

## Supplementary Material

### **Making Gd<sub>2</sub>O<sub>3</sub> Nanofibers Flexible by Grain-Boundary Toughening**

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Supporting Information contains:

Supplementary Methods

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## Supplementary Methods

**Materials:** Polyvinylpyrrolidone (PVP,  $M_w=1,300,000$ ), N-propanol (99%), methyltrimethoxy-silane (MTMS, 98%), and phosphoric acid ( $H_3PO_4$ ) were all from Shanghai Aladdin Reagent Co., LTD. Gadolinium nitrate ( $Gd(NO_3)_3$ , 99.99%) comes from Shanghai Maclin Biochemical Technology Co., LTD. The citric acid (99.5%) comes from Shanghai Yien Chemical Technology Co., LTD. Anhydrous ethanol ( $C_2H_6O$ ,  $\geq 99.8\%$ ) comes from Sinopod Chemical Reagent Co., LTD. In addition, deionized water is provided by the Heal Force NW system.

**Preparation of GNFs:** First, 5.6 g  $Gd(NO_3)_3$  and 2 g citric acid were added to 20.062 g of deionized water and stirred until the state was clarified. Then 40.138 g of n-propanol and 4.2 g PVP were added. After stirring at room temperature for about 12 h, gadolinium precursor solution was obtained. Then, under the conditions of suitable temperature ( $22\pm 3$  °C) and relative humidity ( $42\pm 3\%$ ), 10 ml precursor solution was absorbed by an injection needle and bubbles were expelled. The spinning parameters such as voltage (20 kV), receiving distance (20 cm), and perfusion speed ( $1\text{ mL h}^{-1}$ ) were set for electrospinning. The precursor fibrous membrane was dried under vacuum at 100 °C for 0.5 h and then calcined in an air Muffle furnace at 600°C for 1 h. The polymer is completely pyrolyzed, and at the same time, the high temperature promotes the formation and growth of  $Gd_2O_3$  grains and finally produces flexible GNFs.

**Preparation of  $Gd_2O_3$  nanofibrous aerogel:** Firstly, 0.15 g GNFs and 0.05 g  $SiO_2$  nanofibrous membrane were cut into small pieces and put into 200 g deionized water. The fiber dispersion solution was obtained by homogenizing the membrane with a high-speed dispersing machine (IKA T25, IKA Group, Germany) at a speed of 13000 rpm for 20 min. Then, 1.9 g Methyltrimethoxysilane, 19 g  $C_2H_6O$ , 7.6 g water, and 0.02 g  $H_3PO_4$  were respectively stirred to a clarified state. The solutions were mixed and continued to be stirred for 0.5 h to obtain silica sol. Then, 1.365 g silica sol was dropped into the fiber-dispersing solution and stirred for about 2 h to obtain a stable and uniform fiber-dispersing mixed sol system. Pour into the mold, pre-freeze, and set with liquid nitrogen, remove the frozen block, and immediately transfer to the freeze dryer (Pilot-6, Beijing Boyikang Equipment Co., LTD.) vacuum freeze drying for 24 h, and finally obtain  $Gd_2O_3$  nanofibrous aerogel.

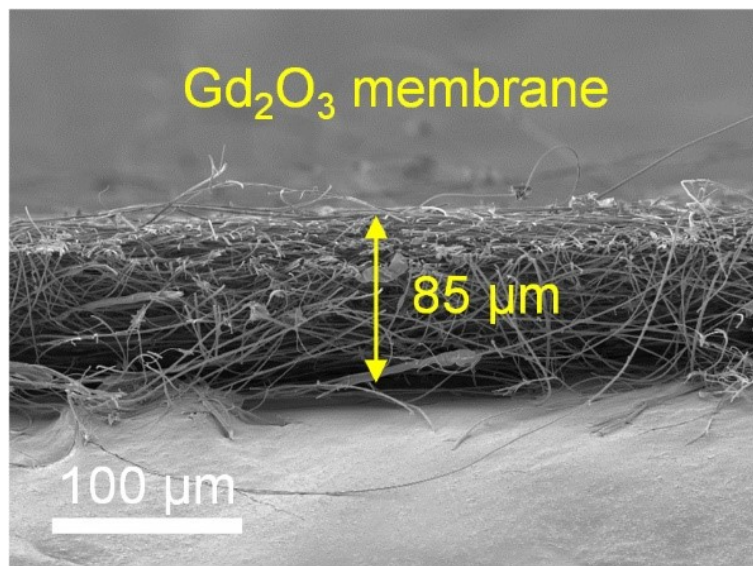
**Neutron absorption simulation:** The simulation program is FLUKA, version 4-3.3.

