Synthesis, Characteristics, Oil Adsorption, and Thermal Insulation Performance of Cellulosic Aerogel Derived from Water Hyacinth

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ABSTRACT: Cellulosic aerogel from water hyacinth (WH) was synthesized to address the dual environmental issues of water hyacinth pollution and the production of a green material. Raw WH was treated with sodium hydroxide (NaOH) with microwave assistance and in combination with hydrogen peroxide (H2O2). The results from X-ray diffraction (XRD), Fourier transform infrared (FT-IR) spectroscopy, and scanning electron microscopy (SEM) showed that lignin and hemicellulose were markedly decreased after treatment, reducing from 24.02% hemicellulose and 5.67% lignin in raw WH to 8.32 and 1.92%, respectively. Cellulose aerogel from the pretreated WH had a high porosity of 98.8% with a density of 0.0162 g cm⁻³ and a low thermal conductivity of 0.030 W m⁻¹ K⁻¹. After modification with methyl trimethoxysilane (MTMS) to produce a highly hydrophobic material, WH aerogel exhibited high stability for oil absorption at a capacity of 43.3, 43.15, 40.40, and 41.88 (g g⁻¹) with diesel oil (DO), motor oil (MO), and their mixture with water (DO + W and MO + W), respectively. The adsorption remained stable after 10 cycles.

INTRODUCTION

Aerogel materials have been studied and synthesized for a long time and used in many different fields, such as insulation material,¹ nanocomposites, biomaterials, paper, packaging, filtration membranes,²−⁴ and absorption of oil spills.⁵,⁶ They are low-density, nanostructured porous materials with very low thermal conductivity, excellent acoustic insulation properties, high mechanical strength, low density, large specific surface area, and high porosity.⁶−¹⁰ Aerogel materials have been synthesized from inorganic, organic, and inorganic–organic hybrid raw materials, such as resorcinol–formaldehyde, melamine–formaldehyde, silicon (SiO₂), alumina (Al₂O₃), titania (TiO₂), zirconia (ZrO₂), and other oxides.¹¹ However, these materials have disadvantages, such as deformation and difficult synthesis with high-cost processes that consume many chemicals that are released into the environment.⁶−¹⁰ More recently, biobased aerogels fabricated from agricultural sources have attracted attention, with some positive results.¹²−¹⁶ The process of synthesizing cellulose aerogel materials from agricultural waste, with enhanced cross-linking to improve mechanical properties, is now the subject of study and has solved the above problems.¹⁷

Water hyacinth (WH) is a rhizomatous and stoloniferous plant with long pendant adventitious roots. It is free-floating on the surface of water and thrives in tropical countries, including Vietnam. The rapid growth rate of WH (125 tons per surface ha in 6 months)¹⁸ means that it causes serious environmental and health issues in the water ecosystem and water transport network. It is also a source of disease development (mosquitoes). A number of attempts have been made to use WH for handicraft products,¹⁹ fertilizers,²⁰ and biofuels (ethanol, methane, and biogas),²¹ but low product conversion rates and the high cost of pretreatment have hindered progress. However, WH has a high cellulose content (up to 50 wt %), which can be used as feedstock for aerogel cellulose. In addition, WH is highly porous and contains a lot of water; it can be cut into small sizes without drying in the pretreatment step, reducing the energy consumption of the synthesis process.

Currently, many studies have been successful in isolating cellulose/nanocellulose in water hyacinth. Tanpichai et al.²² reported that the cellulose content was 89.5% after heat treatment and alkalization, and Juárez-Luna et al.²³ reported that the cellulose content reached 89.88% through dewaxing, heat treatment, and alkaline peroxide, while Ruan et al.²⁴...
Reported that the cellulose content reached 59.9% after alkalization alone. Cellulose aerogel materials synthesized from agricultural waste that enhanced cross-linking had improved mechanical properties for oil adsorption and thermal insulation. Thai et al.25 reported that the motor oil absorption capacity and thermal conductivity with aerogel cellulose from sugarcane bagasse were 25 g·g⁻¹ and 0.031–0.042 W·m⁻¹·K⁻¹, respectively. Nguyen et al.26 reported that the crude oil absorption capacity of aerogel cellulose synthesized from paper waste was 24.4 g·g⁻¹. In another article, Feng et al.27 reported that the oil absorption capacity of aerogel cellulose from recycled paper was 95 g·g⁻¹. Therefore, aerogel cellulose is now the major subject of many studies and has helped the above problems.

