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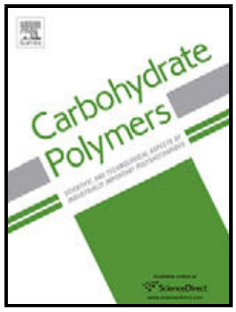

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Transparent, smooth, and sustainable cellulose-derived conductive film applied for the flexible electronic device

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Highlights

- ZnO interface buffer layer alleviate the incompatibility of cellulose and AZO.
- ZnO buffer layer improve the flexibility of cellulose-based conductive film.
- The resultant CZA film could withstand repeated bending treatment.
- A flexible electronic device is successfully fabricated based on the CZA film.

Abstract:

A high-performance flexible conductive substrate is one of the key components for developing promising wearable devices. Concerning this, a sustainable, flexible, transparent, and conductive cellulose/ZnO/AZO (CZA) film was developed in this

study. The cellulose was used as the transparent substrate. The added AZO was as the conductive layer and ZnO functioned as an interface buffer layer. Results showed that the interface buffer layer of ZnO effectively alleviated the intrinsic incompatibility of organic cellulose and inorganic AZO, resulting in the improvement of the performance of CZA film. In compared with the controlled cellulose/AZO (CA) film with 365 Ω/sq sheet resistance and 87% transmittance, this CZA film featured a low conductive sheet resistance of 115 Ω/sq and high transmittance of 89%, as well as low roughness of 1.85 nm Moreover, the existence of conducive ZnO buffer layer enabled the conductivity of CZA film to be stable under the bending treatment. Herein, a flexible electronic device was successfully prepared with the biomass materials, which would be available by a roll-to-roll production process.

Keywords: Cellulose, Conductive film, ZnO buffer layer, Flexible electronic device

1. Introduction

With the rapid development of the market in wearable devices, the utilization of flexible electronic devices has become a predominating trend in the electronics industry (Chen et al., 2020; Lai et al., 2020; Chen et al., 2016; Huang et al., 2019; Jung et al., 2015; Khan et al., 2015; Park et al., 2015; Russo et al., 2011; Agate et al., 2018). A series of promising flexible electronic devices have been developed in the recent decade, such as displays (Kwon et al., 2017), organic light-emitting diodes (OLEDs) (Cao et al., 2014), solar cells (Lee et al., 2017; Mun et al., 2016) and sensors (Yuk et al., 2019; Gao et al., 2020).

Up to now, the conventional flexible substrates are mainly made from polyethylene terephthalate (PET) (Song et al., 2018; Zhou et al., 2016), polycarbonate (PC) (Liu et al., 2009; Park et al., 2008), polyethylene naphthalate (PEN) (Tran et al., 2018), polyestersulfone (PES) (Lei et al., 2013), etc. Even though these substrates feature obvious advantages, like lightweight, chemical stability, and high optical transmittance, the material of the above-mentioned substrates is not sustainable, consuming a large amount of petroleum-derived chemicals. Moreover, these flexible substrates would take hundreds or even millions of years to completely degrade into nature, which inevitably poisons the natural environment and biosystem.

To address the crisis, the focal point of development of the flexible substrate has been shifted to exploit sustainable biomass materials. As one of the most abundant natural biomaterials, cellulose features significant merits, such as biodegradability, renewability, and worldwide availability (Jung et al., 2015; Lei et al., 2019; Li et al., 2014; Wu et al., 2009; Lu & Shen, 2011; Qin et al., 2020; Tong et al., 2020). The cellulose material has been vigorously developed to construct a competitive flexible substrate to minimize the production and usage of the synthesized materials.

Because of the insulation of natural cellulose, the conductive materials should be introduced to produce the conductive cellulose substrate (Du et al., 2017; Ummartyotin & Manuspiya, 2015; Zhang et al., 2018). The commonly used conductive materials are made up of transparent conductive oxides (TCOs), conductive polymer, carbon-based conductive materials, and metal materials (Cuevas et al., 2019; Raghunathan et al., 2017; Gong et al., 2020). Among these conductive materials, the TCOs are characterized by excellent conductivity and high transmittance, which are supportive for producing high-performance electronic devices. The indium tin oxide (ITO) and aluminum-doped zinc oxide (AZO) are the two major TCO materials (Liu et al., 2020; Miao et al., 2014). The shortage and toxicity of indium sources build up an obstacle that limits the further development of ITO in electronic devices (Bae & Kim, 2008; Lei et al., 2013). On the contrary, the AZO features a large volume in nature and non-toxic character, which endow the AZO excellent competitiveness compared to ITO (Hao et al., 2006; Li et al., 2012). The environmentally friendly cellulose/AZO (CA) conductive substrate has been exploited in our group (Liu et al., 2020). However, the difference of intrinsic character between inorganic AZO and organic cellulose lead to the structural incompatibility of these two materials, thus a challenge to prepare a flexible CA conductive substrate is present.

Herein, we developed a flexible CZA conductive substrate by introducing the zinc oxide (ZnO) as an interface buffer layer between the cellulose film and the AZO conductive layer (Figure 1a). The ZnO precursor solution was coated on the cellulose film to construct a stable cellulose/ZnO (CZ) substrate, in which plentiful hydrogen bonding formed between the ZnO and cellulose molecules. The AZO was further deposited on the surface of the ZnO layer, where the AZO can stably grow due to the compatible lattice structure between AZO and ZnO. The prepared cellulose/ZnO/AZO (CZA) film features a high transmittance and conductivity, as well as environmentally

friendly merits. The high-performance cellulose-derived conductive film can be used to fabricate flexible electronic devices (Figure 1b), such as displays and OLEDs.