The particle source used in the simulation is a unidirectional point source of a single energy (1 MeV) NEUTRON, which is vertically incident on the material. The material is a cylinder with a radius of 1cm and a length of 4 cm, the cylinder is filled with Gd<sub>2</sub>O<sub>3</sub>, and the density is set to 0.01, 0.03, 0.05, 0.1, and 1.0 g/cm<sup>3</sup>, respectively. The neutron cross-section library used in the simulation is ENDF-VIII0. Use a USRBIN card to record the flux distribution of neutrons.

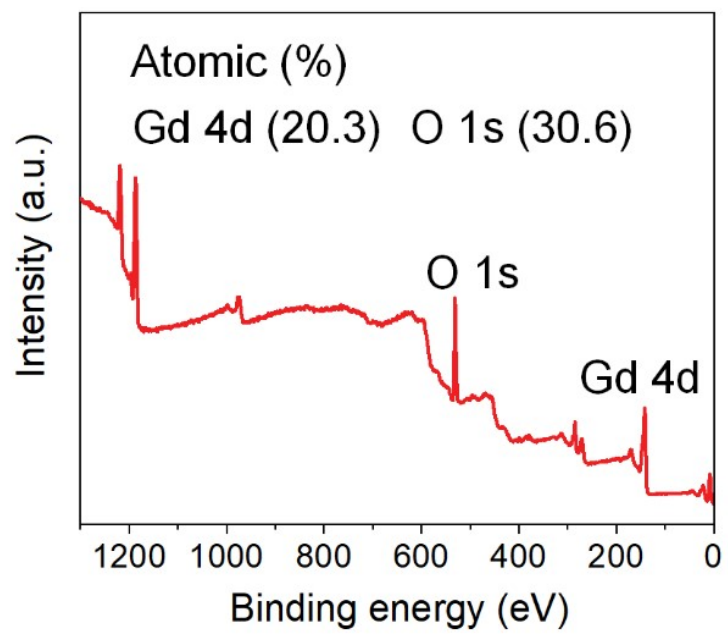
**Characterization:** Scanning electron microscopy (SEM, S-4800, Hitachi, Japan) and energy dispersion spectrometer (EDS, XFLASH 610m, Bruker, Germany) were used to observe the surface morphology and microelements of GNFs. A specific surface area analyzer (BET, ASAP2460, micropolitics Instrument Company, USA) was used to analyze the pore structure in the nanofiber. The flexible characterization of the single fiber was obtained by in-situ bending observation with a transmission electron microscope (TEM, JEM-2100, Nippon Electronics Co., LTD.). The mechanical properties of single fiber were characterized by the SPM mode of Atomic force microscope (AFM, Multimode8, Bruker, Germany). Dynamic thermomechanical analyzer (DMA, Q850, TA Company) was used to test the tensile fracture and buckling cycle of Gd<sub>2</sub>O<sub>3</sub> nanofibrous membrane, and the compression and compression cycle of Gd<sub>2</sub>O<sub>3</sub> nanofibrous aerogel. A thermal constants analyzer (Hot disk TPS 2500S, Swiss Hot Disk Company) was used to determine the thermal conductivity of the aerogel, and an infrared thermal imager (TiS75, American Fluke Company) was used to take infrared thermal imaging photos of the aerogel on the hot table. TEM and X-ray diffractometer (XRD, D8-Advance, Bruker, Germany) were used to test the crystal structure of the fiber, and the grain size was calculated according to equation (1) Scherrer formula:

