanofiller in GPEs [69]. From this research, the amount of 10 wt. % Al<sub>2</sub>O<sub>3</sub> nanofiller in poly (1-methyl 3-(2-acryloyloxy propyl) imidazolium iodide) (PMAPII) resulted in a relatively high DSSC efficiency of 6.51% (AM 1.5 illumination) and a more stable DSSC compared to the ones fabricated with liquid electrolyte. The Al<sub>2</sub>O<sub>3</sub> nanofiller improved inter-layer contact between the GPE and both electrodes (photoanode and counter electrode). This study also found that the Al<sub>2</sub>O<sub>3</sub> nanofiller facilitates ion mobility in the GPEs, resulting in enhanced DSSC efficiency.

Another example that used the aluminium oxide (Al<sub>2</sub>O<sub>3</sub>) nanofiller as an additive in gel polymer electrolytes (GPEs) produced the nanocomposite GPE [70]. This researcher prepared the Al<sub>2</sub>O<sub>3</sub> nanofiller mixed with poly(ethyleneimine) (PEI), poly (ethylene glycol) diglycidyl ether (PEGDE), salt (BMII-I<sub>2</sub>), 4-tert-butylpyridine and mixed solvents of ethylene carbonate (EC)/propylene carbonate (PC). From this work, the addition of Al<sub>2</sub>O<sub>3</sub> nanofiller into GPEs has improved the conductivity. The maximum conductivity ( $\sim 10^{-2} \text{ S cm}^{-1}$ ) and diffusion coefficient ( $\sim 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ ) were attained by the GPEs containing 20 wt. % Al<sub>2</sub>O<sub>3</sub> nanofiller. It was suggested that the Al<sub>2</sub>O<sub>3</sub> nanofiller facilitated the creation of more free volume at the nanoparticle interface, resulting in high charge mobility in the channels of the nanocomposite GPE. However, beyond 20 wt. % Al<sub>2</sub>O<sub>3</sub> nanofiller, the GPE's conductivity and diffusion coefficient decreased due to the aggregation of Al<sub>2</sub>O<sub>3</sub> nanofiller within it. In the dye-sensitized solar cells fabrication, the short-circuits current density,  $J_{sc}$  and efficiency ( $\eta$ ) had improved for nanocomposite GPE containing 20 wt. % Al<sub>2</sub>O<sub>3</sub> nanofiller. The high ionic conductivity influenced the short-circuits current density,  $J_{sc}$ , and promoted high DSSC efficiency. EIS analysis was performed to study the effect of Al<sub>2</sub>O<sub>3</sub> nanofiller on the mechanisms of the DSSCs. The inclusion of Al<sub>2</sub>O<sub>3</sub> nanofiller in the nanocomposite GPEs had reduced the electrochemical reaction at the platinum counter electrode ( $R_{ct1}$ ) and the charge transfer reaction at the photo anode ( $R_{ct2}$ ), such that there was an increase in the charge transport and inter-contact area at the GPE and electrode interface.

In addition, the gel polymer electrolyte (GPE) was prepared by dissolving alkyl-modified nanomicas (AMNM) with salt, poly (ethylene glycol) (PEG), 4-tert-butylpyridine (TBP) and 3-methoxypropionitrile (MPN) [71]. DSSCs performance utilizing the GPEs incorporating 3 wt. % of AMNM displayed enhanced short-circuits current density,  $J_{sc}$  and efficiency. The author explained that the amorphous structure of GPEs was expanded by adding the AMNM, thus reducing the diffusion resistance of the redox couple ( $I^-/I_3^-$ ), as proven by the EIS analysis. EIS spectrum of the DSSC

displayed three semicircles (as referring in Figure 5):  $R_{ct1}$  indicating charge transport existing at the platinum counter electrode (displayed as the first curve),  $R_{ct2}$  indicating the photoanode-gel polymer electrolyte interface (displayed as the second curve) and the  $R_{diff}$  indicating the Warburg diffusion activity in the gel polymer electrolyte (displayed as the third curve). The two capacitors parallel with  $R_{ct1}$  and  $R_{ct2}$  indicate the capacitance of the Helmholtz double layer at the electrode. The  $R_{diff}$  decreased from  $\sim 50.0$  to  $\sim 22.0 \Omega$  with an increasing weight percentage of AMNM in the GPEs.



**Figure 5.** (a) The DSSC's equivalent circuit (b) The Nyquist plot for DSSC utilizing gel electrolytes containing AMNM. Reproduced with permission from Elsevier [71].

**Table 1.** Summary of findings of GPE with filler as an additive.

No.	GPE	$J_{sc}$ (mA cm <sup>-2</sup> )	$V_{oc}$ (V)	$ff$	$\eta$ (%)	Conductivity, $\sigma$ (S cm <sup>-1</sup> )	References
1.	PVdF-HFP:PEO:TiO <sub>2</sub>	2.61	-	-	2.71	-	[64]
	PVdF-HFP:PEO:SiO <sub>2</sub>	4.39	0.80	0.86	3.03	-	
2.	PVdF-HFP:PEO:EC:PC:NaI:I <sub>2</sub> :ZnO	19.48	0.624	0.602	7.33	$8.36 \times 10^{-3}$	[65]
3.	PVdF-HFP:PEO:EC:PC:NaI:I <sub>2</sub> :SiO <sub>2</sub>	27.31	0.564	0.613	9.44	$8.84 \times 10^{-3}$	[66]
4.	SiO <sub>2</sub> :TBAI:I <sub>2</sub> :MSN	11.01	0.751	0.66	5.45	-	[67]
5.	PVdF-HFP:PEO:LiI:I <sub>2</sub> :SiO <sub>2</sub>	13.63	0.60	0.595	4.85	$\sim 1.0 \times 10^{-3}$	[68]
6.	PMAPII:Al <sub>2</sub> O <sub>3</sub>	-	-	-	6.51	-	[69]
7.	PEI:PEGDE:LiI:I <sub>2</sub> :Al <sub>2</sub> O <sub>3</sub>	14.50	$\sim 0.75$	-	6.34	$\sim x10^{-2}$	[70]
8.	PEG:BMII:I <sub>2</sub> :AMNM	15.20	0.76	0.58	6.70	-	[71]

