# Ultralight, elastic, magnetic and superhydrophobic cellulose nanofibril based aerogel with layer-support structure designed for both excellent oil-water separation and efficient PM2.5 removal


**Yu Miao[a], Haichao Xu[a], Enfu Wang[a], Yipeng Liang[a], Zepeng Wang[a], Kuichuan Sheng[b], Chunping Dai[c], Wenbiao Zhang[a,*], Jingda Huang[a,b,*]**

a College of Chemistry and Materials Engineering, Zhejiang A&F University, Hangzhou 311300, China

b College of Biosystems Engineering and Food Science, Zhejiang University, Hangzhou 310030, China.

c Faculty of Forestry, University of British Columbia, Vancouver, Canada.



## Abstract

It is unquestionably a great challenge to develop elastic cellulose nanofibril (CNF) based aerogels satisfying both excellent oil-water separation and efficient PM2.5 removal. In this work, the CNF based aerogel with a layer-support structure was design through CNF as the aerogel scaffold and compositing with bamboo activated charcoal (BAC) and Fe3O4 as the functional particles, followed by hydrophobic modification. The resultant aerogel displays ultra-low density (30.12 mg/cm3), good specific surface area (25.38 m2/g), magnetism (9.44 emu/g) and superhydrophobicity (water contact angle (WCA) of 153.8° and slide angle (SA) of 7°). Benefiting from high internal porosity of the layer-support structure, the system shows excellent absorption efficiency (16.79-51.93 g/g) for various oils and filtration efficiency (97.85 ± 1.2%) for PM2.5 with a low pressure drop (58 Pa). As well as it also shows outstanding mechanical properties and able to maintain a high recovery of 75.4% at 50% strain (corresponding 25.29 kPa) due to the flexibility of the "support" structure given by methyltrimethoxysilane (MTMS). Based on its simple preparation and excellent properties, the magnetic and superhydrophobic CNF/Fe3O4@BAC composite aerogel shows great potential in multifaceted separation technique and recycling of natural resources.


## Forming and working mechanism of modified composite aerogel

As shown in Fig. 1E, the resulting CNF based aerogel displays excellent oil absorption due to its good pore structures from layer-support structures and BAC itself.

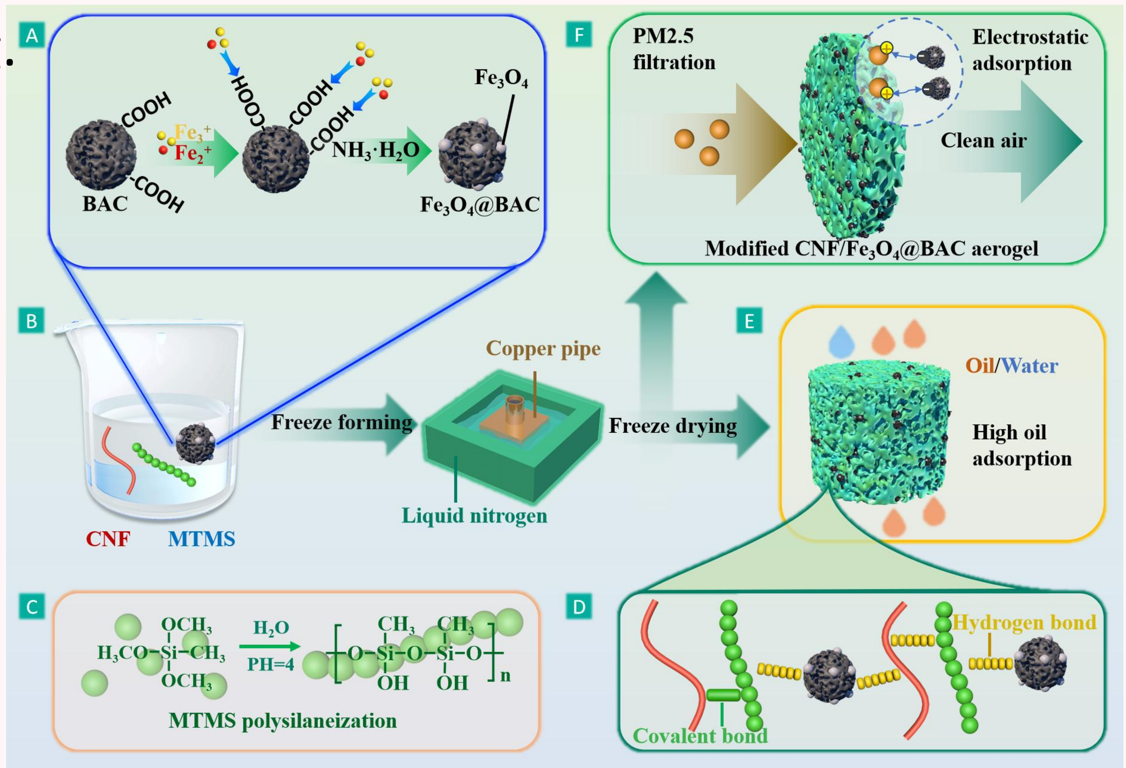


*Fig. 1. Formation process and working mechanism of modified CNF/$Fe_3O_4$@BAC composite aerogel.*

