

Fabrication and Characterization of Hydrophobic Aerogels Containing Bacterial Cellulose Coated with Copper Species via Mild Reduction

Ha V. Le^{1,2,*}, Hanh H. M. Nguyen^{1,2}, Trang T. P. Nguyen^{1,2}, Truc T. T. Nguyen^{1,2}, Khoi D. Dang^{1,2}, Kien A. Le³, Khoa D. Nguyen^{1,2}, Hoan T. Phan^{1,2}



Use your smartphone to scan this QR code and download this article

ABSTRACT

In this study, bacterial cellulose (BC) was coated with copper species via a room-temperature hydrazine-mediated reduction reaction of copper(II) acetate in a suspension phase of *nata de coco*, which was treated by freeze-drying, yielding hydrophobic lightweight aerogels. Structural and textural characteristics of the prepared aerogels were discovered using several techniques including X-ray diffraction (XRD), thermogravimetry analysis (TGA), water contact angle measurement and isothermal nitrogen physisorption. In detail, XRD results indicated the formation of the metallic copper phase in the aerogel while the high cellulose crystallinity was remained unchanged. No oxidized copper phases were detected in the material. Via the TGA profiles, the Cu loading was determined to be in the range from 3.9 to 13.4 wt.%, depending on the copper(II) acetate amount used for the reduction reaction. These values were generally lower than the theoretical Cu contents probably due to the unexpected Cu losses during the preparation procedure. In addition, increasing the Cu content in the BC aerogel led to a significant decrease in the specific surface area with the presence of Cu in the porous structure. On the other hand, as expected, the hydrophobicity of the BC aerogel was significantly enhanced with the Cu content. Indeed, the Cu-coated BC aerogels with high copper contents (> 6.2 wt.%) were hydrophobic, showing a large water-contact angle of up to 138°. Therefore, the resulting hydrophobic aerogels well interacted with water-immiscible organic solvents including diesel oil and cyclohexane with adsorption capacities varied from 20 to 30 g/g. The successful fabrication of the hydrophobic aerogels upon the simple surface modification of abundant bacterial cellulose with Cu species can introduce novel and efficient biomass-based material for the treatments of oil-based liquids in the aqueous environment.

Key words: hydrophobic aerogel, bacterial cellulose, coating, copper, mild reduction

¹Faculty of Chemical Engineering, Ho Chi Minh City University of Technology (HCMUT), 268 Ly Thuong Kiet Street, District 10, Ho Chi Minh City, Vietnam

²Vietnam National University Ho Chi Minh City, Linh Trung Ward, Thu Duc City, Ho Chi Minh City, Vietnam

³Institute for Tropicalization and Environment, 57A Truong Quoc Dung Street, Phu Nhuan District, Ho Chi Minh City, Vietnam

Correspondence

Ha V. Le, Faculty of Chemical Engineering, Ho Chi Minh City University of Technology (HCMUT), 268 Ly Thuong Kiet Street, District 10, Ho Chi Minh City, Vietnam

Vietnam National University Ho Chi Minh City, Linh Trung Ward, Thu Duc City, Ho Chi Minh City, Vietnam

Email: lvha@hcmut.edu.vn

History

- Received: 20-2-2024
- Accepted: 23-5-2024
- Published Online:

DOI :



1 INTRODUCTION

2 In the era of industrialization, inevitable water pollution
3 caused by hydrophobic organic solvents and oil
4 spills threatens both ecosystems and human health^{1,2}.
5 These incidents inflict substantial burdens due to lost
6 resources, cleanup efforts, and potential disruptions
7 to industries reliant on clean water³. As a result,
8 the consequences prompted the necessity for effective
9 and innovative remediation techniques to minimize
10 their environmental impact and ensure public health.
11 At present, the common strategies employed to cope
12 with including adsorption, chemical treatment, incineration
13 and biotreatment¹. Among them, adsorption
14 is deemed a promising approach to capture the contaminants
15 since the process is simply, cost-effective
16 and does not generate secondary pollution³. It is,
17 therefore, essential to design an effective adsorbent.
18 In particular, the ideal sorbents should exhibit high
19 trapping efficiency, high uptake rate, commercial via-

20 bility, environmental friendliness, and facile recyclability⁴.
21 Aerogels are an outstanding class of porous materials,
22 with an extremely low bulk density, a very high porosity,
23 and a low thermal conductivity^{5,6}. In particular, carbon
24 nanotubes (CNTs), graphene, as well as biomass-derived
25 materials⁷⁻¹⁰ have been the outstanding precursors for
26 aerogels fabrication towards their application in oil spill
27 cleanup and water treatment. However, the limitations for
28 the application of CNTs and graphene are high precursor
29 cost, complex fabrication procedures or the need for
30 specialized equipment⁵. In contrast, biomass-based
31 materials can offer distinct advantages, including sustain-
32 ability, biodegradability and inherent safety¹¹. Recently,
33 aerogels derived from bacterial cellulose have attracted
34 attention of the scientists as a potential material for
35 environmental treatments owing to their low cost, sustain-
36 ability, low density, high porosity and biodegradability^{12,13}.
37 The oil
38
39

Cite this article : Le H V, Nguyen H H M, Nguyen T T P, Nguyen T T T, Dang K D, Le K A, Nguyen K D, Phan H T. **Fabrication and Characterization of Hydrophobic Aerogels Containing Bacterial Cellulose Coated with Copper Species via Mild Reduction.** *Sci. Tech. Dev. J. – Engineering and Technology* 2024; (1):1-9.