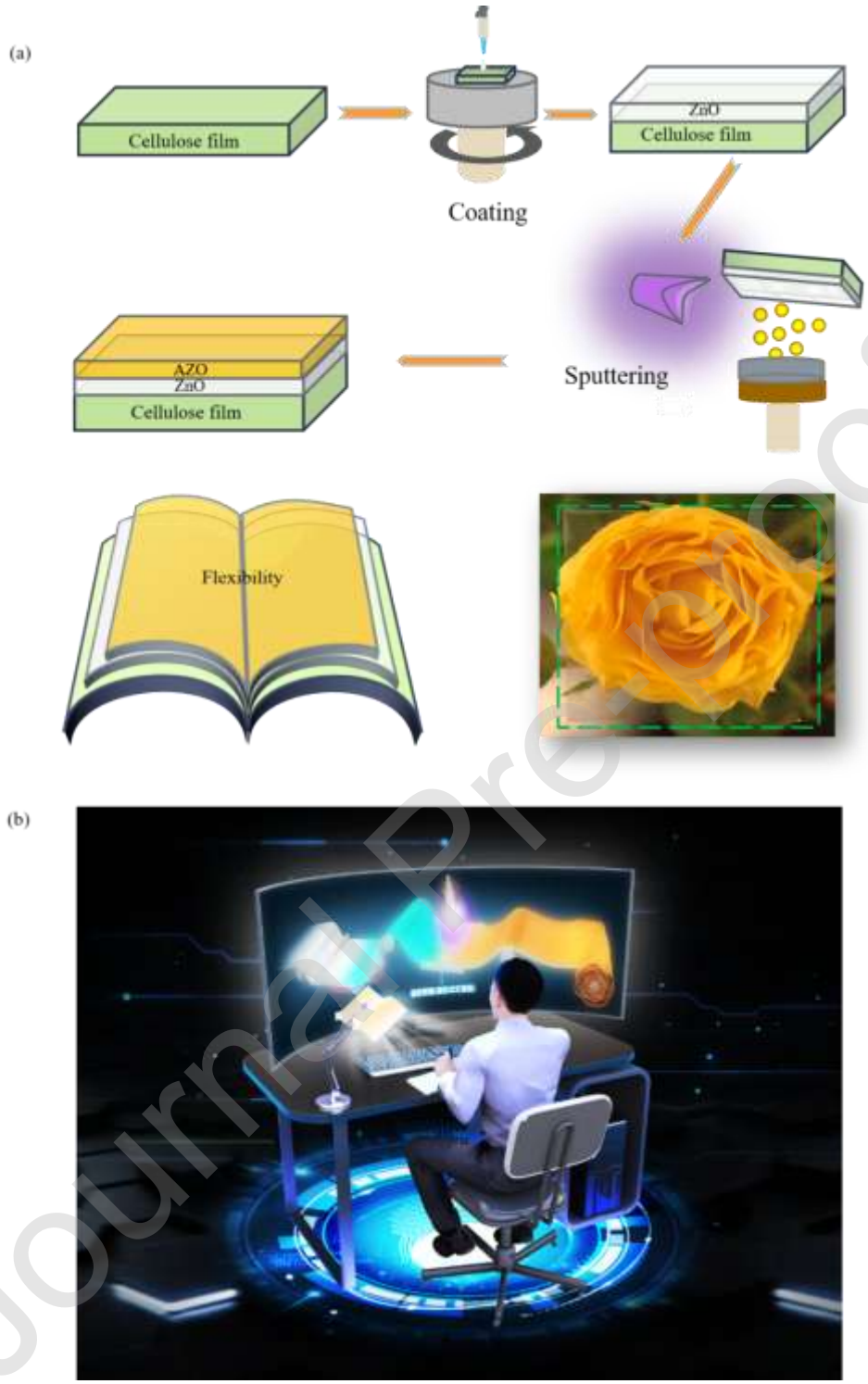

Figure 1. Flexible CZA conductive film (a) fabrication process and (b) the resultant electroluminescent device.

2. Experiments

2.1. Materials

The bamboo dissolving pulp fibers with 1 mm length, 40 μm width, and 35% crystallinity were obtained from Fujian Qingshan Paper Co., Ltd. 1-allyl-3-methylimidazole chloride ([AMIm]Cl) was obtained from Lanzhou Yulu Fine Chemical Co., Ltd. Zinc acetate dihydrate was obtained from Sinopharm Chemical Reagent Co., Ltd. Ethylene glycol methyl ether and ethanol were from Tianjin Zhiyuan Chemical Reagent Co., Ltd. Aluminum doped zinc oxide (AZO) target (ZnO: Al_2O_3 = 98: 2) was obtained from Zhongnuoxincai Corporation.

2.2. Preparation of cellulose film

A 30 g of [AMIm]Cl powder was weighed into a three-necked flask with continuous heating at 80°C for 30 min. Subsequently, 0.5 g of cellulose fiber of dried bamboo dissolving pulp with α-cellulose content of 95% and DP of 650 was slowly added into the resulting [AMIm]Cl solution with heating and stirring for 30 min to dissolve the cellulose fiber. After the reaction was completed the cellulose solution was placed about 30 min to degas. The resultant cellulose solution was scraped into a gel film at a speed of 20 cm/min by using a coater (GBC-A4, Korea GIST Corporation). At last, the prepared cellulose gel film was immersed in the deionized water to completely remove the [AMIm]Cl solution (Liu et al., 2020; Li et al., 2018). The nature of the resultant cellulose film should be more amorphous (Zheng et al., 2019).

2.3. Preparation of cellulose/ZnO film (CZ film)

1 g of zinc acetate dihydrate was weighed and dissolved with 10 ml of ethylene glycol methyl ether. Subsequently, 0.2 ml of ethanol was added into the resultant solution as a stabilizer to prepare the zinc oxide precursor solution with stirring for 24 hours. The collected ZnO precursor solution was spin-coated on the surface of the cellulose film by a desktop homogenizer at 2500 rpm for 40 seconds to obtain a CZ film.

