Cellulose materials with high light transmittance and high haze

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Abstract

Cellulose is a glucose-based macromolecular polymer that is insoluble in water and most organic solvents. It makes up over half of the carbon content in the plant kingdom and it is the most abundant polysaccharide found in nature. With the gradual depletion of non-renewable resources and growing environmental problems, the research of cellulosic materials has become one of the most pressing concerns for scientists. In recent years, cellulose composite film with high transmittance and tunable haze has attracted increasing attention, which has potential applications in flexible lighting and displays, anti-reflection and anti-glare coatings, etc. In this article, the latest progress in recent years in the preparation and applications of cellulose materials with high light transmittance and high haze was reviewed. An overview of applications of cellulose materials with high light transmittance and high haze was discussed, including nano cellulose films, regenerated cellulose films, cellulose derivative films and others.

Introduction

Cellulose is the most abundant natural polymer on the earth. It is inexhaustible and the most precious natural renewable resource for mankind. In the process of the establishment and development of polymer physics and chemistry, cellulose has made significant contributions. Cellulose has great application potential in materials, chemistry and other fields such as catalyst (Bahsis et al. 2018), surface grafting (Cui et al. 2021; Mekonnen et al. 2021), cellulose aerogels (He et al. 2021; Luo et al. 2021; Zhang et al. 2021), click-scaffolds (Nada et al. 2018), sensor (Nawaz et al. 2018; Orelma et al. 2019; Pei et al. 2021; Wang et al. 2021), barrier films (Belbekhouche et al. 2011; He et al. 2021), rheological modifiers (Aaen et al. 2019; Boluk et al. 2011; MC. Li et al. 2015), bactericidal materials (Ma et al. 2021; Nie et al. 2021; Ringot et al. 2009), fluorescent materials (Tian et al. 2016), mechanical reinforcements in nanocomposites (Alonso-Lerma et al. 2021; Siqueira et al. 2009; Siró and Plackett 2010; Winter et al. 2021), and spinning (Elsayed et al. 2020; Hauru et al. 2016; Liu et al. 2019; Reyes et al. 2020; Zhou et al. 2021), etc. Furthermore, the porous structure of cellulose paper not only simplifies the manufacturing process, but also improves the performance of the device. Therefore, the idea of combining the excellent properties and functions of cellulose to design green electronic products has attracted great interest. Since cellulose contains a large number of intra- and inter-molecular hydrogen bonds, materials made of cellulose have excellent mechanical properties (Fujisaki et al. 2014) and excellent thermal stability (Hsieh et al. 2013). Meanwhile, the film made of cellulose has high transparency and high haze, and its transmittance and haze can be controlled by various methods. It is very important in the manufacture of optoelectronic devices that require customized optical properties (Zhang et al. 2019). Interest in the use of cellulose in printed electronics has recently been growing.

Haze is the ratio of the projected light intensity that deviates from the incident light by more than 2.5° to the total projected light intensity, expressed as a percentage (Li et al.2018). Materials with high light transmittance and high haze have certain light management functions. Such materials are important components of many optical electronic devices (Fitz-Gerald et al. 2000; Gao et al. 2018; Haghanifar et al.

2018). If natural polymer materials can be used to replace traditional materials to prepare high light transmittance and high haze materials, the environmental problem caused by electronic waste is expected to be alleviated to a certain extent (Lorenzen 2014). Cellulose has become an ideal raw material to prepare products with high light transmittance and high haze. The material with high haze and high light transmittance has extremely strong scattering ability for incident light. Tuning optical performance including transparency and optical haze plays a critical role in preparing optoelectronic devices with varying optical requirements. Materials with high haze and high light transmittance are also more suitable for solar cells (Brongersma et al. 2014; Li et al. 2020; Preston et al. 2013; Zou et al. 2020), because a high degree of scattering will increase the optical length, thereby increasing the light absorption in the active layer of the solar cells. High-haze materials play important roles in many optoelectronic devices, such as liquid crystal displays or other electronic products that use light-emitting diodes as light sources (Joo and Shin 2010; Kuo et al. 2009; Park and Khang 2016; Song et al. 2013). In addition, the high haze and high light transmittance material is antiglare, making it suitable for outdoor displays and other devices that work in bright environments (Chung 2003). In addition to the aspects mentioned above, high-haze materials also have important application values in the fields of electronic substrates (Bai et al. 2015; Guo et al. 2015; Kim et al. 2015; Roth et al. 2015).

Nowadays, there is increasing attention about using cellulose as raw material to prepare materials with high light transmittance and high haze. This green biodegradable material can be applied to various optical equipment materials. This paper aims to make a systematic introduction and summary of the preparation methods as well as optical properties of various cellulose materials with high light transmittance and high haze. In the present review, cellulose materials with high light transmittance and high haze are classified into nano cellulose films, regenerated cellulose films, cellulose derivative films and other cellulose films. The strategies and applications of cellulose materials with high light transmittance and high haze are systematically introduced.

Nanocellulose Film

During the past few decades, researchers have found that preparing some natural polymers into nanomaterials can give them new properties (Yang et al. 2020). The nanocellulose obtained via chemical or mechanical treatment can be divided into two categories: cellulose nanocrystals and cellulose microfibrils, also called cellulose nanofibrils (Hoeng et al. 2016). In addition to the natural advantages of abundant reserves and renewable recycling, nanocellulose has the advantages of fine nanostructure, good mechanical strength and low thermal expansion coefficient (Hu et al. 2016; Nogi 2013). Film material composed of nanocellulose fibrils has the advantages of fast ion diffusion and high temperature resistance. In addition, nanocellulose has a three-dimensional network porous structure, which can be combined with inorganic nanoparticles (Hokkanen et al. 2018), metal ions and their oxides (Alipour et al. 2020; Chang et al. 2020; Choi and Jeong 2019), carbon materials (Xing et al. 2019; Xu et al. 2020), and conductive polymers (Lay et al. 2016) to prepare multifunctional composite materials. It has been reported that nanocellulose films with high light transmittance and high haze have attracted increasing attention in the preparation of flexible electronic devices.

Zhu et al. (Zhu et al. 2013) focused on adjusting the diameter of the nanocellulose fibers (NFCs) and the bulk density of the nanocellulose film to obtain high light transmittance and high haze. They used TEMPO oxidant to treat cellulose and obtained nanocellulose films with various fiber diameter, and the density of the nanocellulose film was controlled by pressure and drying temperature. Finally, they prepared nanocellulose films with fiber diameter of 25 μm, 50 nm, and 10 nm, and nanocellulose films with thicknesses of 52 μm, 40 μm, and 36 μm and fiber diameter of 10 nm. The film made from 25 μm cellulose fibers has a lower specular transmittance (37%) and a lower diffuse transmittance (5%) at a wavelength of 550 nm. For the nanocellulose film made from 50 nm NFCs, the diffuse transmittance is 92% and the specular transmittance is 19%. The cellulose film made from 10 nm NFCs has a diffuse transmittance of 93% and a specular transmittance of 71%. For fibers with diameter of 50 nm and 10 nm, the optical transmittance can reach 92–93%. The haze of nanocellulose films with a fiber diameter of 25 μm, 50 nm and 10 nm at 550 nm is about 77%, 49% and 20%, respectively. It can be seen that they have higher light transmittance than plastics as well as extremely high haze, which makes them applicable in many fields. In addition to fiber diameter, the bulk density also affects its optical properties. When the total amount of cellulose is the same, as the bulk density increases, both the specular reflectance and the diffuse transmittance increase. With the bulk density increasing, the effective index of the cellulose film increases and the porosity of the paper decreases, which reduces scattering and increases light transmittance. The decrease in scattering results in the increase in transmittance and the decrease in haze value. Therefore, changing the bulk density can also adjust the optical properties of the nanocellulose film. These special optical properties of nanocellulose films provide unexampled possibilities for the development of green optoelectronic devices.