In this work, WH was used to form cellulose aerogel using a simple and low-cost technique to create a high-value-added material for the treatment of oil spills. The results show a promising and practical approach to synthesizing WH into aerogel cellulose using poly(vinyl alcohol) (PVA), which

![Figure 1](https://doi.org/10.1021/acsomega.1c03137)

**Figure 1.** Effect of NaOH concentration at 30 min (a), treatment duration with microwave assistance with a NaOH concentration of 2 w/w % (b) on raw WH, and H₂O₂ concentration (c) in the bleaching process of treated WH.

![Figure 2](https://doi.org/10.1021/acsomega.1c03137)

**Figure 2.** XRD patterns (a) and FT-IR spectra (b) of raw WH (1), WH treated with a 2 w/w % NaOH solution for 30 min (2), and WH treated with NaOH 2 w/w % for 30 min and then with H₂O₂ 17 w/w % for 10 min (3).
enhances the extensibility and strength of the three-dimensional structure of the material. The product exhibits ultraflexibility, very high porosity, ultra-low-density, low thermal conductivity, superhydrophobicity after being coated with MTMS, and excellent oil absorption capability. Therefore, aerogel cellulose made from WH has the potential to be used in thermal insulation, as well as in the treatment of oil spills. In this study, a microwave was used to remove lignin and hemicellulose from WH instead of using a mixture of toluene/ethanol solvents or other solvents. This method has not been previously described, and it can reduce both environmental pollution and cost.

**RESULTS AND DISCUSSION**

**Effect of Pretreatment Conditions on the Removal of Lignin and Hemicellulose.** The concentrations of cellulose, hemicellulose, and lignin in raw WH were found to be 61.61, 24.02, and 8.44%, respectively. Figure 1a,b shows the effect of NaOH concentration and the pretreatment time on the removal of hemicellulose and lignin. It can be seen that the cellulose content in the pretreated sample increased significantly from 62 to 79–80% with a small amount of NaOH. Increasing the NaOH concentration above 2 w/w % had little effect on cellulose recovery. Regardless of the amount of NaOH and the pretreatment time, there was still around 10% lignin in the pretreated sample. Alkaline hydrolysis degrades hemicellulose and lignin by destroying the aryl–ether, carbon–carbon, and aryl–aryl bonds between lignin and hemicellulose.28–30 The bulging fibers induce dissolution of both hemicellulose and lignin. The effect of treatment time is shown in Figure 2b, and it can be observed that when the treatment time is increased from 20 to 30 min with a concentration of NaOH of 2 w/w %, there is a noticeable change, but there is little change when the time is increased to 40 min. The best conditions of the treatment step to save costs and maximize efficiency can be determined from these results, namely, a NaOH concentration of 2 w/w % and a 30 min treatment time. Under these conditions, the cellulose content increased to 82.34% and the contents of hemicellulose and lignin were markedly reduced to 11.14 and 2.84%, respectively. After treatment with NaOH and microwave assistance, WH was bleached with H2O2 for 10 min to remove the remaining lignin and hemicellulose.31,32 H2O2 is a strong oxidizing agent, especially in alkaline solution. Figure 1c shows that the cellulose content increased slightly from around 84 to 88% on increasing H2O2 from 13 to 17%, at the expense of hemicellulose and lignin, and remained almost constant at 88% with further increase in H2O2 concentration. It can be concluded that the combination treatment of NaOH with microwave assistance and H2O2 can markedly reduce the lignin and hemicellulose from WH to 1.92 and 8.32 wt %, respectively, producing a very high content of cellulose fibers for the cellulose aerogel.

