Review on the Role of Polymers in Luminescent Solar Concentrators

This manuscript is dedicated to Professor Mitsuo Sawamoto's outstanding achievements in polymer chemistry and recognizes his recent retirement from 40 years of exceptional service to Kyoto University.

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ABSTRACT: The use of luminescent solar concentrators (LSCs) offers an alternative approach to integrating photovoltaic technologies into the built environment. The research on LSCs has bloomed in the past decade in terms of searching for novel device architectures, developing new luminescent species, and employing unique host materials. This article will provide a concise review on LSCs and focus on the polymer host materials used in LSCs. Finally, we provide a brief outlook on the future development of this research area, particularly on the polymers used as host materials and luminescent species for LSCs. © 2018 Wiley Periodicals, Inc. J. Polym. Sci., Part A: Polym. Chem. **2019**, *57*, 201–215

KEYWORDS: luminescent; polymers; solar concentrators; solar energy; waveguides

INTRODUCTION Generating electricity from renewable energy resources within our community has gained a lot of attention in recent years because it provides a feasible way to realize a society with sustainable development without affecting the environment. Harvesting solar energy by photovoltaic (PV) devices is widely considered the most promising approach.[1–3] However, integration of solar panels into the built environment has proven difficult because solar panels operate optimally under direct light, while in the built environment, the sunlight is diffused and nonuniform, caused by scattering and reflection by buildings, trees, and clouds, which reduces the performance of solar cells.[4,5] In addition, solar panels are typically black and nontransparent. The lack of aesthetics makes the installation of solar panels unfavorable in public areas.

The integration of PV devices into the built environment motivates researchers to use luminescent solar concentrators (LSCs) as a potential solution. The concept of LSCs was introduced in the 1970s with the intent to reduce the cost of solar cells.[6,7] However, the continuous decrease in the cost of solar cells in later decades disabled its original design purpose, and as a result, LSCs did not attract lot of attention afterward. Recently, the research on LSCs has regained its momentum primarily due to the vigorous development of materials research and the urgency of global sustainable development. The research on LSCs has increased to 458 publications in the past decade. This number is over four times higher than the number of publications before 2009 (Fig. 1).

The key development of LSCs has been summarized in several key reviews over the period of research since the 1970s.[8–14] There are also a handful of specific reviews focusing on the luminescent species that are utilized in the LSCs.[15–17] Additionally, LSCs have also demonstrated a huge potential for education for the comprehensive concepts including materials science, spectroscopic optics, and art design.[18] In this article, we will first provide a brief overview on LSCs that includes the operation mechanism, device architectures, and materials. Next, we will focus on the role of polymers in LSCs. Polymers are often used as key components in LSCs. However, the topic on polymers used in the LSC is rarely discussed. In this article, the role of polymers primarily as host materials and luminescent species in the LSC will be highlighted.

AN OVERVIEW ON LSC

Operation Mechanism

The conventional LSC architecture consists of a plastic luminescent waveguide and solar cell attached to the edges. High-energy photons in sunlight are absorbed by the luminescent species and converted into low-energy photons within the plate. The waveguide traps a fraction of low-energy photons and delivers them to the edge of the plate by total internal reflection. The radiation energy at the edge of the waveguide is concentrated and readily utilized by the edge-attached solar cells (Fig. 2).

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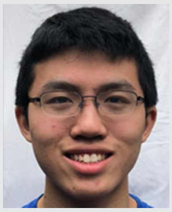

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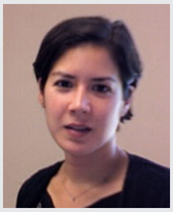

Generally, as a cost-effective approach, this design should allow the solar cells to produce more electricity (P_{LSC}) than those with the same solar cell area under the direct sunlight (P_{cell}). This character is quantified by concentration (C) and this value can exceed 100 in theory.[19,20] The power conversion efficiency of LSC (η_{LSC}) is defined the same as solar cells, which is the ratio of the output electrical energy (P_{LSC}) to the incident radiation energy ($H_{\mathrm{in}} \times A_{\mathrm{LSC}}$). Note that the power conversion

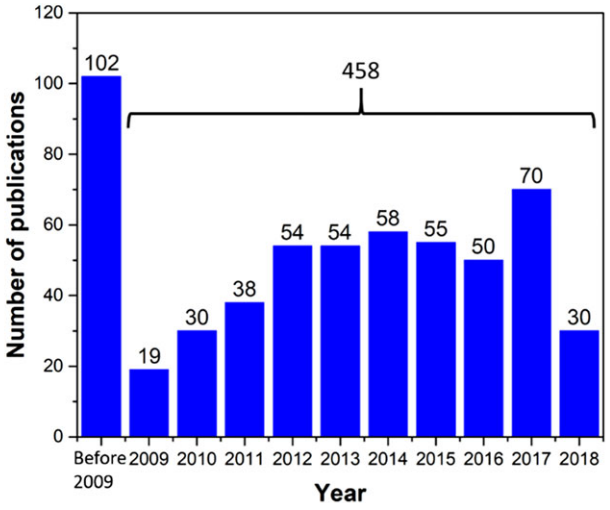

FIGURE 1 Publications per year on the research on LSCs. The results were obtained by Google Scholar search using the key words "luminescent solar concentrator" and "luminescent solar concentrators" within the title. Patents and citations are not included. The number of publications in 2018 is counted until May 23, 2018.

efficiency of LSC cannot exceed the power conversion efficiency of solar cells due to several loss mechanisms occurring within the waveguide.[21–23] The relationship between C and η_{LSC} is connected by geometric gain (G) and the power conversion efficiency of the solar cell (η_{cell}) (eq. 1). The ratio between η_{LSC} and η_{cell} is generally donated by optical collection probability (P). P is sometimes called optical efficiency (η_{opt}) that can be approximately measured by experiments (eq. 2).

$$C=\frac{P_{\mathrm{LSC}}}{P_{\mathrm{cell}}}=\frac{\eta_{\mathrm{LSC}}H_{\mathrm{in}}A_{\mathrm{LSC}}}{\eta_{\mathrm{cell}}H_{\mathrm{in}}A_{\mathrm{cell}}}=\left(\frac{\eta_{\mathrm{LSC}}}{\eta_{\mathrm{cell}}}\right)\times\left(\frac{A_{\mathrm{LSC}}}{A_{\mathrm{cell}}}\right)=P\times G \qquad (1)$$

$$P=\frac{P_{\mathrm{LSC}}}{P_{\mathrm{cell}}}\times\frac{1}{G}=\frac{I_{\mathrm{sc,LSC}}V_{\mathrm{oc,LSC}}\mathrm{FF}_{\mathrm{LSC}}}{I_{\mathrm{sc,cell}}V_{\mathrm{oc,cell}}\mathrm{FF}_{\mathrm{cell}}}\times\frac{1}{G}\approx\frac{I_{\mathrm{sc,LSC}}}{I_{\mathrm{sc,cell}}}\times\frac{1}{G} \qquad (2)$$

In the above equation, I_{sc} (A), V_{oc} (V), FF, and EQE are short circuit current, open circuit voltage, fill factor, and external quantum efficiency of a photovoltaic device, respectively. Note that solar cells are assumed to exhibit minimal change in V_{oc} and FF when attached to the luminescent waveguide.