The optical properties of the nanocellulose film can be tailored by varying the diameter of the fibers and the bulk density of the film. Furthermore, adding other substances to the cellulose can also obtain materials with high light transmittance and high haze.

Fang et al. (Fang et al. 2014) investigated a facile method to regulate the optical properties of paper. The paper was prepared by vacuum filtration, and the haze could be adjusted from 18–60% by adjusting the weight ratio of the TEMPO-oxidied wood fiber to the NFCs, while retaining its transmittance over 90%. The total light transmittance of transparent films with different contents of NFCs exceeds 90%. In addition to excellent optical transparency, the transmission haze of transparent paper substrates in the visible wavelength range has also increased from 18–60%. Because the micron-sized TEMPO oxidized fibers have a wide range of light diffusion characteristics, the transparent paper without NFCs has the highest transmission haze (60%). With the NFCs content in the paper increasing, the haze gradually decreases. Such cellulose films with adjustable optical properties have great application potential. They can be widely used in light diffusion films and photovoltaic devices, such as outdoor displays and solar cells.

It can be seen that mixing TEMPO oxidized cellulose with NFCs can get a high light transmittance and high haze film, and the haze of the film can be adjusted by controlling the number of NFCs while maintaining a high light transmittance. Moreover, mixing spherical cellulose nanoparticles with NFCs can also produce films with the similar properties. Li et al. (Li et al. 2019) prepared a series of CNPs (cellulose nanoparticles) with various morphologies, such as fibrous (CNFs), short rods (CNCs-1), rods (CNCs-3) and spherical (CNCs-2 and CNC_S-4). Cellulose nanocellulose films were prepared using CNFs to achieve an anti-trade-off between transparency (89% at 600 nm) and haze (77%) performance. Meanwhile, by mixing CNFs with CNCs-2 and adjusting their ratio, the optical haze of the film can be accurately controlled to any continuous adjustable value in the range of 5–77%. The nanocellulose film prepared in this way achieves both high transparency and high haze. Figure 1 shows the optical properties of nanopapers containing cellulose nanoparticles with different morphologies. The CNFs and CNCs-2 films show the highest and lowest optical haze, respectively. There are many voids and laminar gaps in the CNF cellulose film which can cause large light scattering. However, the size of CNCs-2 is small, it can fill these gaps to reduce light scattering. Therefore, by a simple mixing CNFs with CNCs-2, a nanocellulose film with continuously adjustable haze can be obtained. As the CNF/CNCs-2 molar ratio gradually increases from 1:4 to 4:1, the optical haze value increases from 23–60%. Therefore, the haze of the film can be easily adjusted in a range of 77–11% with this method while maintaining a high transparency. These nanocellulose films with high light transmittance and adjustable haze can be widely used in light diffusers and flexible optoelectronic fields.

(Fig. 1 should be inserted here.)

Direct mixing the solutions or suspensions of two substances would affects the uniformity and optical properties of the resulting film. A simple, effective and low-cost method to prepare a highly transparent and hazy film was reported by Yang et al. (Yang et al. 2019) by coating TEMPO-oxidized wood fiber suspension onto a TEMPO-oxidized cellulose nanofibril gel. The film exhibits up to 85% light transmission and 62% haze, while showing high tensile strength and excellent thermal stability. The microstructures of the two sides of the highly hazy transparent films were different: one was smooth, while the other was rough. As a reference, they directly mixed TEMPO-oxidized wood fibers and cellulose nanofibers to prepare a film sample. The final results show that the film prepared by the "coating" method presents more excellent optical properties than the film prepared by the "blending" method. This work provides a method for preparing green optoelectronic devices. In addition, ultrasonic treatment of TEMPO oxidized cellulose fibers (Lin et al. 2019) or surface modification of cellulose with hydrocarbons/dimethylformamide (Yu et al. 2018) can also increase light transmittance and haze.

Yang et al. (Yang et al. 2018) used a simple deposition process of micro-scale oxidized wood fibers to adjust the surface morphology of the cellulose nanofiber film. As the upper surface roughness increases, the haze of the film increases. The total transmittance of the obtained film is in the range of 83–88%, showing a low haze of 3.8% to a high haze of 62.3%. In addition, the lower surface of the cellulose nanofiber film is very smooth, which meets the requirement for electronic and optoelectronic applications.

The choice of raw materials may also affect the haze of the nanocellulose film. Zhou et al. (Zhou et al. 2018) systematically studied the optical properties of highly transparent paper made of micro-cellulose fibers with different fiber morphologies. They used the TEMPO-oxidation system to process different fibers, including Northwood fibers, Eucalyptus wood fibers and Manila hemp fibers. The fiber morphology before and after TEMPO oxidation was analyzed in detail. Film made of Northwood fibers, Eucalyptus wood fibers and Manila hemp fibers has a haze of 84%, 81% and 78%, respectively, at a wavelength of 550 nm. By analyzing the nanocellulose films prepared from different types of cellulose, it is found that the fiber diameter has an important influence on the transmission haze. The diameter of Northwood fibers, Eucalyptus wood fibers and Manila hemp fibers treated by TEMPO oxidation system is 45, 26 and 20 nm, and the fiber length is 0.62, 0.57 and 0.52 mm, respectively. It can be seen that as the average length and width of cellulose fibers increase, their haze increases accordingly. Meanwhile, the preparation method also has an impact on the optical properties of the transparent paper. They found that the haze of transparent film prepared by solution casting method is lower than the transparent film prepared by vacuum filtration method. Since the rougher surface causes stronger light scattering, the transparent film prepared by vacuum filtration presents higher haze. This research provides critical information for the future preparation of high light transmittance and high haze materials for electronic devices.

It is seen that nanocellulose films can be a good candidate as high transparent and high haze materials. One generally uses certain chemical or physical methods to disintegrate cellulose into a suitable size, and then vacuum filters the raw material suspension or casts it to prepare cellulose film with high haze and high light transmittance. Since it is difficult to prepare nano-level nanocellulose fibrils from direct high-pressure homogenization due to the strong intramolecular and intermolecular interaction, chemical treatment is generally required to reduce the hydrogen bond density before disintegrating. TEMPO is usually used to oxidize cellulose before the cellulose disintegrating Usually NFCs of different sizes need to be sieved. In this way one can finally obtain the film with properties of high light transmission and high haze.

Regenerated Cellulose Film

Natural cellulose is not meltable nor soluble in most solvents. Therefore, the processing performance of natural cellulose is very poor. There are very few solvents that can dissolve cellulose, so it is very important to develop new cellulose solvents with strong solubility and low toxicity. At present, there are several kinds of solvents that can dissolve cellulose, including NaOH/CS_2 system (Lue et al. 2007), *N*-Methylmorpholine-*N*-oxide (NMMO) monohydrate (Biganska and Navard 2005), ionic liquid system (Zhang et al. 2005; Yuan et al. 2019; Fukaya et al. 2006), NaOH/urea system (Cai and Zhang 2005; Pei et al. 2013), NaOH dilute solvent method and its direct spinning technology (Yamashiki, et al. 1992). The cellulose film prepared by regenerated cellulose can also have the optical characteristics of high light transmittance and high haze.

