

# Aerogels from Wastes and their Applications



Quoc Ba Thai<sup>1</sup>, Duyen K Le<sup>1</sup>, Thao Phuong Luu<sup>1,2</sup>, Nga Hoang Do Nguyen<sup>1,2</sup> and Hai M Duong<sup>1,3\*</sup>

<sup>1</sup>National University of Singapore, Singapore

<sup>2</sup>Ho Chi Minh University of Technology, VNU-HCM, Vietnam

<sup>3</sup>Ton Duc Thang University, Vietnam

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\*Corresponding author: Hai M Duong, Associate Professor, Mechanical Engineering Department, National University of Singapore, Singapore

## Abstract

Aerogels are not only among the lightest solid materials but also highly porous with great absorption capacity and low thermal conductivity. Unfortunately, the advanced materials have not been widely adopted by industries due to its high production cost. Leading aerogel scientists around the world are actively improving the manufacturing methods of different types of aerogels. This paper presents briefly the development of eco-aerogels using recycled fibers from various industrial wastes such as paper waste, fabric waste, and plastic bottle waste. The cost-effective eco-aerogels possess ultra-low densities, high porosity, super-hydrophobicity, and ultra-low thermal conductivities. The reported fabrication methods can be scaled up for mass production with several industrial applications such as heat and sound insulation, oil spill cleaning.

**Keywords:** Aerogel; Paper Waste; Fabric Waste; Plastic Waste; Cellulose Fiber; Cotton Fiber; Polyethylene Terephthalate Fiber; Heat Insulation; Sound Insulation; Oil Spill Cleaning

## Fabrication Methods and Morphologies of Aerogels from Wastes

### Cellulose Aerogels from Paper Waste

Large amounts of paper waste are constantly generated worldwide. So the paper waste should be recycled to preserve forests and to reduce pollution from landfills and incineration [1,2]. Through separation processes, the paper waste can be converted to recycled cellulose fibers [3-6]. The recycled cellulose fibers from paper waste (0.075 - 0.300 g) and kymene (5 -20  $\mu$ l) are first dispersed in 30 ml of DI water by sonicating the mixtures for 10 min. The suspensions are then placed in a refrigerator at -18 °C for more than 12 h to allow the gelation. The cellulose aerogels are obtained by freeze-drying the obtained gels at -98 °C for two days using a freeze dryer. Thereafter, the cellulose aerogels are further cured at 120 °C for another 3 h to completely cross-link the kymene molecules. After being freeze-dried and coated with MTMS via chemical vapor deposition mentioned above, the recycled cellulose aerogels exhibit ultra-flexibility, high porosity, super-hydrophobicity, and outstanding oil absorption capability. Their macropores are possibly caused by the larger size of the recycled cellulose fibers obtained from the paper waste [6]. The water contact angles are measured on both external surface and cross-section of the MTMS-coated cellulose aerogels. Large contact angles of 153.5° and 150.8°, respectively, are obtained, thus proving that the hydrophobic coating successfully covers the whole aerogel network.

A cost-effective method making the recycled cellulose aerogels by using kymene cross-linker is more advanced than using sodium hydroxide and urea [7-9]. Kymene can strengthen the cellulose aerogels by providing a protection mechanism and reinforcement mechanism [5,6]. It can significantly reduce the toxicity of the initial materials, and the entire synthesis duration down to three days, up to four times less than other reported methods. The advanced fabrication methods and excellent properties of the cellulose-based aerogels can help to solve environmental problems such as oil-spill cleaning and heat loss of buildings. This can greatly save energy and reduce emissions of greenhouse gases.

### Cotton Aerogels from Textile Waste

In 2010, Americans discarded 13.1 million tons of textiles. Only 15 percent was reclaimed for recycling, while more than 11 million tons of textiles were dumped and clogged up approximately 126 million cubic yards of landfill space. Decomposing clothing can release methane, a harmful greenhouse gas and a significant contributor to global warming. Dyes and chemicals in fabric and other components of clothing can leach into the soil and contaminate its surface and groundwater. Most of this material is completely reusable or recyclable, but only if it is proactively collected. There is a massive demand for second-hand clothing in developing countries and other materials can be recycled into

advanced materials such as cotton aerogels for several industrial applications.

