

Supporting Information for

**Facile Fabrication of Robust and Stretchable Cellulose Nanofibers/Polyurethane Hybrid Aerogels**

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Supporting information consists of 5 pages, 4 Tables, 5 Figures and 2 Videos.

**Table S1.** The compositions of the CNF/PU hybrid aerogels

| Samples     | CNF aerogel (wt%) | PU (wt%) | Density (g/cm <sup>3</sup> ) |
|-------------|-------------------|----------|------------------------------|
| CNF aerogel | 100               | 0        | 0.03                         |
| CNF/PU-48   | 52                | 48       | 0.06                         |
| CNF/PU-66   | 34                | 66       | 0.09                         |
| CNF/PU-75   | 25                | 75       | 0.12                         |

**Table S2.** Thermal parameters of the CNF/PU/octadecane composites

| Samples              | PCM content (wt%) | $\Delta H_m$ (J/g) | $\Delta H_c$ (J/g) |
|----------------------|-------------------|--------------------|--------------------|
| CNF/PU-48/octadecane | 92.5              | 223.3              | 222.8              |
| CNF/PU-66/octadecane | 87.1              | 207.6              | 207.8              |

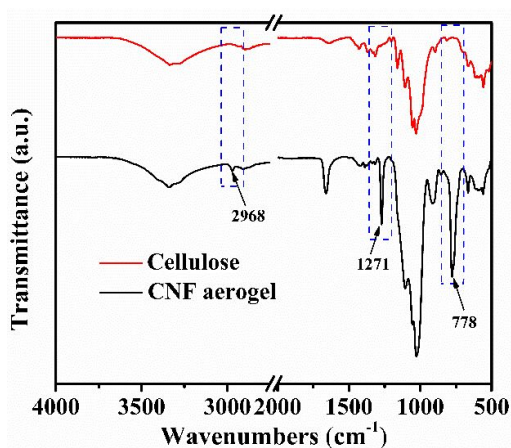
**Table S3.** Thermal conductivity of octadecane and CNF/PU/octadecane composites

| Samples              | Thermal conductivity (Wm <sup>-1</sup> K <sup>-1</sup> ) |
|----------------------|--|
| octadecane           | 0.32±0.01  |
| CNF/PU-48/octadecane | 0.23±0.02  |
| CNF/PU-66/octadecane | 0.25±0.06  |
| CNF/PU-75/octadecane | 0.23±0.03  |

**Table S4.** Thermal parameters of CNF/PU-75/octadecane after different thermal cycling numbers

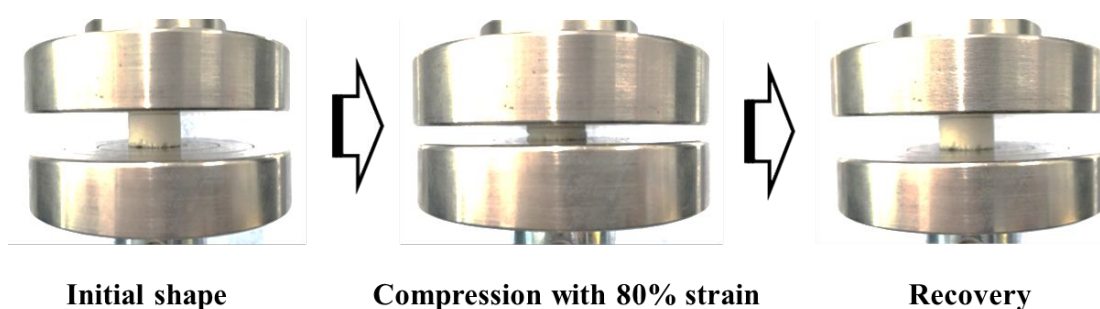
| Cycling numbers   | $T_m$ (°C) | $T_c$ (°C) | $\Delta H_m$ (J/g) | $\Delta H_c$ (J/g) |
|-------------------|------------|------------|--------------------|--------------------|
| 1 <sup>th</sup>   | 37.6       | 16.6       | 190.0              | 189.7              |
| 25 <sup>th</sup>  | 36.6       | 18.1       | 190.1              | 188.9              |
| 50 <sup>th</sup>  | 36.6       | 17.9       | 189.0              | 188.7              |
| 75 <sup>th</sup>  | 36.6       | 18.4       | 188.3              | 187.4              |
| 100 <sup>th</sup> | 36.6       | 18.4       | 187.1              | 186.2              |

$T_m$ : melting temperature,  $T_c$ : crystallization temperature,  $\Delta H_m$ : melting enthalpy,  $\Delta H_c$ : crystallization enthalpy.

