Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Aligned Cellulose/Nanodiamond Plastic with High Thermal Conductivity

Na Song [a,*‡], Donglei Cao [a,‡], Xian Luo [b], Yongqiang Guo[c], Junwei Gu [b, c,*],

Peng Ding [a, *]

[a] Research Center of Nanoscience and Nanotechnology, Shanghai University, 99 Shangda Road Shanghai 200444, P.R. China.

[b] State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi' an, Shaanxi, 710072, P.R. China.

c MOE Key Laboratory of Material Physics and Chemistry under Extraordinary Conditions, Department of Applied Chemistry, School of Science, Northwestern Polytechnical University, Xi' an, Shaanxi, 710072, P.R. China.[1]

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* Corresponding author:

E-mail: snlxf@shu.edu.cn (N. Song), dingpeng@shu.edu.cn (P. Ding), nwpugjw@163.com (J.W. Gu).

‡ These authors contributed equally to this work.

Experimental

Materials.

ND (＞99%) and cellulose were purchased from Sigma-Aldrich Co. (St. Louis, MO). Urea (⩾99%), sodium hydroxide (NaOH, analytically pure) and absolute ethyl alcohol (analytically pure) were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).

Preparation of cellulose/nanodiamond (CND) hydrogel

CND hydrogels were prepared according to a reported method.[1-3] Typically, suitable cellulose powder was added to the solution which containing 7wt % NaOH/urea aqueous prepared by sonication and precooled to 0 °C. Cellulose solution was received after the obtained solution was treated at 0 °C in an ice bath followed by stirring for 2 h. Meanwhile, a certain amount of nanodiamond (ND) dispersed in deionized (DI) water after sonicated for 1 h in a water bath was added to cellulose solution above mentioned and then the mixture was stirred violently to acquire cellulose/ND solution. The mixture solution underwent vacuum filtration for 30 min at 0 °C in an ice bath to remove air bubbles. The obtained mixture solution was spread on poly(tetrafluoroethylene) moulds and subjected to physical regeneration with ethanol and then thorough washing with DI water to construct CND hydrogels with 3.0 mm thickness.

Fabrication of CND plastics.

Cellulose and CND hydrogels were prepared according to our previous work.[2] The resultant hydrogels were sandwiched between stainless steel plates and then hot pressed at 70 °C. After 48 h, CND plastics with 0.2 mm thickness were obtained. The schematic illustration of CND plastics preparation is shown in Fig.1. The cellulose plastics with 0.5 wt% ND, 1 wt % ND, 2 wt % ND, and 5 wt % ND were named 05CND, 1CND, 2CND, 5CND, respectively. Virgin cellulose plastic, named CBP, was also prepared for comparison.

Characterization.

Avatar 370 Fourier transform infrared spectrometer (Nicolet, United Stated) was equipped to get Fourier transform infrared (FTIR) spectra. To investigate morphology and microstructure of the fabricated plastics, scanning electron microscopy (SEM, JSM6700F emission scanning electron microscope) was used. A 200 CX 178 (JEOL, Japan) provided transmission electron microscopy (TEM) images. The specific heat (C_p) and thermal diffusivity (α) of fabricated bioplastics were performed on a Netzsch LFA 447 Nanoflash at 25 °C. Every sample was tested six times to eliminate test errors. Density (ρ) was obtained by a density balance (SARTORIUS, China). Thermal conductivity (TC) was got using the formula:

$$TC = \alpha \cdot C_p \cdot \rho. \tag{1}$$

The laser flash system is introduced in our previous works.[4, 5] Small discs with diameter of 25.0mm and 12.7mm were respectively cut out from the prepared composite as the heat conduction test samples in the direction of "in-plane" and "through-plane". A universal tensile tester (Instron 5569A) was used to get the stress-strain curves at room temperature. Before tested, the samples were cut into 10 × 50 mm strips and Transmission spectrum was obtained using a UV-2501PC (Shimadzu, Japan) UV-visible spectrophotometer. Precision LCR Meter (TH2829A, Tonghui) was used to get electrical resistance. The resistivity can be calculated using the formula.

$$\rho = \frac{RS}{L} \tag{2}$$

Where R is electrical resistance, S and L is area and thickness of samples, respectively.