2.4. Preparation of cellulose/ZnO/AZO (CZA film) and cellulose/AZO conductive film, (CA film)

The CZA and CA conductive films were prepared by sputtering AZO on the as-prepared CZ and cellulose film (4 cm*4 cm). The sputtering treatment was carried out on a magnetron sputtering system (TRP-450, Shenyang Scientific Equipment Corporation, China). In detail, the target AZO and substrate(CZ, cellulose film) were fixed on the sputtering table and sample table, respectively. The distance between the substrate and target was 30 cm. Subsequently, the vacuum degree was kept at 2.5 x 10^{-4} Pa by using a combined mechanical pump and molecular pump. The working environment was at a

1 Pa of high purity Ar. The sputtering temperature was controlled at 80℃. The radio frequency power was set as 100 W. The time of magnetron sputtering was 1 h. The resultant CA and CZA conductive films were stored in a humidity chamber.

2.5. Characterization

The morphologies of samples were investigated with a scanning electron microscope (SEM; SU8010, Hitachi, Japan) at an accelerating voltage of 15 kV. Before the field-emission SEM test, the surface of the films was sprayed with gold particles with a sputter coater (E-1010, Hitachi, Japan). The surface appearance of the sample was characterized by an atomic force microscope (AMF, Bruker Multimode 8). The X-ray diffraction (XRD) profiles of samples were obtained by using a powder X-ray diffraction instrument (X'PERT PRO MPD, Philips, Holland) with Cu Kα at 45 kV. The XRD patterns were collected in the 2θ range of 10°~80° at a scanning speed of 10°/min. The transmittance of the samples was measured by using an ultraviolet/visible spectrophotometer (UV-Vis; UV-1800 SHIMADZU, Japan). The conductive resistance of the CZA and CA conductive film was determined with four-point probe measurements (KDB-3, Kun De Inc., China) as a mean value of five points. Each measurement was done five times and the average was reported.

3. Results and discussion

3.1 Structure and morphologies of the conductive film

As known to all, a phase difference between organic polymer, cellulose, and inorganic oxide, AZO would lead to the structural incompatibility between them. Herein, the incompatible structure and morphologies of CA film were investigated by SEM. As shown in Figure. 2a, the AZO hardly grows well on the surface of the cellulose substrate due to the phase incompatibility. This led to the formation of the uneven size of AZO particles followed by a rough surface (Figure. 2b, c). Of interest, the ZnO was capable to effectively alleviate the difference of phase and structure between organic cellulose and inorganic AZO, resulting in the improvement of morphologies of CZA film. The layer-by-layer structure of CZA film was clearly shown in Fig. 2d, in which the ZnO layer was located between the cellulose substrate and the AZO layer. The CZA film presented a dense and smooth surface (Figure. 2e, and S1), featuring the uniform AZO particles. The contribution of ZnO on optimizing the surface morphology of the AZO layer was also confirmed with the AFM (Figure. 2f). The CZA film featured a low

surface roughness of 1.85 nm, which is 55% lower than that of the CA film of 4.15 nm (Figure. S2).

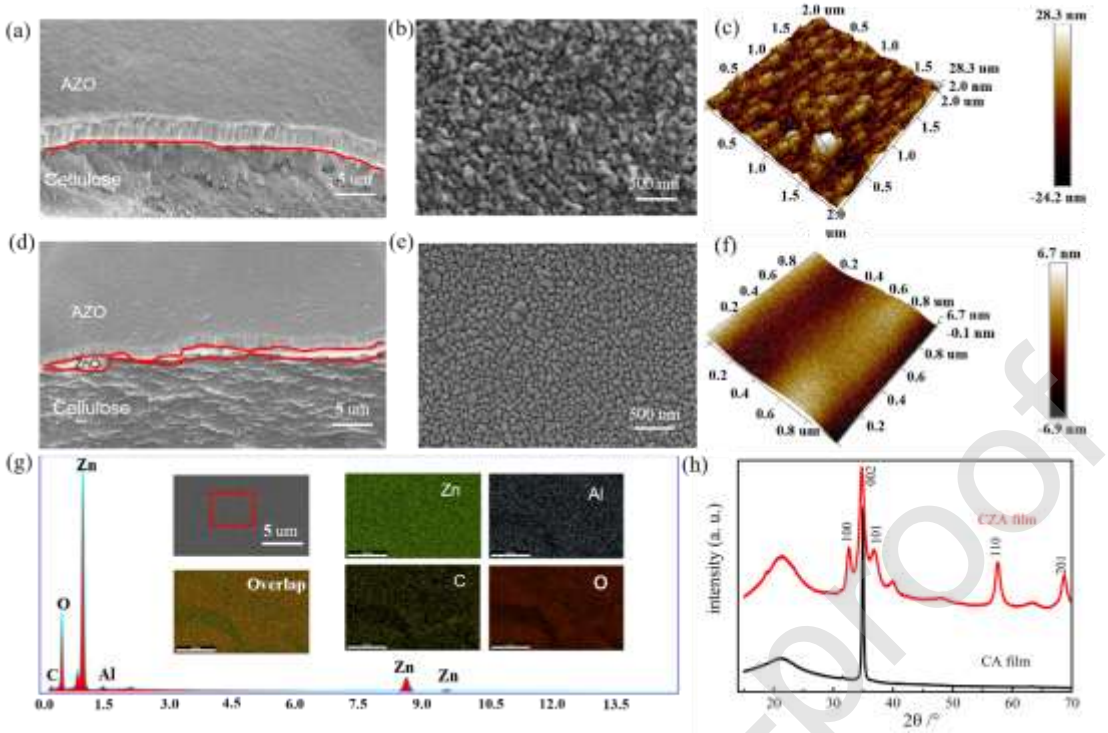

Figure 2. The structure and morphologies of the conductive film. (a-c) SEM images of cross-section, surface, and AFM observation of CA conductive film. (d-f) SEM images of cross-section, surface, and AFM observation of CZA conductive film. (g) elements analysis of CZA conductive film. (h) XRD results of cellulose-based films.