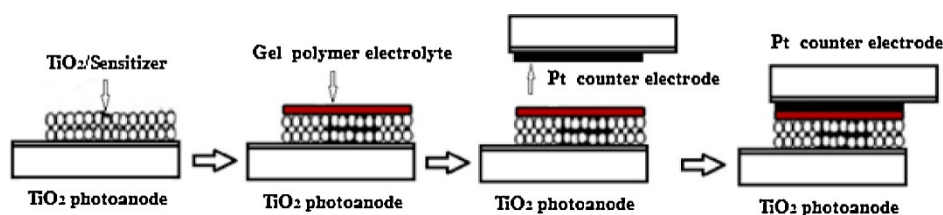


Table 1 summarizes the findings of GPE with filler as an additive. The GPE with the filler shows a high short-current density,  $J_{sc}$ , which depends on the GPEs' conductivity. This phenomenon is due to the addition of filler improving the ions pathway in the gel polymer electrolyte, enhancing the GPE conductivity. The current flow through the DSSC is faster due to the increase of ions mobile in the GPE. The  $V_{oc}$  shows a high value of  $\sim 0.6$  and  $0.80$  V. The high fill factor value of  $\sim 0.6$  to  $\sim 0.90$  described that the contact between the gel polymer electrolyte and photoanode-counter electrode is good [65–68]. In summary, the  $J_{sc}$ ,  $V_{oc}$ ,  $ff$ , and conductivity for GPE using silica ( $\text{SiO}_2$ ) have the highest value. The filler addition in the GPE enhances the conductivity and the DSSCs' performance [66]. Overall, the filler improves the gel polymer electrolytes structures and conductivity [66].

### 3. Plasticizer as an additive in GPE

According to Wang *et al.* [72], plasticizers can increase the ionic conductivity of gel polymer electrolytes (GPEs), thereby revamping the performance of dye-sensitized solar cells (DSSCs). The author prepared GPEs based on polyvinylidene fluoride (PVDF) by mixing it with propylene carbonate (PC)-ethylene carbonate (EC) blend as a plasticizer, sodium iodide (NaI) salt and N-Methyl-2-Pyrrolidone solvent. The weight ratio of PC-EC was kept constant at 1:2. The electrochemical impedance spectroscopy result revealed that the conductivity of the GPE escalated from  $1.0 \times 10^{-3} \text{ cm}^{-1}$  to  $\sim 2.3 \times 10^{-3} \text{ cm}^{-1}$  with the inclusion of a PC-EC plasticizer in the system. In DSSCs, the Warburg impedance,  $Z_w$ , was measured for each DSSC utilizing GPEs containing PC-EC plasticizer. It was evident that the inclusion of the PC-EC plasticizer had lowered the  $Z_w$ , resulting in a decrease in the iodide ion diffusion resistance, therefore facilitating its migration to the electrode. The author also stated that the PC-EC plasticizer allowed the PVDF to mix more thoroughly with the N-Methyl-2-Pyrrolidone, thus expanding the amorphous region and enhancing the chain movement of the gel polymer electrolytes in order to help enhance the iodide ions' mobility and increased ionic conductivity.

Meanwhile, the contact between gel polymer electrolytes and the electrodes was increased. These factors led to a decrease in internal impedance in the GPE and improved DSSC efficiency. Finally, this study also reported that the DSSC (as refer in Figure 6) utilizing GPE containing PC-EC plasticizer had a short-circuit current density,  $J_{sc}$  of  $\sim 5.8 \times 10^{-3} \text{ A cm}^2$ , open-circuit voltage,  $V_{oc}$  of  $0.58$  V, fill factor of 40% and efficiency of 1.32%.



**Figure 6.** DSSC fabrication process. Reproduced with permission from Elsevier [72].

In another study by Chowdhury *et al.* [52], it was also shown that the inclusion of the ethylene carbonate (EC) and propylene carbonate (PC) as a plasticizer in a GPE improved its room-temperature ionic conductivity and afterwards improved the DSSC performance in which that GPE was installed. The GPEs were produced by mixing polyacrylonitrile powder into the EC-PC plasticizer, followed by tetrabutylammonium iodide and iodine ( $\text{I}_2$ ). The author described GPEs as being conventionally

composed of low portions of polymer host in a higher quantity of plasticizer, which EC-PC represents in this work. The glass transition temperature ( $T_g$ ) should be decreased with the existence of a plasticizer in the polymer. The  $T_g$  is a temperature at the glassy/rigid polymer transformed to a soft/rubbery phase. Furthermore, the plasticizer also helped improve segmental motion and expand the gaps left between entangled polymer chains. These modifications lead to increased conductivity of the GPEs and improved efficiency in the DSSC.

