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ADVANCED REVIEW

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Bio-based materials for solar cells

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Abstract

Plant-based materials are emerging as an alternative to conventional components in advanced energy applications. Among these, energy harvesting from sunlight is highly attractive and, in fact, represents the fastest growing energy technology. This review addresses the broad field of solar cell science since plant-based components can be utilized in almost all solar technologies, and in certain photovoltaic technologies, they can fulfill most of the roles in photovoltaic devices. There is strengthened recent interest in developing sustainable materials options as well as new functionalities being developed for bio-based materials. This contribution describes the different options for plant-derived materials in photovoltaics and discusses their deployment feasibility. We focus on performance, lifetime, and embedded energy, all of which are critical to achieve—economically and sustainably–competitive photovoltaic devices. We address the tendency in the current literature for greenwashing, given that not all plant-based solutions are environmentally-sound at the device level. On the other hand, plant-based materials can offer functionalities that cannot be reached with currently used materials.

Abbreviations: Ag Nw, silver nanowire; CNC, cellulose nanocrystals; CNF, cellulose nanofibers; DSC, dye-sensitized solar cell; FTO, fluorine-doped tin oxide; HTM, hole transport material; ITO, indium tin oxide; NCP, nanocellulose paper; OSC, organic solar cell; P3HT, poly (3-hexylthiophene-2,5-diyl); PBDTTT-C, poly[(4,8-bis-(2-ethylhexyloxy)-benzo[1,2-b:4,5-b′]dithiophene)-2,6-diyl-alt-(4-(2-ethylhexanoyl)-thieno [3,4-b] thiophene)-2,6-diyl]; PCBM, phenyl-C61-butyric acid methyl ester; PEDOT:PSS, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; PEIE, polyethylenimine ethoxylated; PEN, polyethylene naphthalate; PET, polyethylene terephthalate; PMMA, poly(methyl methacrylate); PSC, perovskite solar cell; PTB7, poly({4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b′]dithiophene-2,6-diyl}{3-fluoro-2-[(2-ethylhexyl)carbonyl] thieno[3,4-b]thiophenediyl}); PV, photovoltaic; PVF, polyvinyl formal; R2R, roll-to-roll; REI, return of energy investment; RMS, root mean square; Spiro-OMeTAD, $N^2,N^2,N^{2\prime},N^{2\prime},N^7,N^7,N^{7\prime},N^{7\prime}$-octakis(4-methoxyphenyl)-9,9′-spirobi[9H-fluorene]-2,2′,7,7′-tetramine; TCO, transparent conductive oxide; TOCN, TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl) oxidized cellulose nanofiber; TW, transparent wood; UV, ultraviolet.

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KEYWORDS
cellulose, solar cells, substrate, wood-based materials

1 | INTRODUCTION

Bio-based materials, including plant biomass, have been the primary source of energy for humankind throughout its existence. In the context of modern society, solar power is the only renewable energy source with the potential to satisfy the global energy demand. Meanwhile, beyond its traditional energy use, renewable plant-based materials can serve as building blocks for solar cells. The opportunity to utilize plant-based materials is appealing since they can fulfill almost every role in photovoltaic devices, as illustrated in Figure 1. They can be integrated as passive (support, substrate, electrolyte scaffold, or insulator) or active (photo-electric converter, catalyst, or conductor) elements. Plant fibers, paper, wood, and wood composites, woven and nonwoven textiles, as well as emerging nanocellulose in the form of hydro- and organogels, membranes, and thin films, can fit some of the requirements of photovoltaics (Figure 1).

This review discusses different options for plant-derived materials in photovoltaics and their deployment opportunities. Photovoltaics have emerged as a pivotal force in the global energy transition, with significant strides attributed to advancements in materials and manufacturing methodologies. The evolutionary trajectory of solar cell technology encompasses three main generational phases since its invention. First generation solar cells, predominantly composed of monocrystalline and polycrystalline silicon, constitute over 80% of residential solar panels. Notably, the record

