

## Supporting Information

### Coir fiber reinforced PVA aerogels for oil absorption

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### Experimental section

#### 1. Materials

Dichloromethane, n-hexane, xylene, acetone, isooctane, boric acid, nitric acid, ethanol and Hexadecyltrimethoxysilane (HDTMS) were purchased from Li'an Longbohua (Tianjin) Pharmaceutical Chemical Co., Ltd. PVA was purchased from Sinopharm Chemical Reagent Co., Ltd. GNS were purchased from Xianfeng Nanomaterials Technology Co., Ltd. Diesel and gasoline were purchased from an adjacent gas station. Colza oil and coir fiber were obtained from a local market. All experiments used de-ionized (DI) water and chemicals were used as received without further purification.

#### 2. Characterization

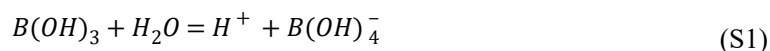
The scanning electron microscope (SEM, QUANTA 650FEG, FEI) was used to characterize the microstructure of the sample surface. X-ray photoelectron spectroscopy (XPS, Thermo Scientific ESCALAB 250Xi) and energy dispersive X-ray spectrometer (EDS, KeveX) were used to explore the elemental composition of the sample. Fourier transform infrared spectrometer (FT-IR, Nexus870) was used for functional group analysis. The contact angles of the samples were measured at room temperature by a JC2000D system with a 5  $\mu$ L droplet. The thermal stability of the aerogel was analyzed by thermogravimetric analysis (TG, NETZSH STA 449C). The analysis was carried out from room temperature to 500 °C at a rate of 10 °C/min in nitrogen atmosphere. All optical photos were taken with the camera.

#### 3. Pretreatment of Coir Fiber

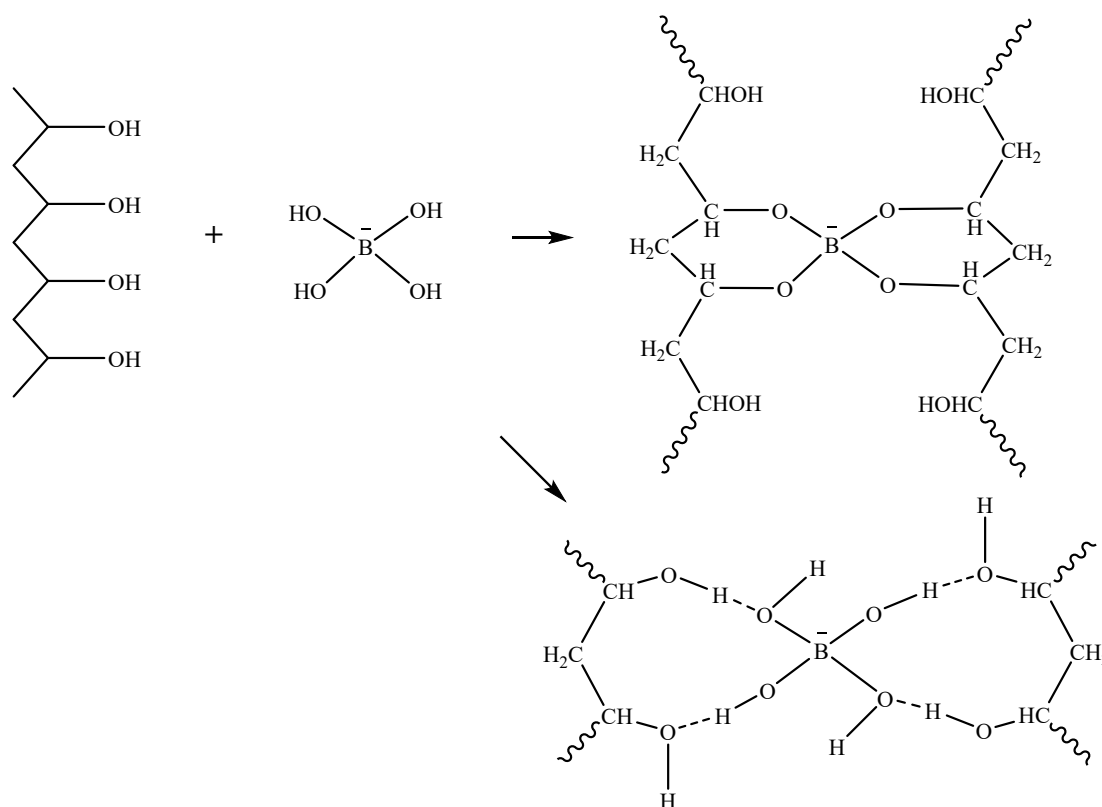
The method of extracting cellulose fiber in this work is to use nitric acid to nitrate and oxidize the part of lignin and hemicellulose. Then, nitrated lignin, oxidized lignin and hemicellulose are dissolved out by ethanol, and the resulting residue is cellulose fiber. The experimental process is as follows: First, pulverize the coir fiber and pass it through a 100-mesh molecular sieve, then mix the short fiber with a nitric acid-ethanol solution (1:2) in a ratio of 1:20 (g/ml). And reflux the mixed solution in a water bath at 100 °C for 2 h. When the fibers turn white, dry the filter residue to obtain cellulose fibers.