$$D = \frac{k\lambda}{\beta \cos\theta}$$

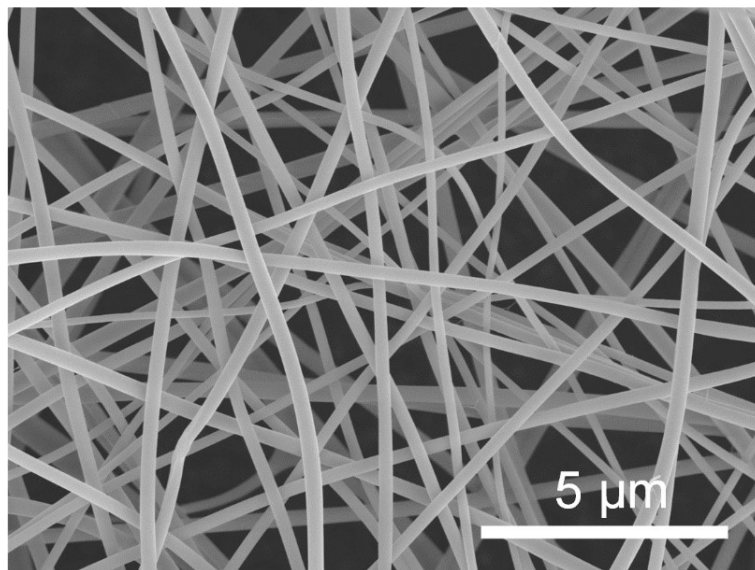
Where  $D$  is the grain size,  $k$  is the constant 0.89,  $\lambda$  is the diffraction wavelength 0.154056nm,  $\beta$  is the half-width and height of the instrument, and  $\theta$  is the diffraction Angle.



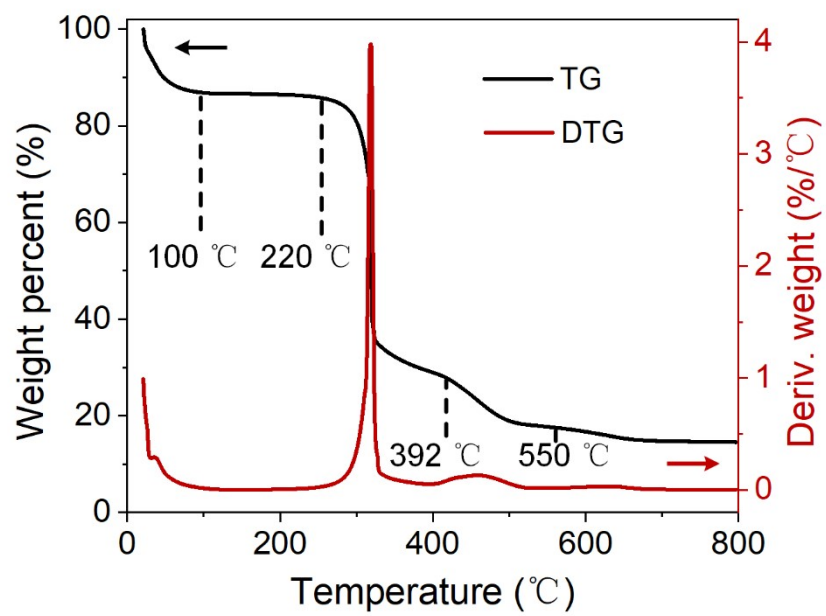
**Figure S1.** Cross-sectional image of the  $\text{Gd}_2\text{O}_3$  nanofibrous membrane.