The work of Zhu et al. (Zhu et al. 2016) provided a precise control of the cellulose fiber dimensions and enabled paper substrates with a diverse range of unique optical properties. They immersed a piece of paper made of regular microfibers in the ionic liquid 1-butyl-3-methylimidazolium chloride (BmimCl). After 15 min, the paper was *in situ* hot-pressed. Finally, the paper became transparent. As shown in Fig. 2, paper treated with ionic liquid achieves a highly transparent (> 90%) and hazy (> 90%), which is comparable to flexible glass and polyethylene terephthalate (PET). The ionic liquid dissolves the cellulose on the surface of the paper, resulting in strong nano-welding between the microfibrils. Under pressure, the dissolved cellulose fills the micropores initially occupied by air. The paper with unique structure achieves high light transmittance and high haze, which may usher in a new era of paper photonics, electronics and optoelectronics.

(Fig. 2 should be inserted here.)

In addition, they studied the use of regenerated cellulose to prepare films with high light transmittance and good haze (Zhu et al. 2013). The energy consumption of regenerated cellulose film is much lower than nanocellulose, and it can be prepared without homogenization. To prepare regenerated cellulose film, they dissolved the cellulose fibers in 1-ethyl-3-methylimidazolium phosphorous methyl ester ionic liquid ($EMIMMeOPO_2H$), then poured the solution into glass-based cell to regenerate and then washed with deionized water. They compared the optical properties of regenerated cellulose film, nanocellulose film and PET film. The nanocellulose film has the highest diffuse transmittance but the lowest specular transmittance. All the films' transmittance is up to 90%, which is great for transparent substrate use. Due to the porous microstructure, nanocellulose film and regenerated cellulose film have higher haze values, while PET has much lower haze. For most of the wavelength ranges investigated, the haze of PET is less than 5%, while that of nanocellulose film and regenerated cellulose film is as high as 50%. Moreover, the regenerated cellulose film shows excellent biocompatibility, which makes it great potential for the application of green and sustainable electronic equipments in the biomedical field.

According to the above research, the haze of the film prepared with pure regenerated cellulose film is not very high. In order to develop the film with higher haze, Hou et al. (Hou et al. 2020) prepared a regenerated cellulose/plain paper composite film by immersing the plain paper in cellulose solution. The light transmittance and haze of this film are very high. Figure 3 shows the optical properties of the composite film. When the composite film is placed close to the picture, the letters below are visible, indicating that it has high light transmittance. When the distance between the composite film and the picture is about 1 cm, the image is blurred, indicating a high haze. Both the composite film and regenerated cellulose film have a high transparency of ~ 90% at 550 nm, which is much higher than ordinary paper (26.4%). The haze of the composite film is 95.2%, slightly lower than ordinary paper (~ 100%), but much higher than regenerated cellulose film (~ 40%). Therefore, the film prepared by immersing ordinary paper in a cellulose solution has excellent optical properties, and its application value in optical equipment is also very considerable. It is worth noting that when the immersion time of the paper in the ionic liquid is different, the light transmittance and haze of the cellulose film change (Chen et al. 2021).

(Fig. 3 should be inserted here.)

The above method of preparing high light transmittance and high haze film from cellulose solution is somewhat complicated, which is not conducive to the applications in the large-scale production process. Li et al. (Li et al. 2019) incorporated a small amount of cellulose with a high degree of polymerization into the straw/AmimCl solution to build an enhanced entangled network, which greatly improved the processability of the solution and the mechanical strength of the regenerated cellulose membrane. The preparation process of this biomass film is shown in Fig. 4a. The process does not involve chemical pretreatment or component separation, but only 5 simple steps including grinding, dissolving, mixing, regeneration in water and drying. Finally, an all-biomass film with certain light transmittance and high haze is obtained. Because the whole preparation process does not involve chemical treatment and component separation, all the components in the wheat straw are completely retained in the final film, resulting a brown and translucent film. The transmittance of straw/cellulose film (80/20 ratio) at 800 nm is about 59%, but the haze is as high as 97%, and it has excellent optical properties (Fig. 4b and Fig. 4c). In addition, the entire preparation process is green and environmental-friendly, in line with the concept of sustainable development, and large-scale production in industry is expected to achieve in this method.

(Fig. 4 should be inserted here.)

Incorporating light-scattering particles into the material is an important method for preparing high-haze materials. When preparing high-haze materials containing scatterers, the type and size of the scatterers are important factors that affect the optical properties of the materials (Liu and Teng 2010). Cheng et al. (Cheng et al. 2020) prepared a cellulose/chitosan composite film with customized transmission haze. Chitosan powders were pre-dispersed in *N*,*N*-dimethylformamide (DMF) and slowly transferred into ionic liquid. Smashed cotton was added and stirred to completely dissolve the cotton cellulose. The suspension was cast on glass and the cellulose/chitosan gel film was regenerated by immersing the coating in water. As shown in Fig. 5 (a-f), the use of the highly transparent and hazy film is an effective solution to avoid decreasing the light brightness or altering any structure of the present commercial LED perse. The Fig. 5(g-j) shows that the haze of the cellulose/chitosan composite film can be tailored by controlling the size and quantity of the chitosan powders. For composite film with 200-mesh chitosan powders, the haze increases with the increase of chitosan content within a certain range. The transmittance of the film is almost the same (> 85%), only slightly lower than that of the cellulose film (> 90%). Compared with the 200-mesh series film with the same amount of chitosan, the 1800-mesh series film has higher transmittance and haze. This result confirms that the smaller the scatterer, the higher the light transmittance and haze of the composite film. Surface roughness is another main parameter that directly affects the transmission haze of the cellulose/chitosan film. As the amount of chitosan powders increases, the surface roughness of the cellulose/chitosan film and their haze increase. However, the transmittance of the cellulose/chitosan film is almost unaffected. Therefore, these cellulose/chitosan films with different optical properties can meet the requirements of different application scenarios. It is worth noting that apart from the raw materials and solvents, there are no other chemical substances in the preparation process, and all solvents can be recycled and reused. This simple and environmentally friendly manufacturing method is also suitable for large-scale continuous production of cellulose films, which is essential to the practical application of cellulose-based degradable optoelectronic products. In addition to chitosan particles, TEMPO-oxidized cellulose (Kasuga et al. 2018) and halloysite nanotubes (Kim et al. 2017) can also be added as a dispersion to the cellulose solution to prepare cellulose films with special optical properties.

(Fig. 5 should be inserted here.)

From the above research work, it is seen that by completely dissolving cellulose in suitable solvent and depositing it in certain coagulating bath, the regenerated cellulose transparent matrix could be obtained, and the undissolved material can be used as a scatterer (undissolved cellulose, insoluble straw powder, insoluble chitosan). The transparent matrix can be various source of cellulose (cotton pulp, wood pulp) and does not require derivatization steps. Moreover, it is convenient to regulate the haze by adjusting the size of the scatterer.

Cellulose Derivative Film

Cellulose derivatives refer to a series of compounds whose hydroxyl groups are partially or completely esterified or etherified. The modified cellulose has more specific functions and they can be applied in various fields (Bella et al. 2017; Eltaweil et al. 2020; Roy and Rhim 2020). In addition, the cellulose derivative materials have strong compatibility with the environment and they are environmental-friendly and renewable. Cellulose materials with high light transmittance and high haze can also be prepared from cellulose derivatives.

Hu et al. (Hu et al. 2013) used carboxymethyl cellulose (CMC) nanofibers to prepare a film with high light transmittance and high haze. They prepared carboxymethylated NFCs by disintegrating wood cellulose fibers in a high-pressure homogenizer. The optical properties of cellulose film are compared with those of polyethylene terephthalate (PET) plastic. There is a significant difference between the transmittance of cellulose film and plastic, mainly because the plastic is composed of a dense structure and therefore light scattering cannot be observed. The cellulose film has a nanostructure and a high porosity, therefore light scattering is observed. The specular transmittance and diffuse transmittance of this nano-paper are different, and this large light scattering can be applied to solar cells.