In Aziz *et al.* [73], diethyl carbonate (DEC) was used as a plasticizer in the gel polymer electrolytes (GPEs), which resulted in improved ionic conductivity and enhanced DSSCs' performance. The GPEs were produced by dissolving polyvinyl alcohol (PVA) as the polymer base in dimethyl sulfoxide (DMSO) solvent. Varied amounts of DEC plasticizer (0, 1.11, 2.57, 4.18 and 5.63 wt%) were added to the mixture together with tetrabutyl ammonium iodide (TBAI), potassium iodide (KI) and iodine crystal ( $I_2$ ). From the electrochemical impedance spectroscopy result, the ionic conductivity has increased from  $\sim 5.2 \times 10^{-3} \text{ cm}^{-1}$  (GPEs without DEC plasticizer) to  $\sim 6.5 \times 10^{-3} \text{ cm}^{-1}$  (GPE with 2.57 wt% of DEC). The improvement in conductivity may be recognized by the added DEC plasticizer raising the dielectric constant of the GPE. The height dielectric constant allowed the ions from the salts to dissociate easier, which therefore boosts ion mobility in the electrolyte. The author also proved that the addition of the diethyl carbonate increased the GPE's conductivity because it reduced the crystalline regions and, at the same time, expanded the amorphous regions in the material. The more amorphous structure of the material allowed ions to travel more easily and freely through it. GPEs incorporated with 2.57 wt% DEC plasticizer had the maximum conductivity and the smallest  $E_a$  of 0.10 eV. The author also provided the measurements of ions' number density ( $n$ ), diffusivity ( $D$ ), and mobility ( $\mu$ ) of GPEs containing various compositions of DEC plasticizer. Measurements from DSSC tests indicated that the addition of DEC plasticizer in the GPE increased the short-circuit current density,  $J_{sc}$  and efficiency,  $\eta$ . The  $J_{sc}$  increased from  $\sim 12.6 \times 10^{-3} \text{ A cm}^2$  to  $\sim 16.0 \times 10^{-3} \text{ A cm}^2$  while efficiency increased from 5.8% to 7.5%. The DEC plasticizer boosted ion mobility in the GPEs, thus increasing conductivity,  $J_{sc}$  and efficiency. Another study utilizing propylene carbonate and ethylene carbonate as plasticizers were carried out by Noor *et al.* [74]. In this work, the GPEs were produced by combining a plasticizer blend of ethylene carbonate (EC)-propylene carbonate (PC) with a constant weight ratio of 1:1 and the poly (vinylidene fluoride-hexafluoropropylene) in acetone. Then, sodium iodide (NaI) and iodine ( $I_2$ ) were added to the mixture, which was heated until a homogenous gel was formed. The authors claimed that adding the EC-PC plasticizer to the GPEs expanded the amorphous region, reducing the GPE's degree of crystallinity overall. Ions are also more mobile in this electrolyte, leading to increased ionic conductivity. The electrical impedance spectroscopy result confirmed that the EC-PC plasticizer had increased the GPE's ionic conductivity at room temperature from  $5.2 \times 10^{-5} \text{ S cm}^{-1}$  (GPE without EC-PC) to  $\sim 1.5 \times 10^{-4} \text{ cm}^{-1}$  (GPEs with 20 wt. % of EC-PC). The authors concluded that the EC-PC plasticizer would boost the dye-sensitized solar cells' performance by improving their ionic conductivity.

Table 2 summarizes the findings of GPE with plasticizer as an additive. The efficiency of the DSSC utilizing the GPE-plasticizer follows all DSSC parameters. The mobility of ions increases, and the current flows through the DSSC faster. The GPE with the plasticizer shows a good  $J_{sc}$ , which depends on the conductivity of the GPE. However, the  $V_{oc}$  shows an average value between 0.4 and 0.65 V. This observation is due to the plasticizer's inability to change the Fermi level of  $TiO_2$  [73]. The fill factor also shows a moderate value which ranges from 0.39 to 0.73. This phenomenon is due to the mild contact between the GPE and both electrodes.

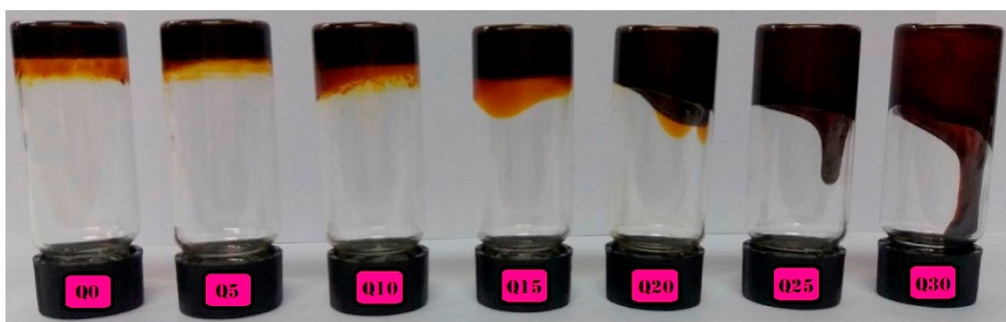
In summary, the conductivity, and DSSCs' parameters of GPE utilizing diethyl carbonate (DEC) have the highest value. This phenomenon is because the GPE already contains the mixed EC-PC. The existence of the DEC improves the conductivity and the DSSCs' performance [73]. Overall, the plasticizer helps the amorphous gel polymer electrolytes by reducing the crystalline regions and enhancing the ions' mobility in the GPE [74].

**Table 2.** Summary of findings of GPE with plasticizer as additive.

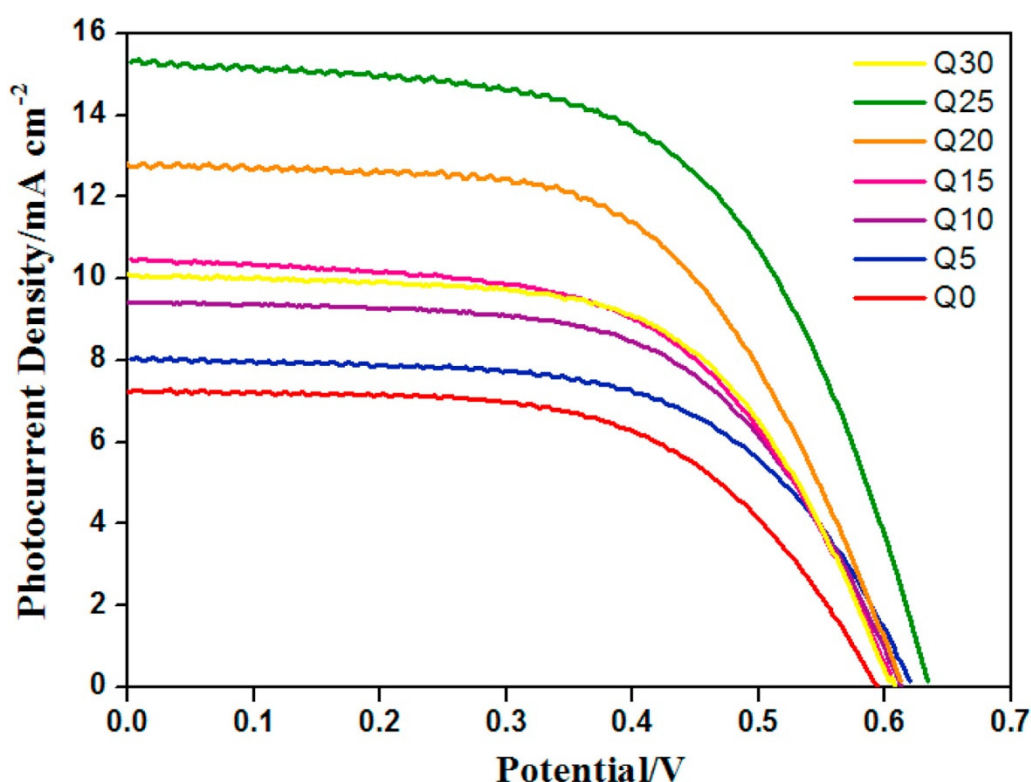
No.	GPE	$J_{sc}$ (mA cm <sup>-2</sup> )	$V_{oc}$ (V)	$ff$	$\eta$ (%)	Conductivity, $\sigma$ (S cm <sup>-1</sup> )	References
1.	PVdF:NaI:I <sub>2</sub> :EC-PC	5.80	0.58	0.39	1.32	$\sim 2.34 \times 10^{-3}$	[72]
2.	PAN:TBAI:I <sub>2</sub> :EC-PC	12.90	0.582	-	3.45	$5.14 \times 10^{-3}$	[52]
3.	PVA:TBAI:I <sub>2</sub> :DEC	16.05	0.64	0.73	7.50	$6.64 \times 10^{-3}$	[73]
4.	PVdF-HFP:NaI:I <sub>2</sub> :EC-PC	2.63	0.47	0.58	0.72	$1.53 \times 10^{-4}$	[74]