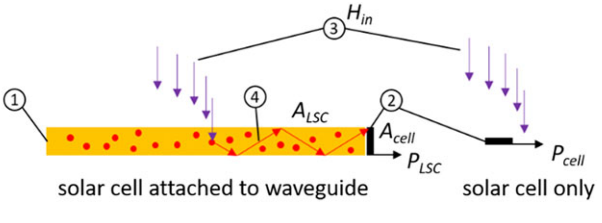

FIGURE 2 A conventional LSC and its operation mechanism: (1) plastic waveguide doped with luminescent species (red dots); (2) solar cell; (3) incident solar radiation; (4) emitted photons within the waveguide due to total internal reflection. H_{in}, incident solar power density (W m^{-2}); A_{LSC}, illumination surface area (m^2) A_{cell}, solar cell surface area (m^2); P_{LSC}: electrical power produced by the LSC (W); P_{cell}, electrical power produced by the solar cell only.

Other LSC Architectures

Beyond the conventional LSC architecture, other designs have been proposed to improve the performance of the device, simplify the fabrication process, or meet specific requirements for the integration into the built environment. Similar to tandem solar cells, tandem LSCs were designed to maximize the light-harvesting capability of the device [Fig. 3(a)].[24–26] In a tandem LSC, each layer can absorb a fraction of the sunlight. Theoretical studies have shown that under optimized conditions, a two-layer tandem LSC can reach a power conversion efficiency of up to 29.5% and a three-layer device can reach a power conversion efficiency of up to 33.6%, which is close to the Shockley Queisser limit for single-junction solar cells.[21,27] A general method to boost the concentration of the LSC is to increase the geometric gain. The cascade LSC architecture has been proposed to this end with minimal increase of the device size [Fig. 3(b)].[28] Strip-shaped luminescent waveguides are attached to the edges of the square-shaped luminescent waveguide. Solar cells are attached to the ends of the strip-shaped waveguides. To achieve radiative energy transfer, spectral match is a necessity for the emission of the square-shaped waveguide and the absorption of the strip-shaped waveguide. Experiments have shown that this design can double or triple the concentration, while significantly reduce the material use of solar cells as compared to the square-shaped LSC only. To simplify the fabrication process, luminescent species can be dispersed in a polymer matrix and coated on a transparent substrate to construct thin-film LSCs [Fig. 3(c)].[29–32] Films are typically in micrometer scale. This approach allows flexible choice of substrates and widens the selection of polymer matrix. Whether the performance of a thin-film LSC is better than a conventional LSC or not is still under debate.[33,34] Beside the top-coated thin-film LSCs, other configurations include bottom-coated LSCs[35] and sandwiched LSCs.[36] Monte-Carlo ray tracing simulation suggests that the bottom-coated LSCs exhibit slightly better performance than the top-coated LSCs.[37] In addition to these flat and rigid LSC architectures, researchers have also explored some non-flat and flexible LSC architectures [Fig. 3(d)]. Cylindrical LSCs have shown a better performance than the flat rectangle LSCs by experiments and simulations.[38,39] Flexible LSCs can be achieved by employing rubber elastomers[40,41] and plastic fibers[42–44] as waveguides. The nonflat and flexible LSCs provide more aesthetic design options in the built environment than the flat and rigid ones.[45]

In addition to the above LSC architectures that are varied in geometric design, changing the configuration of solar cells also produces novel LSC architectures. In conventional LSCs, solar cells only absorb photons emitted from the luminescent species; while a large fraction of photons with the energy between the absorption onset of the solar cells and that of the luminescent species is wasted. To address this severe photon loss, a configuration where solar cells are attached to the

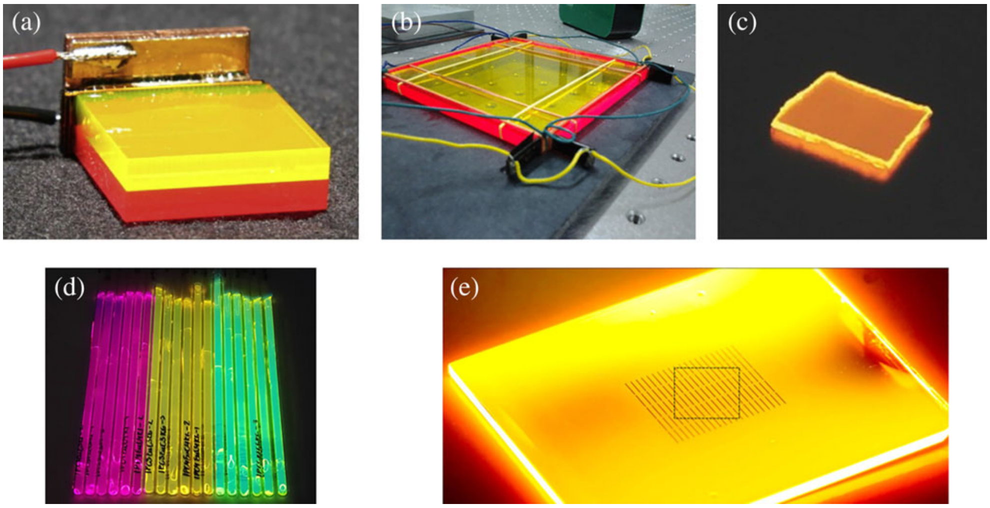

FIGURE 3 Pictures of (a) a tandem LSC (Reproduced from ref. 26, with permission from Copyright © 2009, Elsevier.); (b) a cascade LSC (Reproduced from ref. 28, with permission from Copyright © 2014, AIP Publishing.); (c) a thin-film LSC (Reproduced from ref. 32, with permission from Copyright © 2015, John Wiley & Sons.); (d) cylindrical fiber LSCs (Reproduced from ref. 44, with permission from Copyright © 2018, Elsevier.); and (e) a top-mounted LSC (Reproduced from ref. 46, with permission from Copyright © 2011, Springer Nature.).

surface of the waveguide is proposed [Fig. 3(e)]. Solar cells can be top mounted[46] or bottom mounted.[47,48] The operation mechanism is similar to luminescent downshifting,[49–51] except that the area and thickness of the waveguide are much large than the downshifting layer, and additional mirror reflectors are attached to the edge of the waveguide.

Luminescent Species

There is no doubt that luminescent species play the most important role in the performance of the LSC. Developing novel luminescent species remains the most vigorous yet challenging area in the research of LSCs. Ideal luminescent species should demonstrate the following spectroscopic properties: (1) broad spectral absorption with high absorption efficiency, (2) matching the emitted photons to the spectral response of the edge-attached solar cells, (3) high photoluminescence quantum yield (PLQY), and (4) minimal spectral overlap or large Stokes shift.[12,22] A wide variety of materials have been developed toward the ideal luminescent species. Major categories are organic dyes, quantum dots, and lanthanide ions.