In addition, a film with specific high light transmittance and high haze can also be obtained by mixing CMC with wood cellulose. Hu et al. (Hu et al. 2018) prepared a transparent and high-haze all-cellulose composite film. After simple immersion treatment, this film not only shows superior mechanical properties but also excellent optical properties. CMC is used to improve the transparency and surface smoothness of the film, while lignocellulose fibers with nano-network structure are used as a reinforcing phase to improve the mechanical properties and to increase its optical haze. Moreover, this thin film shows improved optical properties compared with plain paper and pure CMC film. The optical properties of these three films are shown in Fig. 6. The diameter of wood fibers is much larger than the wavelength of visible light, which can cause strong back light scattering, so the common paper exhibits extremely high haze in the wavelength range of visible light. For CMC film, the incident light beam can pass directly through it with almost negligible light scattering effect, resulting in a high transparency and low transmission haze. The total transmittance of the pure CMC film is as high as 92%, and the haze is less than 2%. However, the CMC/lignocellulose composite film presents both high transparency and optical haze as well as excellent mechanical properties and super high surface smoothness, showing up to 90%

light transmittance and > 80% haze. This kind of film with high light transmittance and high haze is expected to be widely used in optoelectronic devices.

(Fig. 6 should be inserted here.)

Due to the introduction of carboxymethyl groups, CMC can be directly dissolved in water. It is therefore convenient to process it into transparent and hazy film.

Other Cellulose Film

In addition to using nanocellulose, regenerated cellulose and cellulose derivatives to prepare high transparency and high haze films, cellulose films with excellent optical properties can also be prepared by using cellulose and other raw materials or using various new processes. Lin et al. (Lin et al. 2021) prepared a poly (ethylene glycol)/cellulose composite film. The haze of the composite film can be adjusted by controlling the molecular weight of poly (ethylene glycol). Yao et al. (Yao et al. 2016) prepared a kind of novel plastic paper. They used ordinary paper as the paper substrate and used a transparent thermosetting epoxy resin as the infiltrating polymer. Due to the mesoporous and fibrous structure of the paper substrate, the epoxy resin can quickly penetrate into the pores and finally form a plastic-paper hybrid material. The refractive index of thermosetting epoxy resin is 1.522, similar to that of cellulose, therefore, it ensures less light scattering at the interface. The introduced thermosetting epoxy resin endows the plastic-paper hybrid substrate with high mechanical strength and excellent solvent stability and protects the inner cellulose paper. Figure 7 shows how ordinary paper is made into plastic paper through the penetration of epoxy resin. This hybrid material combines the advantages of these two substrates. It has unique optical properties: high optical haze (up to 90%) comparable to paper and high total transmittance (up to 85%) comparable to plastics. This new type of material has similar flexibility as paper while strong resistance to water and many solvents. Due to its excellent optical performance, low cost and scalable manufacturing capacity, this plastic paper has great application potential in a series of optoelectronic devices such as solar cells or OLED lighting.

(Fig. 7 should be inserted here.)

Ou et al. (Ou et al. 2017) prepared a highly transparent paper with nanostructures by polishing ordinary paper with $EmimMeOPO_2H$ ionic liquids (ILs). They repeated the ILs-polishing process 3 times to obtain the optimized optical performance. After being polished for the first time, a large distribution of cellulose fibers could still be observed, but no voids between adjacent cellulose fibers were observed. After being polished for the second time, the surface of the cellulose fibers becomes smooth and there is no obvious fibrous profile. After being polished for the third time, the fibrous profile of the cellulose fibers completely disappears without any pores, resulting in a smooth surface for the prepared nanostructured paper. Figure 8 shows a schematic diagram of the ILs polishing process and a photo of the prepared film and its optical performance. When the third ILs polishing process continues, the optical transparency of the nanostructured paper increases to 91%, its haze value decreases to 89%. Compared with glass and plastic, this cellulose film has a short processing time, high haze and high light transmittance, and it may be used in flexible optoelectronic equipment in the future.

(Fig. 8 should be inserted here.)

Cellulose has great promise in the development of renewable and biodegradable devices. Nanocellulose, regenerated cellulose, cellulose derivatives, and other cellulose-based substances can be used to prepare materials with high light transmission and haze. The diameter and bulk density of cellulose are important factors that affect the haze of nanocellulose films. Mixing NFCs of different quality with spherical cellulose particles or TEMPO oxidized cellulose can also obtain highly transparent films with adjustable haze. Moreover, various raw materials and different preparation methods result in significant differences in the optical properties of nanocellulose films. For regenerated cellulose, the regenerated cellulose/paper composite film prepared by immersing ordinary paper in an ionic solution has a higher haze than the regenerated cellulose film. Furthermore, the straw/cellulose composite film also has good light transmittance and high haze, and the preparation method is simple and environmentally friendly. For cellulose derivatives, it can be made into nanofibers or mixed with nanocellulose to prepare films with high light transmittance and high haze. Moreover, when ordinary paper is treated to possess an internal structure similar to nanocellulose as well as a smooth surface, it can also present high light transmittance and high haze.

Materials made of cellulose can achieve similar properties to plastic substrates, and more importantly, they have excellent biocompatibility and degradability. Therefore, biomass materials made from cellulose have great application potential, which is very important for solving environmental and resource problems. We hope that this article will provide ideas for the research and development of cellulose-based high transmittance and high haze materials. Look forward to new contributions to the sustainable development of the world.

Declarations

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Competing Interests

The authors declare that they do not have any conflict of interest.

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Ethical Approval

This article does not contain any studies with human participants or animals performed by any of the authors.

Author Contributions

All authors contributed to the study conception and design. Jun Zhang and Xuejing Zheng had the idea for the article. Ruijie Pan, Yixiu Cheng, Ying Pei, Weiguo Tian, Yongchao Jiang and Keyong Tang performed the literature search and data analysis. The first draft of the manuscript was written by Ruijie Pan. Jie Liu, Jun Zhang and Xuejing Zheng critically revised the work.