2. TEM testing of ND

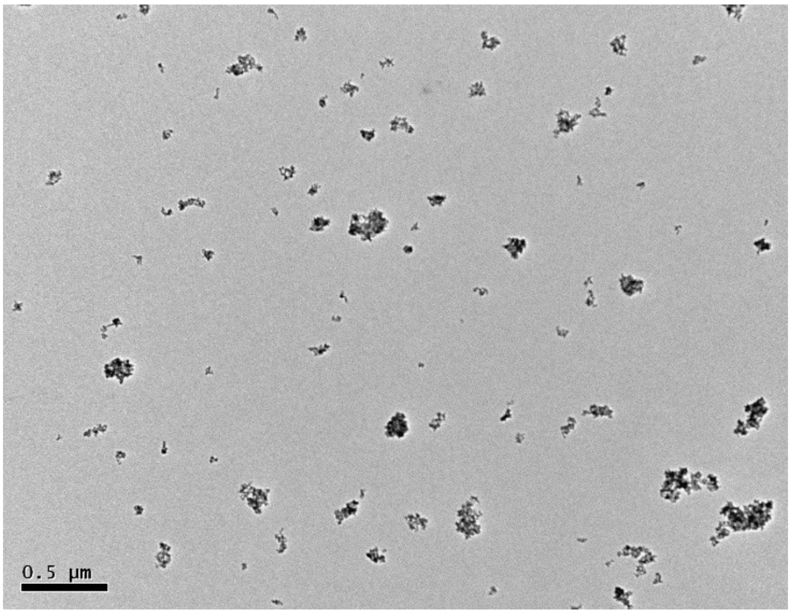

Figure S-1 TEM image of ND

3. Photograph of CND solution and CND hydrogel

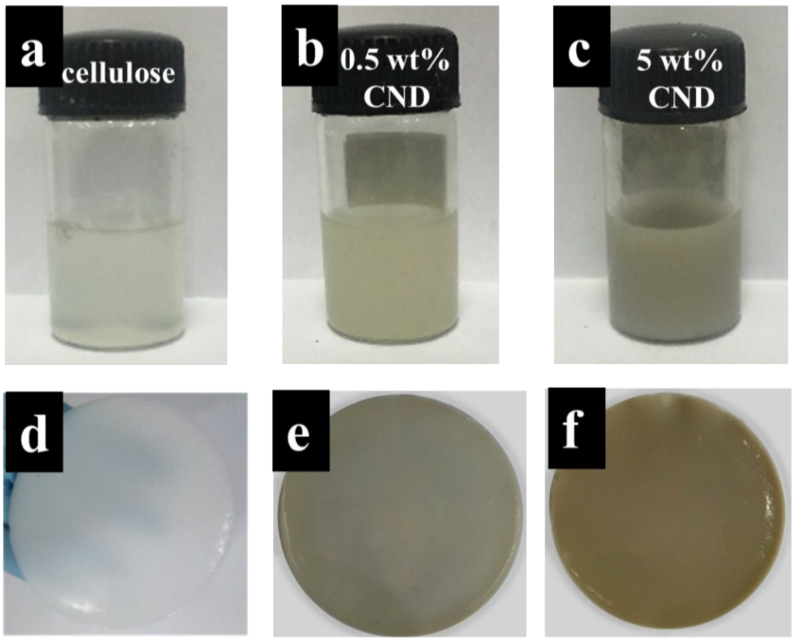

Figure S-2. Photograph of (a) cellulose solution; (b) cellulose solution with 0.5 wt% ND; (c) cellulose solution with 5 wt% ND; (d) cellulose hydrogel; (e) cellulose hydrogel with 0.5 wt% ND; (e) cellulose hydrogel with 5 wt% ND;