The element analysis results in Figure. 2g showed that elements of Zn, Al, C, and O homogeneously distributed on the surface of CZA film, which indicated the uniform growth of AZO. The crystalline structure of cellulose, ZnO, and AZO samples were also investigated (Figure. S3). The ZnO featured a hexagonal phase wurtzite structure, more specifically, the XRD results show the typical characteristic peaks of ZnO at 31.7°, 34.4°, 36.2°, 56.5°, 68.9°, which are the indicator of the lattice plane of 100, 002, 101, 110, and 201. The XRD results of ZnO well match the results of the JCPDF card with index number 36-1451 (Bagheri & Rabieh, 2013). In addition, the obvious peak (002) at 2θ=34.4 demonstrates the c-axis orientation of ZnO, which is perpendicular to the cellulose substrate. Note that, the AZO also features a preferred orientation along c-axis (002) at 34.5°. It is noted that the XRD result of the CZA film sample was like that of the CZ sample, which suggested excellent compatibility between AZO and ZnO in the cellulose substrate. Therefore, it is concluded that the ZnO acted as an interface buffer layer to modulate the growth of AZO, resulting in a desirable structure and morphology of the CZA film.

3.2 Conductivity and transmittance of the conductive film

The optimized structure and morphologies of the conductive layer due to the contribution of ZnO were beneficial for promoting the conductivity of the CZA film. The conductive sheet resistance of CZA film was 115 Ω/sq, which was about 0.68 times lower than that of the original CA film (Figure. 3a). The CZA film also featured a high transmittance of 89%, which was almost consistent with the CA film (Figure. 3a). The high transmittance of CZA film can be well demonstrated by the clear observation of a blooming flower which was covered by the CZA film (Figure. S4). The transmittance curve of CA and CZA film features the fluctuation at 460 and 620 nm (Figure. S4), which can be attributed to the interference of light at the interface of the AZO surface and air (Rezaie et al., 2017). Wherein, there is no fluctuation for CZ film prepared by sol-gel technology and cellulose film may due to their relative dense and compact surface structure (Ko et al., 2017). It also can be observed in Fig. 3b that the CZA film connected a conductive circuit to make the light-emitting diode to light.

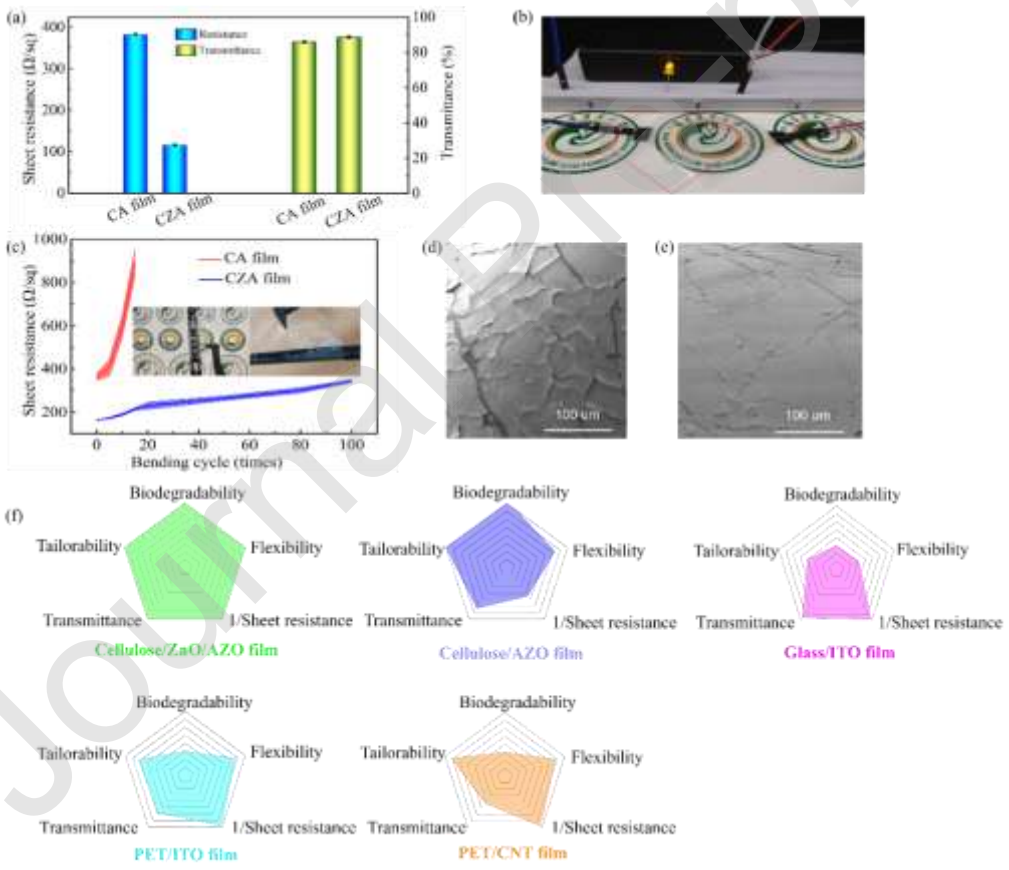

Figure 3. The conductivity and transmittance of the conductive film. (a) Sheet resistance and transmittance. (b) CZA conductive film is a transparent conductor in a closed circuit. (c) The sheet resistance as a function of bending treatment. (d) Surface morphologies of CA conductive film with bending 20 times. (e) Surface morphologies of CZA conductive film with bending 100 times. (f) A radar plot displaying a comparison among cellulose(Liu, et al., 2020), PET(Jing et al., 2014; Park

et al., 2012), and glass substrate (Qin et al., 2016)with different conductive materials (Lee et al., 2011; Sierros et al., 2009).

The ZnO worked as an interface buffer layer endowed the CZA film a high performance of conductivity stability. After a bending treatment of 100 times, the conductive sheet resistance of CZA film was about 350 Ω/sq, which was still lower than that of CA film without bending treatment. However, the bending treatment significantly decreased the conductivity and increased the resistance of CA film, i.e., a 20-time bending treatment enlarged its resistance out of the measurement scope of equipment (Figure. 3c). The bending treatment resulted in the different changes in the conductivity of CZA film and CA film, in which the ZnO was mainly responsible for maintaining the conductivity of the CZA film. The ZnO can strongly connect the intrinsically incompatible cellulose and AZO, which promoted the stability and endurance of the CZA film. Because of the buffer behavior of ZnO, the AZO conductive layer of CZA film was well kept after the bending treatment, even the bending treatment strongly cracked the AZO layer of CA film (Figure. 4d, e, and S5).