Copyright

© VNUHCM Press. This is an open-access article distributed under the terms of the Creative Commons Attribution 4.0 International license.



and oleophilic liquid adsorption performance of sorbents are not only determined by their density and porosity, but is also significantly influenced by surface properties¹⁴. Common strategies for cellulose fibers comprising of chemical vapor deposition, cold plasma treatment and atomic layer deposition are applied with the low-surface-energy alkyl or fluorine functional groups to obtain the hydrophobicity¹⁵⁻¹⁷. However, these methods involve the expensive and toxic organic modifiers. Therefore, the development of a facile and cost-effective approach for the fabrication of hydrophobic cellulose aerogels is of importance. As ideal sorbents for water-immiscible solvents and oil, cellulose-based aerogels need tailoring to tune the aerogel structure toward improved hydrophobicity⁴. Recently, we have demonstrated promising hydrophobic organic solvent adsorption by copper-modified bacterial cellulose aerogels, which was attributed to the copper particles covering polar hydroxyl groups via mild reduction reaction⁵. However, the hydrophobicity of the obtained aerogels was not fully investigated. Herein, the present study focused on a further investigation of the hydrophobic characteristics of copper-coated BC-based aerogels, thereby achieving a complete hydrophobicity and enhancing the selectivity to the oil phase for practical application.

MATERIALS AND METHOD

Materials

Nata de coco pieces with an average BC content of 0.8 wt.% were purchased from the Bich Lien Duong supplier (Ben Tre, Vietnam). A Philips HR2531 hand-blender (650 W) was employed to grind the mixture of *nata de coco* pieces (125 g, containing approximately 1 g of BC) and water (125 g) for 2 min, yielding a suspension phase of *nata de coco*.

In a typical procedure for coating BC with Cu which was based on the previous study with minor modifications, the obtained suspension phase of *nata de coco* was added with $\text{Cu}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ (1 mmol) in a 500-mL Erlenmeyer flask under vigorous stirring for 3 h. Subsequently, 50 equivalents of hydrazine hydrate ($\text{N}_2\text{H}_4 \cdot \text{H}_2\text{O}$) was added dropwise under vigorous stirring, followed by the Cu^{2+} reduction reaction for 15 h. After reaction completion, the Cu-modified BC was collected by gravity filtration and washed repeatedly with water until a neutral pH value was obtained. The resulting mixture was remained on the filter paper for 30 min for further water release and then transferred to propylene boxes, which were frozen at -20°C for 24 hours. The bacterial cellulose aerogels were obtained via freeze-drying,

yielding cylinder-shaped aerogels, which were denoted as “Cell–Cu=1:1” expressing 1 g of BC:1 mmol of $\text{Cu}(\text{CH}_3\text{COO})_2$. Further samples including Cell–Cu=1:2, Cell–Cu=1:3, and Cell–Cu=1:4 were fabricated under similar conditions using 1 g of BC and the varied $\text{Cu}(\text{CH}_3\text{COO})_2$ amount, namely, 2, 3, and 4 mmol, respectively. An aerogel sample named “Cell” was prepared without the modification of Cu for the comparison purpose⁵.

Characterization of the obtained materials

Crystallinity of the materials was discovered by X-ray diffraction measurements on a diffractometer device using Cu radiation (D8 Advance, Bruker, Germany). Morphological photographs of the aerogel samples were achieved on an electron scanning microscope (S-4000, Hitachi, Japan). Thermal behavior of the aerogels was investigated on a thermal gravimetric analyzer (SDT Q600, TA Instruments, USA). Their textural properties were determined by 77 K-isothermal nitrogen adsorption/desorption using a high-performance sorption analyzer (ASAP 2020, Micromeritics, USA).

Adsorption study

The adsorption capacity of the copper-coated aerogels for cyclohexane and diesel oil was discovered. 0.02 g of the aerogel sample was dipped into a glass vial containing 10 mL of the corresponding solvent. The solvent-trapping sample was taken out from the liquid phase but still remained in the vial. Until there were no more solvent drops back to the liquid phase, the sample was completely removed. The cyclohexane solvent adsorption efficiency of the copper-coated aerogels was calculated according to the formula: $Q = (m_1 - m_2)/m_{\text{aerogel}}$ (g/g), where m_1 and m_2 are the total weight of the glass vial containing the tested solvent before and after the adsorption, respectively.

RESULTS AND DISCUSSION

Coating copper particles on the surface of bacterial cellulose fibers and bundles was employed via the Cu(II) to Cu(0) reduction stage by hydrazine in an aqueous phase, which was considered as an effective reducing agent thanks to numerous advantages of high efficiency, fast reaction speed, and room-temperature operation^{5,18}. Due to the addition of hydrazine, the solution color changed from blue of the Cu^{2+} cation to red-brown of the Cu^0 clusters (Figure 1), proving the successful reduction of Cu^{2+} to Cu^0 by hydrazine under ambient conditions. The