3. SEM testing of CND plastics

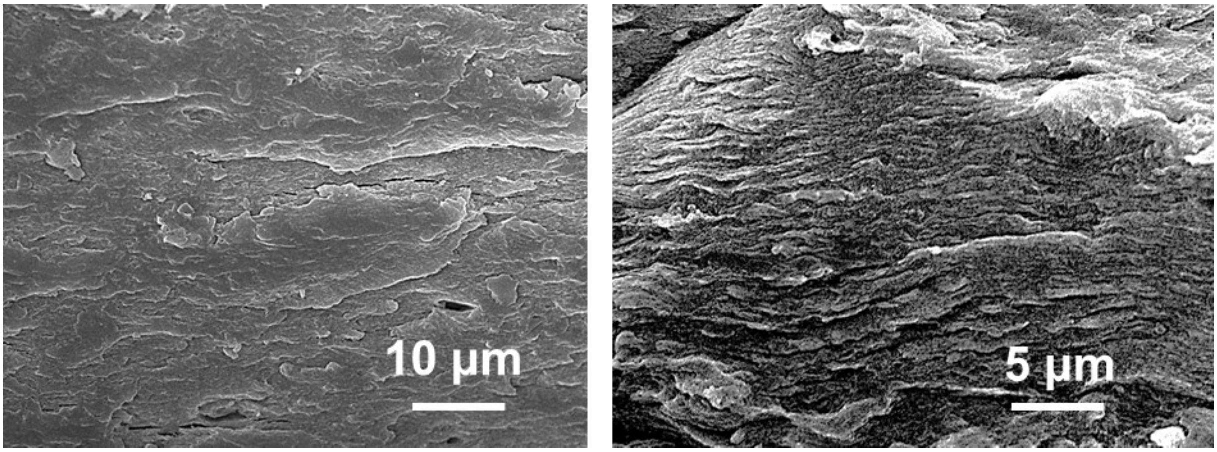

Figure S-3. The cross-sectional SEM images of 05CND with different magnification

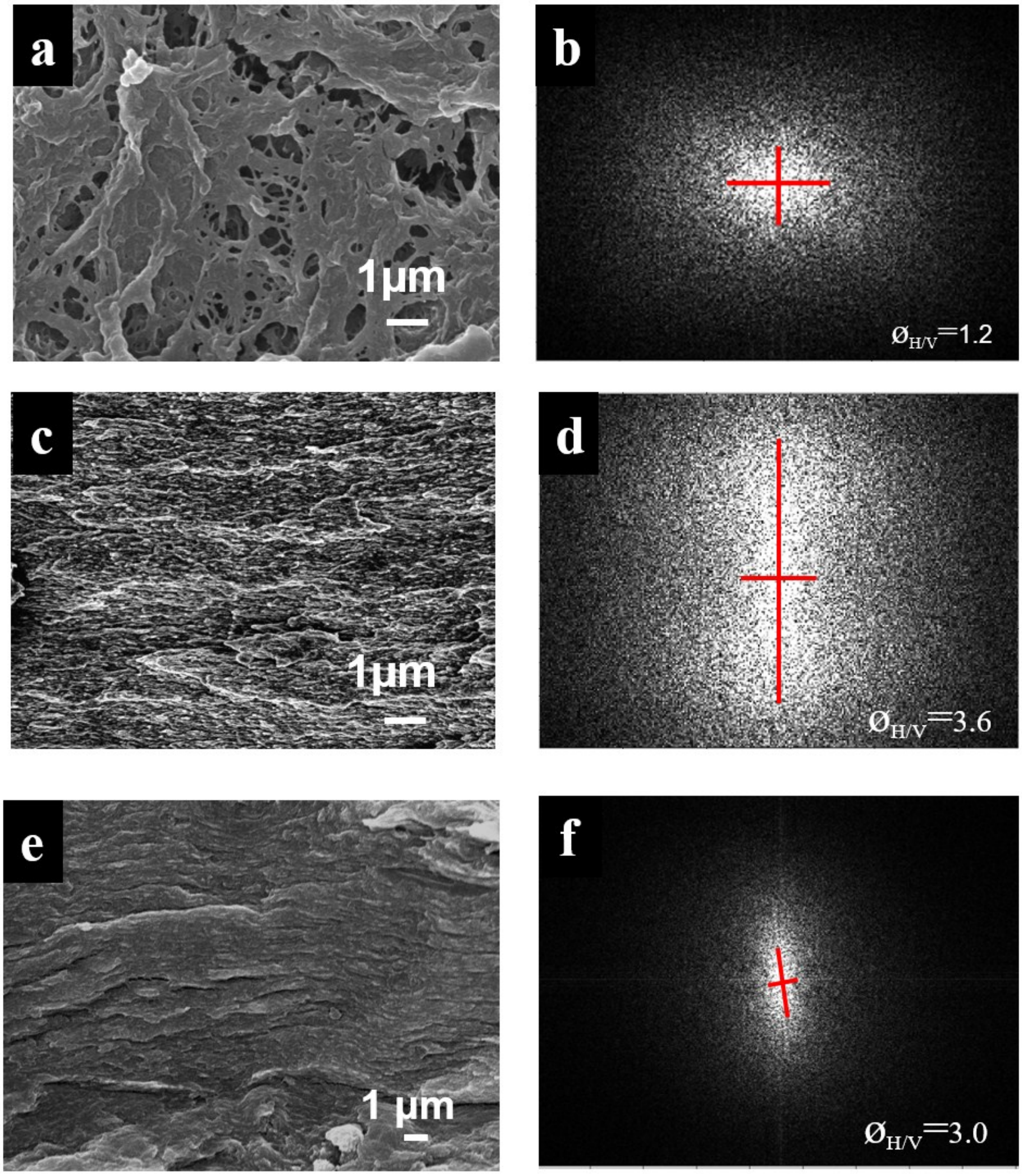

Figure S-4 **Cross-sectional SEM images (a) cellulose hydrogel, (c) CBP, (e) 5 wt% CND and FFT frequency domain images of (b) cellulose hydrogel, (d) CBP, (f) 5 wt% CND.**

Fast Fourier transform (FFT) method was introduced to prove cellulose is oriented. In FFT, the complex spatial patterns that represented by gray values in each pixel of a picture is converted into direction dependent frequency components. FFT is tested on a SEM picture, where the image is transformed into a frequency domain. The intensity of each pixel describes an angular dependence on patterns of spatial alignment where pixels displaying high intensity values are clustered along the orientation of the highest degree of directional anisotropy. Thus, the FFT method is used for analysing the orientation properties of collagen bundles and carbon-fiber reinforced composite.[6]

