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Full paper

Scalable, anisotropic transparent paper directly from wood for light management in solar cells

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ABSTRACT

The growing demand for flexible electronics and solar energy conversion devices has fueled a search for high-quality paper-based materials with excellent mechanical flexibility and optical properties such as high transparency and haze. Despite the tremendous efforts have been dedicated to developing paper-based materials with high transparency or high haze, challenges still remain in achieving both due to the general exclusivity between them. Here, for the first time, we develop a novel anisotropic paper material possessing high mechanical flexibility and fantastic optical properties with both high transmittance (~90%) and high haze (~90%) simultaneously via a simple yet effective "top-down" approach by directly shear pressing the delignified wood material. The anisotropic transparent paper demonstrates a high efficiency as a light management coating layer for GaAs solar cell with a significant efficiency enhancement of 14% due to its excellent light management capability with both high transparency and high haze. The presented "top-down" approach is facile, scalable, cost-effective and "green", representing a promising direction for developing flexible electronics, solar energy conversion devices and beyond.

1. Introduction

Substrate materials with excellent mechanical flexibility, optical properties and biodegradability are urgently needed for the developing of flexible electronics and solar energy conversion devices [1–6]. Plastic as transparent flexible substrate has been ubiquitously used in electronic and optoelectronic devices due to their favorable mechanical properties, simple process technology, light weight and low cost [7–13]. However, plastic is neither biodegradable nor renewable, causing long-term sustainability concerns. Recently, cellulose-based transparent paper has drawn increased attention as an emerging flexible substrate due to its high biodegradability and renewability [2,3,14,15]. Cellulose-based transparent paper is generally fabricated through a "bottom-up" approach involving multiple steps - disintegrating the cellulose fibers using mechanical [16,17], chemical [18–21] or biological [22,23] methods, dispersing into solution and then reconstructing into transparent paper. Despite the advantages of the "bottom-up" approach including fine control of the cellulose structure and hybridizing with various additional components, the multi-step fabrication process would lower the fabrication efficiency, increase the cost, making it less competitive for large-scale applications [24–27].

In addition to mechanical flexibility and biodegradability, light management capability is also vital for flexible electronics and solar energy conversion devices, especially for thin film solar cells [28–31]. High broadband transmittance and haze are highly desirable for light management coating layer used in thin film solar cells, where both the absorption and utilization of sun light can be maximized [32–35]. However, achieving both high transmittance and high haze in a single material still remains a challenge due to the general exclusivity between transmittance and haze. Increasing the transmittance usually causes the sacrifice of haze. So far, only limited successes have been gained in achieving both high transmittance and haze in a single material [36].

Here, for the first time, we develop a simple yet effective "top-down" approach for fabricating an anisotropic flexible paper with both high transparency and high haze by directly shear pressing the pre-delignified wood material. The anisotropic wood-derived paper exhibits great potential as an efficient light management coating layer for GaAs thin film solar cells, demonstrated by a significant enhancement of 14% in solar energy conversion efficiency and 18% in short circuit density. The "top-down" approach presented here is facile, scalable, cost-effective, and "green", representing an attractive direction for developing next-generation flexible electronics and solar energy conversion devices.

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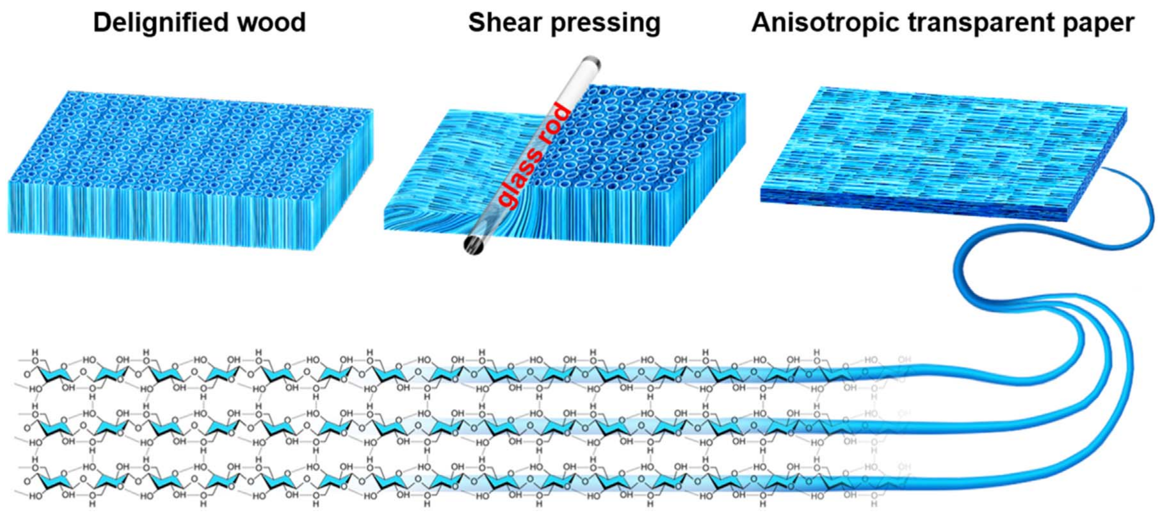

Fig. 1. Graphical illustration of the design concept and "top-down" fabrication process for the anisotropic transparent paper directly from natural wood.