#### 4. Preparation of H-CF/GNS/PVA

PVA is an organic compound that is soluble in water under certain conditions. Its side chain contains many hydroxyl groups, which can form intramolecular hydrogen bonds. Here, boric acid is used as a crosslinking agent for chemical crosslinking. The equation for the reaction between boric acid and water is shown in Eq. (S1):



The boric acid solution forms a diol complex in the presence of an organic polyol, and its reaction process with PVA is as follows:

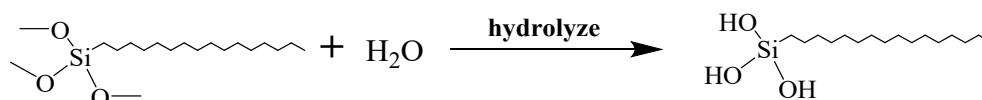


The boronic ester bond formed by the boronic acid condensation reaction is the most widely used dynamic covalent bond, which roughly forms the above two polymers. Then the freeze-thaw method is used for the further cross-linking, which can promote the rearrangement of the molecules. By the way, the folding of the molecular chains makes the structure of the hydrogel semi-crystalline or crystalline.<sup>1</sup>

Heat a certain amount of distilled water to 90 °C, add PVA and adjust the concentration of the PVA solution to 2 wt%. Then add the dissolved boric acid solution, control the ratio of boric acid to PVA at 1:10, stir at the constant temperature to make it fully react, and cool it to the room temperature. Take 3 ml of PVA solution into a glass sample bottle, add 50 mg of pretreated coir fiber and 20 mg of fully ground GNS, stir well, disperse evenly, and freeze it at -20 °C for 12 h. Then thaw it at the room temperature to form CF/GNS/PVA hydrogels, and freeze-dry the hydrogels to obtain CF/GNS/PVA aerogels.

Weigh 2 ml of HDTMS and deionized water (water is evaporated to facilitate the hydrolysis of HDTMS) into two 5 ml glass sample bottles, respectively. Put glass sample bottles and the CF/GNS/PVA aerogel into an airtight container. And the hydrophobic aerogel is prepared by the vapor deposition at 80 °C for 12 h, which is denoted as H-CF/GNS/PVA aerogel. The reaction of

HDTMS hydrolysis to generate silanol is as follows:



The hydroxyl groups on the silanols dehydrate and condense with the hydroxyl groups on the surface of the aerogel to form the low-surface siloxane. Hydrophobically modified aerogels can be used to adsorb oil and organic solvents in water.<sup>2</sup>

