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Recycled Cellulose Aerogels from Paper Waste for a Heat Insulation Design of Canteen Bottles

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Abstract: Exercising in a tropical climate with constant high temperatures and high humidity increases the risk of heatstroke for active people who frequently train outdoors. For these active persons, a cooling source of water nearby can be essential, and this is usually carried in canteen bottles. However, commercially available water canteen bottles have limited thermal insulation capability to keep the liquid content cooled for the required period. This work proposed an engineering solution to enhance the heat insulation performance of water canteen bottles, using recycled cellulose aerogels made from paper waste for the first time as an insulating layer. Recycled cellulose aerogels wrapped around the water canteen bottle provides excellent thermal insulation performance, while not adding significant weight to the bottle. The temperature of the ice slurry in the canteen bottle was measured periodically over four hours with a mercury thermometer. The effects of the static and dynamic conditions on the temperature rate were also quantified. A 1.5 cm thickness of 1.0 wt.% recycled cellulose aerogel wrapped around the canteen bottle can provide an excellent thermal insulation performance with the lowest rise in temperature, achieving a low final temperature of the ice slurry content of 3.5 °C after 4 h. This result is much better than that provided by available commercial bottles under the same conditions.

Keywords: cellulose aerogel; heat insulation; water canteen bottle; dynamic movement

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1. Introduction

A cooling source of water, carried in canteen bottles, can help to prevent dehydration and replace lost fluids for personnel embarking on physical activities. Cold fluids can also mitigate heat injuries by lowering the internal body temperature during the activities. The existing external cooling methods [1] commonly practised by the military are: (i) Cooling the body by spraying it with water or placing ice packs at the groin, neck and armpit areas, (ii) cooling by immersion of the entire body in an ice bath, and (iii) using a body cooling unit (BCU). However, these methods are implemented when heat injury has already occurred. Other unconventional internal cooling methods have no consistent results and are not recommended for practical applications. In addition, current pharmacologic agents cannot cool down the body effectively and should not be the solution to heat injuries [2]. Therefore, it is desirable if the heat injuries can be prevented in the first place.

In order to prevent heat injuries during physical activities, ice slurry can be ingested to reduce the body core temperature [2]. The ice slurry contains a mixture of ice particles and water. It is preferred over cold water because the ice slurry can absorb more heat (through its latent heat of fusion) and reduce the body core temperature more effectively under the same volume [3,4]. There are several commercially thermal insulated bottles like FLOE bottles, readily available on the market today. FLOE is a silicone bottle that is designed to insulate and deliver ice slurry as well as cold liquids to athletes. However, it cannot keep the ice slurry for a sufficient amount of time [5]. Using a thermal jacket made of insulation materials wrapped around the thermal bottle may be a simple and effective engineering solution for heat insulation.

Among the insulation materials, cellulose is perhaps one of the most eco-friendly options [6]. The cellulose fibers are made from recycled cardboard, paper, and other similar materials, which can be collected from various sources [7]. The collected paper waste requires further processing to remove contaminants such as ink and dirt through filtration or mechanical separation techniques [8]. Once the contaminants are removed from the paper waste, the recycled cellulose fibers are obtained in a loose form. However, there are certain downsides to the recycled cellulose fibers, such as allergies that some people may have to newspaper dust. In addition, using loose cellulose fibers for insulation requires some skills [6]. In this study, we prefer cellulose fibers in an aerogel form because the cellulose aerogels is eco-friendly, cost-effective, easy to install and handle, and very stable under ambient conditions [9–12]. Aerogel insulations [13,14] are four times more efficient than glass fibers, cheaper and is the obvious choice for most insulation applications [6]. Heat transfer in porous materials, like aerogels, is based on three processes: Heat conduction in the solid phase, and heat convection through the gaseous phase present in the porous structure of the aerogel, and heat radiation [15–18]. These three factors are combined to calculate the total thermal conductivity, which can be decomposed according to a parallel flux model [18] stated as follows:

$$\lambda_{\text{aerogel}} = \lambda_{\text{solid}} + \lambda_{\text{gas}} + \lambda_{\text{rad}}$$

The contributions of the solid phase (λ_{solid}) and the gas phase (λ_{gas}) can be significantly reduced by decreasing density of aerogels and by narrowing the pore size to less than the mean free path of gas molecules in the air, respectively [15]. Aerogels can enhance heat insulation performance because they are highly porous with a small pore structure and consequently, a very low density [18]. With cellulose aerogels, the concentration of the cellulose fibers influences thermal conductivity via the thermal conductivity through the solid skeleton. Then the sizes of the pores are known to drastically change depending on the concentration of the cellulose fibers, changing the thermal conduction through the gas molecules [19].