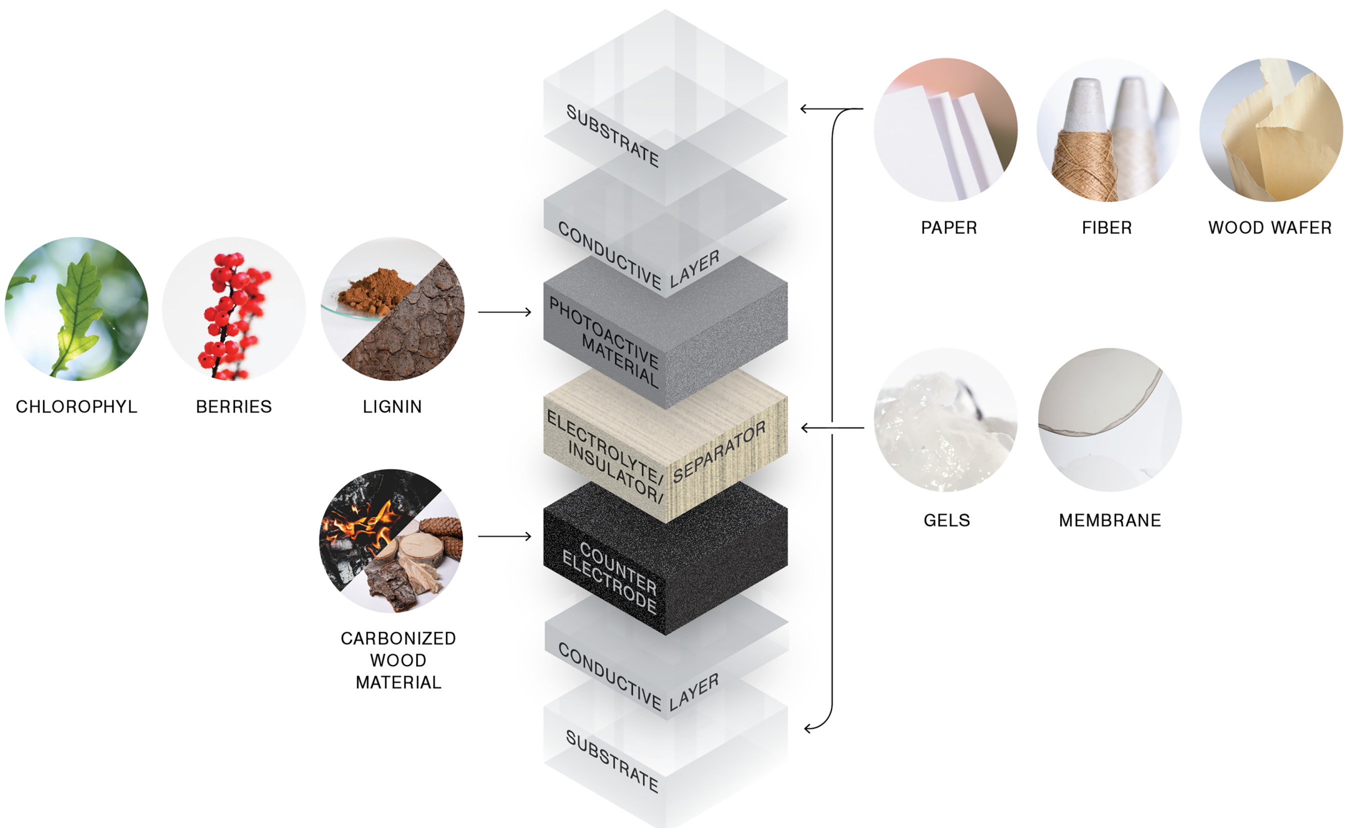

FIGURE 1 Schematic illustration of possible passive and active plant-based components in photovoltaics.

efficiency for crystalline solar cells stands at 27.6% (NREL, 2023). Second-generation solar cells, also known as thin-film solar cells, are made from ultra-thin layers, typically only a few micrometers thick. Solar cells based on amorphous silicon, cadmium telluride (CdTe), and copper indium gallium diselenide (CIGS) are included in this generation.

Currently, considerable research is devoted to the development of third-generation solar cells. These emerging technologies encompass a diverse array of semiconductors, incorporating organic dyes, perovskites, organic polymers, and quantum dots (Kayes et al., 2011). The primary objective for third-generation solar cells is to attain high power conversion efficiency while ensuring cost-effectiveness in production processes through different approaches, such as printing, with perovskite solar cells achieving an efficiency of 26.1% (NREL, 2023).

Plant-based materials can offer functionalities that are not possible for conventional materials. For instance, the optical properties of structures based on wood, cellulose nanofibers, and nanocrystals, can be tailored to surpass those of glass and common plastics. Cellulose aerogel scaffolds offer a way to semi-solidify an electrolyte to reduce performance losses incurred when upscaling dye solar cells and industrial. Furthermore, this review identifies which plant-based materials are already viable for PVs and which are the most promising areas for future development. To avoid creating unrealistic expectations, often leading to *greenwashing*, we introduce a framework to obtain an accurate perspective as far as the benefits that can be achieved by adopting plant-based materials. Technologies are often compared merely based on efficiency or overall costs. These factors omit the aspect of lifetime, which is important to consider when applying plant-based materials since they can impact long-term stability significantly. One option for such a framework is to study which of them would increase the *return on energy investment* (REI), defined as the energy supplied by the PV device over its lifetime normalized by the amount of energy required for its manufacture (embedded energy). To attain sustainable/clean energy production, we propose that plant-based solutions should be critically compared based on the three main factors affecting REI, namely, performance, lifetime, and embedded energy.

Based on the three above mentioned criteria, the following sections evaluate the use of plant-based materials as a PV substrate, counter electrode, electrolyte scaffold, and photoactive component. This is presented as a one-of-a-kind, holistic analysis for consideration of plant-based materials in solar cells. We identify and describe properties of biomaterials that, even if not exploited at present, could be beneficial in PV devices.

2 | SUBSTRATES

2.1 | Solid films

Transparent conductive substrates bring a significant contribution (often over 50%) to material and environmental costs, in particular for emerging photovoltaics (Freitag et al., 2017; García-Valverde et al., 2010; Parisi et al., 2014). Plant-based materials have intrinsically a low embedded energy but savings in energy and cost can be gained since their use requires reduced processing temperatures, provided that opting for low temperature processing retains a good device efficiency. Moreover, they are suitable for roll-to-roll mass production. Ideally, as a substrate, the material should be transparent, stable, lightweight, and highly conductive. Furthermore, to increase device lifetime, the substrates should be good barriers for gas (air, O_2) and water (liquid and vapor) transport. In addition, the substrates should shield the device from external stressors, such as heat and UV light.

Light management characteristics of paper films produced from cellulose fibers and nanofibers (CNF) stem from the unique opportunity to engineer light scattering phenomena via air/fiber interfaces, which can result in light transmission ranging from ultra-high opacity to diffusive or complete transmittance (Jacucci et al., 2021; Toivonen et al., 2018). Low light scattering is possible with CNF films and most of it in the forward direction (i.e., haze), whereas conventional paper, with larger pores increases backscattering (Toivonen et al., 2018). The transparency of cellulose-based films increases by impregnation with refractive index-matching materials (e.g., oils, resins, and waxes) (Jacucci et al., 2021). Recently, an all-cellulose composite material consisting of softwood cellulose nanofibril bundles and regenerated cellulose, was shown for its excellent transparency (90.1%) and transmission haze (95.2%) (Hou et al., 2020). In addition, the promising mechanical and water-resistant properties of related films highlight the untapped potential of the all-cellulose materials for light management in photovoltaics.

Haze has been reported as a desirable optical property in bio-sourced substrates (Fang et al., 2014; Hou et al., 2020; Zhu, Li, et al., 2016). The reason is that light is scattered by the cellulose structure, thus, having a higher chance to interact with the active layer, leading to an increased current produced by the device and its overall efficiency. Haze can also backscatter light, reflecting it away from the active layer and this aspect requires more research from PV application perspectives. Tailoring the refractive index can reduce the share of reflected light and increase the intensity of the light entering the device (Azzam et al., 2014).

Wood can be directly converted into a transparent substrate through a process that includes chemical removal of lignin followed by filling fiber lumens with a refractive index-matching polymer, making the composite transparent, often referred to as transparent wood (TW). Perovskite solar cells (PSCs) have been prepared directly on transparent wood reaching the efficiency of 16.8%, which is quite close to a similar device built on glass with an efficiency of 18.9% (Figure 2) (Li et al., 2019).