140 presence of the BC fibers in the same reaction envi-
 141 ronment led to the development of the Cu crystal on
 142 the fiber surface, affording successful Cu coating.
 143 Pure cellulose aerogels exhibited non-selective affinity
 144 for both water and oil. This limitation can be solved
 145 by introducing copper species to the aerogel, which
 146 modified the surface properties of the aerogels. Cop-
 147 per particles effectively shielded the hydroxyl groups
 148 (-OH) on BC fibers, decreasing hydrophilicity of the
 149 BC aerogels. As described in our previous study, upon
 150 contact with the copper-coated aerogels, the water
 151 droplet was remained on the surface with a water-
 152 contact angle of 133°, preventing water penetration
 153 into the porous structure. In contrast, cyclohexane
 154 was rapidly trapped into the cellulose matrix⁵. The
 155 present work focuses on further investigating the in-
 156 fluence of the Cu content on the material hydropho-
 157 bicity.
 158 XRD analysis was employed to confirm the success-
 159 ful deposition of copper onto the bacterial cellulose
 160 (BC) surface and evaluate its impact on the crystalline
 161 structure (Figure 2). The XRD pattern exhibits two
 162 characteristic diffraction peaks at $2\theta = 14.6^\circ$, 16.7°
 163 $^\circ$ and 22.7° , corresponding to the (1 $\bar{1}$ 0), (110) and
 164 (020) lattice planes of crystalline cellulose, respec-
 165 tively¹⁹. The result indicated that the copper coating
 166 process had negligible impact on the inherent crys-
 167 tallinity of the BC. Notably, no further required toxic
 168 chemicals-involving treatments as compared to plant
 169 cellulose^{20,21}. In addition, the XRD pattern indicated
 170 the successful incorporation of copper particles on
 171 cellulose fibers, as confirmed by the presence of char-
 172 acteristic peaks of $2\theta = 43.5^\circ$, 50.5° and 74.2° , cor-
 173 responding to the lattice planes of (111), (200), (220)
 174 of pure metallic copper phase (JCPDS No. 003-1018),
 175 thus proving the effectiveness of the reduction reac-
 176 tion of Cu²⁺ to Cu⁰ at room temperature^{5,22}. These
 177 results are consistent with previous research by Li and
 178 coworkers on the modification of plant-derived cellu-
 179 lose with copper nanoparticles¹⁸.
 180 Further analysis using TGA revealed the thermal sta-
 181 bility and discovered the copper content of the aero-
 182 gels (Figure 3). The obtained aerogels exhibited the
 183 thermal stability were up to 220 °C with a minor
 184 mass loss of approximately 6% due to the elimina-
 185 tion of adsorbed water from the aerogel matrix, which
 186 was consistent with the previous study of Mohite and
 187 co-workers²³. Above 220 °C, the rapid decomposi-
 188 tion occurred, generating carbon oxides, water and
 189 other gaseous compounds²⁴. As reported in our pre-
 190 vious study, upon the completion of the combustion,
 191 a negligible residual mass of approximately 0.7% for

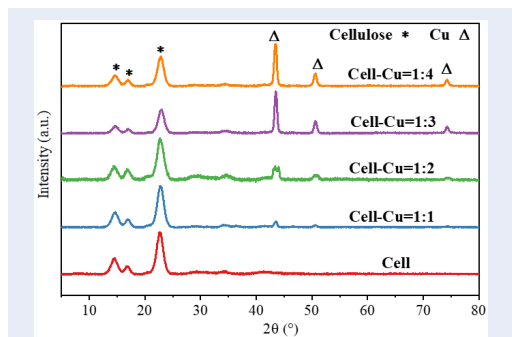


Figure 2: XRD patterns of pristine and Cu-coated bacterial cellulose aerogels.

the pristine BC aerogel was observed, which was at-
 192 tributed to unwashed inorganic additives used in the
 193 preparation of *nata de coco*⁵. For the Cu-containing
 194 aerogel samples, assuming that the residue was cop-
 195 per(II) oxide after the combustion in air, the Cu con-
 196 tent was determined to be 3.9, 6.2, 10.7, and 13.4 for
 197 Cell:Cu=1:1, Cell:Cu=1:2, Cell:Cu=1:3, Cell:Cu=1:4,
 198 respectively (Table 1).
 199

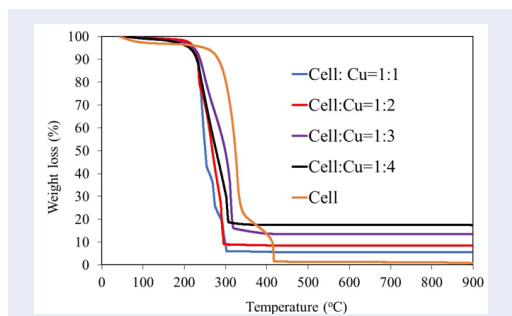


Figure 3: TGA profiles of the Cu-free and Cu-modified BC aerogels⁵.

As can be seen in Table 1, the theoretical copper con-
 200 tent was higher than the TGA-based result, suggest-
 201 ing copper losses probably during the sample prepara-
 202 tion. Under the applied preparation conditions, the
 203 loss reason might be explained based on the two facts
 204 that excess, non-adherent copper species on cellulose
 205 fibers could be leached into water and nano-sized cop-
 206 per particles could pass through the filter paper.
 207 Nitrogen sorption analysis revealed the textural char-
 208 acteristics of the fabricated aerogels (Figure 4). The
 209 isotherms exhibited similar adsorption tendencies at
 210 P/P_o values between 0 and 0.8, indicating negligible
 211 presence of micro- and mesopores. A sharp rise in
 212 adsorption capacity above P/P_o of 0.8 confirmed that
 213 macropore was dominant in the aerogel structures.
 214



Figure 1: The BC suspension phase in water (a); after the addition of copper(II) acetate (b); after the addition of hydrazine for 1 min (c); after the addition of hydrazine for 14 h (d)

Table 1: Copper content in Cu-modified bacterial cellulose aerogels.

Sample	Cu content (wt.%)	
	Theoretical calculation	TGA calculation
Cu:Cell=1:1	6.02	3.90
Cell:Cu=1:2	11.40	6.16
Cell:Cu=1:3	16.10	10.20
Cell:Cu=1:4	20.40	13.39

It should be noted that increasing the copper content deposited on the BC fibers resulted in a significant reduction in the surface area compared with the pristine BC aerogel whose surface area was previously reported to be $45 \text{ m}^2/\text{g}$ ⁵. In detail, for the samples Cell:Cu=1:1 and Cell:Cu=1:2, the surface area was remained stable at approximately $13 \text{ m}^2/\text{g}$. However, further increasing the copper ratio to 10.2 wt.% led to a drastic drop in surface area to approximately $4 \text{ m}^2/\text{g}$ (sample Cell:Cu=1:3), similar to the value observed at the sample Cell:Cu=1:4 (Table 2). This observed trend suggested that copper particles preferentially occupied the pores available in the pristine aerogel, thus reducing its surface area and pore volume¹⁸.