Combining the cellulose fibers from the paper waste can create recycled cellulose aerogels with superior properties. The recycled cellulose aerogels have a very low thermal conductivity of 0.032 $\text{Wm}^{-1}\text{K}^{-1}$ [10–12] comparable to other good thermal insulators such as cotton (0.04 $\text{Wm}^{-1}\text{K}^{-1}$) and sheep wool (0.039 $\text{Wm}^{-1}\text{K}^{-1}$) [19]. The recycled cellulose aerogels exhibit excellent flexibility, as it can be bent through 90 degrees repeatedly without any structural damage [20–22]. Besides, they can be endowed with super-hydrophobic properties by coating with trimethoxymethylsilane (MTMS) at 70°C for 2–4 h [9,11,12].

Some previous studies [9–12] developed cellulose aerogels from recycled cellulose fibers of paper waste mainly for oil spill cleaning, not for heat insulation applications. In this paper, we focus on a thermally insulating applications in water canteen bottles, with potential applications in sporting, occupational, and military training exercises. A high-density polyethylene (HDPE) water canteen is chosen because it is recyclable, rigid, cost-effective, corrosion-resistant, water repellent, non-toxic, and lightweight [23]. As the thermal insulation performance of the commercial water canteen bottles is poor [5], an engineering solution is developed in this work to maintain the ice slurry content at low temperature as long as possible for the duration of physical activities. The water canteen bottles are wrapped around with recycled cellulose aerogels made from paper waste. A 2:1 standard mixture of crushed ice and liquid water is used to test their heat insulation performance. The thermal insulation performance of cellulose aerogel insulated water canteen bottles is also compared to that of typical thermal bottles such as hydro vacuum flasks [24] and FLOE bottles [25], in terms of heat insulation capacity, cost and weight.

2. Materials and Methods

2.1. Materials

Recycled cellulose fibers and Kymene 557H were sponsored by Insul-Dek Engineering Pte. Ltd. (Singapore) and Ashland (Taiwan), respectively. All the solutions were made with deionized (DI) water. Analytical grade MTMS was purchased from Aldrich Sigma (Singapore). All these chemicals were used as received.

2.2. Preparation of Recycled Cellulose Aerogels from Paper Waste

For making 1.0 cm thickness of the 1.0 wt.% cellulose aerogel sample, the amount of mixture should reach a depth of approximately 1.5 cm to cater for shrinkage, loss of water and compensate the cellulose fibers sinking to the bottom of the mold during freezing. A total of 1.2 L of DI water and 12 g of cellulose fibers were required to reach 1.5 cm of depth in the mold. The mixture reactant was stirred thoroughly and placed in an ultrasonic processor for 15 min at 200 W. The ultrasonic processor dispersed the cellulose fibers uniformly into the solution. The obtained suspension was then cooled down to room temperature using a water bath. After that, the Kymene cross-linker (2.4 g of Kymene diluted with 6 mL of DI water) was added to the mixture and sonicated for another 15 min. After the completed sonication process, the mixture sample was poured into the mold and placed in the freezer to freeze overnight. The next step was to place the sample in the freeze dryer at a temperature of −90 °C. After freeze-drying, the sample was placed in the oven for 3 h at 120 °C. After the curing process, the cellulose aerogel samples were placed in an airtight box containing MTMS chemical in the oven at 80 °C for one day. The super-hydrophobic cellulose aerogel sample was obtained, as shown in Figure 1a,b.

2.3. Heat Insulation Static Tests of the Water Canteen Bottle and the FLOE Bottle

The recycled cellulose aerogel was wrapped around the water canteen bottle and the FLOE bottle and secured using thin strips of aluminum foil, as shown in Figure S3. Then they were filled with the ice slurry. Temperatures of the ice slurry were recorded periodically every half an hour over a total experimental period of 4 h using a mercury thermometer. The temperature rate of increase was estimated by taking the difference between the final and initial temperatures of the ice slurry divided by the total test time of 4 h. Following this, the sandwich structure was proposed using the cellulose aerogel in Figure S3b and other textiles to form the insulated jacket in Figure S3c. The details of the design of sandwich structures and the prototype fabrication process of the thermal jacket can be found in Supplements S1 and S2. The main focus of this work was to study the effects of the structures and morphologies of the cellulose aerogels on the heat insulation of the water canteen bottle. In addition, in order to avoid the effects of the textiles and thermal jacket manufacturing technique, prototype 1 in Figure S3a was investigated in this work.