Organic dyes are π-conjugated small molecules. Their spectroscopic properties can be finely tuned by their chemical structure.[52,53] BASF Lumogen F Red 305 (R305) are the most widely used organic dyes in LSC research largely due to its commercial availability, and ideal spectroscopic properties [Fig. 4(a)]. Studies on LSCs using R305 start from small-scale laboratory tests,[54–58] and have been extended to large-scale outdoor and field tests in recent years.[59–64] High-efficiency LSCs (typically η_{LSC} > 4%) have been reported by employing

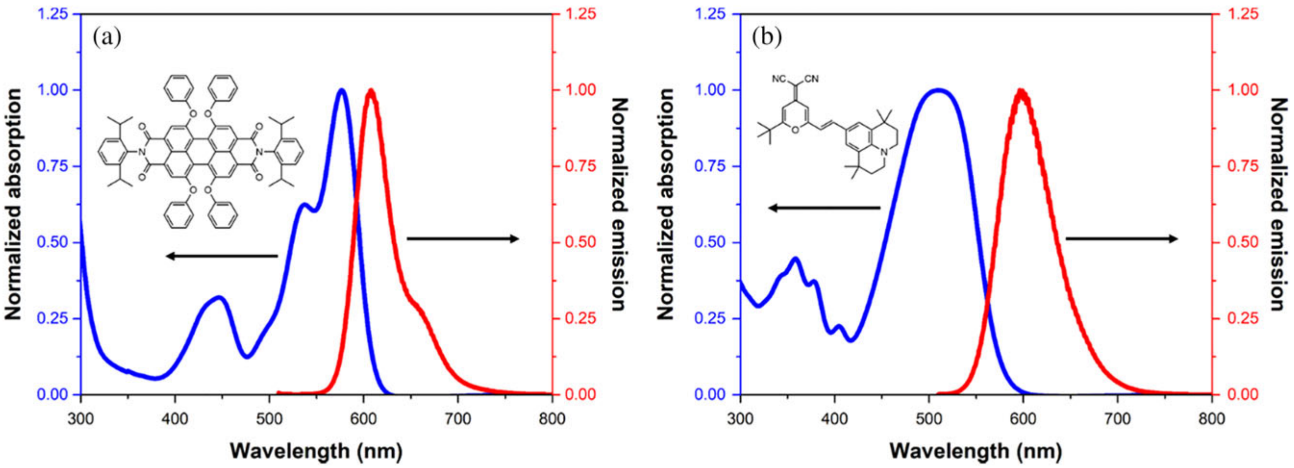

FIGURE 4 Absorption and emission spectra of (a) R305 and (b) DCJTB in chloroform. Inset: molecular structures of R305 and DCJTB.

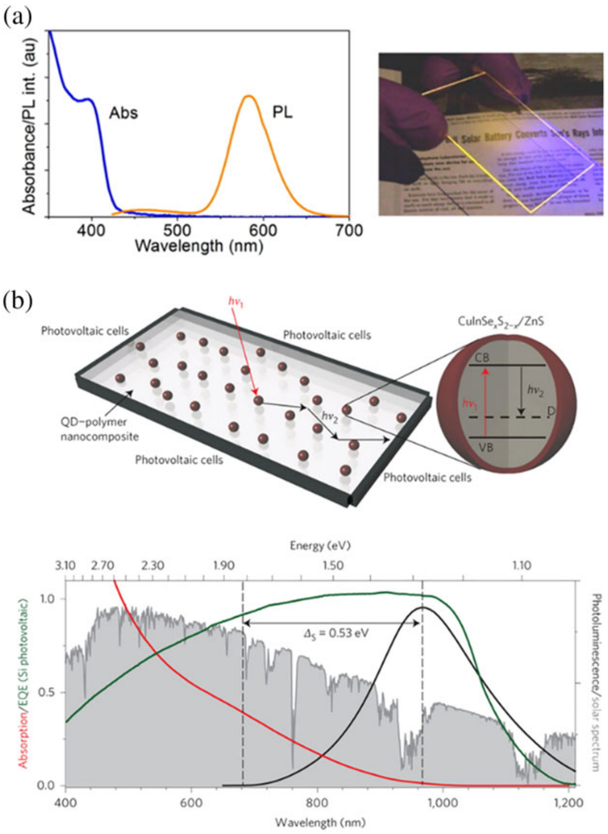

FIGURE 5 (a) Mn^{2+}-doped ZnSe/ZnS QDs: absorption and emission spectra and luminescent waveguide (Reproduced from ref. 85, with permission from Copyright © 2014, American Chemical Society.); (b) CISeS/ZnS QDs with absorption covering full visible spectrum (Reproduced from ref. 87, with permission from Copyright © 2015, Springer Nature.).

R305.[65,66] Moreover, R305 derivatives and structural analogs have been developed and proven efficient for LSC application.[22,67–69] Another representative organic dye is 4-(dicyanomethylene)-2-*t*-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTB) [Fig. 4(b)].[70–72] DCJTB exhibit similar emission wavelength (*ca.* 610 nm) and high PLQY (>0.95) as R305; while its absorption wavelength (*ca.* 500 nm) is shorter than R305 (*ca.* 560 nm) (Fig. 4). The relatively narrow absorption band limits the amount of the light absorbed by DCJTB. On the other hand, it enlarges the Stokes shift. The spectral overlap, and thus reabsorption probability, is minimized. Experiments have shown that LSCs fabricated by DCJTB exhibit comparable performance as those fabricated by R305.[73]

Quantum dots (QDs) are inorganic semiconducting materials with sizes typically below 100 nm. Their spectroscopic properties, especially absorption and emission spectra, are highly size dependent and can be precisely tuned by synthetic conditions.[74–78] Notably, some QDs show multi-exciton generation properties, leading to over unity PLQYs.[79–83] Current state-of-the-art QDs have some unique spectroscopic properties advantageous over organic dyes. For example, the reabsorption issue in LSCs can be effectively and efficiently tackled by employing QDs. Core-shell CdSe/CdS QDs with thick CdS shells exhibit a small spectral overlap, which leads to 45-fold reduction in reabsorption analyzed from absorption and emission spectra.[84] Mn^{2+}-doped ZnSe/ZnS QDs possess well-separated absorption and emission spectra. The fabricated luminescent waveguide only absorbs UV light and therefore the resulting LSCs would be transparent [Fig. 5(a)].[85] Some QDs are capable of absorbing the full visible spectrum of the sunlight and can emit photons at the near-infrared region up to 1000 nm (1.24 eV) while maintaining high PLQYs [Fig. 5(b)].[86–89] The near-infrared emission wavelength matches well to the band gap of monocrystalline silicon (mc-Si) solar cells (E_g = 1.12 eV or 1100 nm).