As shown in Fig.S-4, a、c and e are cross sectional SEM images of cellulose hydrogel, cellulose bioplastic (CBP) and 5 wt% cellulose/nanodiamond plastic (CND), respectively. The FFT white spots of the cellulose hydrogel (Fig.S-4 b) exhibits a circular distribution, while CBP and CND (Fig.S-4 d and f) present an elliptical distribution. The ratio of the scattering vector lengths between the major and minor axis was define as $\Phi_{\mathrm{H/V}}$. Generally, the higher $\Phi_{\mathrm{H/V}}$ represents the higher degree of orientation structure. It can be concluded that CBP and CND exhibits preferable $\Phi_{\mathrm{H/V}}$ compared to the cellulose hydrogel, which means cellulose hydrogel (Fig.S-4 a) is disordered while cellulose plastics (Fig.S-4 c and e) are horizontally oriented.

4. Thermal conductivity and Anisotropy index

Table S-1. Overview of thermal diffusion (α) coefficient and thermal conductivities (λ) of the CBP and CND plastics.

Sample	α (mm^2•s^{-1})		λ (W•m^{-1}•K^{-1})	
	α_X	α_Z	λx	λz
CBP	1.251	0.21	2.59	0.443
05CND	1.358	0.245	3.21	0.580
1CND	1.437	0.168	4.46	0.550
2CND	1.796	0.309	4.51	0.777
5CND	1.854	0.171	5.37	0.496