The increased thickness of the transparent wood significantly reduces the optical transmittance, for instance, an increase from 1.1 mm to 2.2 mm reduces transparency by over 10% at 550 nm (Chen et al., 2019). Also, directionality matters, since TW optical and mechanical properties highly depend on the direction of the cellulose fibers, giving rise to an anisotropic behavior. Optically, there is only a 20% difference in transmittance depending on the incidence of light with respect to the fiber orientation in the visible light range. However, the mechanical strength is twice when stretched along the cellulose fiber's orientation rather than perpendicularly (Zhu, Song, et al., 2016). Thus, one challenge with TW is to maintain sufficient structural integrity while keeping the highest optical performance. Another major challenge of TW films concerns the scalability as well as limited control over the size range of lumen, since the main starting material for TW composites are wood wafers. These challenges can be overcome by creating transparent anisotropic polysaccharide cryogel composites (Zou et al., 2022), which allow a simpler fabrication process (no need for lignin removal) and a versatile tuning of optical properties.

The critical evaluation of composite substrates combining wood-based components and synthetic polymers must consider the stability and lifetime of the resulting photovoltaic devices as well as the environmental impact. It is possible to tailor optical transmission, barrier, processing, and recycling/disposability properties of films based on CNF and CNC. For example, higher elastic modulus as compared to both the filler material and of the delignified wood template of the transparent wood has been demonstrated by using PMMA as impregnation material (Li et al., 2019). Transparent wood also lends itself for functionalization, for instance, to down converting or upconverting nanoparticles. For instance, luminescent transparent wood based on quantum dots has been demonstrated (Fu et al., 2020; Li et al., 2017). A good mechanical stability (bending) of flexible solar cell on nanocellulose substrates has been achieved (Gao et al., 2019). However, solar cells in particular for long-term outdoor use require very high vapor barrier properties. Substrate optimization in terms of moisture penetration is pivotal to increase the device's lifetime.

Aside from high transparency, a substrate for photovoltaic and optoelectronic applications must allow the deposition of functional layers. The fabrication includes the deposition of an electrically conductive layer, which is highly affected by the substrate's surface roughness. High surface roughness is one of the main factors that impact the quality of the deposited electrodes (Hu et al., 2013). An uneven surface can lead to irregular regions with varying electrical conductivity, creating hotspots and lower-than-average conductivity, or, if the roughness is high enough, the substrate can prevent the formation of an electrically continuous layer (Jonda et al., 2000). Furthermore, protrusions on the substrate can cause certain regions of the electrode to penetrate to subsequent layers, short-circuiting the device making it useless.

Cellulose substrates tend to have high surface roughness compared to commonly used substrates like glass, PET, and PEN. Variation in the surface roughness depends on the size of the cellulose fibers used in the formation of the films, going from rough regular paper to nano-sized cellulose films (Jonda et al., 2000; Zhou et al., 2013). Among the smoothest types of cellulose, one can cite films of neat CNC (roughness values as low as 1 nm) (Raghuwanshi &

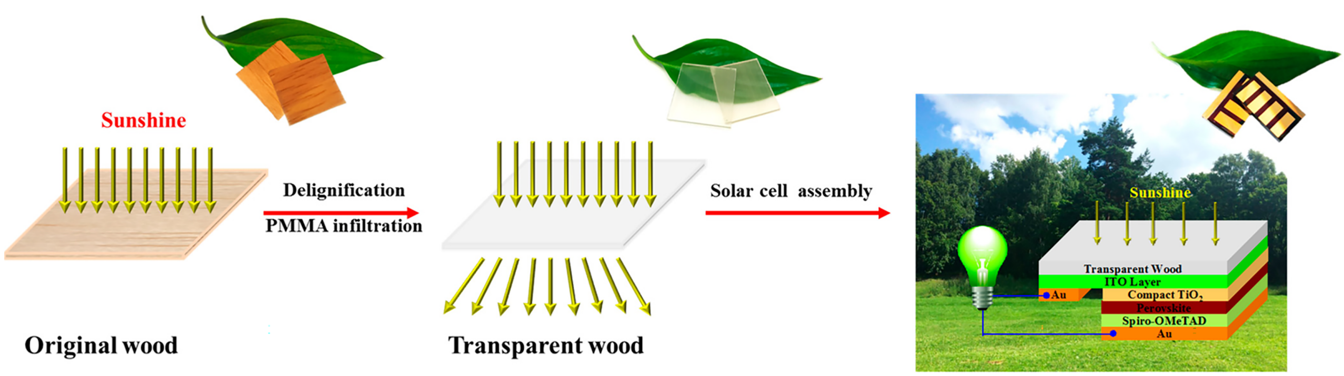

FIGURE 2 Transparent wood used as a substrate for PSCs. *Source*: Reprinted under CC-BY-NC-ND license (Li et al., 2019).

Garnier, 2019). This smoothness is possible due to the CNC compactness, forming very densely packed structures (Leppänen et al., 2022). Although they are very smooth, CNC films suffer from high brittleness, hindering their use in high volume fabrication processes like roll-to-roll manufacturing (R2R). One option is the introduction of CNF, since it tends to create a network of long fibrils providing high elasticity and bendability compared to neat CNC films, but this comes at the expense of potentially increasing the surface roughness (Leppänen et al., 2022).

Another challenge in the deposition of functional layers is the possible effect of liquids, particularly solvents, on the cellulose substrate. Solution-based fabrication methods depend on depositing materials on a substrate through a liquid medium. This liquid tends to be absorbed by the cellulose fibers, swelling the whole film, preventing further deposition of functional layers. To bypass this issue, a smoothening layer can be inserted between the cellulose films and the given functional layer, typically the electrode. This approach (1) can protect the substrate from entering in contact with liquids that swell or dissolve cellulose; (2) can act as a smoothening layer to reduce the overall surface roughness of the substrate; (3) it can increase the substrate gas barrier properties (Vartiainen et al., 2020) (Table 1).