#### 4. Ionic liquid as an additive in GPE

Since the electrolyte's primary role in the DSSC is to facilitate ionic dissociation, ionic conductivity is a necessary trait to look for in designing an electrolyte. To that end, researchers have proposed adding an ionic liquid into their gel polymer electrolyte. Sundararajan V *et al.* [75] reported higher conductivity in their prepared gel polymer electrolyte (GPE) after adding N-butyl-6-methylquinolinium bis (trifluoromethylsulfonyl) imide [C<sub>4</sub>mquin] [NTf<sub>2</sub>] (as refer in Figure 7). In this work, the poly (methyl methacrylate-co-methacrylic) acid-based GPE was prepared by mixing the polymer and salt (NaI-I<sub>2</sub>) in a glass container filled with EC-PC solvents (weight ratio of 1:1). The electrochemical impedance spectroscopy result showed that the system's highest ionic conductivity obtained at ambient temperature was  $\sim 2.3 \times 10^{-3}$  S cm<sup>-1</sup> for the GPE consisting of 25 wt. % [C<sub>4</sub>mquin] [NTf<sub>2</sub>]. This trend of increasing ionic conductivity with an increasing weight percentage of [C<sub>4</sub>mquin] [NTf<sub>2</sub>] suggested that the charge carriers provided by the ionic liquid boosted the ion mobility in the GPEs. Theoretically, both factors dominated mainly for ionic conductivity. In contrast, this GPE (with 25 wt. % of [C<sub>4</sub>mquin] [NTf<sub>2</sub>]) had the lowest activation energy,  $E_a$ , at almost 0.17 eV, which supports the hypothesis that the ionic liquid facilitates ionic mobility, given that the individual ions have less barrier to move through the material. However, ionic conductivity for GPEs-ionic liquid systems decreases at above 25 wt. % of [C<sub>4</sub>mquin] [NTf<sub>2</sub>] due to ion saturation causing the ions to become more sluggish in the GPEs. From Figure 8, when DSSCs containing the GPEs with 25 wt. % of [C<sub>4</sub>mquin] [NTf<sub>2</sub>] were tested, they exhibited good efficiency ( $\sim 5.7\%$ ) with a high  $J_{sc}$  ( $\sim 15.0 \times 10^{-3}$  A cm<sup>-2</sup>). It was demonstrated that the presence of [C<sub>4</sub>mquin] [NTf<sub>2</sub>] ionic liquid at the optimal composition of 25 wt% had improved the total DSSC efficiency by more than 50% compared to the DSSCs without ionic liquid in their GPEs. The inclusion of [C<sub>4</sub>mquin] [NTf<sub>2</sub>] ionic liquid had significantly increased the number of charge carriers, thereby considerably boosting the  $J_{sc}$  and  $\eta$ .



**Figure 7.** GPEs prepared with various amount  $[C_4mquin][NTf_2]$  (From left to right: 0, 5, 10, 15, 20, 25, and 30 wt. %). Reproduced with permission from Elsevier [75].

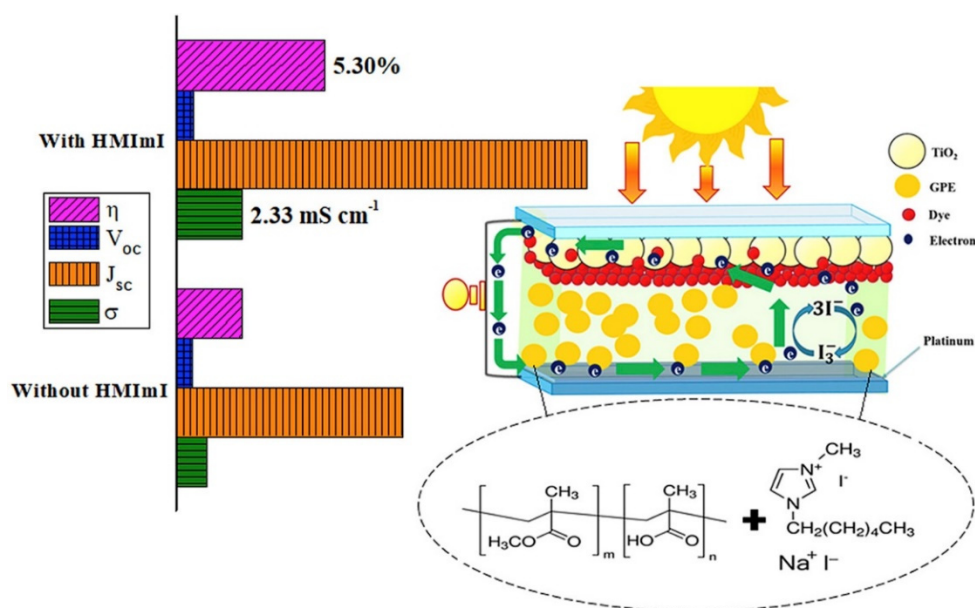


**Figure 8.**  $J-V$  curves of DSSCs utilizing the GPE with different amounts of  $[C_4mquin][NTf_2]$ . Reproduced with permission from Elsevier [75].