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Figures

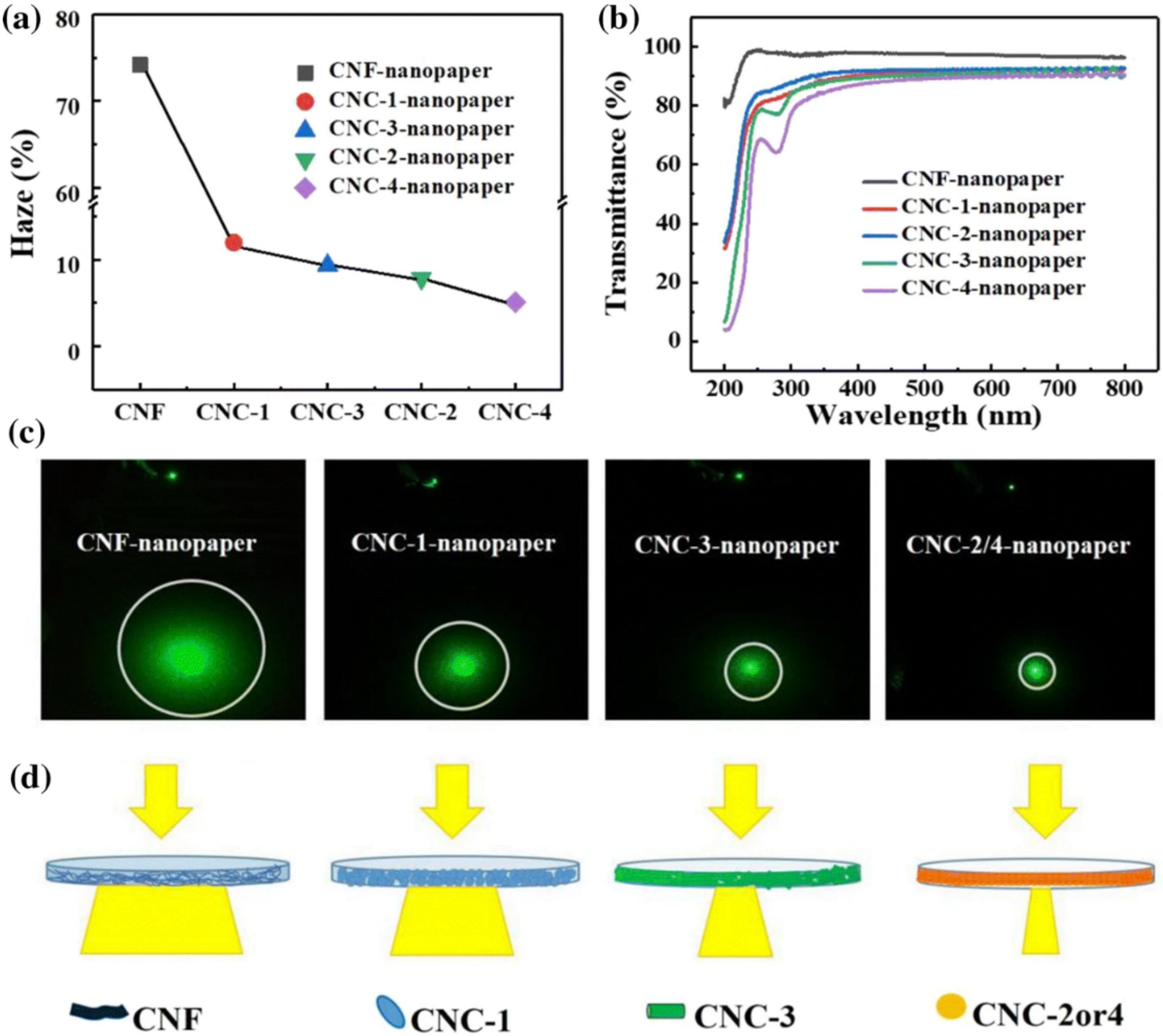

Figure 1

(a) Haze of nanocellulose films for different morphologies (fibrous, short rod, clubbed and spherical) cellulose nanoparticle, (b) UV transmission spectra of different nanocellulose films, (c, d) photos of nanopaper for different morphologies cellulose nanoparticle spreading of green laser (Li et al., 2019).

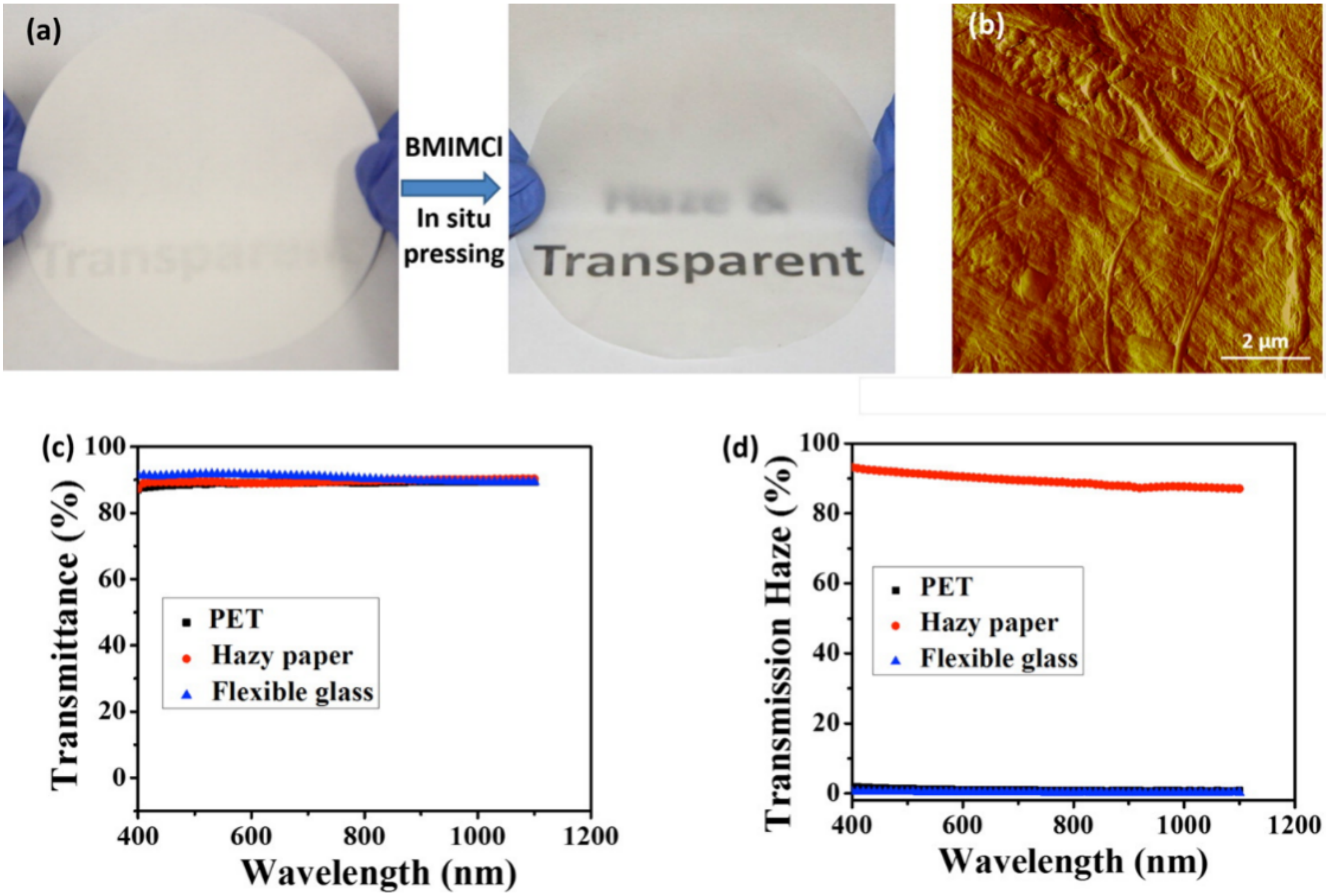

Figure 2

Super hazy paper fabricated by nanowelding exhibits excellent optical properties and surface roughness. (a) In situ pressing the ionic liquid treated paper turns a regular paper into a highly transparent and hazy paper. (b) AFM phase image with a size of 8 × 8 mm^2 showing the surface morphology of super hazy paper. Comparison of total forward transmittance (c) and transmittance haze (d) for super hazy paper, flexible glass, and PET (Zhu et al., 2016).