**Physicochemical Characteristics of WH and Aerogel Cellulose.** Figure 2a shows XRD patterns of raw WH and pretreated WH. The patterns were used to determine the crystallinity index, crystal thickness, and the distance between the lattices (Table 1). It can be seen that the amorphous region (2θ angle of 18°) decreased with pretreatment, whereas the crystalline region (2θ angle of 22°) increased. The crystallinity index increased from 46.44% (raw WH) to 65.45% when pretreated with NaOH at 2 w/w % and 75.40% when pretreated with NaOH at 2 w/w % and H2O2 at 17 w/w % for 30 min, as shown in Figure 2a. The formation of the crystalline structure is due to the interaction between the hydroxyl group and the cellulose.33,34 The decrease in the peaks at 20 angles of 15 and 24.5° and the disappearance of peak 30.5° indicate that calcium oxalate is almost removed when WH is treated with NaOH and with a combination of NaOH and H2O2.35,36 This is because calcium oxalate reacts with strong oxidizing agents, such as hydrogen peroxide or ozone.37 These results show that lignin and hemicellulose are present as amorphous components of raw WH.34,38,39 The results in Table 1 also show the increasing crystal thickness after treatment with NaOH and both NaOH and H2O2, and similar results have been previously reported.33,34 The distance between the lattices (d-spacing) is reduced after the chemical treatment.33

FT-IR results (Figure 2b) show that the OH group and the C–H bond in –CH2– of cellulose (3423.49 and 2925 cm−1), the C–O stretching of the cellulose structure (1060 cm−1),41 and amorphous cellulose vibration or the glucose ring stretch (898 cm−1) increased significantly in the samples after treatment with NaOH and H2O2. This is because of the increasing proportion of cellulose. In addition, the peak at 1736.39 cm−1 is due to the presence of C=O bonds and was also obtained in untreated WH indicating the presence of lignin and hemicellulose; this peak did not appear in the chemical- and bleach-treated materials. There are other peaks: the peak at 1596 cm−1 is associated with C=C; at 1384.29 cm−1 with the C–H and C=O groups of the aromatic ring in lignin; and at 1248 cm−1 with ester, ether, or phenolic compounds.40,45 These peaks change or disappear, confirming the removal of hemicellulose and lignin and the increase in the proportion of cellulose in the chemically treated WH.

It can be observed (Figure 3a) that raw WH has large, tight cellulose fibers with a smooth shape, wrapped with lignin, hemicellulose, and wax. In Figure 3b, SEM results show the decreasing cellulose fiber size, with a clear rolled shape formed after treatment with NaOH; this change comes about because hemicellulose, lignin, and wax are removed from the raw WH during the treatment process. Figure 3c shows the material treated with H2O2 after NaOH, with very small, individual fibers with a small diameter compared to the original WH fibers. These results show that most of the lignin and hemicellulose are removed from the raw WH and are consistent with the results presented in Figure 1. Meanwhile, in treated WH, lignin and hemicellulose are eliminated; therefore, clear cellulose fibers can be observed with small and discrete sizes.

**Aerogel Synthesis.** Figure 4 shows the internal morphology of the aerogel material with different PVA/cellulose ratios. The results indicate that when the ratio of PVA/cellulose is high (Figure 4a with a ratio of 4:1 of the LW1 sample), the arrays of PVA are formed, leading to the low porosity of the material obtained. When the cellulose content increases, more

| samples            | crystallinity index (CI) (%) | crystal thickness (nm) | d-spacing (Å) |
|--------------------|------------------------------|------------------------|---------------|
| raw WH             | 46.44                        | 2.99                   | 0.40          |
| WH treated by NaOH 2% | 65.53                        | 3.86                   | 0.39          |
| WH treated by NaOH 2% and H2O2 17% | 75.40                        | 4.36                   | 0.39          |
Figure 3. SEM image of raw WH (a), WH treated with NaOH (b), and NaOH combined with H$_2$O$_2$ (c).

cellulose fibers can be seen, and they are very evenly distributed when combined with PVA, as shown in Figure 4b (a PVA/cellulose ratio of 4:3 of the LW3 sample), producing a uniform porous material with high porosity that increases its absorption capacity. As shown in Figure 4c, as the cellulose content is increased further (a PVA/cellulose ratio of 4:5 of the LW5 sample), the cross-links are not formed well, leading to a decrease of the porosity of the material because of the low content of PVA and in agreement with the porosity results of these samples shown in Figure 5a. Similar results were obtained in a previous study.46

Figure 5a shows that increasing the cellulose content (by decreasing the ratio of PVA/cellulose) leads to a decrease in density and an increase in porosity. At a PVA/cellulose ratio of 4:3 (LW3), the density was 0.0162 g·cm$^{-3}$ with a porosity of 98.8%; these are the highest values obtained. This is because when the PVA content is high, it not only causes the cross-links to form an aerogel structure but also produces some arrays or blocks of PVA, leading to the porosity being low and the density being high (in samples LW1 and LW2), as shown in Figure 5a and a previous study.46 However, if the cellulose content is continuously increased (LW4 and LW5), because of the saturation of cellulose in water, more cellulose blocks are created, leading to a decrease in the ability to create cellulose aerogel structures and thus reducing the porosity and increasing the density.1 This result affects the absorption capacity of the product.