The layer-by-layer architecture of cellulose substrate, ZnO buffer layer, and AZO conductive layer endowed the CZA film obvious advantages, including high transmittance and conductivity, degradability and renewability, desirable flexibility and tailorability. However, these advantages are hardly integrated into other conductive films, such as glass/ITO film, PET/ITO film, cellulose/CNT film, PET/AZO film, and PET/CNT film (Figure. 3f). Cellulose/CNT film has a lower transmittance which is not beneficial for the fabrication of optoelectronic devices and the roughness is higher (~13 nm). Besides, the interface bonding force is weak between cellulose and CNT (Qi et al., 2013; Soheilmoghaddam et al., 2017) which is same as the PET/AZO film (Jeong et al., 2010).

3.3 CZA film-based electroluminescent device

The flexible, transparent, and conductive CZA film was successfully employed to construct an electroluminescent device. The device featured a typical sandwich structure of CZA/ZnS: Cu/$BaTiO_3$/Ag, in which CZA film was a transparent conductive substrate, copper-doped zinc sulfide acted as a photoactive layer, $BaTiO_3$ functioned as a dielectric layer, and Ag was employed as the back electrode (Figure 4a). Based on this structure, a “scissors hand” pattern-based luminous film was fabricated. More interestingly, the luminous film could work very well even with an arbitrary bending treatment of 90º (Figure 4b and video1). It was expected that the application of CZA

film could be extended to other flexible and bendable devices, like OLEDs, solar cells, touch panels, and electrochromic windows.

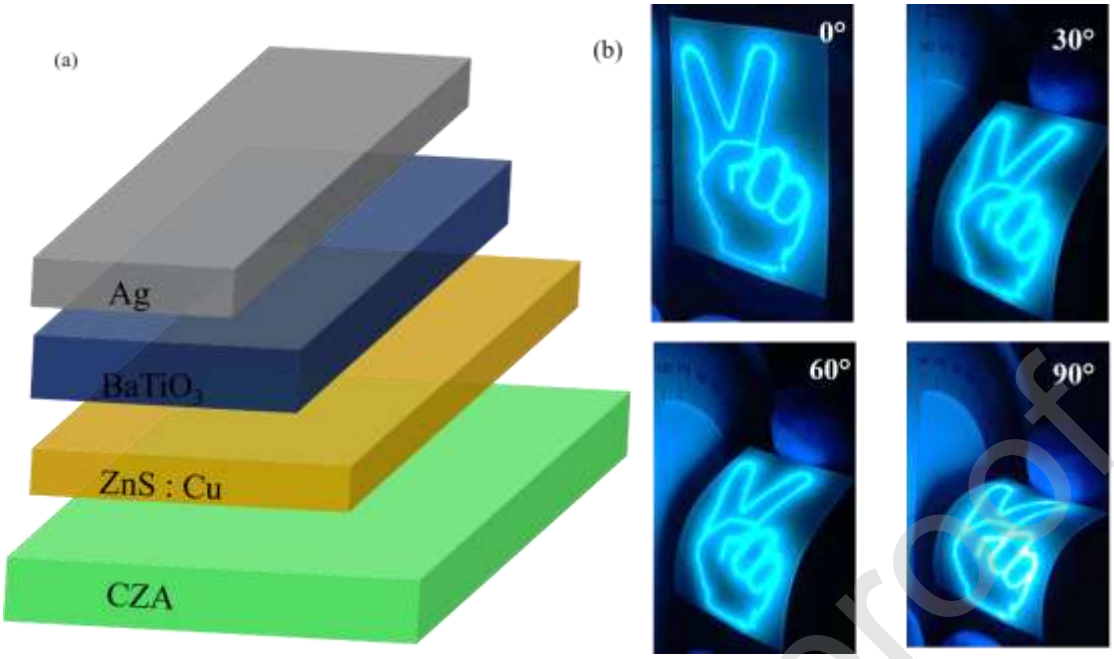

Figure 4. (a) structure and (b) performance of CZA film-based electroluminescent device.

3.4 The roll-to-roll treatment for CZA film

As shown in Figure 5, the newly-developed green and flexible cellulose-derived conductive film can be produced via a scalable roll-to-roll fabrication process. The coating of ZnO and the deposition of AZO also can be easily realized in this process. Besides, the roll-to-roll processable cellulose-derived conductive film with unique performances of high conductivity and light transmittance, flexibility, and environmental friendliness, can be integrated into a series of low-cost electroluminescent devices.

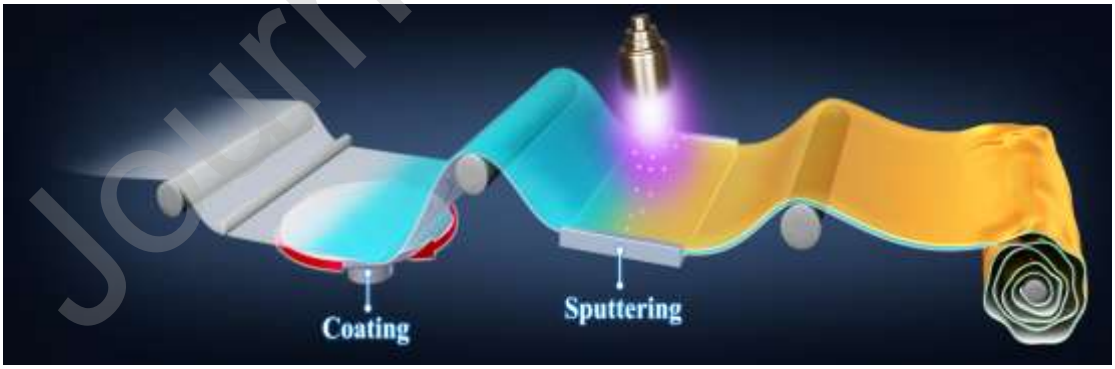

Figure 5. Schematic of the roll-to-roll fabrication of green, flexible, and conductive CZA film.