Lanthanide ions are stable species in the trivalent form. The partially filled 4f inner shell is responsible for the spectroscopic properties of the lanthanide ions. Owing to the rich energy level structure described by the Dieke diagram,[90] lanthanide ions are able to demonstrate major photon quantum processes such as downshifting, quantum cutting, and up-conversion, which are proposed for spectral conversion applications.[91] Nd^{3+}-doped glasses are first examples of LSCs.[92] In the current utilization of lanthanide ions as luminescent species, organic ligands are used to coordinate the lanthanide ions to boost their absorption efficiency based on the energy transfer mechanism.[93] Eu^{3+} complexes are the mostly studied lanthanide ions for the LSC research at the current stage. Most Eu^{3+} complexes exhibit massive Stokes shift and the corresponding LSCs exhibit zero reabsorption; however, the very limited absorption largely limits the realization of high-efficiency LSCs by Eu^{3+} complexes (Fig. 6).[94–99] Lanthanide ions with up-conversion characteristics could help to further increase the performance of the high-efficiency LSCs.[100]

Besides organic dyes, quantum dots, and lanthanide ions, organic–inorganic hybrid perovskite nanocrystals emerge as new luminescent species for the LSC in recent years.[101–104] Various unique spectroscopic properties can be achieved by changing the halide components in these perovskite nanocrystals, including broad absorption,[102] sharp emission,[104] and minimal spectral overlap.[103]

Host Materials

The selection of host materials is another important factor affecting the performance of the LSCs. First, the host materials should be optically transparent in visible and near infrared regions. Second, they should be compatible to the luminescent species and disperse the luminescent species evenly. Third, as the spectroscopic properties of luminescent species—especially emission spectrum and PLQY—are sensitive to the medium,[105,106] a good host material should allow the luminescent species to exhibit the optimal spectroscopic properties to the LSCs. Fourth, ideal host materials should also possess easy processability for waveguide or thin-film fabrication. For host materials used as waveguides, the refractive index (n) is an additional consideration. It can be used to estimate the

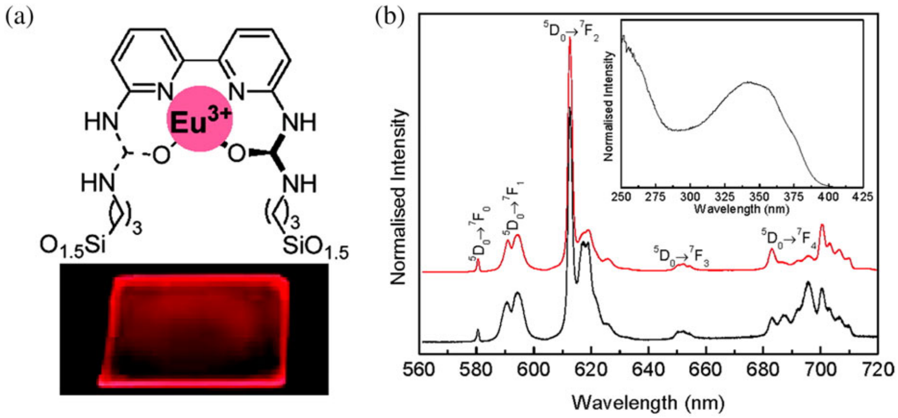

FIGURE 6 (a) An $\mathrm{Eu^{3+}}$ complex for luminescent waveguide and (b) absorption (inset) and emission spectra of the $\mathrm{Eu^{3+}}$ complex (Reproduced from ref. 94, with permission from Copyright © 2011, American Chemical Society.).

surface reflection (R) and trapping efficiency (η_{trap}). Under normal incidence, Fresnel equations can be simplified as follow to represent R (eq. 3).

$$R=\left(\frac{n-1}{n+1}\right)^2 \tag{3}$$

Not all luminescent light is trapped inside the waveguide, surface loss is common for the LSCs. According to the Snell's law, integrating outside the critical angle (θ_c) in spherical coordinates produces η_{trap} (eq. 4).[107]

$$\eta_{\text{trap}}=\frac{\int_0^{2\pi} d\varphi \int_{\theta_c}^{\pi-\theta_c} \sin\theta d\theta}{\int_0^{2\pi} d\varphi \int_0^{\pi} \sin\theta d\theta}=\cos\theta_c=\sqrt{1-\frac{1}{n^2}} \tag{4}$$

Note that the above equation assumes that luminescent species emit photons isotropically within the waveguide. However, theoretical studies have shown that even if the luminescent species are isotropically dispersed within the waveguide, the emission is anisotropic. This decreases η_{trap} by ~4%.

If being used as a waveguide, the best host materials should possess a refractive index optimized for the balance between the surface reflection and trapping efficiency, which requires $(1 - R)\eta_{\text{trap}}$ to be maximized. This leads to a refractive index of 2.0.

Another important factor that needs to be considered is the light scattering of the host material. Light scattering is characterized in many studies; however, the origin of the light scattering is not very clear at the current stage.[103,108] Possible light scattering sources in host materials could be local defects, such as microsize air bubbles, polymer aggregates, and rough waveguide surface, which are inevitably formed during device fabrication.[34,109] In small devices, light scattering may contribute to increasing the device performance,[110] whereas for large-area LSCs, it may be deleterious to the device performance, due to the interference of the photon transport within the waveguide.[34] Based on the literature search, no evidence shows that light scattering by host materials is deeply studied in LSC research. Research efforts in this area are urgently needed.

If only considering the optical transparency, glass might be the perfect material for being the host material. However, huge drawbacks are associated with glass in terms of the compatibility of luminescent species and difficulty in process and fabrication. Polymers are currently the most widely used host materials for the LSCs because of high transparency and easy processability (Fig. 7).[111] Polyacrylates, polysiloxanes, and polycarbonates are three popular polymer scaffolds as host materials in the LSCs. Polyacrylates possess refractive indices of *ca.* 1.5. They can be thermally or UV polymerized from the acrylate monomers,[22,112] or they can be thermally treated to other shapes.[45] They are typically rigid in bulk form or can be

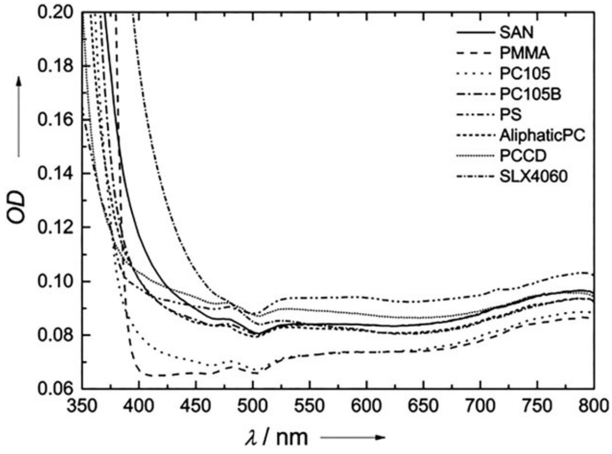

FIGURE 7 Measured optical density (OD should be considered as absorbance) for some host materials at thickness of 3.2 mm. SAN, styrene acrylonitrile resin; PMMA, poly(methyl methacrylate); PS, polystyrene; and five different formulations of PC (PC105, PC105B, AliphaticPC, PCCD, and SLX4060) (Reproduced from ref. 111, with permission from Copyright © 2017, John Wiley & Sons.).

flexible if made into fibers.[113,114] Polysiloxanes possess refractive indices of 1.4–1.5. Though the trapping efficiency of polysiloxanes would be lower than that of polyacrylate, the most impressive advantage of polysiloxanes is the high mechanical compliance that enables bulk flexibility and stretchability.[40,115] Polycarbonates possess refractive indices of *ca.* 1.7. They exhibit the same transparency as polyacrylates but provide higher trapping efficiency than polyacrylates and polysiloxanes.[111,116]

Details about the polymers as host materials will be discussed in Section 3.1.