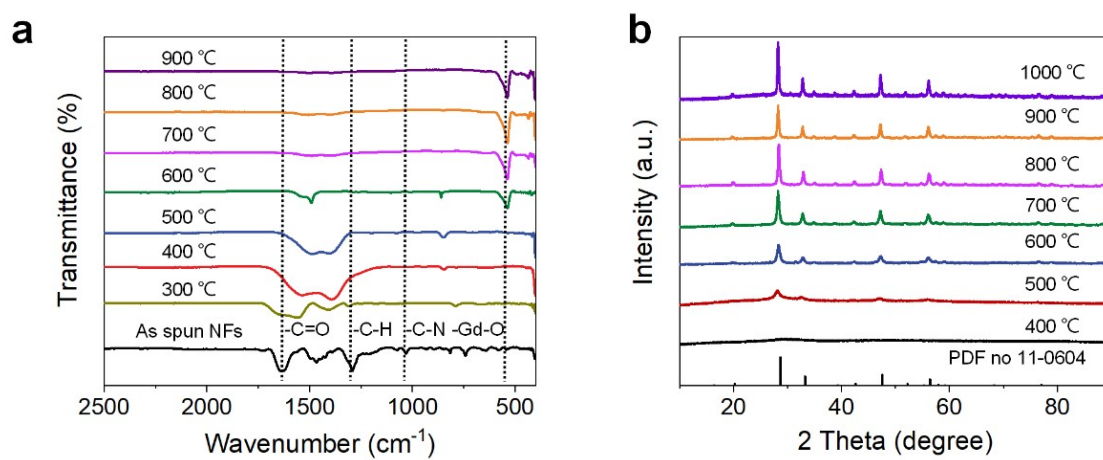
**Figure S2.** XPS spectra of GNFs.



**Figure S3.** SEM image of PVP/Gd(NO<sub>3</sub>)<sub>3</sub> precursor nanofibers.

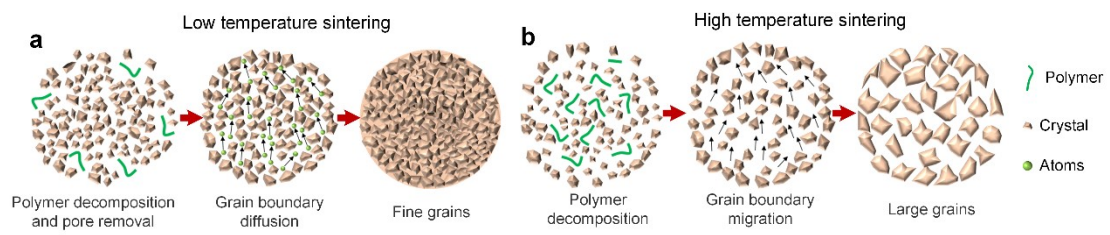


**Figure S4.** TG curve of PVP/Gd(NO<sub>3</sub>)<sub>3</sub> precursor nanofibers.

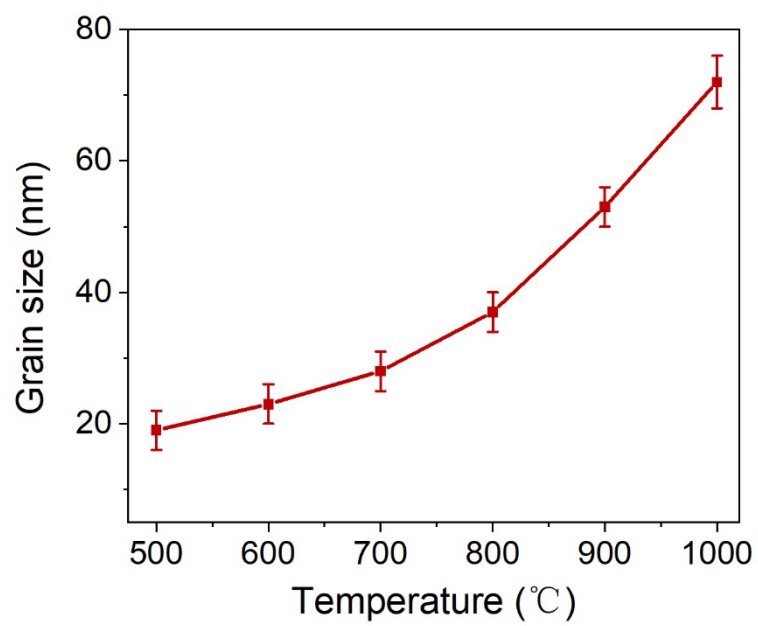


**Figure S5.** a) FT-IR and b) XRD pattern of PVP/Gd(NO<sub>3</sub>)<sub>3</sub> hybrid nanofibers calcined at different temperatures.