Conclusions

In this study, we developed a green, flexible, and transparent cellulose-derived

conductive film with a sandwich structure of cellulose/ZnO/AZO. The ZnO functioned as a buffer layer to modulate the growth of AZO on the cellulose substrate, resulting in a desirable structure and morphology of CZA film with low sheet resistance of 115 Ω/sq and high transmittance of 89%. The ZnO also promoted the interface bonding of cellulose substrate and AZO conductive layer in the developed system. With the ZnO buffer layer, the conductivity of CZA film is well maintained after the bending treatment, while it was almost absence for the control CA film without the ZnO. More specifically, the sheet resistance was 360 Ω/sq from 115 Ω/sq for the CZA film with 100 times bending, and the CA film became almost electronic insulation with 20 times bending, which was of the indicator of high flexibility of CZA film. The high-performance CZA film was also furtherly utilized to exploit a flexible electroluminescent device. Our study opens a new route for the development of a green, flexible, and transparent cellulose-based electroluminescent device.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

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References

Agate, S., Joyce, M., Lucia, L., & Pal, L. (2018). Cellulose and nanocellulose-based flexible-hybrid printed electronics and conductive composites – A review. *Carbohydrate Polymers*, *198*(June), 249–260. https://doi.org/10.1016/j.carbpol.2018.06.045

Bae, J. H., & Kim, H. K. (2008). Characteristics of Al doped ZnO co-sputtered InZnO anode films prepared by direct current magnetron sputtering for organic light-emitting diodes. *Thin Solid Films*, *516*(21), 7866–7870. https://doi.org/10.1016/j.tsf.2008.05.035

Bagheri, M., & Rabieh, S. (2013). Preparation and characterization of cellulose-ZnO nanocomposite based on ionic liquid ([C4mim]Cl). *Cellulose*, *20*(2), 699–705. https://doi.org/10.1007/s10570-012-9857-3

Cao, W., Li, J., Chen, H., & Xue, J. (2014). Transparent electrodes for organic optoelectronic devices: a review. *Journal of Photonics for Energy*, *4*(1), 040990. https://doi.org/10.1117/1.jpe.4.040990

Chen, D., Liang, J., & Pei, Q. (2016). Flexible and stretchable electrodes for next generation polymer electronics: a review. *Science China Chemistry*, *59*(6), 659–671. https://doi.org/10.1007/s11426-015-5520-9

Chen, W., Zhang, D., Yang, K., Luo, M., Yang, P., & Zhou, X. (2020). Mxene (Ti3C2Tx)/cellulose nanofiber/porous carbon film as free-standing electrode for ultrathin and flexible supercapacitors. *Chemical Engineering Journal*, *October*, 127524. https://doi.org/10.1016/j.cej.2020.127524

Cuevas, A., Campos, B. B., Romero, R., Algarra, M., Vázquez, M. I., & Benavente, J. (2019). Eco-friendly modification of a regenerated cellulose based film by silicon, carbon and N-doped carbon quantum dots. *Carbohydrate Polymers*, *206*, 238–244. https://doi.org/10.1016/j.carbpol.2018.10.074

Du, X., Zhang, Z., Liu, W., & Deng, Y. (2017). Nanocellulose-based conductive materials and their emerging applications in energy devices - A review. *Nano Energy*, *35*(January), 299–320. https://doi.org/10.1016/j.nanoen.2017.04.001

Gao, J., Wang, L., Guo, Z., Li, B., Wang, H., Luo, J., Huang, X., & Xue, H. (2020). Flexible, superhydrophobic, and electrically conductive polymer nanofiber composite for multifunctional sensing applications. *Chemical Engineering Journal*, *381*(June 2019), 122778. https://doi.org/10.1016/j.cej.2019.122778

Gong, Q., Li, Y., Liu, X., Xia, Z., & Yang, Y. (2020). A facile preparation of polyaniline/cellulose hydrogels for all-in-one flexible supercapacitor with remarkable enhanced performance. *Carbohydrate Polymers*, *245*(May), 116611. https://doi.org/10.1016/j.carbpol.2020.116611

Hao, X. T., Tan, L. W., Ong, K. S., & Zhu, F. (2006). High-performance low-

temperature transparent conducting aluminum-doped ZnO thin films and applications. *Journal of Crystal Growth*, *287*(1), 44–47. https://doi.org/10.1016/j.jcrysgro.2005.10.040

Huang, S., Liu, Y., Zhao, Y., Ren, Z., & Guo, C. F. (2019). Flexible Electronics: Stretchable Electrodes and Their Future. *Advanced Functional Materials*, *29*(6), 1–15. https://doi.org/10.1002/adfm.201805924

Jeong, J. A., Shin, H. S., Choi, K. H., & Kim, H. K. (2010). Flexible Al-doped ZnO films grown on PET substrates using linear facing target sputtering for flexible OLEDs. *Journal of Physics D: Applied Physics*, *43*(46). https://doi.org/10.1088/0022-3727/43/46/465403

Jing, M. xiang, Han, C., Li, M., & Shen, X. qian. (2014). High performance of carbon nanotubes/silver nanowires-PET hybrid flexible transparent conductive films via facile pressing-transfer technique. *Nanoscale Research Letters*, *9*(1), 1–7. https://doi.org/10.1186/1556-276X-9-588

Jung, Y. H., Chang, T. H., Zhang, H., Yao, C., Zheng, Q., Yang, V. W., Mi, H., Kim, M., Cho, S. J., Park, D. W., Jiang, H., Lee, J., Qiu, Y., Zhou, W., Cai, Z., Gong, S., & Ma, Z. (2015). High-performance green flexible electronics based on biodegradable cellulose nanofibril paper. *Nature Communications*, *6*(May). https://doi.org/10.1038/ncomms8170