The cotton pads are cut into small strands (0.5 × 4 cm) and then are mixed in 200 ml deionized water. Then, the mixed dispersion is homogenized using a juice blender for 15 min. Afterwards 66.6 μL PAE solution is added into the above dispersion, which later goes through a sonication process at 140 W for 5 min. The homogenized dispersion is frozen at -18 °C for 24 h and then dried in vacuum at -98 °C for 96 h to obtain the monolith aerogels. Both cotton fibers and cellulose fibers are squeezed due to the volume expansion from water to ice. Finally, these aerogels are cured at 120 °C for another 3 h. The cotton aerogels are prepared using the same method without the addition of the cellulose fibers [10, 11]. In order to obtain the hydrophobic aerogels for oil absorption, all obtained aerogels are coated with MTMS at 70 °C for 12 h to undergo a silanization process. The silanization process is carried out based on the reaction between hydroxyl groups and alkoxy groups in MTMS [12]. The cotton aerogels present macropores with the diameter larger than 50 μm. As for the cotton aerogels, the increase of the cotton fiber concentration causes more packed structure. A large contact angle over 130° of the cotton aerogels coated with MTMS indicates their hydrophobic properties. The contact angle slightly decreases with the increase of the cotton fiber concentration. This occurs because the homogenous silanization process may be less effective [10-12].

### Polyethylene terephthalate Aerogels from Plastic Bottle Waste

In the 1980s, polyethylene terephthalate (PET) is used popularly for the production of disposable soft drink bottles. Most plastic wastes are toxic and create a direct hazard to the environment. They are seen as noxious materials due to their substantial fraction by volume in the waste stream. Due to their high resistance against the atmospheric and biological agents, the plastic wastes are considered to be non-biodegradable [13]. Most plastics including PET can take up to hundreds of years to decompose, and usually end up in oceans and landfills. If current production and waste management trends continue, an estimated 12,000 million tons of plastic wastes will be in landfills or in the natural environment by 2050 [14]. The plastic wastes, known as marine debris in the ocean, can potentially lead to entanglement and ingestion by marine life and effectively kill the marine life through starvation and debilitation [15]. Landfills wise, the plastic wastes within landfills may not pose a significant risk to groundwater contamination if managed correctly. However, such worrying trends can eventually lead to land scarcity as well as wastage of land area that can be allocated for other useful purposes, and also potentially destroy the aesthetics of the city. In order to overcome the above-mentioned issues, the aerogel development for thermal and acoustic insulation applications can be a sustainable and environmental-friendly solution by

utilizing the recycled PET (rPET) fibers from the PET plastic bottles.

In order to make the plastic aerogels, initially the recycled PET (rPET) fibers are immersed with aqueous NaOH and then heated at 80 °C for 1 hour to produce carboxyl and hydroxyl groups on their surface. The PET fibers are washed thoroughly with DI water and then added into the mixture of polyvinyl alcohol (PVA), glutaraldehyde (GA) and DI water. The GA cross-linker with the addition of HCl (controlled at pH ~ 3) can improve the interaction between the rPET fibers and PVA. The GA cross-linker with the addition of HCl can improve the interaction between the rPET fibers and PVA. HCl solution can be added in the above mixture to accelerate the crosslinking reaction. The obtained mixture is sonicated for 30 minutes and then heated in the oven for 3 hours at 80 °C for the cross-linking reaction and then frozen in the fridge. The frozen suspension is freeze-dried for 2 days to make the rPET aerogel. The hydrophilic rPET aerogels can be converted into the superhydrophobic ones by coating MTMS on their highly porous network structures in the oven for 24 hours at 70 °C [16]. The rPET aerogels are formed through the hydrogen - and ester-bonds between PVA and the rPET fibers [17,18] and the acetal bridges from GA cross-linkers [19].

The porous structures of the rPET aerogels having different rPET fiber contents, fiber deniers and fiber lengths are shown in Figure 3.2. All of the rPET aerogels have the macropores of over 50 nm, the high porosities of 98.3 - 99.5%, and ultra-low densities of 7 - 26 mg/cm<sup>3</sup>. When the rPET fiber content increases, the aerogel density increases, and the porosity decreases. So the pore sizes and the number of the pores within the aerogel structures reduce. Adding more PVA content can reduce the porosity and the pore sizes of the rPET aerogel as well. If the rPET fiber deniers increase, the greater diameters of the rPET fibers result in the higher porosities and larger pore sizes of the rPET aerogels. After coating with MTMS, the MTMS-coated rPET aerogels become super-hydrophobic and have the water contact angles of 120.7° - 149.8° Their super-hydrophobicity is from the methyl groups of MTMS replacing the hydroxyl groups on their surface [20-21].

### Applications of Aerogels from Wastes

#### Oil Absorption Application

A 5w40 motor oil is used to investigate the oil absorption capabilities of the recycled cellulose aerogels. The cellulose aerogel is loaded on the top of the mixture and quickly absorbs the motor oil within 7 min. The maximum absorption capacity of 95 g/g is achieved with the 0.25 wt.% cellulose aerogel due to its lowest density and highest porosity (99.4 %). The significant enhancement of the absorption capacity may be largely ascribed to the reduced densities and increased porosities of the cellulose aerogels. Without using urea and hydroxide residuals, the densities of the cellulose aerogels cross-linked by the kymene

are consequently lowered with their weights. The cotton aerogel with a low concentration of 0.25 wt.% has the highest absorption capacity over 100 g/g, much larger than those of the commercial sorbents. The increase of the cotton fiber concentration from 0.25 to 0.75 wt.% leads to the decrease of absorption capability of the cotton aerogels due to their raised density and lower porosity. The 0.5 wt.% cotton aerogel has the highest absorption speed.