2.4. Heat Insulation Dynamic Tests of the Water Canteen Bottle and the FLOE Bottle

The water canteen bottle was wrapped with 1.5 cm thickness of the 1.0 wt.% cellulose aerogel and then placed in a water canteen sleeve. This work was necessary to maintain the integrity of the recycled cellulose aerogel during walking or running. Periodic running movement was simulated using the treadmill to keep the constant speed of 10 km/h for 10 min with a 30 min rest period over a total duration of 3 h. Periodic walking movement was also conducted to simulate a route march. Similarly, the treadmill was utilized to keep the constant speed of 5 km/h, and the temperature was taken periodically over 3 h. Temperatures of the ice slurry were recorded periodically every half an hour over the total experimental period using a mercury thermometer.

3. Results and Discussion

3.1. Morphology of the Cellulose Aerogels

The chemical Kymene 557H wet strength resin is added to enhance the strength of the material by facilitating the formation of cross-linkages between the paper fibers. The chemical contains quaternary ammonium groups that absorb onto negatively charged paper fibers and continue to form cross-linkages between them. Kymene additives work during the curing process when the functional groups on the Kymene polymer react with cellulose fiber to form a covalent bond. The polymer molecules cross-link, forming a network in the cellulose web that provides strength during the gelation process. Kymene wet-strength products can also reinforce existing fiber-to-fiber bonds, which further enhance the strength of the cellulose aerogel. Therefore, the strength of the material continues to increase even after aerogel sample completion [9,26]. The water contact angles in Figure 1c,d prove the uniform coating of MTMS on the whole surface of the cellulose aerogel and its super-hydrophobic property with the values in a range of 149–155°. The scanning electron microscope (SEM) used in Figure 1b confirms that the sample had a highly porous network of uniform fibers of roughly 8-µm diameter. This implies that cellulose fibers are bonded uniformly via hydrogen bonding and covalent bonding to form a porous 3D network. The cellulose aerogel had a pore size in the range of 40–200 µm and high porosity of over 95%. The values were lower than cellulose aerogel made from nanocellulose fibers [20,21] probably because of the macroporous structure of the recycled cellulose aerogel compare to the nanoporous network of the nanocellulose aerogels or bacterial cellulose aerogels.

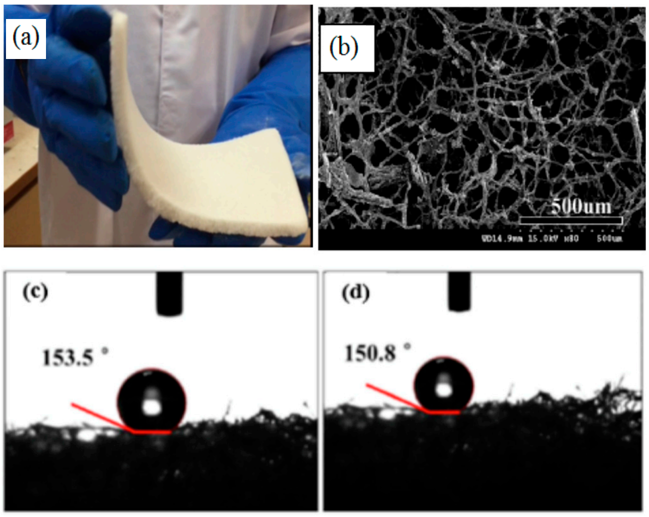

Figure 1. (**a**) An A4-size flexible cellulose aerogel, (**b**) morphology of highly porous trimethoxymethylsilane (MTMS) coated recycled cellulose aerogel, and water contact angles on (**c**) the external surface (153.5°) and (**d**) the internal surface (150.8°), respectively.

3.2. Heat Insulation Performance of the Water Canteen Bottle and the Commercial FLOE Bottle With and Without the Wrapped Cellulose Aerogel

Figure 2a compares the heat insulation performance of the water canteen bottle and the commercial FLOE bottle without the cellulose aerogel wrapped as the insulation layer. Under the static test, the water canteen bottle and the FLOE bottle performed poorly: The temperature of the ice slurry increased dramatically from −1 °C to room temperature after 4 h. The FLOE bottle did not have better heat insulation performance than the water canteen bottle, and it was not a good alternative for the water canteen bottle in terms of heat insulation capacity, cost and weight as shown in the Supplement Document.