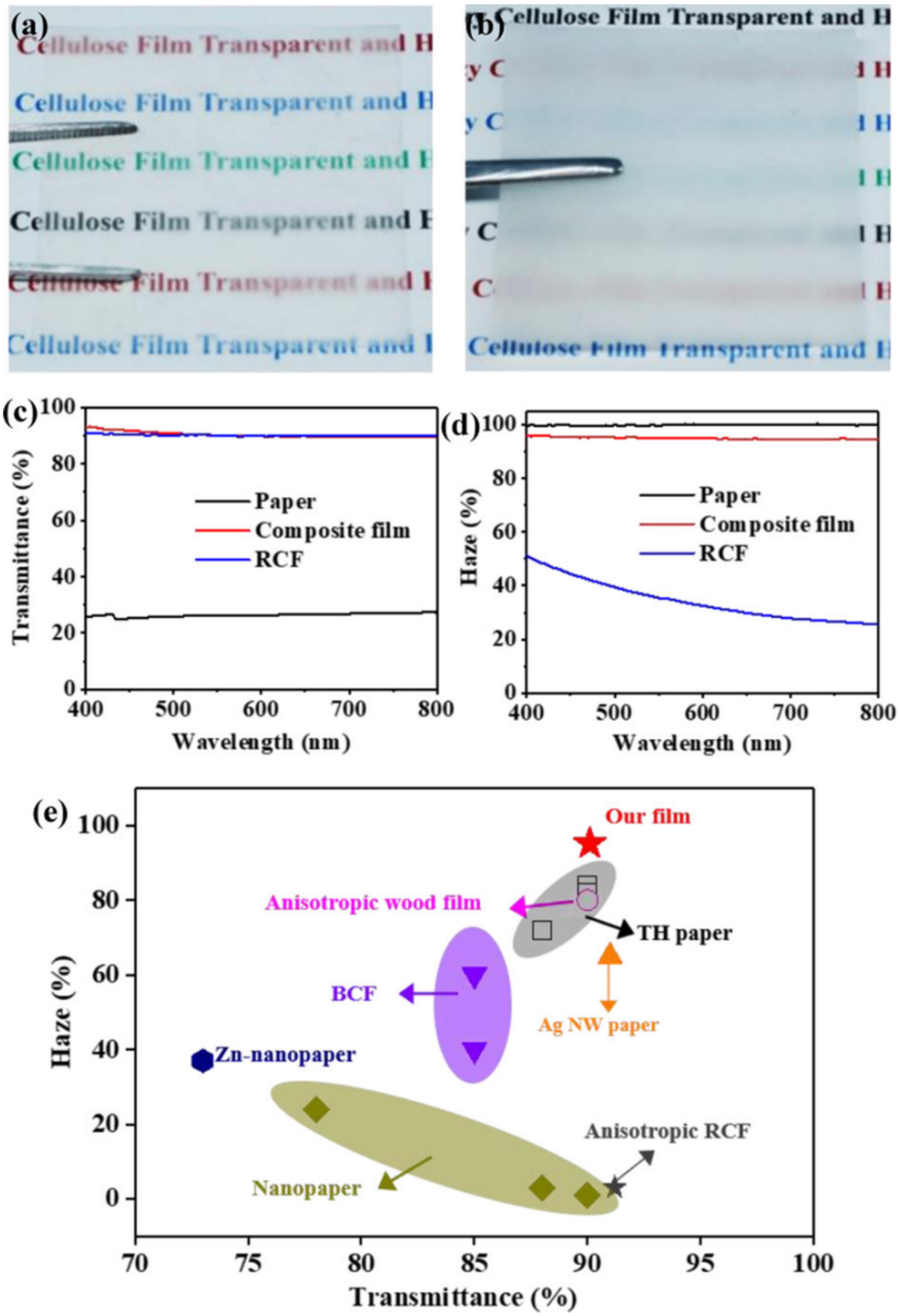

Figure 3

Optical properties of common paper, our composite film, and RCF. (a) Highly transparent and hazy all-cellulose composite film with close contact to the color letters underneath to indicate its high light transmittance. (b) The same composite film at 1cm away from the substrate to indicate its high transmission haze. (c) Total transmittance and(d) transmission haze of our composite film, common paper, and RCF (Hou et al., 2020).

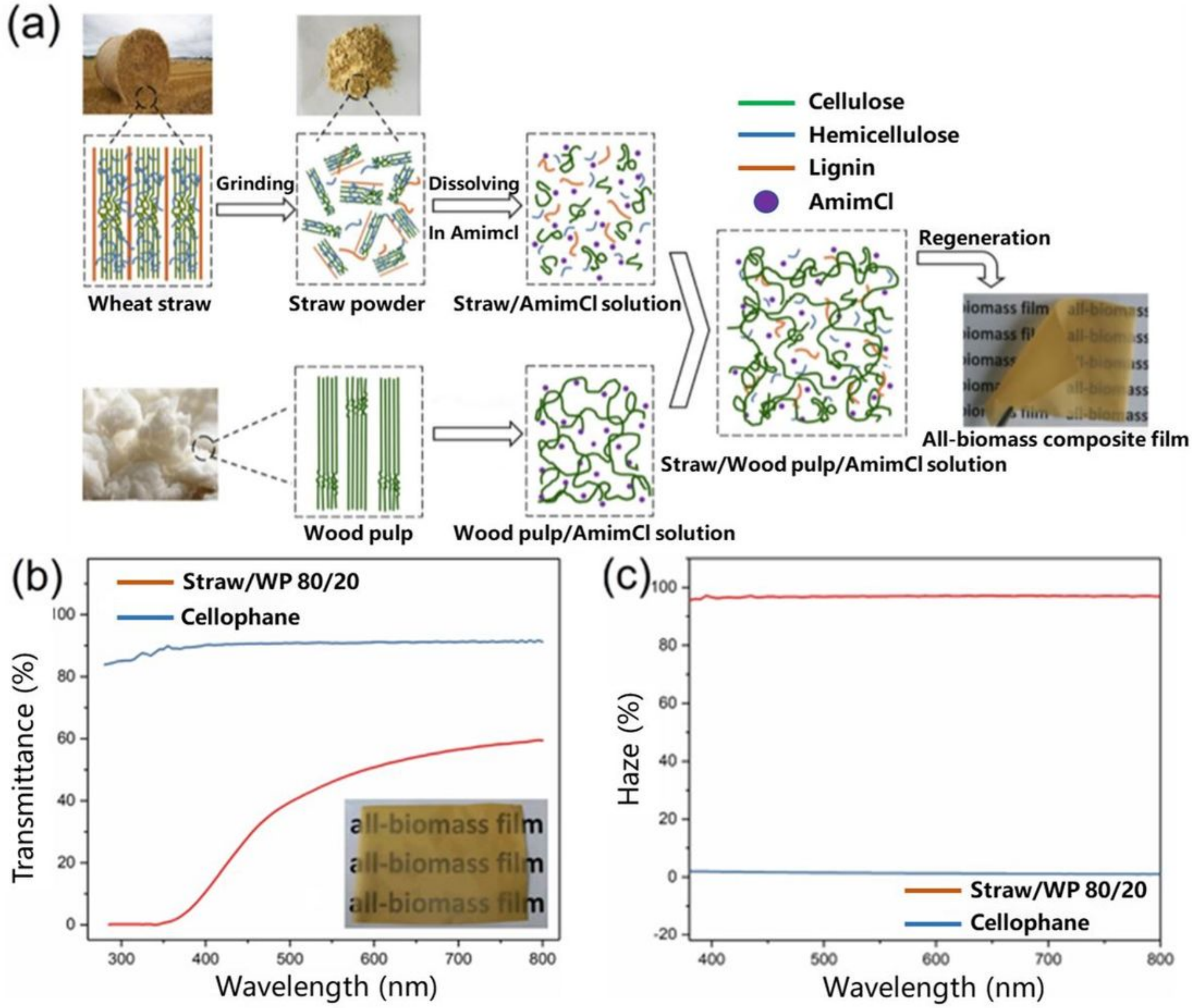

Figure 4

(a) Schematic illustration of the preparation procedure of straw-based all-biomass films. (b) UV−vis transmission spectra of the straw/WP film (straw/WP, 80/20) and pure cellulose film. The insert image is a photograph of the straw/WP film (straw/WP, 80/20). (c) Transmission haze of the straw/WP film (straw/WP, 80/20) and cellophane in the visible region. (Li et al., 2019).