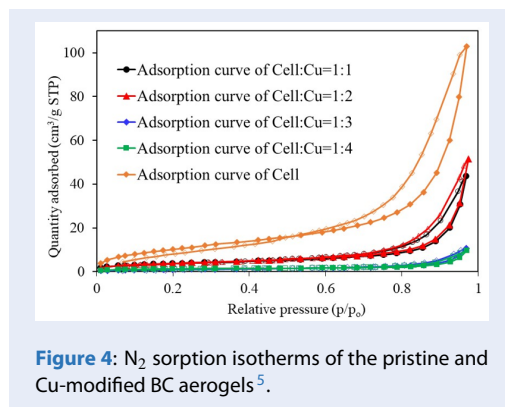


Figure 4: N_2 sorption isotherms of the pristine and Cu-modified BC aerogels⁵.

Water contact angle analysis revealed the surface characteristics of copper-loaded aerogels (Figure 5). The Cell:Cu=1:1 aerogel completely interacted with water, indicating insufficient copper particles to encapsulate the highly polar hydroxyl groups of bacterial cellulose. In contrast, the samples Cell:Cu=1:2, Cell:Cu=1:3, and Cell:Cu=1:4 exhibited increasing contact angles, particularly 115.6° , 131.4° , and 137.6° , respectively, implying the inconsiderable interaction between water and copper-coated cellulose fibers. Water droplets could be remained on the aerogel surface. As a result, the aerogels Cell:Cu=1:3 and Cell:Cu=1:4 could float on water. The observed behavior demonstrated the good hydrophobicity of Cell:Cu=1:3 and Cell:Cu=1:4 aerogels. This study suggested a critical copper loading threshold for accomplishing hydrophobicity, highlighting the potential for tuning the surface properties via controlling copper content incorporation.

The oleophilic-liquid adsorption capacity of the BC aerogels was discovered via using cyclohexane solvent as a typical hydrophobic phase (Figure 6). The obtained aerogels exhibited good adsorption due to the low surface energy of cyclohexane, enabling facile

penetration into the web-like skeleton. Notably, the copper-free BC-aerogels demonstrated the highest adsorption capacity, reaching 37.7 g/g . However, introducing copper particles led to a decrease in adsorption capacity. In particular, at a 6.02% copper loading (Cell:Cu=1:1), the adsorption capacity decreased by 21%. A further increase in copper content (Cell:Cu=1:3) resulted in a 17.2% lower adsorption efficiency compared to Cell:Cu=1:1, but only 4% lower than Cell:Cu=1:2. The lowest adsorption capacity was observed for the Cell:Cu=1:4, which was a significant reduction compared to the copper-free and lower copper-loading samples. In fact, pristine cellulose aerogels possess the ability to adsorb different liquids but with an unclear selectivity in terms of polarity, which could inhibit the removal efficiency of immiscible liquid from aqueous medium^{18,25}.

Extended investigation into the adsorption capacity of BC-based aerogels for diesel oil revealed a similar trend observed for cyclohexane, namely a decrease in adsorption efficiency with increasing copper content. This might be owing to the reduction in pore size upon copper incorporation, consequently constraining the available volume within the porous structure. As expected, the Cell:Cu=1:1 sample exhibited the highest adsorption capacity, followed by relatively similar values for Cell:Cu=1:2 and Cell:Cu=1:3, with an obvious drop observed for Cell:Cu=1:4. This suggested that factors beyond pore size, namely hydrophobicity and hydrophilicity balance also determined adsorption behavior. It should be noted that the Cell:Cu=1:1 sample remained hydrophilic, promoting oil adsorption, while the Cell:Cu=1:4 sample turned hydrophobic, potentially promoting oil interaction. However, its significantly lower surface area inhibited its overall adsorption capacity. These mutual influences might be responsible for the decrease in adsorption tendencies observed, aligning with the proposed explanation regarding pore size limitations.

Nanocellulose aerogel modified with hexacycltrimethoxysilane exhibited an excellent adsorption performance for cyclohexane, reaching approximately 100 g/g while multifunctional polyimide aerogels revealed the similar cyclohexane adsorption capacity of 33 g/g ^{26,27}. In contrast, the sodium alginate/graphene oxide/silicon oxide aerogel modified with methyltrimethoxysilane possessed lower uptakes for the removal of organic solvent and oils, namely 22 and 23 g/g for cyclohexane and diesel oil, respectively²⁸. It should be noted that manufacturing these materials required costly precursors and complicated processes which could inhibit the application cope in capturing oil and