When both the water canteen bottle and the FLOE bottle were covered with 1.5 cm thickness of the 1.0 wt.% recycled cellulose aerogel in Figure 2b, their temperature trends showed that their heat insulation performance improved significantly. When the water canteen bottle was wrapped around with the cellulose aerogel, the temperature of the ice slurry inside reduced from 20 °C (without the wrap around) to 3 °C, 6.7 times after 4 h. After 4 h under the static condition, the temperatures of the ice slurry inside the insulated water canteen bottle and the insulated FLOE bottle were 3 °C and 8 °C, respectively. The rate of change in the temperature (dT/dt) for the insulated water canteen bottle was 0.02 °C/s, half that of the insulated FLOE bottle (0.04 °C/s). A smaller thermal rate value indicated a more gradual temperature rise and hence, better thermal insulation. With the same cellulose aerogels wrapped, the water canteen bottle had better heat insulation performance than that of the FLOE bottle. The difference between the heat insulation performances of the two bottles may be dependent on their material usage and engineering design.

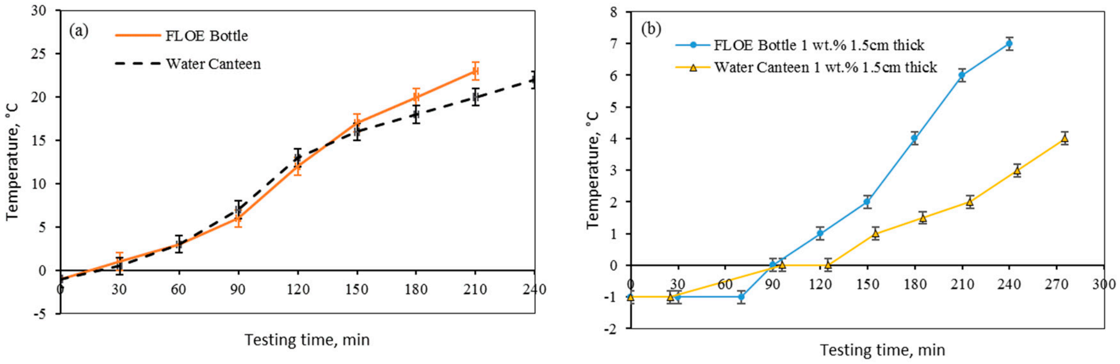

Figure 2. The heat insulation performance of the water canteen bottle and the FLOE bottle (**a**) without wrapped the cellulose aerogel, and (**b**) wrapped with a 1.5 cm thickness of 1.0 wt.% cellulose aerogel as the insulation layer.

3.3. Effects of the Aerogel Thickness on the Heat Insulation Performance of the Water Canteen Bottle

Figure 3 shows the effects of the cellulose aerogels and their thickness on the heat insulation performance of the water canteen bottle containing the ice slurry. The un-insulated water canteen bottle showed the worst heat insulation performance as the temperature of the ice slurry increased dramatically from −1 °C to 22 °C after 4 h. Once the water canteen bottle was wrapped with the recycled cellulose aerogel, its heat insulation performance improved significantly. For example, the water canteen bottle wrapped with 1.5 cm thick cellulose aerogels prepared from the 0.6 wt.% suspensions kept the temperature of the ice slurry below 3.5 °C after 4 h. This happened due to the very low thermal conductivities of the cellulose aerogels (K = 0.04 $Wm^{-1}K^{-1}$) [10–12]. As can be seen from Figure 3, the increase of cellulose aerogel thickness led to an enhancement of the thermal insulation performance of the water canteen bottle due to the increase of thermal resistance with the thickness of the materials. A higher thickness meant less heat flow through the materials, and so did a lower conductivity. The thermal resistance was proportional to the thickness of the layer of the insulation materials. Besides, the temperature rate of the water canteen bottle wrapped with cellulose aerogels decreased from 0.07 °C/s, 0.05 °C/s to 0.02 °C/s with increasing insulation thickness, from 0.5 cm, to 1.0 cm and 1.5 cm, respectively. The lower the temperature rate, the slower the heat transfer through the aerogel. Thus, the thicker cellulose aerogels enhanced the heat insulation performance of the water canteen bottle because the thermal resistance per unit-exposed area was proportional to the insulation thickness and inversely proportional to insulation thermal conductivity.