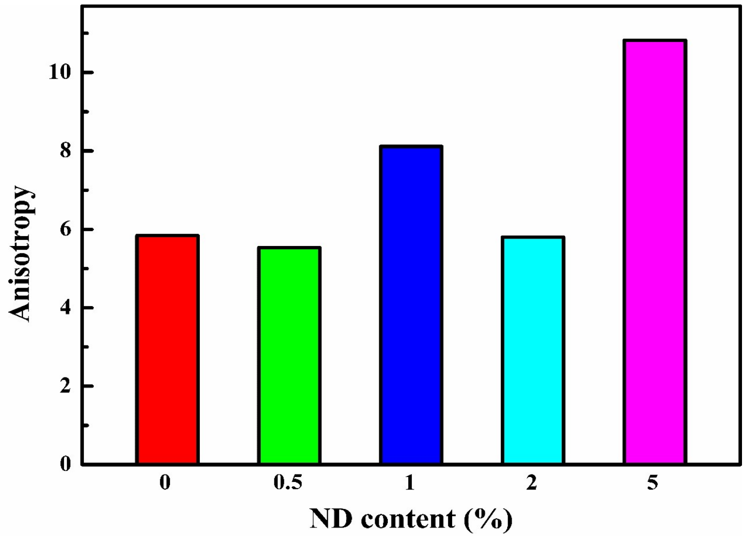

Figure S-5 Anisotropy index (AI) of CBP and CND plastics with different ND content. (AI= λ_x / λ_z, λ_x = in-plan TC, λ_z = through-plan TC)[4, 7]

5. Transparency

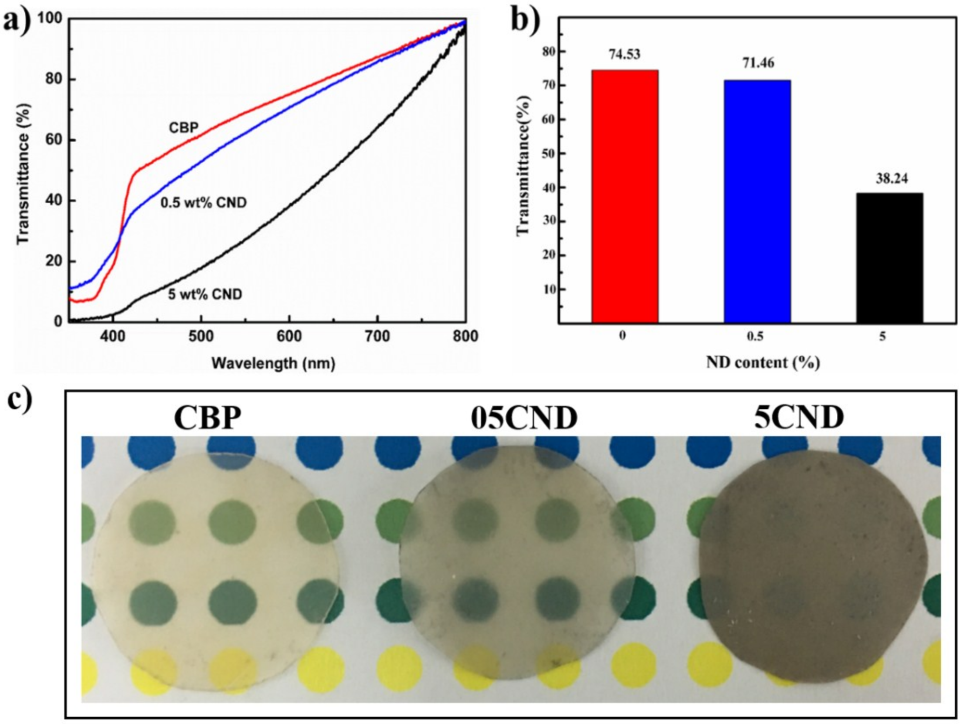

Fig. S-6 (a) UV-vis spectroscopy of CBP and CND plastics; (b) Transmittance of CND plastic with different ND content at 600nm. (c) Physical photographs of CBP, 05CND, 5CND plastics.

The transparency of composites filled with graphene or carbon black becomes poor even at low filler loadings, which is the same with our previous work.[2] As a contrast, CND composites maintain translucent state at 5 wt%. Obviously, the transparency of the composite gradually decreases with the ND content increases (shown in Fig. 5c). Ultraviolet-visible (UV-vis) spectroscopy was introduced to further characterize the transmittance of composite plastics. UV-vis spectroscopy (Fig. **S-6**) presents that pure CBP shows good transparency with 75% transmittance at a wavelength of 600 nm, indicating CBP has a very uniform structure.[2] The transmittance at the wavelength of

600 nm can still be maintained 71.5% as weight fraction of ND is 0.5 wt%, slightly decreasing compared with CBP. But the transparency of the composite plastic decreased significantly with the content of ND reaching to 5 wt%. This phenomenon occurs because the filler ND causes the incident light to be scattered when the composite was irradiated by incident light.[8]

Reference

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