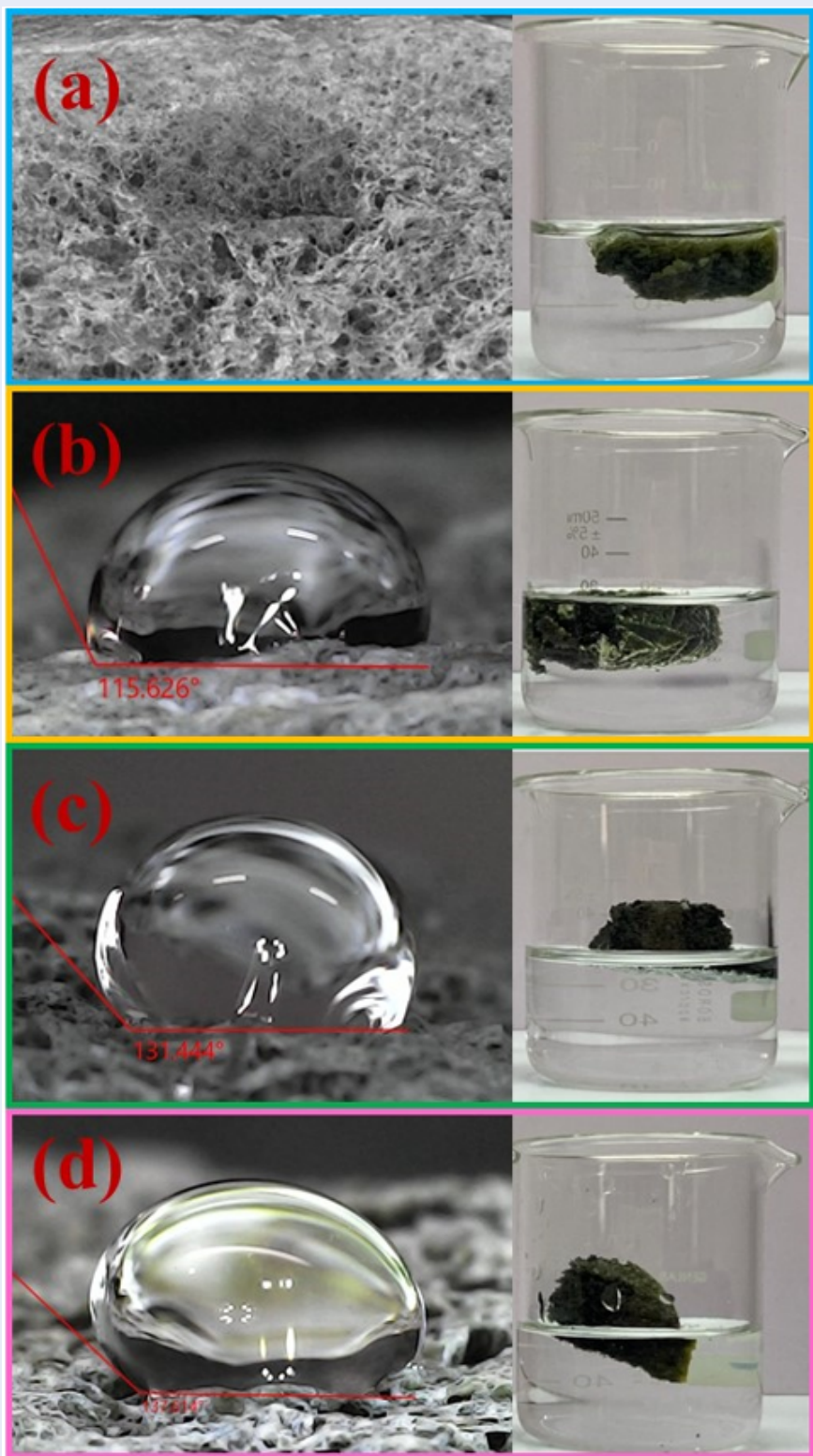


Figure 5: Photographs of water contact angle and water interaction for the Cu-containing BC aerogel samples Cell:Cu=1:1 (a); Cell:Cu=1:2 (b); Cell:Cu=1:3 (c); Cell:Cu=1:4 (d).

Table 2: BET specific surface area of pure and Cu-modified BC aerogels⁵.

Sample	Specific surface area (m ² /g)
Cell:Cu=1:1	13.4
Cell:Cu=1:2	13.9
Cell:Cu=1:3	4.05
Cell:Cu=1:4	4.59
Cell	45.0

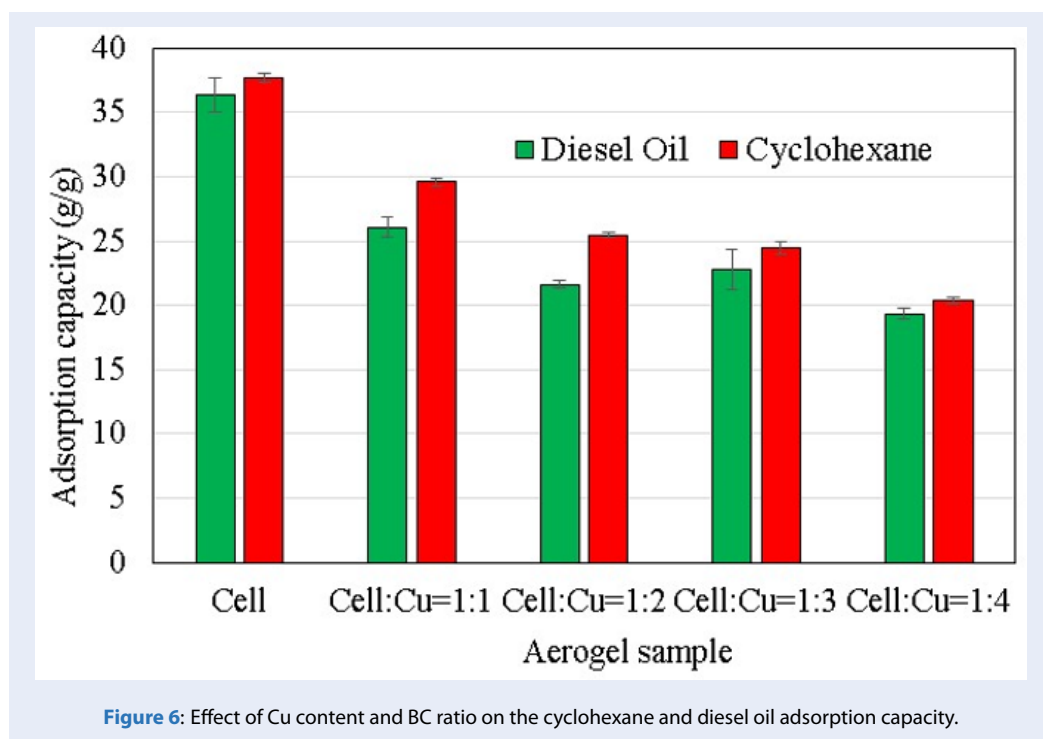


Figure 6: Effect of Cu content and BC ratio on the cyclohexane and diesel oil adsorption capacity.