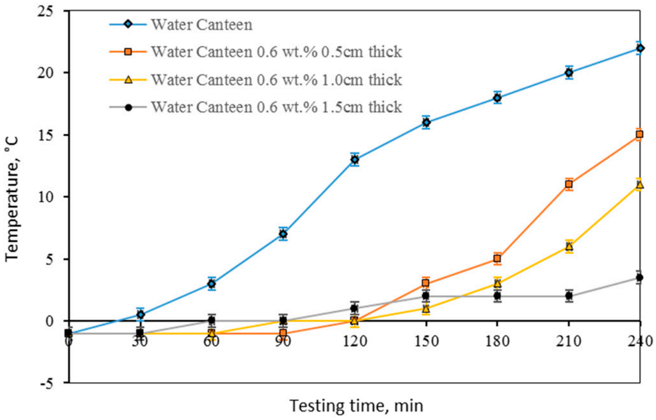

Figure 3. Effects of the cellulose aerogel as the insulation layer and their thickness on the heat insulation performance of the water canteen bottle.

3.4. Effects of the Various Aerogel Compositions on the Heat Insulation Performance of the Water Canteen Bottle

The heat insulation performance of the water canteen bottle wrapped with the recycled cellulose aerogels having different cellulose compositions at the same 1.5 cm thickness is compared in Figure 4. The 1.0 wt.% cellulose aerogels had a slightly better insulation performance than the 0.6 wt.% cellulose aerogels. However, there was only a slight 0.8 °C difference of the final temperature of the ice slurry within the same water canteen bottle. Hence, the cellulose aerogel composition may not have had any significant effect on the thermal insulation performance of the water canteen bottle. The 1.0 wt.% cellulose aerogel was chosen over the 0.6 wt.% one because it was more mechanically robust.

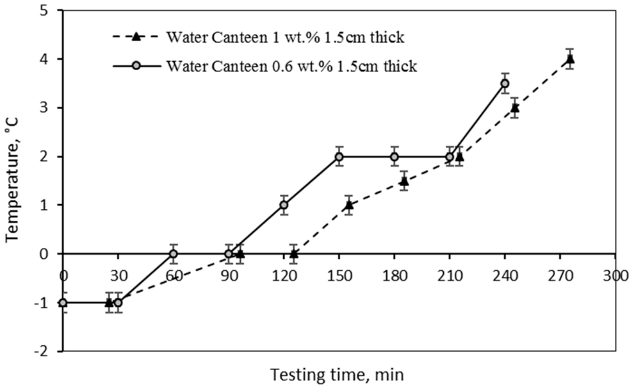

Figure 4. Effects of various cellulose aerogel compositions on the heat insulation performance of the water canteen bottle wrapped with the same 1.5 cm thickness of the cellulose aerogel.

3.5. Effects of Internal Temperature

The effect of internal temperature on the temperature rate of the water canteen bottle wrapped with cellulose aerogel has been investigated by filling the water canteen bottle with the ice slurry and cold water. In both cases, the water canteen bottle was wrapped with 1.5 cm thickness of the 1.0 wt.% cellulose aerogels. The results show that the internal temperature while the water canteen bottle was being filled with the ice slurry slowly increased compared to when the water canteen bottle was filled with the cold water after 6 h, as in Figure 5. The temperature rate for the water canteen bottle wrapped with cellulose aerogels increased from 0.02 °C/s to 0.05 °C/s when the internal temperature increased by controlling the ice slurry/chilled water content. It can be explained that the lower initial temperature and the ice particles of the ice slurry can counter heat transfer and delay temperature rise through both the recycled cellulose aerogel and the water canteen bottle effectively. The cellulose aerogel layer exhibited a better temperature rate on the water canteen bottle filled with the ice slurry.

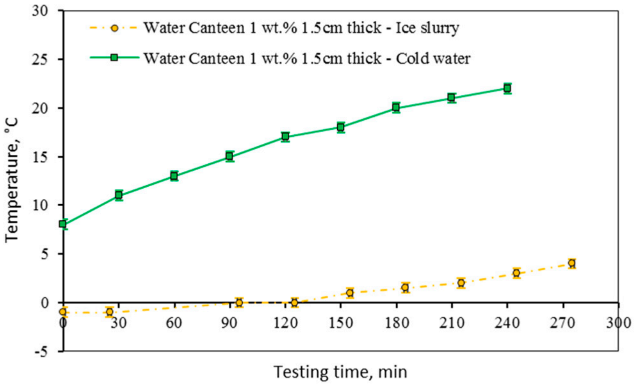

Figure 5. Effects of internal temperature on temperature rate of the water canteen bottle wrapped with 1.5 cm thickness of 1.0 wt.% cellulose aerogel.