2.2 | Woven substrates

PV textiles have great potential to produce self-powered, flexible devices (Mather & Wilson, 2017). Textiles represent scaffolds with a hierarchical, multi-level structure (from fiber, yarn, and fabric to actual wearables). There are various approaches to fabricate PV textiles: coating or doping the filaments with distinct PV constituents that can be further knitted or woven into a textile (interlaced configuration, Figure 3). Alternatively, the collective filaments or yarns can be used as the substrates to form an entire solar cell, for example, by coating all necessary layers, (coaxial configuration, Figure 3). So far, PV textiles have been broadly researched to enhance their photovoltaic efficiency. However, there is still a major gap between high-performance systems and their practical applications (Zhai et al., 2022).

Very few attempts have been made to apply biopolymers as a woven substrate for PV (Ebner et al., 2017). Most efforts to develop PV textiles are based on metal, glass, or synthetic polymer wires and filaments (Ebner et al., 2017; Hatamvand et al., 2020; Yun et al., 2016). These substrates are suitable for PVs because they are typically homogeneous, continuous, and present smooth surfaces. However, metal and glass wires are not flexible enough to meet the requirements for further fabric processing and they are prone to break under conditions of practical textile use. By contrast, natural fibers, usually soft and flexible, are relatively short and must be processed into a continuous yarn to turn them into a textile. Such yarns present uneven surface textures, which is disadvantageous for many coating procedures. So-called manufactured cellulosic fibers (MCFs), such as viscose or Tencel™ filaments, circumvent such obstacles. MCFs are prepared by dissolving cellulose and spinning them into uniform, continuous filaments with diameters typically ranging from 10 to 30 μm (Wojnowska-Baryła et al., 2022). The increasing awareness of the environmental impact of the textile sector has led to the development of alternative, more benign spinning technologies (Hummel et al., 2015; Wojnowska-Baryła et al., 2022). Their large-scale commercialization would reduce the environmental impact of fiber production while at the same time offering options to introduce a certain functionality. For instance, by producing hollow filaments, double structures are accessible, making it possible to locate one electrode on the inside of the fiber and the other on the outside—turning the fiber into a bi-functional system, that is, as a substrate and as an insulating layer (Reyes et al., 2022). Coaxial spinning offers the alternative to prepare shell-core architectures with compatible inner and outer layers (Lundahl et al., 2018). This method involves the layer-by-layer assembly of distinct structures onto a fiber-type substrate, including the outer layer of an electrode material. In a twisting configuration (Figure 3), a wire-shaped electrode is twisted around a fiber that is already coated with functional layers. Such methods have already been successfully utilized in fiber-based PVs fabrications (Hatamvand et al., 2020). In addition, MCFs allow the incorporation of substances directly into the fiber to add given functionalities, such as conductivity (Haslinger et al., 2020; Lund et al., 2018).

Due to their reduced manufacturing costs and modular adjustability, third-generation photovoltaic devices, such as PSCs and organic solar cells, are promising candidates for fiber and filament-shaped PV structures. However, one of the main challenges is that the multilayer structure of these cell types requires evenly applied coatings, at a commercially viable speed, and at operating temperatures limited by the moderate thermal stability of bio-based fibers. Moreover, fiber-based PVs possess limitations such as significant performance loss by increasing the length of the textile PV, which cannot be fully overcome by replacing a conventional substrate with a bio-based counterpart (Zhai et al., 2022). Additionally, subsequent weaving and knitting processes required to turn the constituent PV filaments into a flexible textile generate high stress through friction and bending. Thus, inter-layer adhesion and flexibility are critical to maintain the integrity of all the PV components. For instance, transparent conductive oxide (TCO)-free DSCs have been proposed to avoid the shortcomings of the brittle TCO layer (Yun et al., 2016). Further, the stability of the devices under deformations such as

TABLE 1 Solar cells built on bio-based substrates.

Substrate	Transparency of the conductive electrode (%)	Surface roughness of the substrate (nm)	Conductive layer	Conductive layer sheet resistance ($\Omega\ sq^{-1}$)	Device configuration	Efficiency (%)	Active area (mm^2)	Reference
Perovskite solar cells								
Paper	–	<20	Au	0.24	Paper/Au/SnO_2/meso-TiO_2/$CH_3NH_3PbI_3$/Spiro-OMeTAD/MoO_3-Au-MoO_3	2.7	10	(Castro-Hermosa et al., 2017)
CNC	80	–	PEDOT:PSS	200	CNC/PEDOT:PSS/$CH_3NH_3PbI_3$/PCBM/Au	5.41	4	(Yusoff & Nazeeruddin, 2017)
NCP	91.71 at 550 nm	2.151 (RMS)	PEDOT: PSS	–	Paper/Acrylic resin/PEDOT:PSS/PEDOT:PSS 4083/$CH_3NH_3PbI_3$/PCBM/Al	2.25	–	(Gao et al., 2019)
Regenerated cellulose	80 at 550 nm	–	Ag NWs	29	Cellulose—Ag NWs/PEDOT:PSS/$CH_3NH_3PbI_3$/$PC_{61}BM$/Al	4.49	6	(X. Ma et al., 2019)
TW	86 at 550 nm	30	ITO	–	TW/ITO/Compact TiO_2/Perovskite/Spiro-OMeTAD/Au	16.8	–	(Li et al., 2019)
Organic solar cells								
CNF	92.8 at 600 nm	–	Ag NWs	43	CNF/Ag NWs/PEDOT:PSS/P3HT/PCBM/Al	3.2	9	(Nogi et al., 2015)
TOCN	78.5 at 550 nm	28 (RMS)	Ag NWs	2.62	TOCN/AgNWs/ZnO/PM6:Y6/MoO3/Ag	7.47	4	(Lin et al., 2021)
Paper	–	2.6	Ag	0.3 ± 0.1	Paper/PVF/Ag/ZnO/P3HT:PCBM/PEDOT:PSS	3.37	16	(Rawat et al., 2019)
					Paper/PVF/Ag/ZnO/PTB7:PCBM/PEDOT:PSS	6.44	16	
	–	2.6 ± 0.2	Ag	–	Paper/PVF/Ag/ZnO/P3HT:PCBM/PEDOT:PSS	2.38	10,800	(Jayaraman & Iyer, 2020)
					Paper/PVF/Ag/ZnO/PTB7:PCBM/PEDOT:PSS	4.23	10,800	
CNC	55–75 at 550 nm	1.8 ± 0.6 (RMS)	Ag-PEIE	–	CNC/Ag-PEIE/PBDTTT-C:PCBM/MoO_3/Ag	2.7	–	(Zhou et al., 2013)