306 organic solvents. Therefore, the present study could
 307 offer a cost- and step-efficient procedure to yield hy-
 308 drophobic aerogels for the environmental treatments.
 309

310 CONCLUSIONS

311 In conclusion, this study successfully accomplished
 312 surface-modified BC aerogels via a feasible liquid-
 313 phase reaction and subsequent freeze-drying. The hy-
 314 drophobic property of the aerogel can be obtained due
 315 to the Cu coating. Notably, increasing the Cu content
 316 can significantly improve the hydrophobicity of the
 317 aerogel while its surface area was declined. On the
 318 other hand, the adsorption capacity for cyclohexane
 319 and diesel oil in the range of 20-30 g/g demonstrated
 320 the high potential of this composite aerogel toward
 321 cleaning-up oil spills in the aqueous environment. Fo-
 322 cusing on this objective, the selective adsorption of

the oil phase in the presence of water or in an emul- 323
 sion phase would be intensively investigated. 324

ACKNOWLEDGMENT 325

We acknowledge Ho Chi Minh City University of 326
 Technology (HCMUT), VNU-HCM for supporting 327
 this study. 328

ABBREVIATION 329

- BC: Bacterial cellulose 330
- SEM: Scanning electron microscope 331
- TGA: Thermogravimetry analysis 332
- XRD: X-ray diffraction 333

CONFLICTS OF INTERESTS 334

The authors declare that they have no competing in- 335
 terests. 336

337 **AUTHORS' CONTRIBUTIONS**

338 **Ha V. Le:** Conceptualization, Methodology, Data cura-
 339 tion, Writing – review & editing. **Hanh H. M.**
 340 **Nguyen:** Investigation, Methodology, Writing – orig-
 341 inal draft. **Trang T. P. Nguyen:** Investigation, For-
 342 mal analysis, Methodology. **Truc T. T. Nguyen:** In-
 343 vestigation, Formal analysis, Writing – original draft.
 344 **Khoi D. Dang:** Methodology, Data curation. **Kien**
 345 **A. Le:** Methodology. **Khoa D. Nguyen:** Writing – re-
 346 view & editing. **Hoan T. Phan:** Funding acquisition,
 347 Project administration, Writing – original draft.

348 **REFERENCES**

349 1. Thai QB, Nguyen ST, Ho DK, Tran TD, Huynh DM, Do NHN, et
 350 al. Cellulose-based aerogels from sugarcane bagasse for oil
 351 spill-cleaning and heat insulation applications. *Carbohydrate*
 352 *Polymers*. 2020;228:115365; Available from: <https://doi.org/10.1016/j.carbpol.2019.115365>.
 353
 354 2. Ganesamoorthy R, Vadivel VK, Kumar R, Kushwaha OS, Ma-
 355 mane H. Aerogels for water treatment: A review. *Journal of*
 356 *Cleaner Production*. 2021;329:129713; Available from: <https://doi.org/10.1016/j.jclepro.2021.129713>.
 357
 358 3. Yu C, Lin W, Jiang J, Jing Z, Hong P, Li Y. Preparation of a porous
 359 superhydrophobic foam from waste plastic and its applica-
 360 tion for oil spill cleanup. *RSC Advances*. 2019;9(65):37759-
 361 67; Available from: <https://doi.org/10.1039/C9RA06848A>.
 362
 363 4. Sai H, Fu R, Xing L, Xiang J, Li Z, Li F, et al. Surface Mod-
 364 ification of Bacterial Cellulose Aerogels' Web-like Skeleton
 365 for Oil/Water Separation. *ACS Applied Materials & Inter-*
 366 *faces*. 2015;7(13):7373-81; Available from: <https://doi.org/10.1021/acsami.5b00846>.
 367
 368 5. Nguyen HHM, Tan KVM, Van TTT, Nguyen HN, Phan ANQ, Tran
 369 ATT, et al. Preparation of Cu-modified bacterial cellulose aere-
 370 gels derived from nata de coco towards the enhanced adsorp-
 371 tion of hydrophobic organic solvents. *Journal of Porous Ma-*
 372 *terials*. 2023;30(4):1195-205; Available from: <https://doi.org/10.1007/s10934-022-01413-z>.
 373
 374 6. Haimer E, Wendland M, Schlufter K, Frankenfeld K, Miethe P,
 375 Potthast A, et al. Loading of Bacterial Cellulose Aerogels with
 376 Bioactive Compounds by Antisolvent Precipitation with Su-
 377 percritical Carbon Dioxide. 2010;294(2):64-74; Available from:
 378 <https://doi.org/10.1002/masy.201000008>.
 379
 380 7. Gui X, Li H, Wang K, Wei J, Jia Y, Li Z, et al. Recyclable car-
 381 bon nanotube sponges for oil absorption. *Acta Materialia*.
 382 2011;59(12):4798-804; Available from: <https://doi.org/10.1016/j.actamat.2011.04.022>.
 383
 384 8. Shi L-A, Ge J, Hu B-C, Ma T, Zhao H, Song Y-H, et al. Joule-
 385 heated carbonized melamine sponge for high-speed absorp-
 386 tion of viscous oil spills. *Nano Research*. 2021;14(8):2697-
 387 702; Available from: <https://doi.org/10.1007/s12274-020-3274-y>.
 388
 389 9. Li L, Hu T, Sun H, Zhang J, Wang A. Correction to "Pressure-
 390 Sensitive and Conductive Carbon Aerogels from Poplars
 391 Catkins for Selective Oil Absorption and Oil/Water Separation".
 392 *ACS Applied Materials & Interfaces*. 2017;9(42):37509-; PMID:
 393 29019390. Available from: <https://doi.org/10.1021/acsami.7b14699>.
 394
 395 10. Yue X, Zhang T, Yang D, Qiu F, Li Z. Hybrid aerogels de-
 396 rived from banana peel and waste paper for efficient oil ab-
 397 sorption and emulsion separation. *Journal of Cleaner Produc-*
 398 *tion*. 2018;199:411-9; Available from: <https://doi.org/10.1016/j.jclepro.2018.07.181>.
 399
 400 11. Yin T, Zhang X, Liu X, Li B, Wang C. Cellulose-based aerogel
 401 from Eichhornia crassipes as an oil superabsorbent. *RSC Ad-*
 402 *vances*. 2016;6(101):98563-70; Available from: <https://doi.org/10.1039/C6RA22950F>.