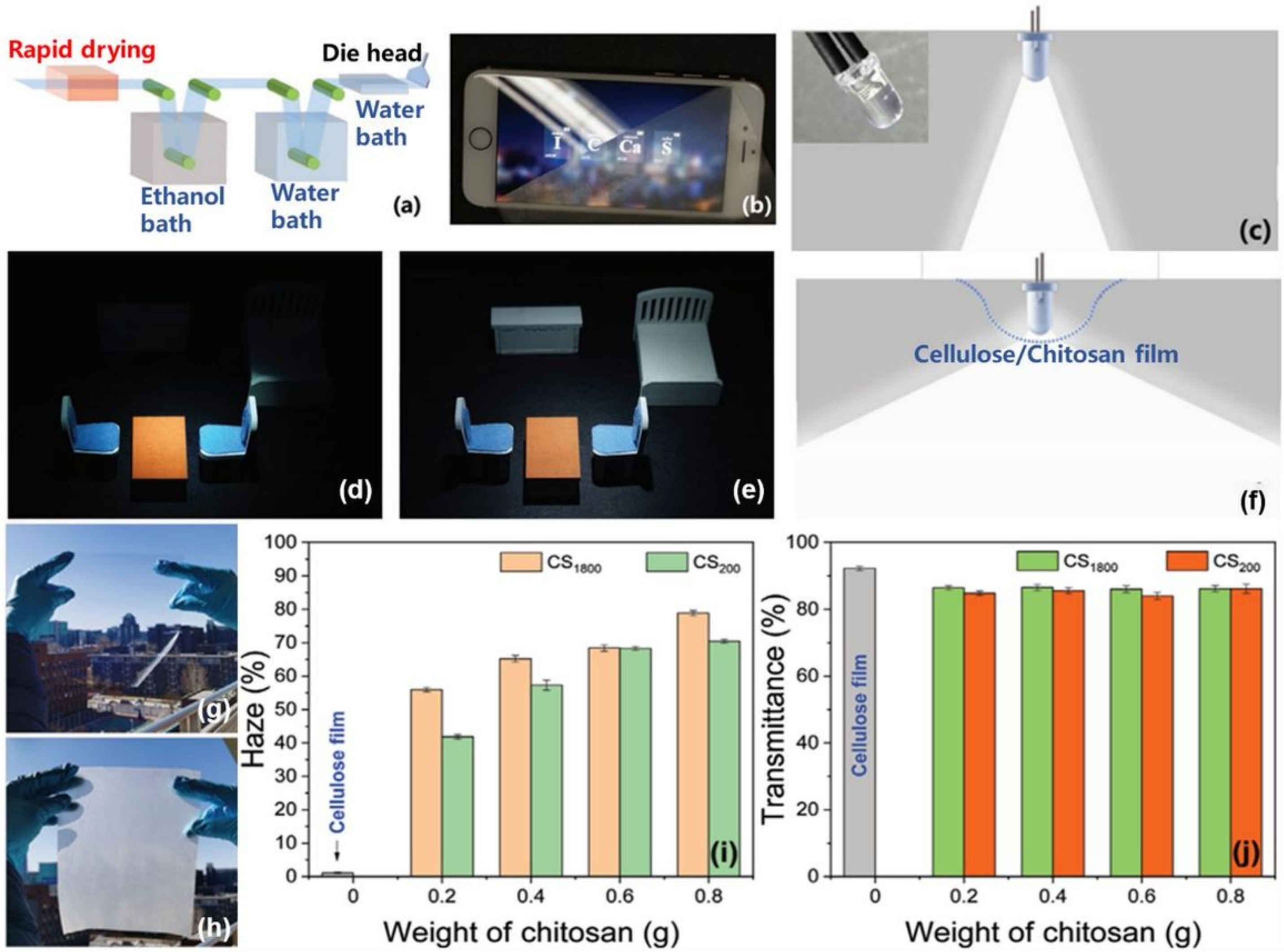

Figure 5

(a) a schematic illustration of the cellulose/chitosan composite films continuous production line; (b) anti-glare effect of the hazy cellulose/ chitosan film on the screen of smart phone; (c) the light scattering diagram of the naked LED bead (insert, the commercial LED bead) or (f) LED bead covered with the hazy film; the illuminating conditions of the room model installed with (d) naked LED bead or (e) LED bead covered with hazy film; (g,h) the digital images of the transparent cellulose film and the hazy cellulose/chitosan composite film; (i,j) the statistically average haze and transmittance of cellulose film and different cellulose/chitosan composite films ranged from 400 to 800 nm. (Cheng, Tian, Mi, Zheng, & Zhang, 2020).

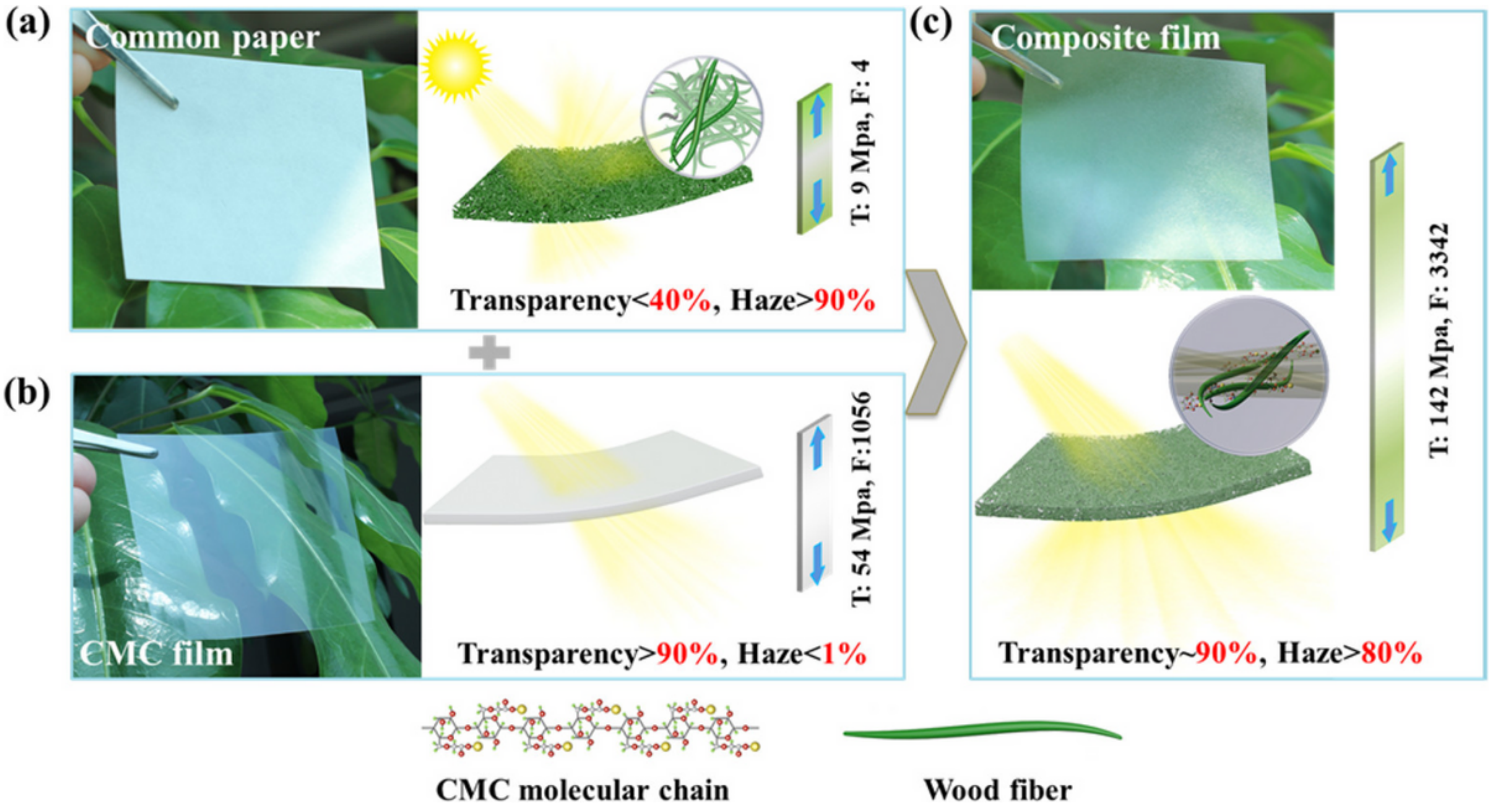

Figure 6

Schematic showing the incorporation of molecular CMC into common paper to prepare mechanically robust, transparent, and hazy all cellulose composite film with excellent mechanical properties. (a) Opaque but hazy common paper with low mechanical properties. (b) Transparent and clear CMC film with medium mechanical properties. (c) Transparent and hazy all-cellulose composite film with excellent tensile strength and folding times (Hu et al., 2018).