The thermal conductivity of the aerogel cellulose samples with different PVA/cellulose ratios was tested to examine their suitability for thermal insulation applications. Figure 5b shows that when the cellulose concentration increases, starting with sample LW1, the thermal conductivity of the sample decreases from 0.034 and reaches the lowest value of 0.030 W·m$^{-1}$·K$^{-1}$ with sample LW3. This finding is because when the cellulose concentration increases, the cellulose aerogel has a relatively complete structure, with lower density and higher porosity, as shown in Figure 5a. In fact, because the gas molecules tend to adhere on the surface of the solid molecules of the material, this limits their ability to transfer heat through the pores inside the material. Therefore, formation of uniform pores of the right size increases the insulation capacity of the material.47 It was confirmed that thermal conduction connections between particles made up the conduction path, and, in low-density and high-porosity materials, the number of pores of the size of the mean free-motion path of air molecules was higher.48,49 Then, when the cellulose concentration continued to increase from sample LW4 to sample LW5, decreasing the porosity and increasing the density of the material, thermal conductivity increased to 0.036 W·m$^{-1}$·K$^{-1}$ in sample LW5. The porosity and thermal conductivity of cellulose aerogels from water hyacinth are similar to those of samples from other cellulosic sources (shown in Table 2).

The N$_2$ adsorption–desorption isotherm of the powder LW3 sample is shown in Figure 6. The N$_2$ isotherm indicated a type II adsorption profile, which is characteristic of macroporous materials.53 The plot of BJH pore size distribution (PSD) shows that the pores were mostly distributed in the mesoporous range of 15–60 Å with a pore diameter of 26.6 Å. This pore size range is encouraging since the materials are advantageous for any application, especially in adsorption. The sample had a BET surface area of 20 m$^2$·g$^{-1}$, approximate to the results of aerogels in a previous publication,54 and a total
pore volume of 0.048 cm³ g⁻¹. The low specific surface area of the powder LW3 sample indicates that the aerogel framework is nonporous and stable. This porosity is different from the overall porosity of the material. The aerogel material has a porosity of up to approximately 99%, and the macropores from the frameworks were formed between the cellulose and PVA. These pores help the material to have good insulation and good oil absorption capacity.

The result of thermogravimetry and differential scanning calorimetry (TG-DSC) in Figure 7a shows that a slight reduction (around 6 wt %) was observed from 70 to 120 °C, which can be explained by the loss of water from the cellulose aerogel. The largest extent of weight loss was found from 250 to 350 °C and is due to the decomposition of the aerogel cellulose and also the degradation of PVA. This phenomenon corresponds with a strongly exothermic reaction of the material, as shown by the DSC curve, indicating that a large amount of heat is emitted by the samples. The remaining mass of around 1.21 wt % remains almost constant when the temperature is above 650 °C, corresponding to the decomposition of some inorganic compounds in the initial material. It can be concluded that the material is thermally stable up to about 250 °C, showing its potential to be used in insulation materials. This result is comparable with those of previous studies.

Figure 4. SEM images of aerogel cellulose from WH with different PVA/cellulose ratios: (a) 4:1, (b) 4:3, and (c) 4:5.

Figure 5. Effect of cellulose concentration on the density and porosity (a) and thermal conductivity (b) of aerogels.
Table 2. Comparison of the Thermal Conductivity of Various Cellulose Aerogels

| type of cellulose aerogel       | density (g cm⁻³) | porosity (%) | thermal conductivity (W m⁻¹ K⁻¹) |
|--------------------------------|------------------|--------------|---------------------------------|
| water hyacinth aerogels (this study) | 0.016–0.018 | 97.2–98.8     | 0.030–0.036                     |
| sugarcane bagasse aerogels     | 0.016–0.112     | 91.9–98.9     | 0.031–0.042                     |
| regenerated cellulose aerogels | 0.009–0.137     | 91.0–99.0     | 0.040–0.075                     |
| bacterial cellulose–silica aerogels¹| 0.007–0.229 | 89.0–99.6     | 0.031–0.037                     |
| bacterial cellulose–silica aerogels²| 0.007–0.201 | 90.3–99.6     | 0.029–0.037                     |