Solar Cells

In LSCs, the luminescent waveguide is responsible for concentrating the radiation, while the edge-attached solar cells are responsible for converting the concentrated radiation into electricity. Both theoretical and experimental studies have shown that solar cells can exhibit improved performance under concentrated radiation or optimal light conditions.[117,118] Detailed balance-derived power conversion efficiency for single-junction solar cells achieves an efficiency value of *ca.* 33% under 1 sun condition (AM1.5G, 10^3 W m^{-2}) and can boost up to *ca.* 40% under 100 suns (10^5 W m^{-2}) or up to *ca.* 45% under 10,000 suns (10^7 W m^{-2}).[117] Moreover, solar cells can exhibit higher power conversion efficiencies under indoor conditions than that usually measured under solar simulators.[118] This is because the indoor light is visible light, and solar cells respond optimally to visible light. Therefore, the performance of solar cells is expected to be improved when the solar cells are attached to the luminescent waveguide. Note that concentration (C, eq. 1) is an effectiveness factor for an LSC. Devices with $C > 1$ are considered effective LSCs because solar cells output more power than they do under the same incidence condition. A high C can make the LSC more cost-effective than using the solar cells.

It is clear that the power conversion efficiency of the LSC depends on the power conversion efficiency of the solar cell (eq. 1). A variety of solar cells have been applied to LSCs. Silicon solar cells are the most widely used because of their highly commercial availability and can be easily cut into strip size for waveguide attachment.[66,73,119–121] LSCs with efficiencies up to 4.2% have been reported using monocrystalline silicon (mc-Si) solar cells with an efficiency of 16.8%.[66] Those with efficiencies up to 5.3% have been reported using polycrystalline silicon (pc-Si) solar cells with an efficiency of 12.0%.[73] Amorphous silicon (a-Si) solar cells with 10% efficiency have been also applied to the LSCs and a previous study showed an LSC efficiency of 0.91%.[119] The second-generation thin-film solar cells have also been reported for LSCs, especially for high-efficiency devices.[26,65] They include the current record of a 7.1% efficiency LSC using 24.4% efficiency GaAs solar cells[65] and a 6.7% efficiency tandem LSC using 15.4% efficiency GaInP solar cells.[26] Moreover, research-focused third-generation solar cells have also been explored but the reports are very limited. Dye-sensitized solar cells have been attached to luminescent waveguides, but their overall LSC efficiency is below 1%.[122,123] Organic solar cells are promising candidates for LSC applications.[124,125] Current technologies have realized printing processes for solar cell fabrication.[126,127] The printing technologies may enable the solar cells to be directly printed on the edge of the luminescent waveguide. The photon loss due to the unmatched refractive index between the waveguide and solar cells and the absorption by the index-matching agents can be reduced or eliminated by this approach.

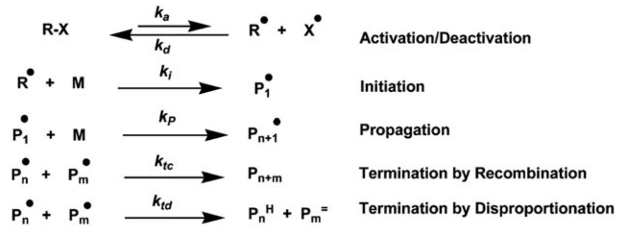

FIGURE 8 General scheme of a free-radical polymerization (Reproduced from ref. 129, with permission from Copyright © 2014, Royal Society of Chemistry.).

POLYMERS AS HOST MATERIALS

Polyacrylates

Polyacrylates are polymers synthesized from monomers containing the chemical structure of acrylate, which contains a carboxylate conjugated to a vinyl group. A general method for synthesizing polyacrylates involves free-radical polymerization (Fig. 8).[128,129] Free radicals are formed through thermal decomposition or photolysis of radical initiators. The newly formed free radicals subsequently undergo propagation by reacting with the vinyl C=C bond of the monomer to form and transfer the radicals on to the monomers. Propagation continues between unreacted monomer and the carbon free radicals extending the growth of the polymer chain, until termination occurs. Polymer growth terminates upon the occurrence of radical combination, disproportionation, or interaction with inhibitors.

Poly(methyl methacrylate) (PMMA) is a synthetic polymer generated from the methyl methacrylate monomer (MMA) (Fig. 9). It can be obtained from its monomers *via* different processes of polymerization. The monomers polymerize using the common methods of free radical and anionic initiations by bulk, solution, suspension, and emulsion polymerization.[130]

PMMA is the standard polymer matrix for the LSC due to its relatively low cost, high transparency (92% for visible

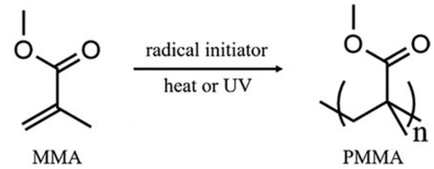

FIGURE 9 Polymerization of MMA to form PMMA.

light),[131] high refractive index (n = 1.49), satisfactory photostability,[13] chemical resistance, and excellent performance in all-weather conditions.[132] PMMA has little absorption in the visible region,[133] but it begins to show absorption at the wavelength of 730 and 800 nm,[134] which is due to the C–H bond vibrational absorption in this region.[135] As a host material, its absorption has little effect on optical efficiency for those compounds that emit in visible region, such as R305 and DCJTB. However, the host material absorption will have larger effect on the compounds that emit further into the red/infrared. To reduce the absorption, the alkyl group could be replaced by heavy atoms such as fluorine or deuterium.[136]

In addition to its promising optical properties, PMMA has excellent thermal properties. Although PMMA and glass could both be used as waveguides, PMMA can absorb and release heat at a lower rate than glass due to their thermal conductivity difference. Thermal conductivity of glass ranges from 0.8 to 1.0 W mK^{-1} while PMMA is between 0.17 and 0.20 W mK^{-1}.[48] Thus, PMMA-based LSCs have small increase in temperature with the changing of ambient temperature.

In terms of mechanical properties, PMMA has a high Young's Modulus (1.8–3.1 GPa)[137] and a low elongation at break (2.5%).[138] Additionally, PMMA does not shatter upon rupture, and is one of the hardest thermoplastics with high scratch resistance.[139] These properties will be beneficial to its application in LSCs as waveguides.

The general protocol to fabricate the PMMA-based luminescent waveguide involves mixing the luminescent species with MMA monomer and radical initiator into a flask that is treated by sonication. The flask is then moved into a heated water bath at 85 °C for prepolymerization. Stirring the mixture or frequently shaking the flask is very necessary. When the mixture becomes as viscous as glycol, it is then quickly cooled in an ice bath and loaded into a mold, that is put into an oven at a temperature around 50 °C for 2 days. Luminescent waveguide is obtained after removing the mold.[140]

Moreover, PMMA has also been used in thin-film LSCs. Common methods to achieve thin-film waveguides include spin coating[141] and doctor blading.[142] In these processes, PMMA particles are mixed with luminescent species in solvent such as chloroform or toluene and deposited onto the substrate, forming thin films after drying the materials.[143–145]

As most of the organic dyes used in LSCs are soluble in MMA, PMMA is a good host material for organic dyes.[16] PMMA is also compatible with rare-earth complexes with polar ligands. However, PMMA has an unfavorable interaction with QDs, which can result in the aggregation of QDs. To disperse the QDs, polylauryl methacrylate (PLMA) is used. PLMA will be discussed later in the article.