2. Results and discussion

Wood is one of the most abundant resources on earth and widely used in green electronics, biological devices, energy storage and cellulose industry [37–39]. Here we used Basswood as the starting material to directly fabricate anisotropic transparent paper (see Experimental for synthesis details). Note that different types of wood possess similar anisotropic microstructures, so our "top-down" approach can also be used to prepare anisotropic transparent paper from other kinds of wood. Fig. 1 graphically illustrates the design concept and "top-down" fabrication process of the anisotropic transparent paper. Typically, the Basswood was first cut into slices vertically to its growth direction, then treated with NaClO solution to remove the lignin, resulted in the delignified wood with white color. The delignified wood was then shear pressed into paper by rolling a glass rod upon the delignified wood slice with a constant pressure. Compared with the traditional "bottom-up" paper fabrication process, the "top-down" process demonstrated here is quite straightforward, less time-consuming and more cost-effective. It is worth noting that the intrinsic alignment of cellulose fibers in natural wood can be well inherited by shear pressing, resulting in an anisotropic structure of the wood-derived paper. More attractively, the wood-derived paper is super flexible, highly transparent yet with high haze, holding great promise for solar cell application.

The delignification efficiency is a major factor that determines the total efficiency of the whole "top-down" process (shear pressing is not the efficiency-determined step for this process is very quick, usually finished in a few minutes). Based on this consideration, we carried out control experiments to investigate the delignification efficiency by using vertically-cut (radial) wood and parallelly-cut (longitudinal) wood slices. Fig. 2a displays the schematic of the delignification process of the radial and longitudinal wood slices. Compared with longitudinal wood, radial wood has shorter channels, making it easier for the NaClO solution to enter the wood channels, and the decomposed products to transport into the outside mother solution. This can be confirmed by the color evolutions of the two samples during the delignification process recorded by digital camera (Fig. 2b and Fig. S1). The color of the wood slice gradually changed from yellow to white with increasing delignification time, indicating that the content of lignin diminished gradually. The radial wood slice became completely white in about three hours, which is 3 times shorter than that of the longitudinal wood (Fig. 2c), suggesting the high delignification efficiency of radial wood.

The morphologies and microstructures of the original wood and derived paper materials are characterized by scanning electron microscopy (SEM). Fig. 3a-c shows the photo and SEM images of the original radial wood without delignification treatment. The original radial wood is yellowish (inset in Fig. 3a) due to the existence of lignin. Multiple channels with different sizes along the tree growth direction can be observed. After delignification treatment, the color of the wood sample become white and the structure become more porous due to the removal of lignin (Fig. S2). In addition to the shear pressing method, we have also performed vertical pressing as control experiments to investigate the influence of pressing method in the structure of the resultant paper. Vertical pressing of the delignified radial wood results in isotropic transparent paper, where the cellulose fibers are randomly distributed (Fig. 3d-f). By vertical pressing, the vertically aligned channels are completely crushed into a dense isotropic transparent paper. On the contrary, the alignment of cellulose fibers can be well preserved by shear pressing (Fig. 3g-i and Fig. S3), which can be observed both on the top surface and cross section of the anisotropic transparent paper.

The anisotropic microstructure leads to an interesting anisotropic light management capability. A single mode green laser with collimated light spot was perpendicularly incident on the transparent paper with a spot size of around 200 μm. For the isotropic transparent paper, the scattering pattern is circular owning to the random fiber orientation (Fig. 4a). On the contrary, a large divergence angle for anisotropic transparent paper where cellulose fibers are aligned is observed across the alignment direction due to the light diffraction. Consequently, the transmittance scattering pattern is elliptical for the anisotropic transparent paper (Fig. 4d). The scattered light intensity distributions in the x and y directions are shown in Fig. 4b and e for isotropic transparent paper and anisotropic transparent paper, respectively. It can be observed that the scattering intensity of isotropic paper in x and y directions at different scattering angle is similar, which can be attributed to the random distribution of cellulose fibers (Fig. 4b). However, for the anisotropic paper, lower refractive index fluctuation in the y direction was obtained due to the aligned cellulose fibers in this direction (Fig. 4e). Small angle X-ray scattering (SAXS) was further employed to confirm the anisotropy of shear pressed transparent paper. From Fig. 4c we can see that the obtained SAXS pattern for vertically pressed transparent paper is uniform annulus, suggesting that cellulose fibers in the vertically pressed paper are randomly distributed with an isotropic feature. Unlike the vertically pressed paper, the SAXS pattern of the shear pressed transparent paper is an asymmetric annulus, confirming that the cellulose fibers in the shear pressed paper are orientated along with the shear press direction.

In addition, the polarization effect of the anisotropic transparent paper was further evaluated by a polarizing microscope from Olympus