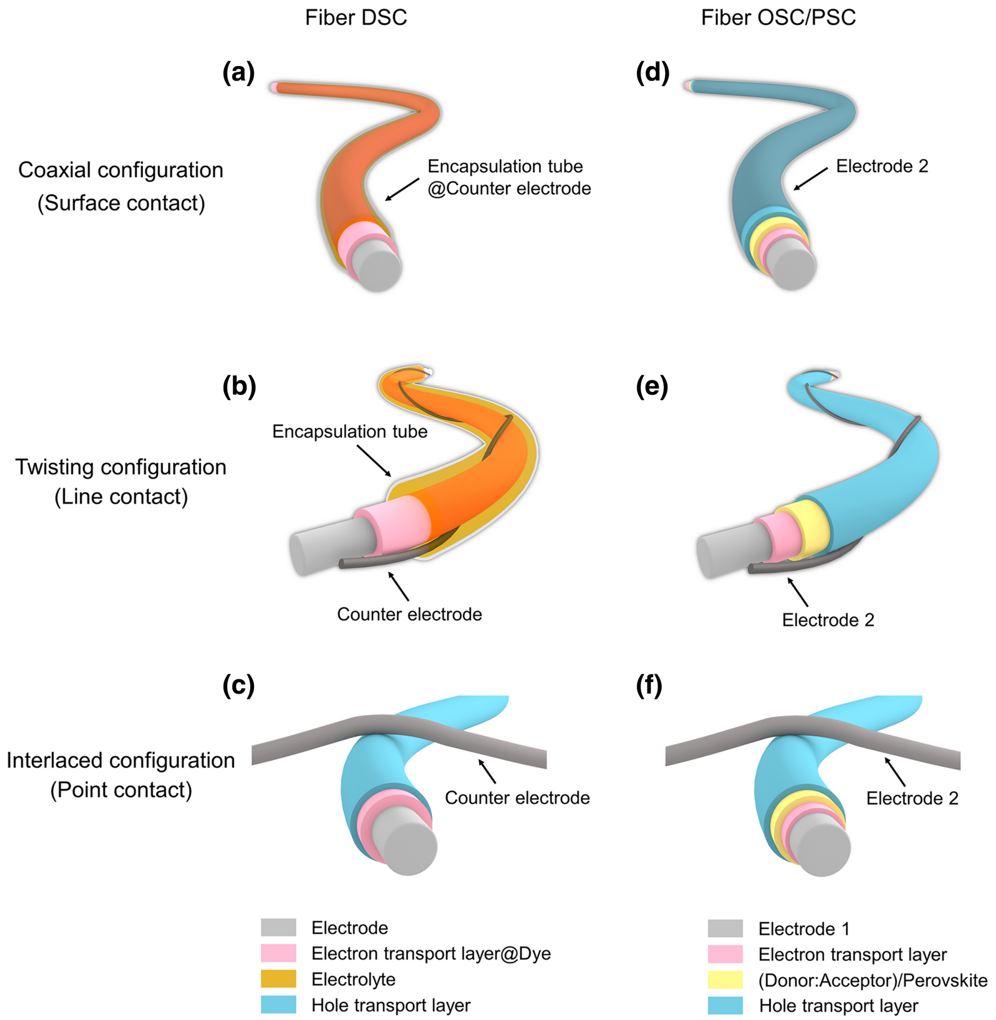

FIGURE 3 Schematics of different configurations of fiber-type PVs for DSCs (a–c) and OSCs/PSCs (d–f); a coaxial fiber DSC (a), a twisting fiber DSC (b), an interlaced fiber DSC (c), a coaxial fiber OSC/PSC (d), a twisting fiber OSC/PSC (e), and an interlaced fiber OSC/PSC (f). *Source*: Image reprinted with permission from Zhai et al. (2022).

bending and stretching is important and should be thoughtfully evaluated. Lastly, if assembled into a wearable textile, possible effects of weathering and wear abrasion have to be evaluated. In particular chemo-mechanical stresses exerted during laundering can have a considerable impact on the macromolecular integrity of the fiber matrix (Wedin et al., 2019). Furthermore, if any breakage of the system occurs, potentially hazardous materials such as lead, or liquid electrolyte could be released to the environment. Such factors need careful consideration for any future development.

3 | CATALYSTS AND CONDUCTIVE LAYERS

Products derived from wood and plant pyrolysis can be incorporated in the conductive and catalyst layers, thus offering an opportunity to replace rare and/or expensive metals. In DSCs, the total embedded energy of the device could be reduced by almost 30% by substituting the Pt-coated fluorine doped tin oxide (FTO) counter electrode with a catalytic and conductive carbon layer (Parisi et al., 2014). In PSCs, carbon electrodes can replace both the Au-based layer and the hole-transport material (HTM), thus reducing overall fabrication costs and enabling printability of the entire cells (Raminafshar et al., 2018). Furthermore, due to the inertness and stability of the carbon layers and the elimination of the ion migration of the Au contact to the perovskite layer, utilizing carbon electrodes enhances the long-term stability of the PSCs (Hadadian et al., 2020). The application of biocarbons in photovoltaics started from DSCs. Therein, plant-based sources of carbon have included for instance wood (Xu et al., 2018), fallen foliage (Cha et al., 2018), recycled paper (Xu et al., 2018), and other plant biopolymers (Cha et al., 2018; P. Ma et al., 2018; Tiihonen et al., 2021; Xu et al., 2018). When carbonized, plant-based materials offer a variety of nanostructures. Another characteristic of biocarbons is their varying amounts of trace metals based on the type of plant and soil in which they grow. The implications of the varying composition of the biocarbons have not yet been systematically investigated.