It is well known that PMMA possesses a high mass density of the acrylate group, and thus it is a relatively polar polymer. QDs are usually coated with nonpolar hydrophobic organic ligands, such as trioctylphosphine oxide (TOPO),[146] tributylphosphin (TBP),[89] and dodecanethiol (DDT)[147] to maintain high PLQYs. It is obvious that this polar environment of PMMA is not favorable to QDs due to the aggregation in such polar media. In most cases, the aggregation significantly affects the spectroscopic properties of the QDs, shifting the absorption and emission spectra, and significantly decreasing or completely quenching the PLQY.[148,149] To address this issue, poly(lauryl methacrylate-*co*-ethylene glycol dimethacrylate), abbreviated as P(LMA-*co*-EDGMA), is selected as an alternative to PMMA as the host material for the QD-based LSCs because it possesses similar high transparency and the same refractive index as PMMA.[150] There are two components in P(LMA-*co*-EDGMA) (Fig. 10). Component LMA has a dodecyl chain, which can interact with the hydrophobic ligands of the QDs. This interaction helps the QDs disperse well without any loss of PLQY.[150] To create a stronger polymer, LMA needs to be copolymerized with another component EGDMA, which serves as a crosslinker in the polymer.

p(LMA-*co*-EGDMA) can be used to make conventional LSCs in which the polymer also serves as a waveguide.[10,15,16,84,150] In the general protocol to make the waveguide, the monomer precursor of LMA-*co*-EDGMA is mixed at a mass loading of ~20%, followed by adding QDs. Afterward, UV initiator is added to the mixture. To obtain a clear solution, ultrasound treatment is needed. Finally, the mixture is illuminated by the UV lamp in a glass mold.[89]

Moreover, p(LMA-*co*-EGDMA) can be applied to the fabrication of thin-film LSCs. Specific protocol to make the QDs/P(LMA-*co*-EGDMA) composite thin-film devices is as following. The QDs solution is added to the polymer precursor solution. The mixture solution should be treated with ultrasound to obtain a clear solution. Several droplets of the resulting mixture are placed onto a clean borosilicate glass coverslip, which is then covered by a second coverslip. The resulting glass/solution/glass samples were photopolymerized with UV light under nitrogen for 1–3 h to form a solid film with a thickness of ~100 μm.[85,151]

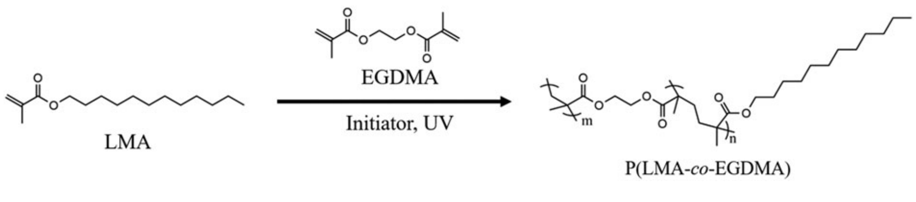

FIGURE 10 p(LMA-*co*-EGDMA) synthesized from LMA and EGDMA.

By using LMA-co-EGDMA as the host material, the performance of the LSC may be enhanced especially for large-scale devices because of the low attenuation coefficient and optical scattering portability of the polymer matrix.[152]

Polysiloxanes

As discussed in the previous section, although LSCs potentially makes the application of photovoltaics favorable in the built environment, the conventional designs using rigid materials such as rigid polyacrylates or glasses might limit some application where flexible shapes are needed. Switching from rigid substrates into flexible ones becomes the most promising solution. Since 2012, studies have shown that polysiloxanes, which is also known as silicones, is a novel flexible waveguide material to enable such application.[40,56,57,68,110,115,153–155] Additionally, their good optical[56,153] and mechanical properties[40,115,156] and good photostability make them competitive to polyacrylates and polycarbonates. In this section, we will mainly discuss two commercialized polysiloxanes:polydimethylsiloxane (PDMS) and ureasil, which are widely used as waveguides in LSCs.

PDMS is the primary material under the category of polysiloxanes. The Si–O backbone provides PMDS with good mechanical properties, such as flexibility and mechanical resistance,[156] chemical resistance, and thermal stability[40] (Fig. 11). Compared to the preparation of ureasil-based LSCs, which will be discussed later, the fabrication of PDMS-based LSCs is pretty simple, typically, by mixing the dye molecule solution with the mixture of the prepolymer and the curing agent (Sylgard 184, Dow Corning) at 10-to-1 ratio. After curing in the mold, it forms a flexible slab, which can be constructed into a circular shape.[153] As for optical properties, except the highly transparency, PDMS also possess low refractive index (n = 1.41) with a low reflection possibility (R = 2.9%).[157] LSCs with a high power conversion efficiency of 5.23% ± 0.01% were obtained, which was due to the segregated dyes in the PDMS waveguides inducing strong scattering effects in the long-wavelength range.[110] Moreover, compared to PMMA, an increased Stokes shift in PDMS was observed, which will reduce the reabsorption and further increase the performance of the LSCs.[40] One must take into account of the concentration of the dye molecules. At higher concentrations, quenching effects are apparent, which is ascribed to the lower solubility of the dye into the PDMS.[57]

Ureasil is another siloxane-based polymer that has been applied as waveguide materials into the LSCs due to high transmission,[56] flexibility,[115] and stability.[97] The chemical structure of ureasil is an organic–inorganic hybrid that consists of poly(ethylene oxide) (PEO) or poly(propylene oxide) (PPO) block copolymer chains and a siliceous network. Based on the structure and composition of the Jeffamine precursor, ureasils can be further classified into di-ureasil hybrids and tri-ureasil hybrids (Fig. 12).[68] After reacting with 3-(triethoxysilyl)propyl isocyanate (ICPTES), di-ureasil forms a linear structure, while tri-ureasil forms a tripodal structure. A general protocol to prepare the ureasil-based luminescent waveguide is by adding the stock solution of dye molecules into the di-ureasil (e.g., d-UPTES) or tri-ureasil (e.g., t-UPTES) solution before the gelation, which is catalyzed by hydrochloric acid.

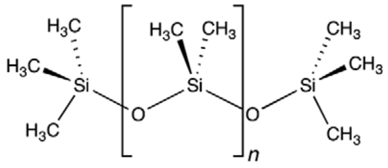

FIGURE 11 Chemical structure of PDMS.

The formation of the silica network *via* sol–gel chemistry by hydrolysis and condensation allows for properties of both components, such as mechanical, thermal, and photostability.[56,158] Although there are limited papers discussing the mechanical properties on ureasil-based LSCs, the alternating Si–O network enables them to possess better flexibility and stretchability compared to PMMA. For d-U(600) which is prepared with Jeffamine ED 600 (Fig. 12), the initial degradation temperature showed it could withstand more than 320 °C. This temperature is beyond the typical operation of organic electronic devices, which is 65–140 °C.[56] No obvious optical degradation was observed after exposure to accelerated UV light.[56,97] The transmission could reach as high as 80% with the refractive index around 1.5 in the visible/NIR region (Fig. 13). This relatively high transmission and good value of the refractive index can effectively reduce the optical losses. When compared to PMMA, ureasil can absorb at wavelengths below 360 nm. Transmission as high as 80% may not be ideal for large LSCs, because trapped light will be attenuated fast within the waveguide.