402 12. Phan HT, Nguyen KD, Nguyen HHM, Dao NT, Le PTK, Le HV.
 403 Nata de coco as an abundant bacterial cellulose resource
 404 to prepare aerogels for the removal of organic dyes in wa-
 405 ter. *Bioresource Technology Reports*. 2023;24:101613; Avail-
 406 able from: <https://doi.org/10.1016/j.biteb.2023.101613>.
 407
 408 13. Liebner F, Haimer E, Wendland M, Neouze M-A, Schlufter
 409 K, Miethe P, et al. Aerogels from Unaltered Bacterial
 410 Cellulose: Application of sCO₂ Drying for the Prepa-
 411 ration of Shaped, Ultra-Lightweight Cellulosic Aero-
 412 gels. 2010;10(4):349-52; PMID: 20166232. Available from:
 413 <https://doi.org/10.1002/mabi.200900371>.
 414
 415 14. Liu H, Geng B, Chen Y, Wang H. Review on the Aerogel-Type Oil
 416 Sorbents Derived from Nanocellulose. *ACS Sustainable Chem-*
 417 *istry & Engineering*. 2017;5(1):49-66; Available from: <https://doi.org/10.1021/acsschemeng.6b02301>.
 418
 419 15. Korhonen JT, Kettunen M, Ras RHA, Ikkala O. Hydrophobic
 420 Nanocellulose Aerogels as Floating, Sustainable, Reusable,
 421 and Recyclable Oil Absorbents. *ACS Applied Materials & In-*
 422 *terfaces*. 2011;3(6):1813-6; PMID: 21627309. Available from:
 423 <https://doi.org/10.1021/am200475b>.
 424
 425 16. Zheng Q, Cai Z, Gong S. Green synthesis of polyvinyl al-
 426 cohool (PVA)-cellulose nanofibril (CNF) hybrid aerogels and
 427 their use as superabsorbents. *Journal of Materials Chemistry*
 428 *A*. 2014;2(9):3110-8; Available from: <https://doi.org/10.1039/C3TA14642A>.
 429
 430 17. Shi J, Lu L, Guo W, Sun Y, Cao Y. An environment-friendly
 431 thermal insulation material from cellulose and plasma modifi-
 432 cation. 2013;130(5):3652-8; Available from: <https://doi.org/10.1002/app.39615>.
 433
 434 18. Li Z, Zhong L, Zhang T, Qiu F, Yue X, Yang D. Sustainable, Flex-
 435 ible, and Superhydrophobic Functionalized Cellulose Aerogel
 436 for Selective and Versatile Oil/Water Separation. *ACS Sustain-*
 437 *able Chemistry & Engineering*. 2019;7(11):9984-94; Available
 438 from: <https://doi.org/10.1021/acsschemeng.9b01122>.
 439
 440 19. Gong J, Li J, Xu J, Xiang Z, Mo L. Research on cellulose
 441 nanocrystals produced from cellulose sources with various
 442 polymorphs. *RSC Advances*. 2017;7(53):33486-93; Available
 443 from: <https://doi.org/10.1039/C7RA06222B>.
 444
 445 20. Raghav N, Sharma MR, Kennedy JF. Nanocellu-
 446 lose: A mini-review on types and use in drug de-
 447 livery systems. *Carbohydrate Polymer Technolo-*
 448 *gies and Applications*. 2021;2:100031; Available from:
 449 <https://doi.org/10.1016/j.carpta.2020.100031>.
 450
 451 21. Iguchi M, Yamanaka S, Budhiono A. Bacterial cellulose-a
 452 masterpiece of nature's arts. *Journal of Materials Science*.
 453 2000;35(2):261-70; Available from: <https://doi.org/10.1023/A:1004775229149>.
 454
 455 22. Liu A, Shi Z, Reddy RG. Mechanism study of Cu-Zn al-
 456 loys electrodeposition in deep eutectic solvents. *Ionic*.
 457 2020;26(6):3161-72; Available from: <https://doi.org/10.1007/s11581-019-03418-2>.
 458
 459 23. Mohite BV, Patil SV. Physical, structural, mechanical and ther-
 460 mal characterization of bacterial cellulose by G. hansenii
 461 NCIM 2529. *Carbohydrate Polymers*. 2014;106:132-41; PMID:
 462 24721060. Available from: <https://doi.org/10.1016/j.carbpol.2014.02.012>.
 463
 464 24. Yang H, Yan R, Chen H, Lee DH, Zheng C. Characteris-
 465 tics of hemicellulose, cellulose and lignin pyrolysis. *Fuel*.
 466 2007;86(12):1781-8; Available from: <https://doi.org/10.1016/j.fuel.2006.12.013>.
 467
 468 25. Hopson C, Villar-Chavero MM, Domínguez JC, Alonso MV,
 469 Oliet M, Rodriguez F. Cellulose ionogels, a perspective
 470 of the last decade: A review. *Carbohydrate Polymers*.
 471 2021;274:118663; PMID: 34702482. Available from: <https://doi.org/10.1016/j.carbpol.2021.118663>.
 472
 473 26. Shang Q, Chen J, Hu Y, Yang X, Hu L, Liu C, et al. Facile Fabrica-
 474 tion of Superhydrophobic Cross-Linked Nanocellulose Aero-
 475 gels for Oil-Water Separation. 2021;13(4):625; PMID: 33669607.
 476 Available from: <https://doi.org/10.3390/polym13040625>.
 477
 478 27. He X, Zhang L, Meng D, Wu J. From hydrogel to aere-
 479 gel: A green fabrication of multifunctional polyimide ab-