The water contact angle of aerogel cellulose was tested using a Drop Shape Analyzer DSA100E, and the results are shown in Figure 7b. It can be seen that the aerogel cellulose material is highly hydrophobic after MTMS treatment, while all oil drops were absorbed completely. When water was applied to the aerogel cellulose, a water contact angle of 142° was observed on the left-hand side and 142.1° on the right-hand side of the drop. This result shows the potential of the aerogel cellulose synthesized from WH to be used for oil spill treatment.

Absorption Capacity for Diesel Oil, Motor Oil, and Their Mixture with Water. The absorption capacity of aerogel cellulose samples obtained from WH was tested with diesel oil (DO) and motor oil (MO) using samples LW1, LW2, LW3, LW4, and LW5, which were prepared as described, and the results are shown in Figure 8a.

The results in Figure 8a show that the LW3 sample has the highest absorption capacity of 38.45 g·g⁻¹ with DO and 43.30 g·g⁻¹ with MO. The difference in absorption capacities can be explained by the difference in the porosity and the density of the materials.⁵⁸ The samples from LW1 to LW3 with decreasing cellulose concentration have absorption capacities significantly increasing from 33.27 to 38.45 g·g⁻¹ with DO and 35.66 to 43.3 g·g⁻¹ with MO. These results can be explained by the effect of the porosity of the as-prepared aerogels on the absorption capacity, which increases with an increase in porosity.⁵⁹ Then, if the cellulose concentration is increased further, the absorption capacity decreases to 35.2 g·g⁻¹ with DO and 37.25 g·g⁻¹ with MO using sample LW5; this result can be explained by the decreasing porosity and the increasing density of the samples.⁵⁸

The results shown in Figure 8b follow the same pattern of absorption capacity of the prepared samples on DO + W and MO + W as those of the previous case. LW3 also has the highest absorption capacity with DO + W and MO + W, with values of 38.59 and 42.93 g·g⁻¹, respectively. These results show the high potential of the synthesized aerogel cellulose modified with MTMS from WH to be used in oil spill treatment. To investigate the selectivity of the sample-synthesized material, sample LW3 was used because of its good performance. The absorbed oil was squeezed out, and the water content in the oil was determined after drying at 110 °C. The result shows that there was only 0.012 w/w % of water in the recovered oil, indicating that the prepared material has high selectivity to absorb oil from water.⁶⁰

Reusability. The reusability of aerogel cellulose materials from WH for MO absorption was tested with the LW3 sample, which has the highest oil absorption capacity, by determining changes in capacity over a number of cycles. After 3 min of submergence in MO, the sample was compressed by a 5 kg loading capacity for each cycle. This process was repeated 10 times. The results of the absorbing–releasing testing cycle are shown in Figure 9a. It can be seen that there was a significant deterioration in the oil absorption capacity from 43.3 g·g⁻¹ in cycle 1 to 26.1 g·g⁻¹ in cycle 2. However, from the second cycle to the tenth cycle, the absorption capacity does not change a great deal, reaching 21.6 g·g⁻¹ in the last cycle. The
reason for the large drop in absorption capacity after the first cycle is the massive structural collapse after the first compression, which leads to a huge loss of pores as the medium for retaining the oil; the structure is almost stable after other cycles.61,62

To confirm this explanation, an experiment on the ability of the samples to self-restore their shape was set up. Sample LW3 was tested using a loading capacity of 5 kg for 1 min, after which the sample was allowed to self-restore for 3 min.63,64

The initial height of the sample was 21.5 mm; after the shape self-restoration process, it was 8.5 mm, as shown in Figure 9b. After the first compression, the height change was not significant. The sample height after 10 compression cycles with the same process was 7.5 mm. This result reflects the same trend as that of the oil adsorption capacity of the material samples, as presented in Figure 9a.