Some studies have demonstrated the stability of bio-sourced carbon in a counter electrode-counter electrode cell in cyclic voltammetry (Cha et al., 2018; P. Ma et al., 2018). The stability of complete DSCs with biocarbons was tested in 3000 h light soaking test, which revealed that the degradation of electrolyte with the biocarbon electrolyte was even slower compared to the conventional Pt catalyst (Tiihonen et al., 2021).

Carbon materials derived from different plants such as Aloe vera, corn stalk, and *Phragmites australis* have been used as electrodes in PSCs (Gao et al., 2020; C. Liu et al., 2021). A comparison of four types of biocarbons used as electrodes revealed that parameters such as interfacial contact, sheet resistance, and morphology of the bio-carbons influence photovoltaic performance. This is scribed to the charge transport and reducing charge recombination. Aging tests have been promising as well: PSCs with biocarbon in storage for 2000 h retained 87% of their initial performance, surpassing significantly conventional PSCs based on gold electrode (Gao et al., 2020).

The efficiency of carbon-based PSCs is still far below that of PSCs that use gold and HTM due to the low conductivity of the carbon electrode and intrinsically poor contact at the interface with the perovskite. The quality of the interface between perovskite and carbon layer was improved by incorporating N, O co-doped porous carbon electrode based on KOH-activated soybean dregs in PSCs, which showed enhanced efficiency and stability (H. Liu, Geng, et al., 2022).

Recently, a large-scale (10 × 10 sq.cm) was demonstrated with HTM-free PSCs, with an interface-engineered graphitic carbon counter electrode derived from *Eichhornia crassipes* invasive plant chops. An efficiency of 6.32% and long-term stability over 50 days under different conditions such as continuous illumination and ambient air conditions were shown (23°C under 77% humidity) (Pitchaiya et al., 2022).

Bio-carbon can be prepared from low-cost sources and the processing is based on simple methods: Figure 4 shows an example of a process used to prepare a carbon counter electrode for DSCs from brewery residue (Tiihonen et al., 2021). However, the carbonization process in general requires treatment at very high temperatures (e.g., brewery waste was treated at 800°C). The porous carbon layers can be used in addition to catalyst layer also as a conducting layer. In practice, the layer needs to be typically so thick rendering it opaque, and thus it does not suit all applications (e.g., semitransparent solar cells) (Bogachuk et al., 2022; Tiihonen et al., 2021). High transparency might be achieved by optimizing approaches for enhancing the conductivity of the biocarbon electrodes.

4 | ELECTROLYTE AND INSULATOR LAYERS

In emerging photovoltaics, the electrodes are separated by an electrolyte/separator layer which enables fast transport of charge carriers (i.e., ion diffusion). The suitability of an electrolyte hinges on its (a) capacity as interfacial contact between the anode and cathode, (b) long-term stability during operation, and (c) minimal absorption of visible light. Plant-based materials, being intrinsically insulating, have been used in the formulation of electrolyte gelators and as solid-state porous electrolyte scaffolds. While both functions allow the electrolyte to be held together during the cell assembly, additional functionalities are introduced in the case of porous scaffolds. For instance, those based on

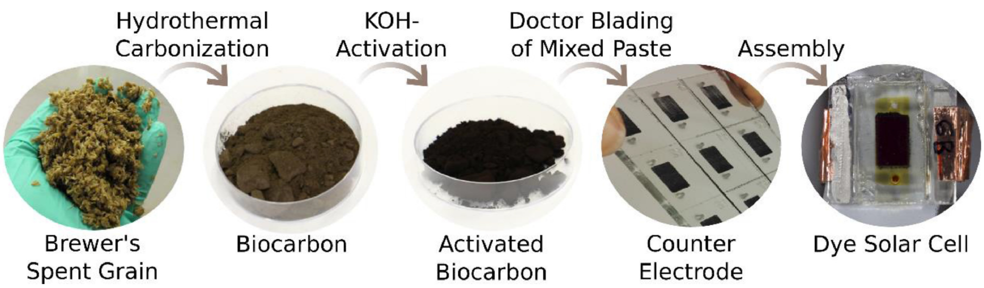

FIGURE 4 Process from brewery residues to dye solar cells with activated biocarbon counter electrodes. *Source*: Reprinted with permission (Tiihonen et al., 2021).

cellulosic self-standing films (Poskela et al., 2019) or flexible printed substrates (Or et al., 2019) may display a high porosity (>95%) while being mechanically robust.

Agents to solidify the electrolyte are usually added in small quantities with no significant environmental effects. Thus, the main concern is associated with the effect these materials have on the device manufacturing process, performance, and lifetime. Various plant-based materials, such as cellulose, seaweed, starch, and guar gum, have been utilized to gel electrolytes as discussed in research papers and recent reviews specialized in polymer electrolyte development (Farhana et al., 2021; Gunasekaran et al., 2020; Hasan et al., 2020); a thorough listing of performance of these materials in presented by Hasan et al. (2020). Owing to the extremely high porosity that can be achieved, CNF and CNC scaffolds can produce little to no charge transfer losses (Or et al., 2019; Poskela et al., 2019). Furthermore, they enable increased efficiency by endowing a more homogeneous distribution of electrolyte components on the photoelectrode (Figure 5) (Borghei et al., 2018; Kaschuk et al., 2019; Poskela et al., 2019; Weerasinghe et al., 2017)—a problem that can otherwise lead to significant losses (as high as 35%) in the overall performance of large-area devices (Miettunen et al., 2018). The next generation electrolyte supports should add functionality and increase the stability of the device by inhibiting unwanted cell side reactions or by capturing impurities. Plant-based biopolymers and their structures offer an opportunity to materialize such demands; for example, they can absorb moisture and scavenge radicals. High porosity and solvent retention properties of the biopolymers can enhance the stability of the DSCs (Mahalingam et al., 2022).

5 | PHOTOACTIVE MATERIALS AND CHARGE SELECTIVE LAYERS

Even though the production of the photoactive materials (i.e., dyes) for PV devices is highly energy intensive, they constitute less than 0.001% of the total mass of the device (Greijer et al., 2001). Since their contribution to the total embedded energy of the whole unit is typically very small (around 2%) (Parisi et al., 2014), the performance and the lifetime of the dyes define their REI.