- 473 sorbents. European Polymer Journal. 2017;89:461-7;Available
474 from: <https://doi.org/10.1016/j.eurpolymj.2017.02.039>.
- 475 28. Yang Y, Chen X, Li Y, Yin Z, Bao M. Construction of a Super-
476 hydrophobic Sodium Alginate Aerogel for Efficient Oil Ab-
477 sorption and Emulsion Separation. Langmuir. 2021;37(2):882-
478 93;PMID: 33415974. Available from: [https://doi.org/10.1021/](https://doi.org/10.1021/acs.langmuir.0c03229)
479 [acs.langmuir.0c03229](https://doi.org/10.1021/acs.langmuir.0c03229).

Tổng hợp và phân tích vật liệu aerogel kỵ nước chứa cellulose vi khuẩn được phủ đồng thông qua phản ứng khử êm dịu

Lê Vũ Hà^{1,2,*}, Nguyễn Huỳnh Mai Hạnh^{1,2}, Nguyễn Thị Phương Trang^{1,2}, Nguyễn Thị Thanh Trúc^{1,2}, Đặng Đình Khôi^{1,2}, Lê Anh Kiên³, Nguyễn Đăng Khoa^{1,2}, Phan Tuấn Hoàn^{1,2}



Use your smartphone to scan this QR code and download this article

TÓM TẮT

Trong nghiên cứu này, cellulose vi khuẩn (bacterial cellulose, BC) đã được bao phủ với đồng thông qua phản ứng khử ở nhiệt độ phòng giữa đồng(II) acetate và hydrazine trong hệ phân tán của thạch dừa trong nước, sau đó được sấy đồng khô để tạo thành các aerogel siêu nhẹ kỵ nước. Các đặc trưng cấu trúc của aerogel đã được xác định bằng các kỹ thuật khác nhau bao gồm phương pháp nhiễu xạ tia X (XRD), phân tích nhiệt trọng lượng (TGA), đo góc thấm ướt và hấp phụ nitrogen đẳng nhiệt. Kết quả XRD đã chứng minh sự hình thành của pha đồng kim loại trong aerogel trong khi độ tinh thể cao của cellulose vẫn được duy trì. Hơn nữa, các pha đồng khác không xuất hiện trong vật liệu. Bằng phân tích TGA, hàm lượng đồng có mặt trong vật liệu đã được xác định trong khoảng 3.9 đến 13.4% tùy thuộc vào hàm lượng đồng(II) acetate được sử dụng trong phản ứng khử. Các giá trị này nhìn chung thấp hơn so với hàm lượng đồng theo lý thuyết do sự thất thoát trong quá trình tổng hợp vật liệu. Việc tăng hàm lượng đồng trong aerogel đã dẫn đến diện tích bề mặt của vật liệu giảm đáng kể với sự có mặt của đồng trong cấu trúc mao quản của vật liệu. Tuy nhiên, như mong đợi, tính kỵ nước của vật liệu đã được cải thiện đáng kể khi tăng hàm lượng đồng. Các mẫu aerogel chứa hàm lượng đồng cao (> 6.2 wt.%) kỵ nước với góc thấm ướt lên đến 138°. Do đó, các aerogel kỵ nước tương tác tốt với các dung môi hữu cơ không tan trong nước như diesel oil và cyclohexane với hiệu quả hấp phụ từ 20 đến 30 g/g. Việc tổng hợp thành công các aerogel kỵ nước dựa trên sự biến tính bề mặt BC với đồng có thể cung cấp những vật liệu hấp phụ mới và hiệu quả trên cơ sở sinh khối hướng đến việc xử lý các chất lỏng dạng dầu trong môi trường nước.

Từ khóa: aerogel kỵ nước, cellulose vi khuẩn, bao phủ, đồng, phản ứng khử êm dịu

¹Khoa Kỹ thuật Hóa học, Trường Đại học Bách khoa Tp. HCM, 268 Lý Thường Kiệt, Quận 10, Thành phố Hồ Chí Minh, Việt Nam

²Đại học Quốc gia Thành phố Hồ Chí Minh, Phường Linh Trung, Thành phố Thủ Đức, Thành phố Hồ Chí Minh, Việt Nam

³Viện Nhiệt Đới Môi Trường, 57A Trường Quốc Dung, Quận Phú Nhuận, Thành phố Hồ Chí Minh, Việt Nam

Liên hệ

Lê Vũ Hà, Khoa Kỹ thuật Hóa học, Trường Đại học Bách khoa Tp. HCM, 268 Lý Thường Kiệt, Quận 10, Thành phố Hồ Chí Minh, Việt Nam

Đại học Quốc gia Thành phố Hồ Chí Minh, Phường Linh Trung, Thành phố Thủ Đức, Thành phố Hồ Chí Minh, Việt Nam

Email: lvha@hcmut.edu.vn

Lịch sử

- Ngày nhận: 20-2-2024
- Ngày chấp nhận: 23-5-2024
- Ngày đăng:

DOI:



Trích dẫn bài báo này: Hà L V, Hạnh N H M, Trang N T P, Trúc N T T, Khôi D D, Kiên L A, Khoa N D, Hoàn P T. **Tổng hợp và phân tích vật liệu aerogel kỵ nước chứa cellulose vi khuẩn được phủ đồng thông qua phản ứng khử êm dịu.** *Sci. Tech. Dev. J. - Eng. Tech.* 2024; ():1-1.