Ureasil can be easily processed as monolith,[56,68,155] film,[97,159] and coating.[43] When incorporated with QDs, it can also prevent the formation of clusters or aggregates[97,155] and facilitates more effective dispersion of the polymer chains due to its more open and distorted internal framework.[68] Moreover, ureasil is also a photoactive component. Thus, it is possible for energy to transfer between the ureasil host and/or the donor chains to the acceptor chains of the dye molecules, which minimize reabsorption losses and covers a broader wavelength range of the solar spectrum.[68] However, this photoactive property also limits the performance of the LSC devices as the light trapped in ureasil will be absorbed instead of transferring to the edge.

Polycarbonates

Polycarbonates are polymers containing carbonate groups in their chemical structures. They are a group of thermoplastic polymers that become pliable or moldable above a specific temperature and solidify upon cooling. The main polycarbonate is produced by bisphenol A (BPA) and phosgene ($COCl_2$) (Fig. 14). Due to high-temperature (near 300 °C) processing and the toxicity of the starting materials, bulk polycarbonates are rarely synthesized in the laboratory. Reports on polycarbonates used as both host and waveguide material for luminescent species in the LSCs are rare.[66,111,160] Luminescent species-doped

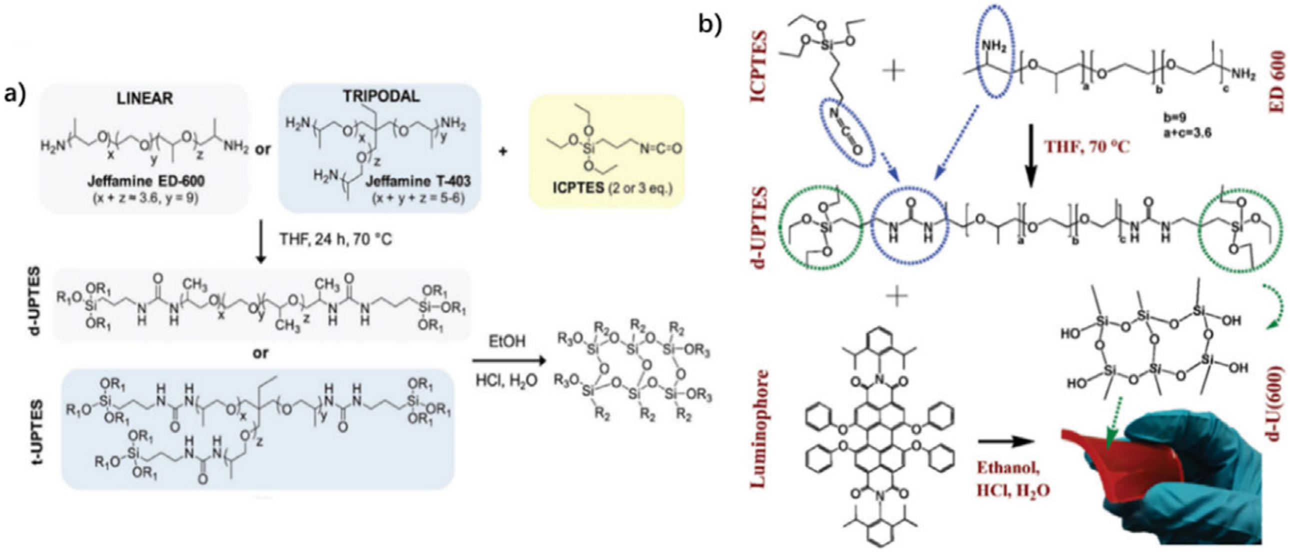

FIGURE 12 (a) Two types of ureasil and a general preparation protocol (Reproduced from ref. 68, with permission from Copyright © 2009, Royal Society of Chemistry.). (b) The structure of a di-ureasil and its preparation protocol (Reproduced from ref. 56, with permission from Copyright © 2015, John Wiley and Sons.).

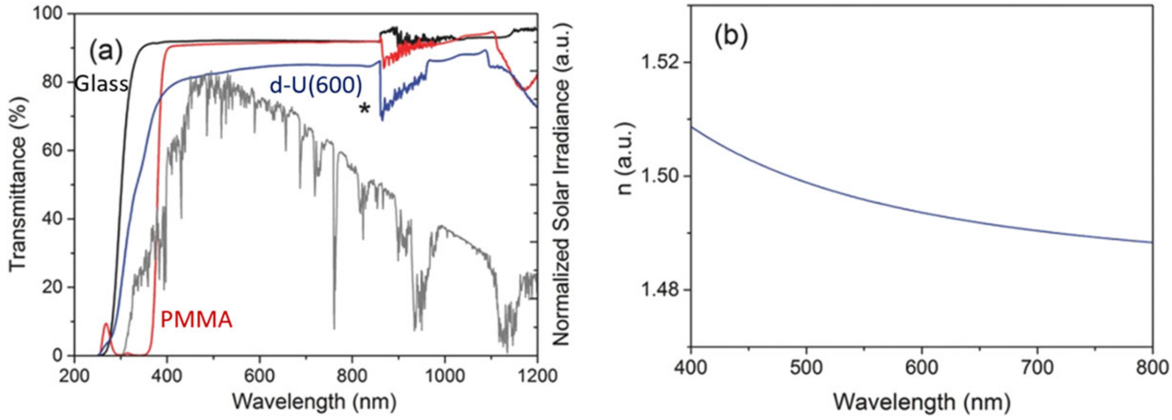

FIGURE 13 (a) Optical transmittance in the UV/Vis/NIR region of waveguide slabs made of d-U(600) (blue), PMMA (red), and glass (black). The AM 1.5G solar irradiance is shown in the background (gray). *Change of lamp. (b) Refractive index of d-U(600) across the visible region (400–800 nm) (Reproduced from ref. 56, with permission from Copyright © 2015, John Wiley & Sons.).

polycarbonates-based waveguides are usually processed under a high temperature by injection molding.[66,111] In current LSC research, polycarbonates are usually used as waveguide for thin-film LSCs, attributing to a high refractive index of *ca.* 1.7, capable of trapping near 81% emitted light inside the waveguide.[31,161] In the near-infrared region, polycarbonates exhibit similar absorption properties as polyacrylates due to carbon–hydrogen (C–H) stretching (Fig. 15).[162,163]

Other Polymers

Besides polyacrylates, polysiloxanes, and polycarbonates, there are other polymers that are used as host materials in

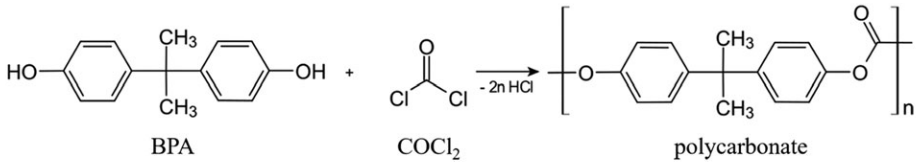

FIGURE 14 Synthesis of polycarbonate from BPA and COCl$_2$.