3.6. Effects of the Static and Synamic Conditions

The temperature rate of the water canteen bottle wrapped with 1.5 cm thickness of the 1.0 wt.% cellulose aerogels under the static and dynamic conditions has been evaluated, as shown in Figure 6. The temperature trend for the running condition (round dots) shows a steep temperature increase from 1 °C to 15 °C after 2 h. Due to the shaking motion during running, turbulence was introduced within the water canteen bottle. This turbulence may have sped up the melting of the ice particles. Once all ice particles were melted, the heat was no longer absorbed in melting the ice particles, and the heat was transferred directly to the liquid and raised the temperature quickly. However, the shaking motion under the walking condition (triangle dots) was not as vigorous as that of under the running condition. The temperature had a more stable trend without any steep increase. Since the temperature of the ice slurry did not cross 0 °C after 3 h under the walking condition, there were still ice particles within the bottle that acted as buffers and this minimized temperature rise. Interestingly, the thermodynamic of the water canteen bottle under the walking condition was slightly better than that under the static condition. This event may have been due to the uniform distribution of the ice particles during walking and needs to be investigated further in the future.

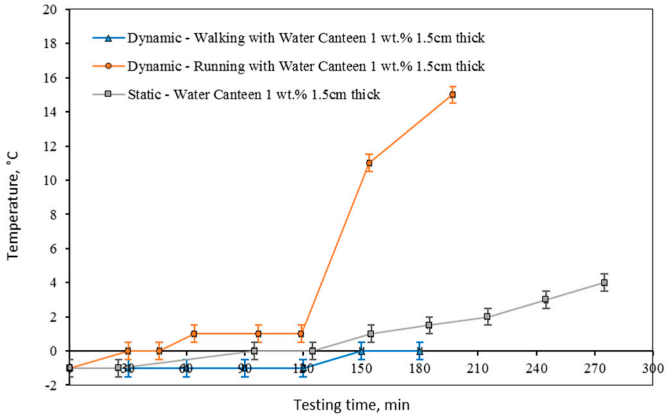

Figure 6. The effects of dynamic conditions on the thermodynamic of the water canteen bottle wrapped with a 1.5 cm thickness of 1.0 wt.% cellulose aerogel.

4. Conclusions

This work demonstrated that eco-friendly cellulose aerogels from paper waste can be used for excellent heat insulation applications. The heat insulation performance of the water canteen bottle wrapped with the cellulose aerogels were affected much by aerogel thickness but less by the cellulose fiber concentrations. Wrapping the military canteen bottle with 1.5 cm thickness of the 1.0 wt.% recycled cellulose aerogels can give the best heat insulation performance with the lowest temperature rate rise and lowest final temperature of the ice slurry. Compared to the commercial bottles, the cellulose-aerogel insulated water canteen bottle is more cost-effective and lightweight. Indeed, the findings in this paper can be applied to athletes training and competing in the heat to prevent heat injuries and optimize performance.

Supplementary Materials: The following are available online at http://www.mdpi.com/2311-5521/4/3/174/s1, Figure S1: Materials of (a) nylon, (b) cellulose aerogel, (c) neoprene are used for the insulated jacket design, and (d) the sandwich structure consists of neoprene as the outmost layer, the cellulose aerogel within and Nylon as the innermost layer, Figure S2: (a) Sketch of the insulated jacket and (b) final insulated jacket on the military canteen, Figure S3: (a) Prototype 1 of the water canteen wrapped with the recycled cellulose aerogel, (b) proposed sandwich structure of the fabric embedded with the cellulose aerogel, and (c) water canteen wrapped with a thermal jacket using the cellulose aerogel. The details of the design of sandwich structures and prototype fabrication process of the thermal jacket can be found in Supplements S1 and S2, Figure S4: Heat insulation performance of the water canteen wrapped with the cellulose aerogel (Prototype 1) and other commercial thermal bottles, Table S1: Comparison among commercial heat insulation bottles and the water canteen wrapped with the cellulose aerogel.

Author Contributions: Conceptualization, H.M.D. and J.K.W.L.; methodology, D.K.L. and Y.Q.X.; formal analysis, T.X.N.; investigation, L.W.Z.; writing—original draft preparation, L.W.Z.; writing—review and editing, Q.B.T. and H.D.M.; supervision, H.D.M.

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