Supplementary Information for

## An all-natural bioinspired structural material for plastic replacement

Qing-Fang Guan<sup>†</sup>, Huai-Bin Yang<sup>†</sup>, Zi-Meng Han<sup>†</sup>, Zhang-Chi Ling, Shu-Hong Yu\*

\* Correspondence to: Shu-Hong Yu (<u>shyu@ustc.edu.cn</u>).

<sup>†</sup> These authors contributed equally to this work.



Supplementary Figure 1 | Atomic force microscope (AFM) image of TEMPO-oxidized cellulose nanofiber (CNF).



Supplementary Figure 2 | Characterization of TiO<sub>2</sub> coated mica microplatelet (TiO<sub>2</sub>-mica). a, Scanning electron microscope (SEM) image of micro-sized TiO<sub>2</sub>-mica platelets. The average lateral size of TiO<sub>2</sub>-mica is about 25  $\mu$ m, and the thickness is about 1  $\mu$ m. b, The statistical lateral size of the TiO<sub>2</sub>-mica microplatelet, showing the lateral size ranging from 4  $\mu$ m to 60  $\mu$ m. c, SEM image of the surface of TiO<sub>2</sub>-mica microplatelet, showing the TiO<sub>2</sub>-mica consisted of TiO<sub>2</sub> nanograins with diameters ranging from 10 to 100 nm on its surface. d, Transmission electron microscope (TEM) image of the micro-sized TiO<sub>2</sub>-mica platelet. e, TEM image of the surface of TiO<sub>2</sub>-mica microplatele, showing the TiO<sub>2</sub> nanograins were evenly coated on the surface of TiO<sub>2</sub>mica. f, XRD pattern of TiO<sub>2</sub>-mica microplatelets.



Supplementary Figure 3 | The comparison of the unique connected-nanograin structure between natural nacre and the all-natural bioinspired structural material. a, Surface of *Anodonta woodiana*. b, Surface of the all-natural bioinspired structural material.



Supplementary Figure 4 | Comparison of Fourier transform infrared spectroscopy (FT-IR).



Supplementary Figure 5 | Comparison of loss factor (tan  $\delta$ ). The curves show that Tan  $\delta$  of the all-natural bioinspired structural material is more stable than widely used petroleum-based plastics under temperature ranging from 30 °C to 190 °C.



**Supplementary Figure 6 Comparison of thermal conductivity.** Comparison of thermal conductivity of the all-natural bioinspired structural material with typical polymers<sup>1</sup>.



**Supplementary Figure 7 Comparison of thermal diffusivity.** Comparison of thermal diffusivity of the all-natural bioinspired structural material with typical polymers<sup>1</sup>.



Supplementary Figure 8 | Comparison of thermal and mechanical properties of all-natural bioinspired structural material with typical polymers, metals, and ceramics. a, Ashby diagram of specific modulus versus specific yield strength<sup>1,2</sup>. b, Ashby diagram of thermal expansion versus specific fracture toughness<sup>1,2</sup>.



**Supplementary Figure 9 | Comparison of hardness.** Comparison of Shore D hardness number of the all-natural bioinspired structural material with other widely used materials. Error bars represent standard deviation.

Properties	This work	PA
Ultimate strength (MPa)	281	90-165
Modulus (GPa)	20	2.62-3.2
Fracture toughness ( $K_{\rm Ic}$ , MPa m <sup>1/2</sup> )	6.7	2.22-5.62
Hardness (HSD)	~94	~83
Thermal expansion coefficient (10 <sup>-6</sup> K <sup>-1</sup> )	~7	~147
Thermal conductivity (W m <sup>-1</sup> K <sup>-1</sup> )	~0.53	~0.24

Supplementary Table 1 | Comparison of mechanical and thermal properties of all-natural bioinspired structural material with polyamide (PA)<sup>1</sup>.

## **Supplementary References**

- 1. M. Ashby, *Materials Selection in Mechanical Design*. (Elsevier, Oxford, 2011).
- 2. Guan Q. F., et al. Lightweight, tough, and sustainable cellulose nanofiber-derived bulk structural materials with low thermal expansion coefficient. *Sci. Adv.* **6**, eaaz1114 (2020).