Safaruddin *et al.* [76] reported that introducing ionic liquid (IL) into a gel polymer electrolyte (GPE) did have a substantial effect on its ionic conductivity and DSSC efficiency. The author also reported that the different imidazolium-based ionic liquids were used in each GPE, such as 1-methyl-3-propylimidazolium iodide (MPII), 1-butyl-3-methylimidazolium (BMII), and 1-hexyl-3-methylimidazolium iodide (HMII). The GPEs were prepared by dissolving poly (ethylene oxide) (PEO), as the polymer base, into the mixing solvents of ethylene carbonate (EC) and propylene carbonate (PC). Then, sodium iodide (NaI) as salt, iodine ( $I_2$ ), and an ionic liquid (MPII, BMII, and HMII) were added to the mixture, and the mixture was heated at a constant temperature of 80 °C until a homogenous gel was formed. EIS study showed that the GPEs' conductivity with ILs increased. The bulk resistance,

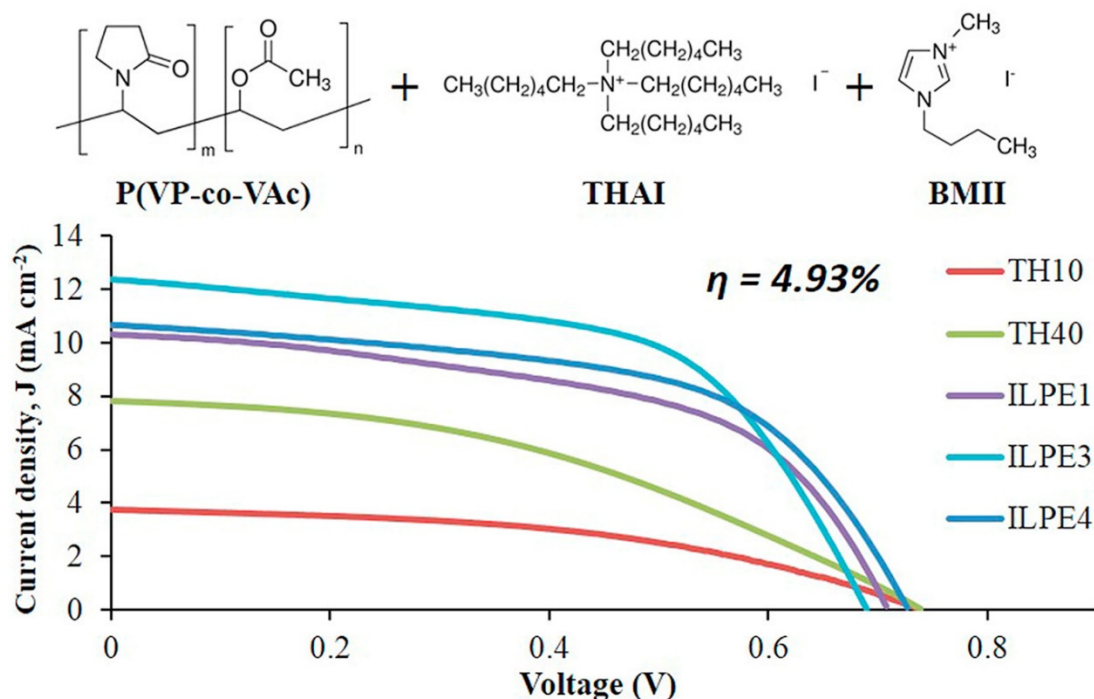
$R_b$ , decreased with the presence of ionic liquid in the GPEs. It was also found that the GPE containing MPEI had a maximum conductivity of  $\sim 9.4 \text{ mS cm}^{-1}$  compared to the GPEs-BMII ( $\sim 8.5 \text{ mS cm}^{-1}$ ) and GPEs-HMII ( $\sim 7.7 \text{ mS cm}^{-1}$ ) because the MPEI ionic liquid has the shortest alkyl group of imidazolium compared to BMII and HMII ionic liquid. The author also elaborated that the ionic liquids with the longer alkyl group of imidazolium (BMII and HMII) would reduce the ionic conductivity of the GPEs, simultaneously increasing their viscosities, resulting in lowered ion mobility. From the characterization of the DSSC fabricated using these GPEs, GPE-MPEI showed the best performance with the highest  $J_{sc}$  and  $\eta$  of  $\sim 25.0 \times 10^{-3} \text{ A cm}^{-2}$  and  $\sim 9.4\%$ , respectively, compared with the DSSC utilizing GPE-BMII and GPE-HMII. The EIS results of the fabricated DSSC showed that GPE-MPEI had the lowest value of  $R_{ct1}$  (charge transport resistance at the counter electrode/electrolyte) and  $R_{ct2}$  (charge transport resistance at photoanode/electrolyte), suggesting that the charge transport activity was high at both interlayers. Overall, this work showed that essential enhancements had been achieved in the gel polymer electrolytes by using imidazolium iodide ionic liquid as an additive.