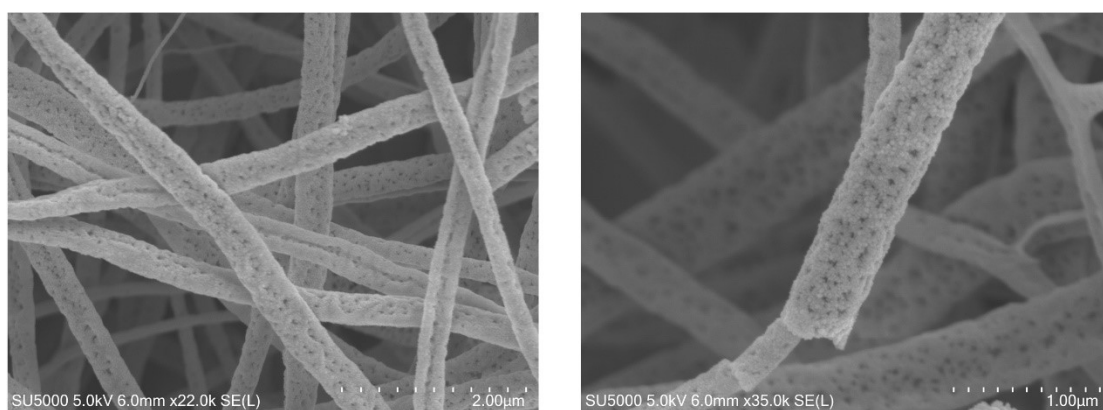




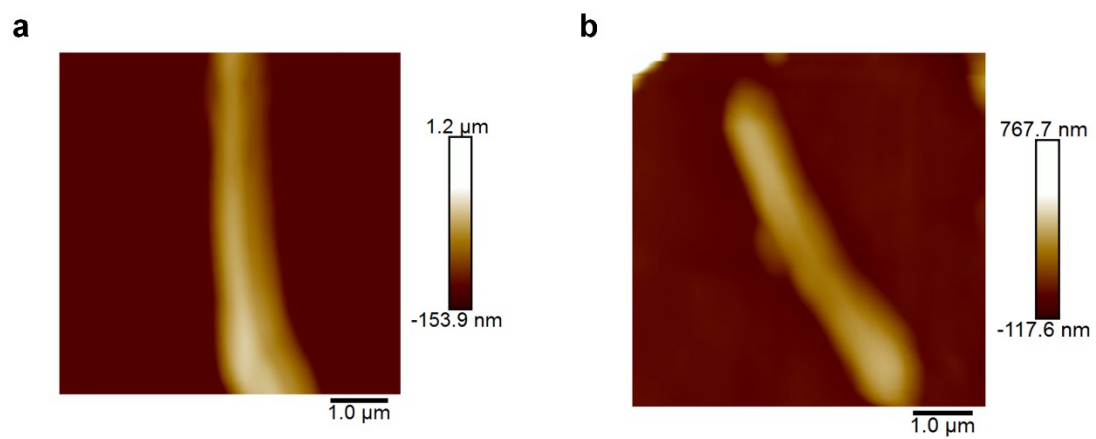
**Figure S6.** Diagram of a) low-temperature sintering and b) high-temperature sintering processes.



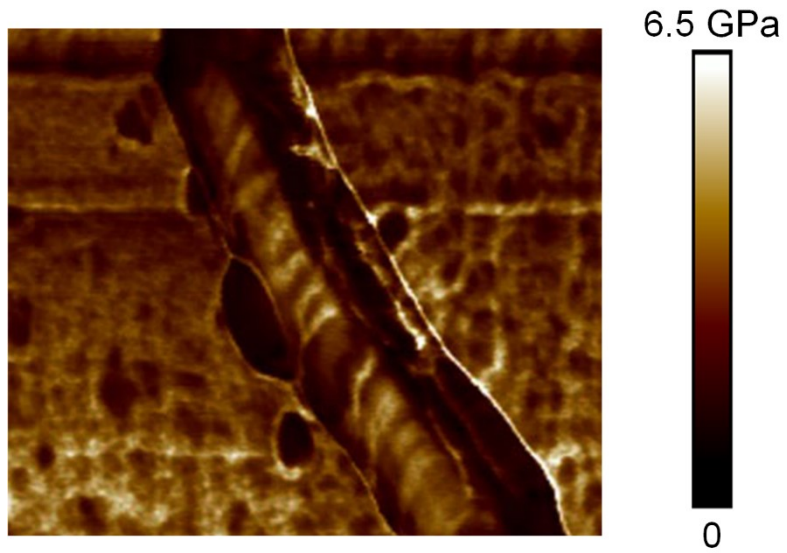
**Figure S7.** Grain size of GNFs calcined at different temperatures.