Moreover, the layer-support structure gives the system good mechanical properties because of its enough space for compression and rebound and the support wrapped in MTMS providing excellent flexibility, meeting the reuse performance. The introduced $Fe_3O_4$ supplies the system with magnetic properties. In the air filtration process (Fig. 1F), PM2.5 is blocked by both the networks of the system and the electrostatic adsorption of the negatively charged $Fe_3O_4$@BAC. In addition, the layer-support structure could reduce pressure drop. Based on the above preparation mechanism and construction strategy, the resulting CNF/$Fe_3O_4$@BAC aerogel achieves a balance between excellent oil absorption capacity and efficient air filtration.

## Results - Oil absorption capacity

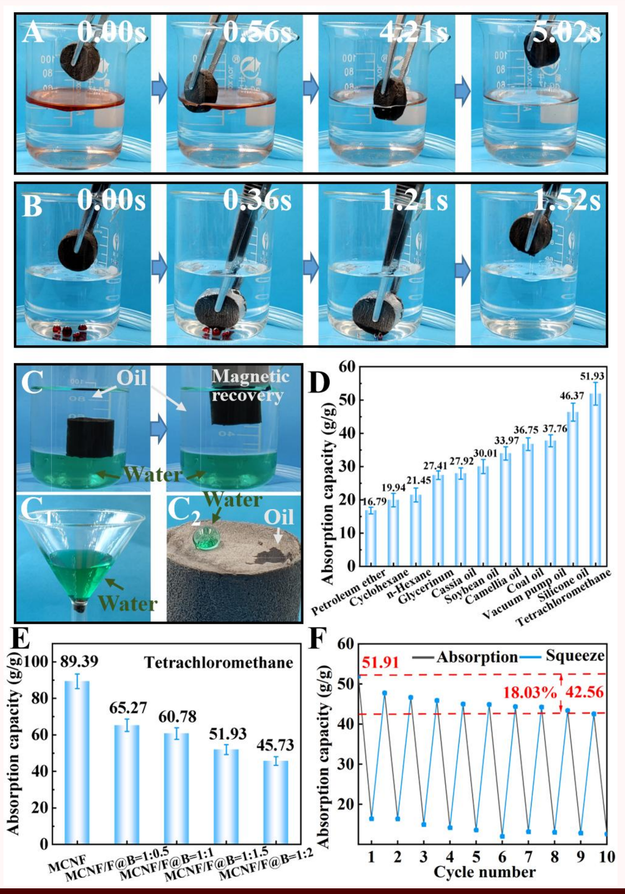

*Fig. 2. Numerical images displaying absorption capacity for (A) n-hexane and (B) dichloromethane (both stained by sudan red) from water. Status of modified CNF/$Fe_3O_4$@BAC aerogel in oil-water mixture (C) before and after magnetic attraction. (D) Absorption capacity of modified CNF/$Fe_3O_4$@BAC aerogel for different oils. (E) Oil absorption capacity for by different CNF based aerogels. (F) Cyclic absorption capacity for tetrachloromethane of aerogel. before and (f, g) after filtration test.*

- The modified CNF/$Fe_3O_4$@BAC aerogel complete absorption of light oil (5.02 s) and heavy oil (1.52 s) in a very short time (Fig. 2A, B).
- Depending on their density and viscosity, the absorption for different oils (organic solvents) is 16.79-51.93 g/g (Fig. 2D).
- It shows an excellent oil recovery and reuse capacity, only decreasing by 18.03% after 10 cycles (Fig. 2F).

## Results - Filtration capacity

- The filtration efficiency of the aerogel (1: 1.5) reaches the maximum of 97.85%, pressure drop is 58 Pa (Fig. 3A-C).
- The aerogel had excellent stability in the high temperature filtration process (Fig. 3E).
- The modified CNF/$Fe_3O_4$@BAC aerogel still maintained high filtration performance (>95%) after washing for 5 times. The pressure drop only increases to 97 Pa at the end of 5 cycles and Qf keeps at 0.031 $Pa^{-1}$, indicating good reusability (Fig. 3G).