Plant-derived photoactive dyes can be extracted from leaves, roots, barks, fruits, and flower petals, extensive performance table can be found in the literature (Calogero et al., 2015; Maddah et al., 2020; Rahman et al., 2023; Richhariya et al., 2017). However, such chromophores often suffer from narrow absorption bands, resulting in a low overall conversion efficiency (usually <1%, maximum 4.2%) (Kumara et al., 2017; Richhariya et al., 2017). Natural dyes, even when combined to achieve different absorption spectra, have persistently shown almost one order of magnitude smaller efficiency, almost 1.5% for DSCs sensitized with pomegranate juice and 1.3% for DSCs sensitized with shisonin and chlorophyll, as examples (Rahman et al., 2023) compared to synthetic dyes, 15% (Ren et al., 2023). Because of this and their

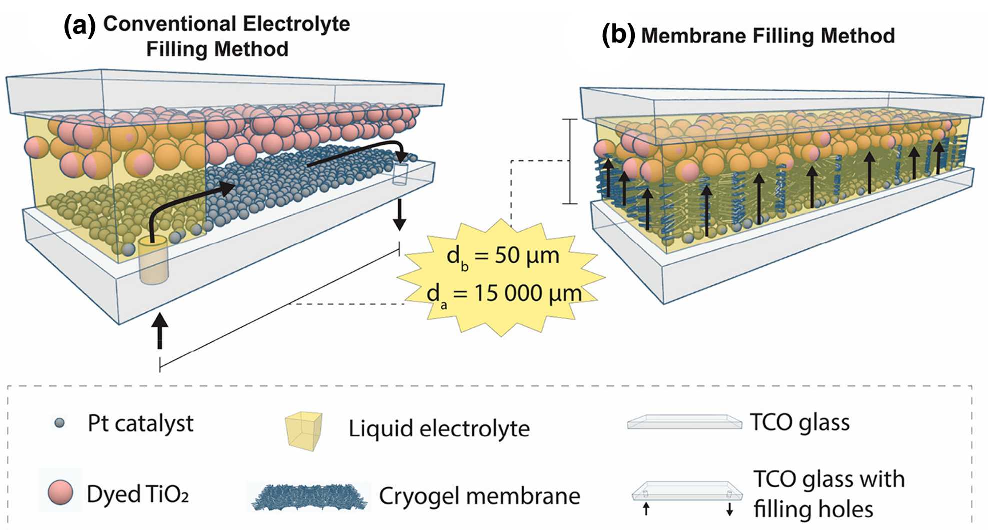

FIGURE 5 Electrolyte filling based on nanocellulose aerogels as an electrolyte scaffold (Poskela et al., 2019). *Source*: Reprinted under CC-BY license.

poor stability (Kumara et al., 2017), the utilization of natural dyes was withered while the utilization of other plant-based components has increased in the past few years.

TiO_2 is the most common semiconductor material used as a photoanode in DSCs. In addition to TiO_2, other metal oxides have been used as photoanode of DSCs. Nevertheless, the electron transport in these materials is affected by their low electrical conductivity. One approach used for enhancing the properties of metal oxides is using them as composites with bio-carbons. The composite of La_2MoO_6 nanoparticles with bio-carbon synthesized by hydrothermal approach showed better electron transfer and photovoltaic performance compared to those of La_2MoO_6 photoanode due to the narrowing of the band gap and more catalytic active sites resulting from a higher specific surface area (Munusami et al., 2021).

The active material in PSCs is a mineral structure that cannot be replaced by plant-derived materials. Nevertheless, plant-based materials have been used in strategies for enhancing the quality of the light-absorbing perovskite layer, surface passivation, or to modify the interface between the active material and the charge transport layers. Plant-based materials are rich in various functional groups, including carbonyl and amino groups. These groups can chelate the undercoordinated Pb^{2+} ions, preventing non-radiative recombination. For example, the light-absorbing perovskite layer's crystal quality and UV/O_3 resistance were improved by using a plant-based L-theanine as additive. It enhanced the crystallization process and passivated the undercoordinated Pb^{2+} ions resulting in an efficiency increase from 22.9% to 24.58%. Moreover, due to the modified surface morphology and hydrophobicity of perovskite film with L-theanine, the devices demonstrated better stability under ambient conditions. The unencapsulated solar cells with the L-theanine additive maintained 89% of the initial efficiency after 97 days, while the efficiency of the reference devices decreased by 50% over the same period (Li et al., 2023). The incorporation of biomaterials as photoactive components or additives in this layer in PCSs is still in its infancy and it is difficult to evaluate their full potential as a viable replacement for conventional photoactive materials.

The characteristics of charge-selective layers, such as carrier selectivity and stability, play a major role in the efficiency and long-term stability of a photovoltaic device (Wang et al., 2019). The common charge-selective layers are fabricated by expensive synthesis processes. Bio-based materials derived from inexpensive substrates can be low-cost alternatives to be used as charge selective layers. For instance, a bio-nanocomposite made from the *Calotropis procera* plant was employed in PSCs and demonstrated suitable energy levels to function as a hole selective layer. The resulting PSCs showed 9.8% efficiency which was 59% higher than that control device without hole transport layer (Arjmand et al., 2022). Because of the abundant functional groups in plant-derived materials, they are suitable candidates for modification of the interfaces between the charge selective layer and light-harvesting layer. The interface between perovskite and the electron transport layer was modified by incorporating the natural bio-functional molecule cobalamin into the electron transport layer. The functional groups in cobalamin passivated the interface due to coordination and electrostatic bonding. Additionally, they enhanced perovskite crystallization and reduced non-radiative recombination by passivating the interface defects. The efficiency of the devices was improved from 18.8% (control devices without interface modifier) to 20.6%. (Pang et al., 2023).

Amino acid biomaterials with carboxylic and amino functional groups have been used for interfacial or surface modification of the ZnO electron selective layer in organic solar cells. The surface modification enhanced the efficiency of the organic solar cells by affecting different parameters, including the energy levels, work function, and wettability of the electron selective layer (C. Liu, Zhou, et al., 2022).