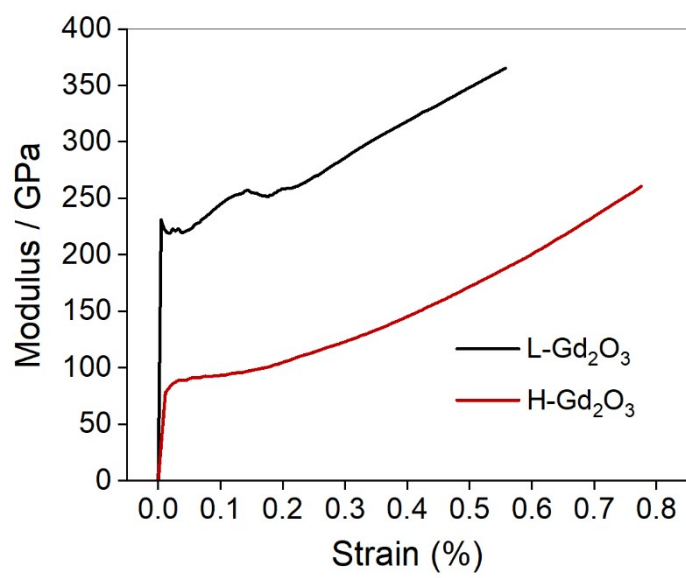
**Figure S8.** SEM images of GNFs sintered at 1000 °C.



**Figure S9.** Three-dimensional AFM images of a) L-Gd<sub>2</sub>O<sub>3</sub> and b) H-Gd<sub>2</sub>O<sub>3</sub>.



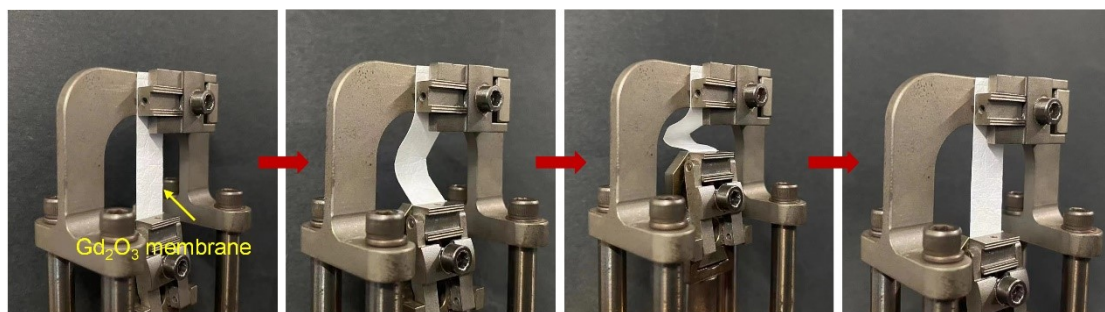
**Figure S10.** Image of elastic modulus mapping of H-Gd<sub>2</sub>O<sub>3</sub>.