Furthermore, the 1-hexyl-3-methylimidazolium iodide (HMIm<sup>+</sup>I<sup>-</sup>) ionic liquid was utilized as an additive to study the conductivity improvement and DSSCs performance [77]. The poly (methyl methacrylate-co-methacrylic acid) based GPEs were prepared as a mixture of polymer, EC, PC, NaI-I<sub>2</sub> and HMIm<sup>+</sup>I<sup>-</sup> ionic liquid. The amount of HMIm<sup>+</sup>I<sup>-</sup> ionic liquid was varied with different weight percentage, % (0, 5, 10, 15, 20 and 25 wt.%). The electrochemical impedance spectroscopy measurements on the prepared GPEs indicated conductivity obtained was  $\sim 2.3 \text{ mS cm}^{-1}$  for GPEs containing 20 wt. % HMIm<sup>+</sup>I<sup>-</sup> ionic liquid. The conductivity initially increased with the increasing amount of HMIm<sup>+</sup>I<sup>-</sup> ionic liquid in the GPE system, proving that the number of mobile charge carriers was enhanced. However, the ionic conductivity dropped slightly when 25 wt. % HMIm<sup>+</sup>I<sup>-</sup> was added in the GPEs. The number of ions accumulation in excess causes the mobile ions to change neutral couples; thus, ions mobility becomes more sluggish in the GPE. On the other hand, the GPE-containing HMIm<sup>+</sup>I<sup>-</sup> ionic liquid, which had the highest conductivity, had the lowest activation energy,  $E_a$ , indicating that the ions could easily move with minimal required energy in the GPEs. From the DSSC parameter study (as refer in Figure 9), short-circuit current density,  $J_{sc}$  showed an increasing trend starting from  $\sim 8.1 \times 10^{-3} \text{ A cm}^{-2}$  (GPE with 0 wt. % HMIm<sup>+</sup>I<sup>-</sup>) to  $\sim 15.0 \times 10^{-3} \text{ A cm}^{-2}$  (GPE with 20 wt. % HMIm<sup>+</sup>I<sup>-</sup>), which suggested that by adding HMIm<sup>+</sup>I<sup>-</sup> ionic liquid, the conductivity of GPEs could improve, which is in direct relationship with  $J_{sc}$ . Furthermore, the DSSCs efficiency,  $\eta$  also exhibited the same increasing trend, from  $\sim 2.3\%$  (GPE with 0 wt. % HMIm<sup>+</sup>I<sup>-</sup>) to  $5.30\%$  (GPE with 20 wt. % HMIm<sup>+</sup>I<sup>-</sup>). The author concluded that when the concentration of HMIm<sup>+</sup>I<sup>-</sup> ionic liquid is increased, so would the  $J_{sc}$  increase, indicating the dominant contributor to DSSCs efficiency.



**Figure 9.** Mechanism of DSSCs fabrication with HMIIm<sup>+</sup>I<sup>-</sup> ionic liquid. Reproduced with permission from Elsevier [77].

In another study, the author studied the effect of 1-butyl-3-methylimidazolium iodide (BMII) as an ionic liquid on the ionic conductivity of gel polymer electrolytes and DSSCs [78]. They prepared the poly(1-vinylpyrrolidone-co-vinylacetate)-based gel polymer electrolytes, including polymer, salt (tetrahexylammonium iodide) and BMII. The electrochemical impedance spectroscopy (EIS) analysis showed that the ionic conductivity increased with increasing weight fraction of BMII ionic liquid in the GPEs, from  $\sim 0.6 \times 10^{-3} \text{ S cm}^{-1}$  (GPE without BMII) to  $1.05 \times 10^{-3} \text{ S cm}^{-1}$  (GPE with 80 wt. % of BMII), which is an increase of about 40%. The BMII ionic liquid contributes many ionic species to the material, leading to a higher ionic transport rate. However, the ionic conductivity decreased when higher quantities of IL were added to the GPE, which is influenced by the formation of ion pairs which hindered the polymers chain's segmental motion, hence, causing the ions to move more sluggishly. The author stated that adding BMII had expanded the amorphous state and reduced the crystalline region of the GPEs, which is indicative of higher mobility for ions, which will lead to increased ionic conductivity. The GPE with 80 wt. % BMII had the lowest activation energy,  $E_a$ , and the highest conductivity. Lower activation energy promoted better ion transport which led to enhanced ionic conductivity. In the DSSC assembly (as refer in Figure 10), the crucial parameters such as short-circuit current density,  $J_{sc}$  and efficiency, % were improved with the presence of ionic liquid in the gel polymer electrolytes sample. The short-circuit current density,  $J_{sc}$  value, was reported from  $\sim 7.8 \times 10^{-3} \text{ A cm}^{-2}$  (GPE without BMII) and increased to  $\sim 12.4 \times 10^{-3} \text{ A cm}^{-2}$  (GPE with 80 wt. % BMII). This increasing pattern can be explained by the previous EIS study where the ionic conductivity increased with increasing ion concentration (from BMII) in the GPEs, while the  $J_{sc}$  also followed this trend. The author also understood that the existence of BMII ionic liquid had enhanced the ion mobility, improving in the  $J_{sc}$  and DSSC efficiency, reaching the highest value of  $\sim 5\%$ .

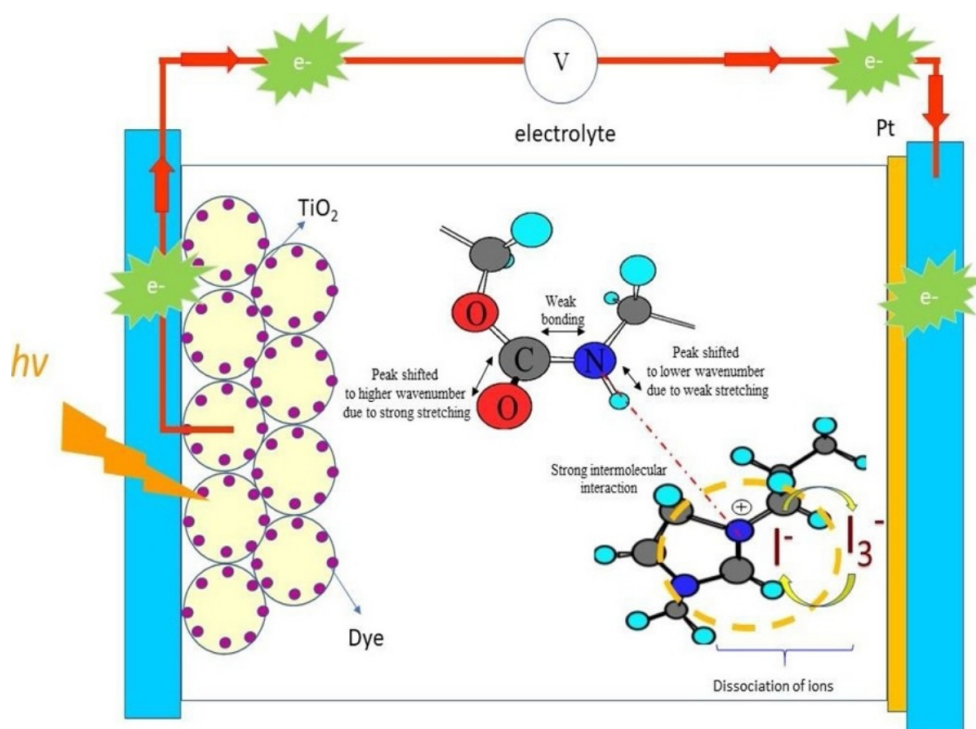


**Figure 10.**  $J$ - $V$  curves for DSSCs fabrication with BMII ionic liquid. Reproduced with permission from Elsevier [78].