Due to the insufficient charge mobility and selectivity of the carriers, biomaterials have not yet been thoroughly investigated for application as charge selective layers. However, biomaterials that are abundant in functional groups might be used to achieve interface engineering of the charge selective layers. Furthermore, there is a paucity of studies on how charge selective layers based on biomaterials impact solar cell stability.

6 | BIO-BASED MATERIALS AFFECTING RECYCLING IN PHOTOVOLTAICS

At the end of their life photovoltaics pose a recycling challenge. A major hurdle to recycling photovoltaics is making it profitable. While in developed countries recycling could be enforced with legislation, overall financial profitability is the best guarantee for reaching a high rate of recycling. Generally, precious metals are considered as a main economic incentive for recyclers due to their high value and expected supply shortage. For example, Ag represents only 0.1% of the total mass in silicon panels but accounts for 47% of the relative materials value (IRENA & IEA-PVPS, 2016).

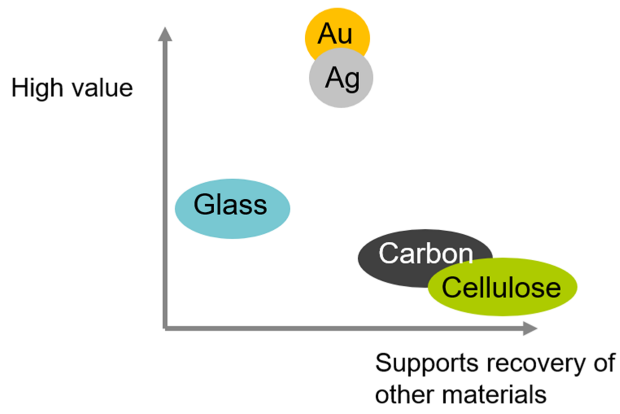

FIGURE 6 The value of the materials and how they support recovery of other materials.

Furthermore, according to forecasts the photovoltaics manufacturing will consume up to 10% of Ag in the world by 2050 (Dias et al., 2016).

According to recent studies substrates play a major role in the viability of recycling process. For instance, when using conventional conductive glass substrates, the concentration of rare metals (e.g., Au, Ag, Pt) in the device is too small for their economically viable recovery (Akulenko et al., 2023; Miettunen & Santasalo-Aarnio, 2021). Plant-based substrates can be simply burned, which concentrate the rare metals and facilitate their recovery; additionally, the process acts as an energy source (Figure 6) (Kaschuk et al., 2022). For instance, in DSCs based on plant-based substrates, rare metal concentrations could exceed the threshold for economically viable recovery, by around 100-fold (Miettunen & Santasalo-Aarnio, 2021). A similar impact of combustible substrate is shown for PSCs. However, along with a significant increase in the concentration of precious metals, there is an equally high increase in the concentration of toxic lead (Akulenko et al., 2023). This high lead concentration can impose additional challenges for the implementation of the recycling process.

In addition to alternative substrates, researchers strive for the substitution of precious metals such as Pt, Au, and Ag with more affordable and abundant materials. Carbon is one of the promising candidates to decrease manufacturing costs and overall module prices (Celik et al., 2020; Espinosa et al., 2013; Miettunen & Santasalo-Aarnio, 2021). Moreover, end-of-life treatment of such devices, especially if the substrate is also bio-based, can be limited to simply one-step incineration. The unintended consequence of using low cost and abundant carbon is the decreased incentive to recycle since there is less high value materials to recover (Figure 6).

7 | CONCLUSION

The main driving force in adopting plant-based materials in solar cells is the need to replace expensive, energy-intensive, rare, and/or non-renewable device components. In this regard, it is pivotal to have a holistic view of the main criteria for economic and environmental viability of PV devices: their REI. REI analysis can be based on the estimation of the power conversion efficiency, the lifetime, and the embedded energy of the applied components. Based on this analysis, plant-based components fall into three categories:

1. *Nearing implementation*: Carbon electrode materials and electrolyte gelators offer similar or even higher performance or/and lifetime compared to conventional counterparts, and they have the potential to offer medium to small improvements in embedded energy of the entire device. PSCs with carbon electrodes are growing interesting for commercial uptake, while DSCs still remain in a niche position.
2. *Further research is needed*: Plant-based substrates have the possibility to achieve superior optical performance and major reduction in embedded energy, but stability needs to be improved. Surface roughness, oxygen and water transmission should be reduced to a minimum—all without compromising their structural integrity. Plant-based substrates are interesting for novel PV applications such as printed electronics.

3. *Major challenges, minor gains*: Photoactive materials have great challenges in both efficiency and lifetime and, on the device level, the potential to decrease embedded energy is marginal. DSCs with natural dyes have applications for highly specialized purposes, for example, education.

Beyond merely replacing solar cell components, plant-based materials also offer functionalities that cannot be reached by conventional ones. For instance, the optical properties of structures based on CNF, CNC, and wood can be tuned to surpass those of glass and plastics. Another example is cellulose aerogel scaffolds that can offer a way to semi-solidify an electrolyte to reduce major performance losses related to upscaling of dye solar cells. Plant-based materials also have untapped possibilities in photovoltaics, for instance MCFs could be functionalized to serve in PV textiles, which is another new area of research. Moreover, utilizing bio-based materials as a substrate can bring significant advantages of reducing the environmental impact of devices and improving the recovery of expensive and rare metals.

AUTHOR CONTRIBUTIONS

Kati Miettunen: Conceptualization (lead); formal analysis (lead); project administration (supporting); supervision (lead); writing – original draft (lead); writing – review and editing (lead). **Mahboubeh Hadadian:** Formal analysis (supporting); project administration (lead); supervision (supporting); writing – original draft (supporting); writing – review and editing (supporting). **Joaquín Valdez García:** Formal analysis (supporting); writing – original draft (supporting); writing – review and editing (supporting). **Alicja Lawrynowicz:** Formal analysis (supporting); writing – original draft (supporting); writing – review and editing (supporting). **Elena Akulenko:** Formal analysis (supporting); writing – original draft (supporting). **Orlando J. Rojas:** Writing – review and editing (supporting). **Michael Hummel:** Formal analysis (supporting); writing – original draft (supporting); writing – review and editing (supporting). **Jaana Vapaavuori:** Formal analysis (supporting); writing – original draft (supporting); writing – review and editing (supporting).

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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