■ CONCLUSIONS

WH can be used to synthesize aerogel cellulose after treatment with NaOH with microwave assistance and in combination with H2O2 and then modified with MTMS to produce a highly hydrophobic material that can be used for oil adsorption. Aerogel cellulose from WH also has low thermal conductivity, showing its potential as an insulation material. The capacity and the reusability of aerogel cellulose modified with MTMS to absorb DO and MO and their mixtures with water highlight its potential for oil spill treatment. The results in this study confirm that aerogel cellulose produced from WH not only solves environmental problems in waterways but also offers a new, low-cost, “green” material that has the potential to be used in many industrial fields.

■ EXPERIMENTAL SECTION

Material Preparation. The chemicals used include sodium chlorite (NaClO2, Himedia, 80–83%), acetic acid (CH3COOH, Xilong, ≥99.5%), sulfuric acid (H2SO4, Prolabo VWR, 95%), acetone (CH3OCH3, Xilong, purity 99.5%), poly(vinyl alcohol) (C2H4O)n, Merck, Mw approx. 145 000, methyl trimethoxysilane (C2H12O3Si, Sigma-Aldrich, 98%), hydrogen peroxide, (H2O2, Xilong, purity ≥30%), and sodium hydroxide (NaOH, Xilong, ≥96%). Motor oil 20W-40 4T and DO 0.05S-II were used for test absorption.

Raw WH was collected from rivers in Long An province, Vietnam, and washed with water to remove soil and other impurities. Raw, wet WH was then cut into small pieces, ground using a blender, filtered, and dried at 70 °C for 48 h to obtain WH fibers less than 0.1 mm in length (passed through a sieve with holes of 0.1 mm), as shown in Figure 10.

To remove lignin and hemicellulose, 2 wt % dried water hyacinth were mixed with a solution of NaOH at various concentrations (1, 2, 3, and 4 w/w %) in a 1 liter glass flask and ultrasonicated for 15 min. Then, the mixture was heated in a microwave (EM-G2135W, AQUA) at medium power (385

Figure 8. Oil absorption capacity of samples prepared with different PVA/cellulose ratios. (a) Pure oil and (b) oil/water mixture.

Figure 9. Reusability of the LW3 sample in MO absorption (a) and images (b) of the initial sample LW3 (1) compressed with 5 kg of loading capacity for 1 min (2) and after free self-restoration for 3 min (3).
W) for set times (10, 20, and 30 min) and stirred at a speed of 400 rpm, with a condenser tube support to identify the most cost-effective combination of treatment time and removal efficiency. A known amount of H₂O₂ aqueous solution at various concentrations (13, 15, 17, 19, and 21 vol %) was added to the cooled, microwave-treated mixture with the pH adjusted to around 11 using 5 M NaOH; the mixture was then heated in the microwave for a further 10 min. As soon as the mixture had cooled to room temperature, it was removed by placing the sample in a vacuum oven for another 24 h.

The porosity of the cellulose aerogel material is calculated by:

\[ \Phi = \left( 1 - \frac{\rho}{\rho_o} \right) \times 100\% \]  

where \( \rho \) is the dry sample density (g/cm³) and \( \rho_o \) is the density of the original solid sample (g/cm³).

The surface morphologies and elements of the obtained cellulose-based aerogels were observed using a field emission scanning electron microscope (Hitachi, S-4800). The crystal-line structures of the samples were identified using an X-ray diffractometer (Bruker D8 Advance diffractometer). Fourier transform infrared (FT-IR) analysis was performed in the range of 400—4000 cm⁻¹ using an FT-IR spectrophotometer (Tensor 27 FT-IR, Bruker, Germany) to identify the functional groups of samples. The water contact angle was measured on an OCA-20 (Dataphysics, Germany) to quantify the hydrophobicity of aerogel cellulose after coating with MTMS. The thermal conductivity of the samples was measured using a thermogravimetric analysis (TGA) technique on a Shimadzu DTG60H. The thermal conductivity of the samples was measured at room temperature using a C-Therm TCi thermal conductivity analyzer (C-Therm Technologies, Canada).

The oil absorption capacity of aerogel is calculated by:

\[ q = \frac{m_t - m_0}{m_0} \]  

where \( q \) is the oil absorption capacity of the aerogel at time \( t \) (g·g⁻¹) and \( m_t \) (g) and \( m_0 \) (g) are aerogel weights after and before oil absorption, respectively.

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Notes
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