Ng HM *et al.* [79] studied the usage of 1-methyl-3-propylimidazolium iodide (MPII) ionic liquid to reinforce the ionic conductivity of gel polymer electrolyte and improve DSSCs performance. This research work on the poly [1-vinylpyrrolidone-co-vinyl acetate] based GPEs by combining polymer, salt (KI), iodine ( $I_2$ ) in the dual solvents of ethylene carbonate (EC) and propylene carbonate (PC). For the DSSC assembly, the GPEs were sandwiched between two electrodes (TiO<sub>2</sub>-dye electrode and platinum counter electrode). The GPEs contained 15 wt. % MPII ionic liquid showed the maximum conductivity of  $\sim 3.0 \text{ mS cm}^{-1}$ . The author described that adding IL into the material revamps the GPEs' conductivity by increasing the number of mobile ions.

Furthermore, the ionic liquid has a plasticizing effect which helps improve the polymer segmental mobility by softening the polymer backbone chains. In this manner, it will create more conduction pathways for the charge carrier's movement through the free zone of the GPE. As expected, GPEs with the maximum conductivity had the lowest  $E_a$  (activation energy), which showed that the ions required the minimum value to begin moving through the material. In the DSSCs, the inclusion of MPII ionic liquid in the GPEs had improved the short-circuit current density,  $J_{sc}$  (from  $\sim 8$  to  $\sim 12 \times 10^{-3} \text{ A cm}^{-2}$ ) and conversion efficiency,  $\eta$  (from  $\sim 3$  to 4.67%). It was found that the increase of  $J_{sc}$  was due to the increasing MPII ionic liquid concentration. This change was believed to contribute to increased iodide ionic conductivity, and the intercalation of the cations into the photoanode. This work performed EIS analysis for DSSCs to support the result. The DSSC using GPEs consisting of 15 wt. % ionic liquid had a lower ohmic serial resistance ( $R_s$ ) than the DSSC utilizing GPEs without MPII ionic liquid. The addition of the MPII positively changed the titanium dioxide photoelectrode's conduction band, which helped the excited electrons to recombine with the  $I_3^-$  in the GPE.

Another study used MPII ionic liquid to enhance ionic conductivity and DSSCs efficiency in a palm-based polyurethane GPE [80]. The GPEs was produced by mixing MPII ionic liquid at various proportion (from 10–30 wt. %) with palm kernel oil-based monoester-OH (PKO-p), methylene diphenyl diisocyanate (MDI) and ethylene carbonate (EC). The ionic conductivity increased with the increasing amount of MPII ionic liquid in the GPEs as analyzed by the electrochemical impedance spectroscopy (EIS) measurement. Concurrently, the concentration of free charge carriers ( $I^-$  ions) from the MPII ionic liquid increased due to the high contribution of the ion-dissociation in the GPEs. The DSSC (as refer in Figure 11) data analysis,  $J_{sc}$  and  $\eta$  were improved for the GPEs containing MPII ionic liquid. MPII and  $I_2$  were used as  $(I^-/I_3^-)$  in the GPEs, with both having low ionization energy and large atomic diameter, contributing to their ease of ion dissociation. Therefore, the DSSC efficiency is improved with the presence of MPII ionic liquid.



**Figure 11.** DSSC fabrication with MPII ionic liquid. Reproduced with permission from Elsevier [80].

Another study also showed that the MPII enhanced dye-sensitized solar cell efficiency and its ionic conductivity [81]. The P(VA-co-PE)-based GPEs contained 10 wt. % MPII exhibited a maximum ionic conductivity ( $\sim 4.0 \times 10^{-3} \text{ S cm}^{-1}$ ) and the lowest activation energy,  $E_a$ , of almost 0.14 eV. MPII increases the ion concentration in the GPEs, contributing to the higher ionic conductivity. In the DSSC fabrication, the existence of MPII also improved the  $J_{sc}$  and  $\eta$ . DSSC utilizing GPEs without MPII was measured at only  $\sim 7.0 \times 10^{-3} \text{ cm}^{-2}$  and  $\sim 3.4\%$  for  $J_{sc}$  and efficiency, respectively. However, after the GPEs were added and incorporated with the MPII at the optimum composition of 10 wt. %, both DSSC parameters were enhanced to  $10 \times 10^{-3} \text{ A cm}^{-2}$  and  $\sim 4.4\%$  for  $J_{sc}$  and efficiency, respectively. This phenomenon may be attributed to the addition of MPII providing the ideal ion concentration in the GPEs, leading to the high  $J_{sc}$  and DSSC performance. Shi *et al.* [82] explained that the inclusion of