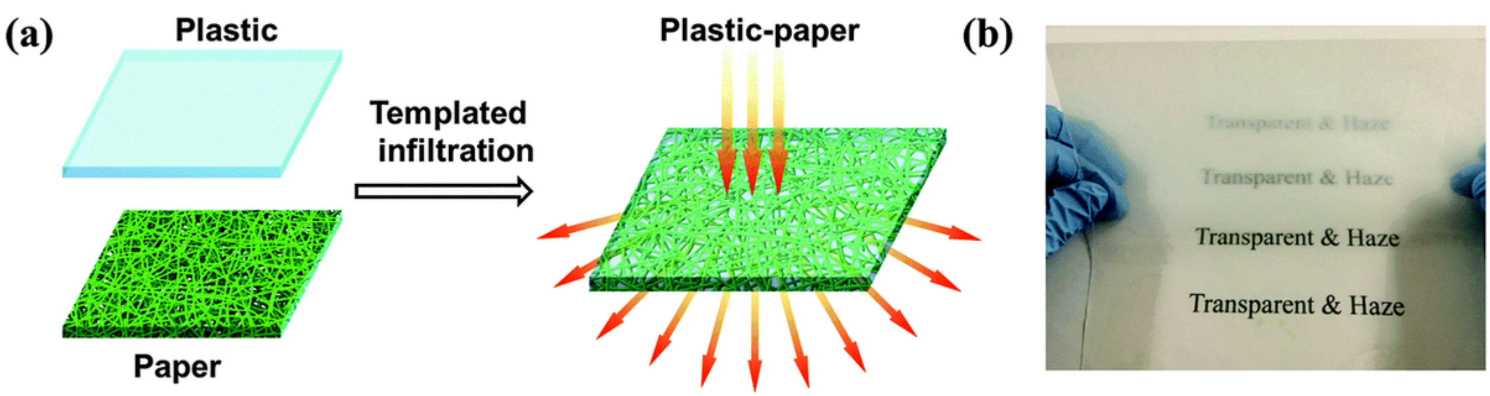

Figure 7

(a) Schematic showing the combination of plastic and paper to form a plastic–paper substrate via templated polymer infiltration. (b) Images of a plastic–paper by PDMS templated epoxy-resin infiltration into porous paper. The letters behind the plastic–paper displays different readability, which is due to the high transparency and high haze of the plastic–paper (Yao et al., 2016).

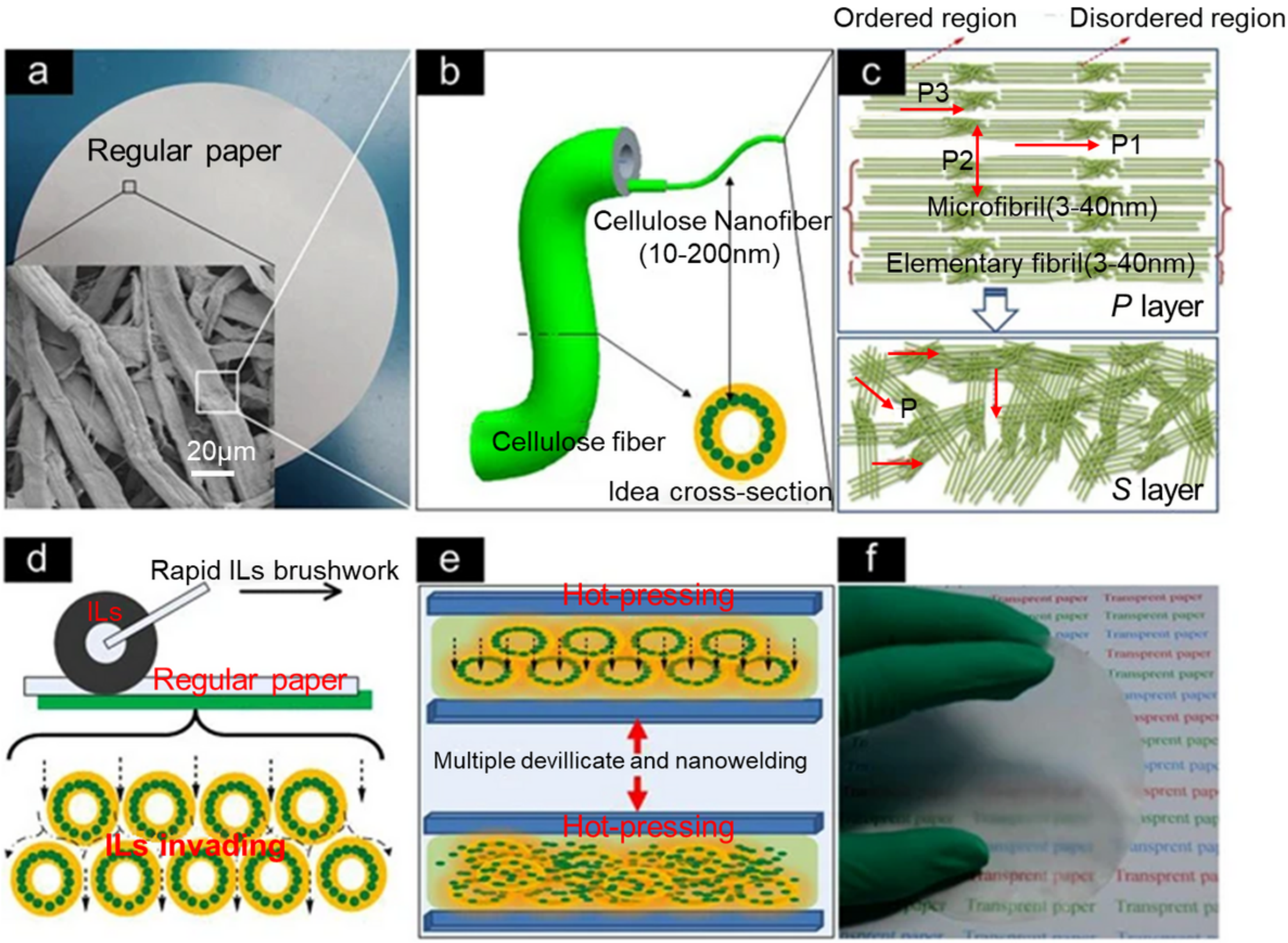

Figure 8

(a) The invading models of ILs in fibrous cellulose matrix under a hot-pressing process. (b) Formation of cellulose nanofibers and its nanowelding process. (c) Digital picture of the nanostructured paper. Optical transmittance curve (d) and optical haze (e, 550nm) of the different nanostructured paper samples ranging from 400 nm to 1100 nm were compared. (f) Digital picture of the nanostructured paper (Ou et al., 2017).

Supplementary Files

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