Khan, S., Lorenzelli, L., & Dahiya, R. S. (2015). Technologies for printing sensors and electronics over large flexible substrates: A review. *IEEE Sensors Journal*, *15*(6), 3164–3185. https://doi.org/10.1109/JSEN.2014.2375203

Ko, H. U., Kim, H. C., Kim, J. W., Zhai, L., & Kim, J. (2017). Fabrication method study of ZnO nanocoated cellulose film and its piezoelectric property. *Materials*, *10*(6). https://doi.org/10.3390/ma10060611

Kwon, J. H., Choi, S., Jeon, Y., Kim, H., Chang, K. S., & Choi, K. C. (2017). Functional Design of Dielectric-Metal-Dielectric-Based Thin-Film Encapsulation with Heat Transfer and Flexibility for Flexible Displays. *ACS Applied Materials and Interfaces*, *9*(32), 27062–27072. https://doi.org/10.1021/acsami.7b06076

Lai, H., Li, W., Xu, L., Wang, X., Jiao, H., Fan, Z., Lei, Z., & Yuan, Y. (2020). Scalable fabrication of highly crosslinked conductive nanofibrous films and their applications in energy storage and electromagnetic interference shielding. *Chemical Engineering Journal*, *400*(January), 125322.

https://doi.org/10.1016/j.cej.2020.125322

Lee, C., Lai, K., Lin, C., Li, C., Ho, K., & Wu, C. (2017). *Nano Energy A paper-based electrode using a graphene dot / PEDOT : PSS composite for fl exible solar cells*. *36*(February), 260–267.

Lee, H. S., Bang, J. O., Lee, H. J., Lee, G. J., Chai, K. H., & Jung, S. B. (2011). Analysis of thermo-mechanical behavior of ITO layer on PET substrate. *Proceedings - Electronic Components and Technology Conference*, 1796–1799. https://doi.org/10.1109/ECTC.2011.5898757

Lei, M., Wu, S., Liang, J., & Liu, C. (2019). Comprehensive understanding the chemical structure evolution and crucial intermediate radical in situ observation in enzymatic hydrolysis/mild acidolysis lignin pyrolysis. *Journal of Analytical and Applied Pyrolysis*, *138*(January), 249–260. https://doi.org/10.1016/j.jaap.2019.01.004

Lei, P. H., Hsu, C. M., & Fan, Y. S. (2013). Flexible organic light-emitting diodes on a polyestersulfone (PES) substrate using Al-doped ZnO anode grown by dual-plasma-enhanced metalorganic deposition system. *Organic Electronics*, *14*(1), 236–249. https://doi.org/10.1016/j.orgel.2012.10.030

Li, J., Yang, H., Huang, K., Cao, S., Ni, Y., Huang, L., Chen, L., & Ouyang, X. (2018). Conductive regenerated cellulose film as counter electrode for efficient dye-sensitized solar cells. *Cellulose*, *25*(9), 5113–5122. https://doi.org/10.1007/s10570-018-1913-1

Li, S., Huang, D., Zhang, B., Xu, X., Wang, M., Yang, G., & Shen, Y. (2014). Flexible Supercapacitors based on bacterial cellulose paper electrodes. *Advanced Energy Materials*, *4*(10), 1–7. https://doi.org/10.1002/aenm.201301655

Li, Y. D., Wang, D. M., Jin, H., Lu, J. P., & Zhou, J. (2012). Effect of film thickness on properties of Al-doped ZnO film as transparent conducting electrodes in OLEDs. *ICSICT 2012 - 2012 IEEE 11th International Conference on Solid-State and Integrated Circuit Technology, Proceedings*, *Dc*, 1–3. https://doi.org/10.1109/ICSICT.2012.6467808

Liu, X., Huang, K., Lin, X., Li, H., Tao, T., Wu, Q., Zheng, Q., Huang, L., Ni, Y., Chen, L., Ouyang, X., & Li, J. (2020). Transparent and conductive cellulose film by controllably growing aluminum doped zinc oxide on regenerated cellulose film. *Cellulose*, *27*(9), 4847–4855. https://doi.org/10.1007/s10570-020-03147-0

Liu, X., Xiao, W., Ma, X., Huang, L., Ni, Y., Chen, L., Ouyang, X., & Li, J. (2020). Conductive Regenerated Cellulose Film and Its Electronic Devices – A Review. *Carbohydrate Polymers*, *250*, 116969. https://doi.org/10.1016/j.carbpol.2020.116969

Liu, Y., Li, Q., & Shao, H. (2009). Properties of ZnO:Al films deposited on polycarbonate substrate. *Vacuum*, *83*(12), 1435–1437. https://doi.org/10.1016/j.vacuum.2009.05.006

Lu, X., & Shen, X. (2011). Solubility of bacteria cellulose in zinc chloride aqueous solutions. *Carbohydrate Polymers*, *86*(1), 239–244. https://doi.org/10.1016/j.carbpol.2011.04.042

Miao, D., Jiang, S., Zhao, H., Shang, S., & Chen, Z. (2014). Characterization of AZO and Ag based films prepared by RF magnetron sputtering. *Journal of Alloys and Compounds*, *616*, 26–31. https://doi.org/10.1016/j.jallcom.2014.07.105

Mun, S., Zhai, L., Min, S. K., Yun, Y., & Kim, J. (2016). Flexible and transparent strain sensor made with silver nanowire-coated cellulose. *Journal of Intelligent Material Systems and Structures*, *27*(8), 1011–1018. https://doi.org/10.1177/1045389X15577651

Park, C., Kim, S. W., Lee, Y. S., Lee, S. H., Song, K. H., & Park, L. S. (2012). Spray coating of carbon nanotube on polyethylene terephthalate film for touch panel application. *Journal of Nanoscience and Nanotechnology*, *12*(7), 5351–5355. https://doi.org/10.1166/jnn.2012.6343