ionic liquid in the GPEs increased the dye-sensitized solar cell performance by increasing the molality amount of the ionic liquid. The study began by preparing the GPEs with three different ionic liquids, namely 1-butyl-3-methylimidazolium chloride, 1-butyl-3-methylimidazolium iodide and 1-butyl-3-methylimidazolium tetrafluoroborate. The molarity of each ionic liquid was varied in the range of 0, 0.2, 0.4 and 0.6 M. Every set of ionic liquid with different molarities was mixed with 0.05 M iodine, acetonitrile, 0.5 M lithium iodide, and 20 wt. % polyethylene glycols (PEG). The DSSC test results showed that the DSSCs utilizing GPEs containing 0.4 M of 1-butyl-3-methylimidazolium chloride had the best performance, with its DSSC parameters being  $V_{oc}$  of 0.81 V,  $J_{sc}$  of  $\sim 10 \times 10^{-3} \text{ A cm}^{-2}$ ,  $ff$  of 0.63 and  $\eta$  of  $\sim 4.9\%$ . The author described how adding ionic liquid into the GPEs increased ion concentration, thereby increasing its conductivity. Thus, in the DSSCs, the  $J_{sc}$  and efficiency also improved. On the other side, the study of molarity variation shows that the ionic liquid must be added to the GPEs at optimal molarity. The ionic conductivity lowers as the molarity is more than 0.6 M, decreasing both  $J_{sc}$  and  $V_{oc}$ , which decreases the DSSC efficiency. Another ionic liquid that has been used for this purpose is 1-glycidyl-3-methylimidazolium chloride (GMIC) [83]. The researcher prepared the gel GPE by mixing the GMIC, cationic starch (CS), dimethyl sulfoxide (DMSO), potassium iodide (KI) and iodine ( $I_2$ ), whereby the mixture eventually turned into a gel form. The conductivity of the GPE with GMIC ionic liquid increased, which improved the DSSC efficiency. The optimized CS-based GPE was utilized in dye-sensitized solar cells, presenting  $\sim 0.5\%$  efficiency.

**Table 3.** Summary of findings of GPE with ionic liquid as additive.

GPE	$J_{sc}$ (mA cm <sup>-2</sup> )	$V_{oc}$ (V)	$ff$	$\eta$ (%)	Conductivity, $\sigma$ (S cm <sup>-1</sup> )	References
1. (P(MMA-co-MAA)):NaI:I <sub>2</sub> : [C <sub>4</sub> mquin] [NTf <sub>2</sub> ]	15.32	0.62	0.59	5.60	$\sim 2.30 \times 10^{-3}$	[75]
2. PEO:EC:PC:NaI:I <sub>2</sub> :MPH	$\sim 25.0$	0.55	-	$\sim 9.4$	$\sim 9.4 \times 10^{-3}$	[76]
3. P(MMA-co-MAA):NaI:I <sub>2</sub> :HMIm <sup>+</sup> I <sup>-</sup>	14.7	0.60	0.60	5.30	$\sim 2.3 \times 10^{-3}$	[77]
4. P(VP-co-VAc):THAI:I <sub>2</sub> :BMH	$\sim 12.4$	-	-	4.93	$1.05 \times 10^{-3}$	[78]
5. P[VP-co-VAc]:KI:I <sub>2</sub> :EC:PC:MPH	11.98	0.59	0.66	4.67	$\sim 3.0 \times 10^{-3}$	[79]
6. PKO-p:MDI:EC:MPH	3.30	0.71	0.36	0.85	$9.07 \times 10^{-4}$	[80]
7. P(VA-co-PE):MPH	9.97	0.67	0.65	$\sim 4.4$	$\sim 4.0 \times 10^{-3}$	[81]
8. PEG:LiI:I <sub>2</sub> : [BMIM]Cl	9.56	0.81	0.63	$\sim 4.9$	-	[82]
9. CS:KI:I <sub>2</sub> :GMIC	-	-	-	$\sim 0.5$	-	[83]

Table 3 summarizes the findings of GPE with ionic liquid as an additive. The GPE with the ionic liquid shows a good  $J_{sc}$ , which depends on the conductivity of the GPE due to the high concentration ( $I^-/I_3^-$ ) in the gel polymer electrolyte [81,82]. However, the  $V_{oc}$  shows an average value between 0.50 and 0.81 V. This observation is due to the ionic liquid's decreased open-circuit voltage ( $V_{oc}$ ) [83]. The fill factor range also shows a moderate value of 0.35 to 0.65. This phenomenon is due to the slight contact between the GPE and both electrodes.

In summary, the  $J_{sc}$ ,  $V_{oc}$ ,  $ff$ , and conductivity for GPE employing 1-methyl-3-propylimidazolium iodide (MPH) ionic liquid have the highest value. The existence of the MPH ionic liquid enhances the conductivity and the DSSCs' performance [76]. Overall, the ionic liquid increases the concentration of ( $I^-/I_3^-$ ) in the GPE and the DSSC performance [81,82].

## 5. Summary, challenges, and outlook

Dye-sensitized solar cells (DSSCs) are a device for converting electricity from solar energy with various advantages such as low manufacturing price, assembly friendly, and excellent benefit features. The high performance of DSSC utilizing the gel polymer electrolyte shows good stability, high conductivity, and no-leakage problem. Various researchers have used different polymers, solvents, and salts for the gel polymer electrolyte in DSSCs. The improvement of GPE for DSSCs fabrication is still challenging. In this review, the GPE with filler as an additive show that the filler will expand the amorphous region, enhancing the ionic conductivity of GPE. The filler improved the free volume, which conducts the polymer chains to be expanded in the GPE, resulting in the creation of micro-channels for ions mobility. This phenomenon is similar to the GPE utilizing the plasticizer as an additive. The ionic conductivity of the GPEs increased with the addition of the plasticizer because it reduced the crystalline regions and, at the same time, expanded the amorphous regions in the material. The more amorphous structure of the material allowed ions to travel more easily and freely through it. However, for the ionic liquid as an additive in the GPE work, the ionic liquid is contributed many ionic species to the material, leading to a higher rate of ionic transport. Overall, the ionic liquid, plasticizer, and filler as an additive in the gel polymer electrolyte (GPE) have improved the DSSCs performance, including the open-circuit voltage,  $V_{oc}$ , fill factor,  $ff$ , short-current density ( $J_{sc}$ ), and efficiency.

Currently, a few challenges include the mixture process between the additives and gel polymer electrolytes, the contribution of additives towards gel polymer electrolyte conductivity, and the additives' impact on the cells' performance. To overcome these challenges, the researchers should be exploring more the preparation skill of the GPE and should be studying more on the perfect combination between the additive and GPE. In the future, these GPEs with various additives are potentially used for other applications, especially in energy storage, including supercapacitors and Li-ion batteries. The high conductivity shown by the gel polymer electrolytes incorporated with additives is highly recommended for the superior performance of the devices.

### Conflicts of interest

There are no conflicts of interest to declare.

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