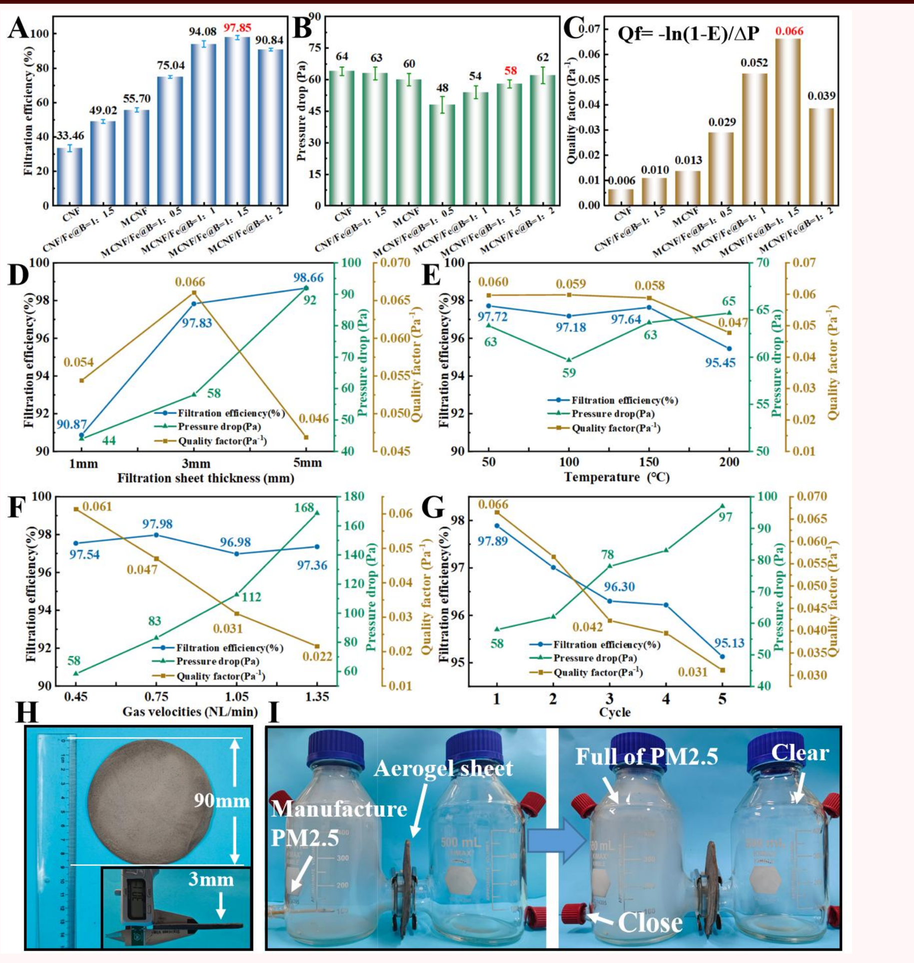


*Fig. 3. (A) Filtration efficiency, (B) pressure drop and (C) Qf for PM2.5 of CNF based aerogel at 0.45 NL/min. Filtration efficiency for PM2.5 of modified CNF/Fe3O4@BAC aerogel sheet under different (D) thickness, (E) temperature treatment for 2 h, (F) gas velocities, (G) cycle. (H) Illustration and (I) simulating PM2.5 filtration process of modified CNF/Fe3O4@BAC aerogel sheet.*

## Conclusions

In summary, the modified CNF/$Fe_3O_4$@BAC composite aerogel with the layer-support structure is developed for both oil absorption and air filtration through a simple freeze-drying and heat drying. Benefiting from the lay-support structure and the enhancement of $Fe_3O_4$@BAC, the resulting aerogel exhibits an ultralow density (30.12 mg/cm³), good magnetic (9.44 emu/g), heat resistance, excellent recoverability, superior filtration efficiency for PM2.5 (97.85±1.2%, 58 Pa) and remarkable oil absorption ability (16.79-51.93 g/g). Furthermore, it maintains high filtration efficiency for PM2.5 at different air flow rates, different temperatures and after 5 washing-filtration cycles. Given the advanced design concept and excellent performance, this study provides a simple, effective and environmentally friendly strategy for constructing multifunctional CNF based aerogels with excellent mechanical strength, superior PM2.5 filtration and remarkable oil absorption capacity.


## Acknowledgements

Natural Science Foundation of Zhejiang Province (No. LY21C160002), Project funded by China Postdoctoral Science Foundation (No. 2021M692806), Scientific Research Development Foundation of Zhejiang A&F University (No. 2020KX0076).


1958 浙江农林大学 ZHEJIANG A&F UNIVERSITY