Park, H. J., Park, J. H., Choi, J. I., Lee, J. Y., Chae, J. H., & Kim, D. (2008). Fabrication of transparent conductive films with a sandwich structure composed of ITO/Cu/ITO. *Vacuum*, *83*(2), 448–450. https://doi.org/10.1016/j.vacuum.2008.04.061

Park, S. C., Biswas, S., Fang, J., Mozafari, M., Stauden, T., & Jacobs, H. O. (2015). Millimeter Thin and Rubber-Like Solid-State Lighting Modules Fabricated Using Roll-to-Roll Fluidic Self-Assembly and Lamination. *Advanced Materials*, *27*(24), 3661–3668. https://doi.org/10.1002/adma.201500839

Qi, H., Liu, J., Gao, S., & Mäder, E. (2013). Multifunctional films composed of carbon nanotubes and cellulose regenerated from alkaline-urea solution. *Journal of Materials Chemistry A*, *1*(6), 2161–2168. https://doi.org/10.1039/c2ta00882c

Qin, G., Fan, L., & Watanabe, A. (2016). Formation of indium tin oxide film by wet

process using laser sintering. *Journal of Materials Processing Technology*, *227*, 16–23. https://doi.org/10.1016/j.jmatprotec.2015.07.011

Qin, X., Duan, C., Feng, X., Zhang, Y., Dai, L., Xu, Y., & Ni, Y. (2020). Integrating phosphotungstic acid-assisted prerefining with cellulase treatment for enhancing the reactivity of kraft-based dissolving pulp. *Bioresource Technology*, 124283. https://doi.org/10.1016/j.biortech.2020.124283

Raghunathan, S. P., Narayanan, S., Poulose, A. C., & Joseph, R. (2017). Flexible regenerated cellulose/polypyrrole composite films with enhanced dielectric properties. *Carbohydrate Polymers*, *157*, 1024–1032. https://doi.org/10.1016/j.carbpol.2016.10.065

Rezaie, M. N., Manavizadeh, N., Abadi, E. M. N., Nadimi, E., & Boroumand, F. A. (2017). Comparison study of transparent RF-sputtered ITO/AZO and ITO/ZnO bilayers for near UV-OLED applications. *Applied Surface Science*, *392*, 549–556. https://doi.org/10.1016/j.apsusc.2016.09.080

Russo, A., Ahn, B. Y., Adams, J. J., Duoss, E. B., Bernhard, J. T., & Lewis, J. A. (2011). Pen-on-paper flexible electronics. *Advanced Materials*, *23*(30), 3426–3430. https://doi.org/10.1002/adma.201101328

Sierros, K. A., Morris, N. J., Kukureka, S. N., & Cairns, D. R. (2009). Dry and wet sliding wear of ITO-coated PET components used in flexible optoelectronic applications. *Wear*, *267*(1–4), 625–631. https://doi.org/10.1016/j.wear.2008.12.042

Soheilmoghaddam, M., Adelnia, H., Sharifzadeh, G., Wahit, M. U., Wong, T. W., & Ali Yussuf, A. (2017). Bionanocomposite regenerated cellulose/single-walled carbon nanotube films prepared using ionic liquid solvent. *Cellulose*, *24*(2), 811–822. https://doi.org/10.1007/s10570-016-1151-3

Song, W., Fan, X., Xu, B., Yan, F., Cui, H., Wei, Q., Peng, R., Hong, L., Huang, J., & Ge, Z. (2018). All-Solution-Processed Metal-Oxide-Free Flexible Organic Solar Cells with Over 10% Efficiency. *Advanced Materials*, *30*(26), 1–8. https://doi.org/10.1002/adma.201800075

Tong, R., Chen, G., Tian, J., & He, M. (2020). Highly transparent, weakly hydrophilic and biodegradable cellulose film for flexible electroluminescent devices. *Carbohydrate Polymers*, *227*(September 2019). https://doi.org/10.1016/j.carbpol.2019.115366

Tran, D. P., Lu, H. I., & Lin, C. K. (2018). Conductive characteristics of indium tin oxide thin film on polymeric substrate under long-term static deformation. *Coatings*, *8*(6). https://doi.org/10.3390/coatings8060212

Ummartyotin, S., & Manuspiya, H. (2015). An overview of feasibilities and challenge of conductive cellulose for rechargeable lithium based battery. *Renewable and Sustainable Energy Reviews*, *50*, 204–213. https://doi.org/10.1016/j.rser.2015.05.014

Wu, R. L., Wang, X. L., Li, F., Li, H. Z., & Wang, Y. Z. (2009). Green composite films prepared from cellulose, starch and lignin in room-temperature ionic liquid. *Bioresource Technology*, *100*(9), 2569–2574. https://doi.org/10.1016/j.biortech.2008.11.044

Yuk, H., Lu, B., & Zhao, X. (2019). Hydrogel bioelectronics. *Chemical Society Reviews*, *48*(6), 1642–1667. https://doi.org/10.1039/c8cs00595h

Zhang, Y., Zhang, L., Cui, K., Ge, S., Cheng, X., Yan, M., Yu, J., & Liu, H. (2018). Flexible electronics based on micro/nanostructured paper. *Advanced Materials*, *30*(51), 1–39. https://doi.org/10.1002/adma.201801588

Zheng, X., Huang, F., Chen, L., Huang, L., Cao, S., & Ma, X. (2019). Preparation of transparent film via cellulose regeneration: Correlations between ionic liquid and film properties. *Carbohydrate Polymers*, *203*(February 2017), 214–218. https://doi.org/10.1016/j.carbpol.2018.09.060

Zhou, W., Chen, J., Li, Y., Wang, D., Chen, J., Feng, X., Huang, Z., Liu, R., Lin, X., Zhang, H., Mi, B., & Ma, Y. (2016). Copper Mesh Templated by Breath-Figure Polymer Films as Flexible Transparent Electrodes for Organic Photovoltaic Devices. *ACS Applied Materials and Interfaces*, *8*(17), 11122–11127. https://doi.org/10.1021/acsami.6b01117