The rPET aerogel with 0.05 wt.% PVA has the maximum absorption capacity of 70.2 g/g due to its lowest density of 0.011 g/cm<sup>3</sup> and highest porosity of 99.2 %. When the rPET fiber concentration increases, the rPET aerogel structure is more packed and its oil absorption capacity is decreased. The absorption capacities of the MTMS-coated rPET aerogels are significantly better compared to the commercial polypropylene sorbents. Their enhanced absorption capacities may be attributed to the lower density and the higher porosity. The pseudo-second-order model is better in accurately predicting the oil absorption behavior of the rPET aerogels. The same 0.5 wt.%, the rPET aerogels can absorb the 5w50 motor oil approx. 3.0 times and 1.5 times faster than cellulose aerogels [4] and cotton aerogels [10], respectively.

### Heat and Sound Insulation Applications

The cellulose aerogels and the cotton aerogels have the low thermal conductivities (0.03 - 0.04 W/m·K), less toxic emissions and use less harmful chemicals during the fabrication [4-6,22]. The cellulose aerogels have the lowest density of 0.04 g/cm<sup>3</sup>, which is lighter than other common insulation materials [6,22]. The cellulose aerogel is used to design an insulated jacket for the military canteens to prolong ice slurry for active military personnel in training or operation [6,22]. It is reported that that the vacuum flask yields the best thermal insulating results with the least temperature increase after 4 h, followed by the military canteen that is wrapped by the insulated jacket. The temperature of the content inside both bottles is approx. 0.1 - 0.2 °C after 4 h, which proves that they can maintain the ice slurry inside chilled. The FLOE bottle and the military canteen without the insulated jacket cannot keep the ice slurry inside after 3 h and 1 h, respectively. The vacuum flask poses some concerns in terms of heavier weight load and higher costs despite their best result. It is not ideal to replace the current military canteens that are used by the soldiers with the vacuum flask bottles. So the thermal jacket using the cellulose aerogels is successfully designed and that the simple but effective engineering solution for the heat insulation of the military canteens is validated [23].

The rPET aerogels exhibit ultra-low the thermal conductivities of 0.035 - 0.038 W/m·K due to their highly porous network. At the ambient temperature and pressure conditions, the trapped air within the highly porous aerogels is a substantial contributor to their low thermal conductivities [24]. The range of the fiber concentrations, deniers and lengths may have no significant effect on the thermal conductivities of the rPET

aerogels. The thermal conductivities (0.035 - 0.038 W/m·K) of the rPET aerogels are lower than those of the silica aerogels (0.036 - 0.417 W/m·K) [25], silica-cellulose aerogels (0.039 - 0.041 W/m·K) [26], significantly lower than the rPET fibers (0.150 W/m·K) [27]. The thermal conductivities of the rPET aerogels are highly competitive to recycled cellulose aerogels (0.029 - 0.032 W/m·K) [28], conventional thermal insulation materials like polyurethane (0.020 - 0.040 W/m·K), mineral wool (0.035 W/m·K) and polystyrene (0.034 W/m·K) [29,30].

For the same thickness of 30 mm, the absorption coefficient of the rPET aerogels increases with increasing the rPET fiber concentration. An increase in the PET fiber concentration can cause more fiber-to-fiber contact, tortuosity as well as fiber entanglement [31]. Consequently, there are greater energy losses to the propagated acoustic waves due to increasing surface friction [32] and internal reflections [33]. As a result, the sound absorption is enhanced. The 2.0 wt.% rPET aerogels demonstrate the best acoustic absorption results across the entire range of frequency. Compared to the commercial Basmel material, the 2.0 wt.% rPET aerogel shows the noise reduction coefficient of 0.46 and can perform 20 % better at frequencies of 2000 - 2500 Hz. For the same volume and density, more amounts of the used rPET fibers with the shorter length can potentially lead to a more tortuous path and can increase the fiber-to-fiber contact areas within the fiber matrix. Thus greater energy losses through friction can lead to better acoustic absorption.

### Conclusion

Eco-aerogels from the paper waste, the textile waste, and the plastic bottle waste are developed successfully. The aerogels generated from both fabrication methods demonstrate stable hydrophobic properties over at least six months. The method is eco-friendlier and cost effective by using fewer toxic solvents and the freeze-drying process. For oil spill cleaning, heat and sound applications, the performance of the eco-aerogels is much better than commercial materials. They can be also used for other potential applications such as medical devices, personal care products, and packaging etc. However, the fabrication cost and time of these aerogels need to be further reduced for industrial scale-up.

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