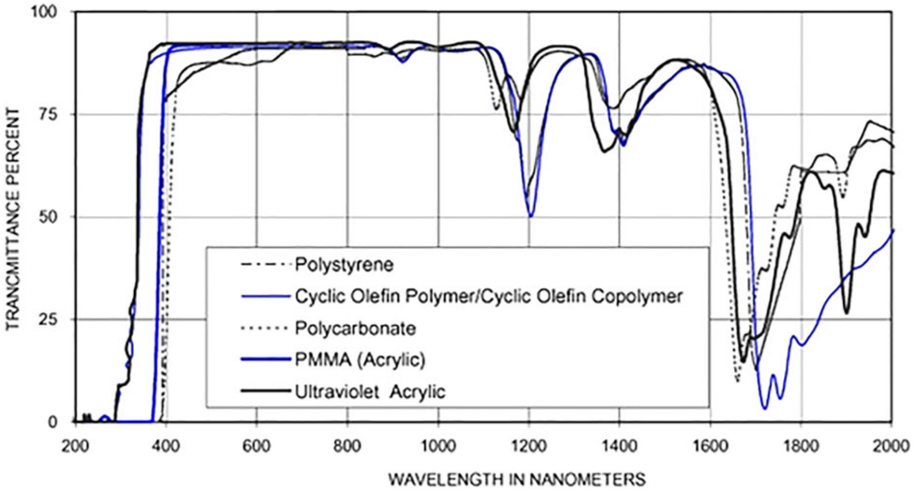

FIGURE 15 Transmission of polycarbonate compared to other polymers.[163]

LSCs. For example, polyacrylamide (PAA) hydrogels have been used for a protein-based luminescent species to provide aqueous mimic conditions.[164] Polyimide (PI) and poly(vinyl alcohol) (PVA) were used to provide specific conditions for dye alignment.[165] Poly(lactic acid) (PLA), a renewable material, has been explored as a transparent matrix for luminescent solar concentrators.[166] Biocompatible and recyclable cellulose nanocrystals have also been explored as low-cost host materials for LSCs.[167] Polyvinylpyrrolidone (PVP) was used as a host material for doctor-blade deposition of QDs onto standard window glass for low-loss large-area LSCs.[168] Epoxy resins have been reported as a host materials for LSCs, attributing to their high photostability, physical strength, and excellent adhesion to a variety of substrates.[169–171]

POLYMERS (MACROMOLECULES) AS LUMINESCENT SPECIES OR ENERGY TRANSFER MEDIA

A significant amount of work on π-conjugated polymers has been carried out in the areas of organic photovoltaics,[125] organic light-emitting diodes,[172] organic field-effect transistor,[173] and organic thermoelectrics.[174] However, there are only few studies about the direct use of π-conjugated polymers as luminescent materials in LSCs. Many π-conjugated polymers exhibit wide absorption band extending to near-infrared regions;[175–177] however, low PLQY (typically <20%) is associated with them due to the fast nonradiative decay rate of excitons. Even though there are reports about π-conjugated polymers with high PLQYs for organic light-

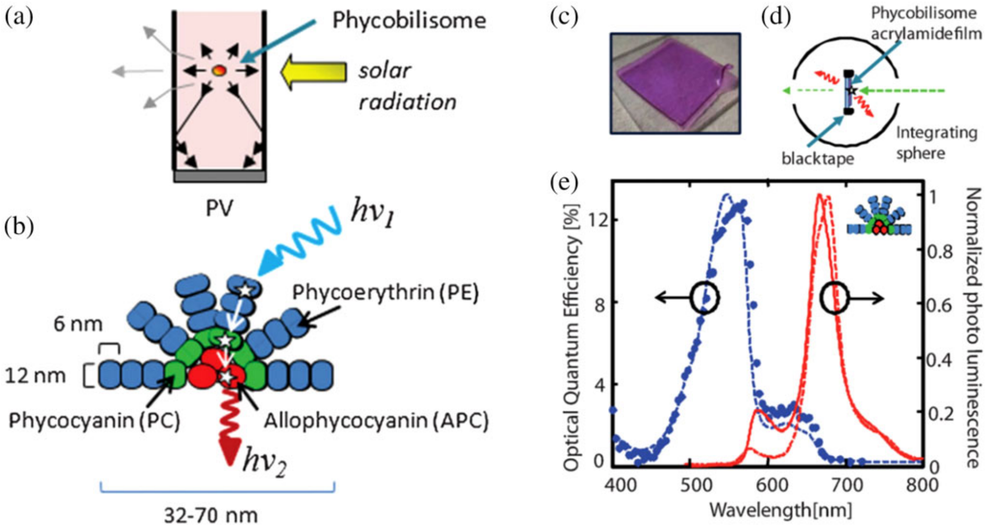

FIGURE 16 (a) Schematic representation of an LSC; (b) schematic representation of the structure of a phycobilisome; (c) photograph of a cast polyacrylamide film that contains intact phycobilisomes consisting of fully coupled PE, PC, and APC biliproteins; (d) schematic representation of the measurement set up used to determine the optical quantum efficiency of the phycobilisome film; and (e) optical quantum efficiency (blue dots) and normalized photoluminescence (red solid line) of the acrylamide phycobilisome (PE-PC-APC) films (Reproduced from ref. 164, with permission from Copyright © 2015, John Wiley and Sons.).

emitting diodes,[178] they typically emit blue light, which is not favorable for LSC application.[179]

The use of phycobilisomes, large water-soluble pigment-protein complexes, may be the first study that reports the use of macromolecules as the luminescent species for LSCs (Fig. 16).[164] In phycobilisome, hemispherically organized rods of phycoerythrin (PE) and/or phycocyanin (PC) biliproteins join a core of allophycocyanin (APC) biliproteins. Förster resonance energy transfer (FRET) occurs within the phycobilisome from PE to PC then to APC, converting short-wavelength photons to long-wavelength photons. The PLQY for PE, PC, and APC are 98%, 51%, and 68%, respectively. Polyacrylamide hydrogels were used as the host material for waveguide fabrication to mimic the native aqueous environment suitable for phycobilisomes.

Another example that includes polymers in an energy system is to use a π-conjugated polymer as energy sensitizers to transfer the energy to an organic dye.[32] The PLQY of the energy transfer system is 79%, a relatively high value compared to other luminescence species in LSCs. In addition, conjugated polyelectrolytes have been explored for luminescent waveguides.[180,181] One report using a polyelectrolyte conjugated with an organic dye unit showed some LSC performance, but still not comparable to the organic dye only system primarily due to the deficient energy transfer.[68] Beside polymers, dendrites and oligomers have also been used for LSCs in other reports. They are based on energy transfer systems.[112,169] There are also other designs such as functionalize the organic dyes to crosslink them with polymers to increase the stability of luminescence species.[69,115]

CONCLUSIONS AND OUTLOOK

In conclusion, there has been significant progress in the research of LSCs over the past decade. A variety of new LSC architectures have been developed and demonstrated advantages over the conventional LSC architecture in terms of device efficiency, optical properties, mechanical properties, and application areas. Great efforts have been made to develop luminescent species and host materials. It is expected that in the near future, with the finding of suitable luminescent species and host materials, the research of LSCs will move to a practical level where large-area devices are made and applied to the built environment.

In addition, looking forward to the practical application of this technology, we believe that much greater attention should be paid to the research of polymers in LSCs. Host materials that are compatible with QDs and retain easy processability and optical transparency would be an area requiring urgent investigation. A lot of research has been done on π-conjugated polymers for organic electronics. They could have very broad absorption spectra extending to the near-infrared region, and can maximize the absorption of LSCs. We hope this article would stimulate the research of polymers on LSCs.

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