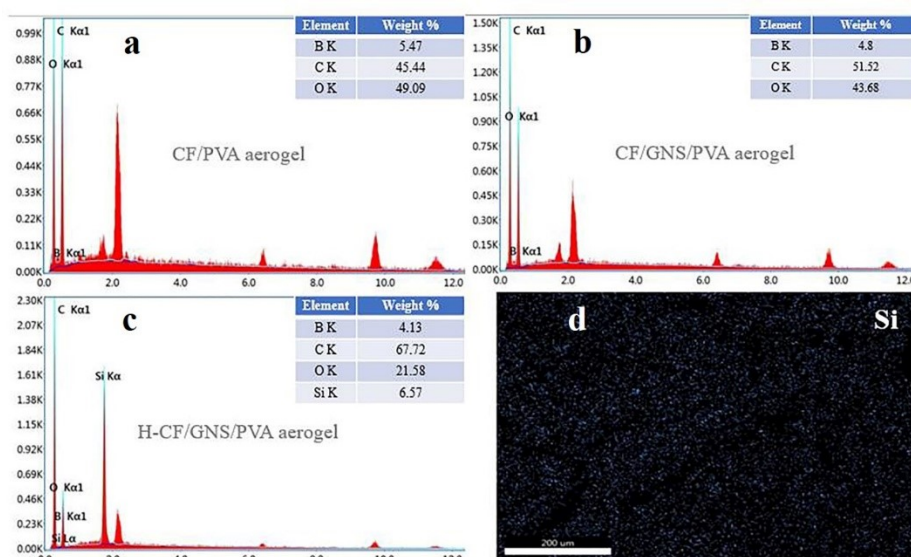
### 5. Oil adsorption capacity of H-CF/GNS/PVA

The adsorption performance of H-CF/GNS/PVA aerogels is evaluated by comparing the quality changes of aerogels before and after the oil adsorption. Immerse the H-CF/GNS/PVA aerogel in common oils and organic solvents (dichloromethane, n-hexane, xylene, acetone, isooctane, diesel, colza oil, gasoline), then take out the aerogel saturated with adsorption, and make sure that the oil and organic solvent do not drip when weighing the mass of the aerogel after adsorption. The adsorption capacity (Q) of aerogels is calculated according to the Eq. (S2):

$$Q = \frac{W - W_0}{W_0} \times 100\% \quad (\text{S2})$$

$W$  (g) is the mass of H-CF/GNS/PVA aerogel after adsorbing oil and organic solvents,  $W_0$  (g) is the original mass of the aerogel.

1. V. de Zea Bermudez, P. P. de Almeida and J. F. Seita, *J. Chem. Educ.*, 1998, **75**, 1410.
2. Z. Zhang, H. Liu and W. Qiao, *Colloid Surface A*, 2020, **589**, 124433.



**Fig. S1** (a) EDS spectrum of CF/PVA aerogel. (b) EDS spectrum of CF/GNS/PVA aerogel. (c) EDS

<b>Sorbent materials</b>	<b>Absorbed substances</b>	<b>Sorption capacity (g g<sup>-1</sup>)</b>	<b>Cost</b>	<b>Ref.</b>
Carbon nanotube sponges	oils and organic solvents	80–180	High	3
Graphene sponge	oils and organic solvents	20–86	Medium	4
Carbonaceous nanofiber aerogel	oils and organic solvents	40–115	High	7
Nitrogen doped graphene foam	oils and organic solvents	200–600	High	5
Graphene/a-FeOOH composite	cyclohexane, toluene, vegetable oil	10–30	High	6
Graphene/CNT aerogel	oils and organic solvents	275–743	High	8
CNF aerogels	oils and organic solvents	106–312	Low	9
Silane-treated PVA/CNF aerogel	oils and organic solvents	44–96	Medium	10
Sodium chlorite-KF carbon aerogel	oils and organic solvents	147–292	Medium	11
RGO/carbon nanofiber aerogel	oils and organic solvents	393–1002	High	12
Cotton-cellulose aerogel	oils and organic solvents	40–90	Medium	13
H-CF/GNS/PVA aerogel	oils and organic solvents	24-53	Low	This work

spectrum of H-CF/GNS/PVA aerogel. (d) Distribution of Si in H-CF/GNS/PVA aerogel.

**Table S1** Comparison of various adsorbents