**Figure S11.** The simulated tensile stress-strain curve of fibers.

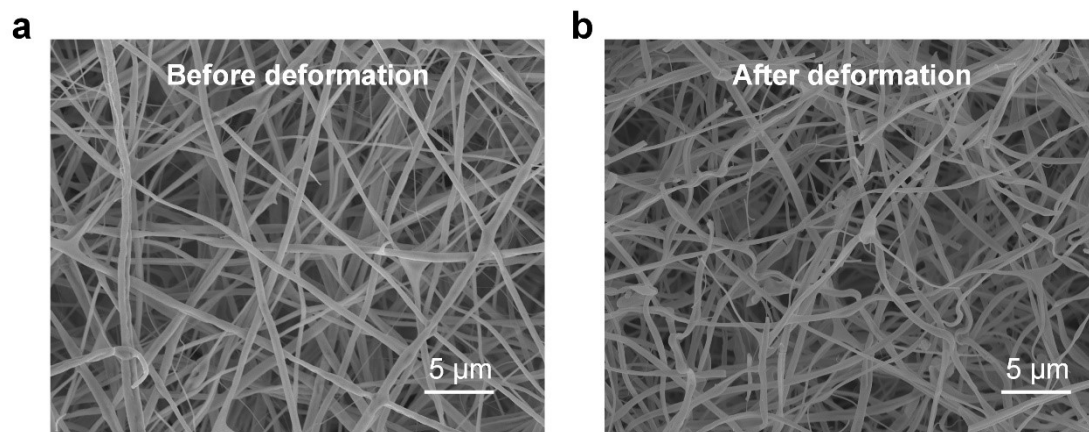


**Figure S12.** Large-scale production equipment.

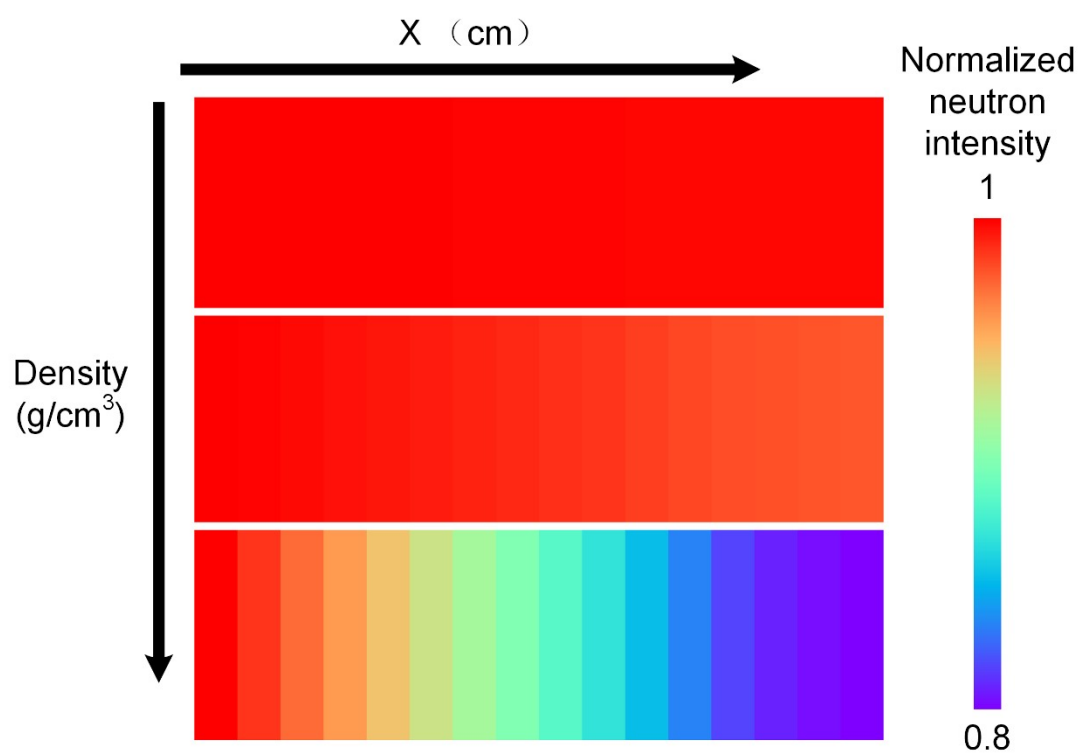


**Figure S13.** Mechanical test diagram.





**Figure S14.** SEM micrographs before and after deformations.



**Figure S15.** Neutron absorption processes in aerogels of different densities.