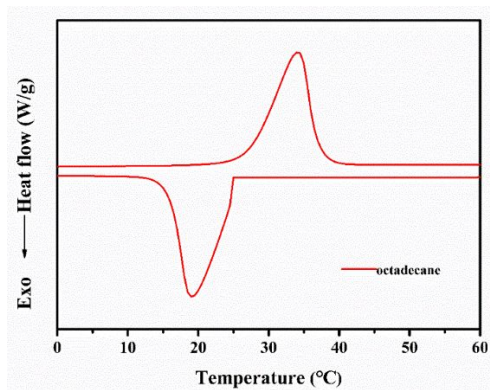


**Fig.S1.** FTIR spectra of the cellulose and the CNF aerogel

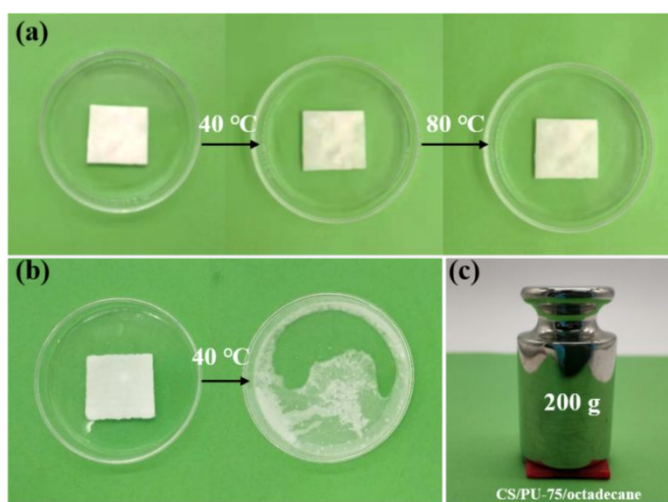
The FTIR spectra of the cellulose and CNF aerogel are compared in Fig.S1. The new peaks at  $778\text{ cm}^{-1}$  in the CNF aerogel spectrum is assigned to C-Si asymmetric stretching vibration. The peak at  $1100\text{ cm}^{-1}$ - $1130\text{ cm}^{-1}$  was belonged to the stretching absorption peaks of the Si-O-Si bonds, overlapping with the peaks of the C-O bonds of cellulose. The new peak at  $1271\text{ cm}^{-1}$  and  $2968\text{ cm}^{-1}$  correspond to the stretching and bending vibration of  $-\text{CH}_3$  in the siloxane compounds. The above results confirmed the successful occurrence of polysiloxane reaction between cellulose and methyltrimethoxy silane.



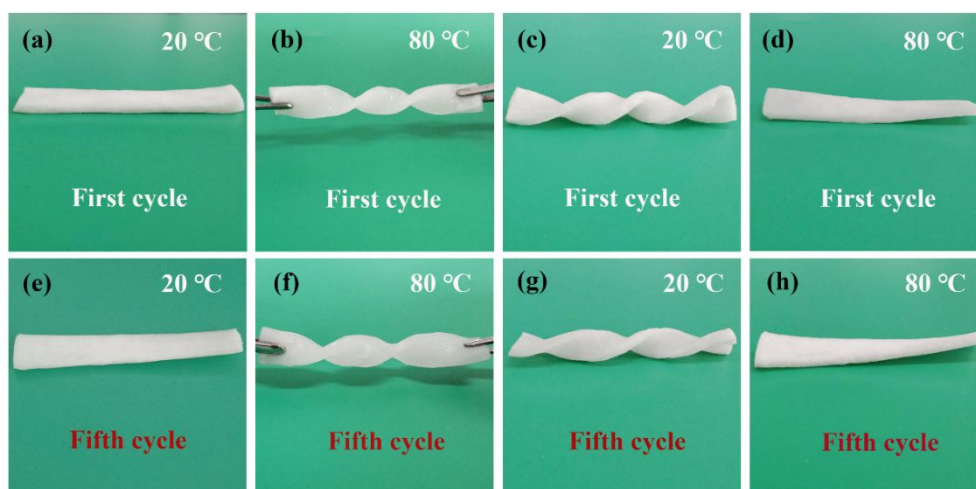
**Fig.S2.** Shape recovery process of CNF/PU-75



**Fig.S3.** DSC curves of octadecane



**Fig.S4.** Shape stability of (a) CNF/PU-75/octadecane and (b) octadecane with increasing temperature. (c) The mechanical stability of CNF/PU-75/octadecane at 40 °C.



**Fig.S5.** Shape memory behavior of the CNF/PU-75/octadecane at 80 °C. a-d Images showed the shape memory process of the CNF/PU-75/octadecane. e-h Images showed the CNF/PU-75/octadecane recovered its permanent shape after five cycles.