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Research paper

Effect of Silane Concentration on the Physical and Mechanical Properties of Bacterial Cellulose Silylated Aerogels

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Abstract

Bacterial cellulose (BC) produced by Acetobacter *xylinum* shows unique physicochemical properties. In this research, silane has been used as coupling agent to coat bacterial cellulose to change the properties of BC from hydrophilic to hydrophobic. Different silane concentrations (0%, 5%, 10% and 15%) were applied to determine the physical and mechanical properties of BC-silylated aerogels. Supercritical fluid extraction (SFE) process was used to get the 3D shape of BC aerogels since the separation of components using carbon dioxide as supercritical fluids occur faster than common extraction. BC membrane was characterized by FESEM and it is shown that porosity of BC membrane was affected with silane concentration. For physical testing, silane concentration has no effect on porosity of the BC aerogels while water absorption and thickness swelling showed that BC membrane coated with 15% silane concentration can be used for water or gas filtration.

Keywords: *Acetobacter xylinum*; *Bacterial Cellulose; silylated aerogels; supercritical fluid extractions; mechanical properties*

1. Introduction

Bacterial cellulose (BC) is another renewable source as it features some outstanding properties and shows higher average molecular weight, chemical purify, fiber strength and degree of hydratization of BC compared to plant cellulose [1]. BC does not consist of hemicellulose and lignin like plant cellulose. Removal of the hemicellulose and lignin are quite difficult, and it will damage the structure of cellulose during the process. These unique properties resulting from the ultrafine structure made by BC made it suitable to be used in textile, food industries, as biomaterial for cosmetic and medicine. The extraordinary structure of BC has been extensively analyzed and characterized by many researchers. It is shown that ultrafine structure of BC can be utilized in multitude application and it resembles collagen network that makes it very efficient for various biomaterial application.

BC can be formed as aerogels which are highly porous, very lightweight materials that exhibit several interesting properties such as extremely low thermal conductivity, large specific surface area and extensive in sound propagation [2]. Aerogels have been mainly investigated as filter materials, carrier for catalysts, storage media for gases especially for fuel cells and heat and sound insulators due to its high porosity and readily accessible interconnected open pore system [3]. Despite its remarkable properties, BC also can be improved by coating it with silane. The silylated BC is hydrophobic so that, it is efficient for water filtration as water can pass through it and not absorb it. The hydrophobic of aerogel is produced in the presence of silane as a coupling agent. The presence of coupling agent leads to chemical modification which is susceptible to react with the fibers and the matrix constitutes a particularly astute way of controlling the quantity and the nature of the groups present at their surface. Moreover, the best mechanical properties for the composite can be achieved by creating covalent bridges between the fibres and the matrix.

To the best of our knowledge, very few studies have been reported which involved only the interaction between silane-based reagents and cellulose and the incidence of this treatment on the mechanical properties of the ensuing composites have been published to date. Because these investigations are highly focused on the results and did not focus on the actual interactions between cellulose and the only silane used, we decided to undertake a more thorough study aimed at unravelling these features. The silanes considered here bear the general formula R-Si-(X)3, where X is an oxyalkyl group and R an organic moiety, which is chosen as a function of the nature of the matrix. The industrial development of these products is mostly due to their application as coupling agent between glass fibres and polymer matrixes and has led to a wide diversity of structures and properties. Therefore, it seemed appropriate to test them on cellulosic fibres, which bear the same potentially reactive groups, i.e., OH functions.

In this paper, the aim is to determine the best concentration of silane by using the supercritical fluid extraction (SFE) method of silylated aerogels used for water filtration. SFE method is used due to its performance and time saving of processing rather than the common vacuum bagging system.

Therefore, the best concentration of silane by using the SFE method is needed to improve the physical and mechanical of silylated aerogel. The effectiveness in water filtration requires high strength to maintain water permeability. Thus, BC is found very suitable and remarkable mechanical properties with its low cost, biocompatibility and sustainable source which have high potential as a component in water filtration membranes.

2. Materials and Methods

2.1. Materials

Inoculum of Acetobacter *xylinum* (Bacterial Cellulose) was used in this experiment. Silane with different concentration (0%, 5%, 10% and 15%) was obtained from Merck, Malaysia. All the other reagents were of analytical grade and were used without further purification.

2.2. Preparation of Bacterial Cellulose

Acetobacter *xylinum* were cultured on the nutrient medium in a laboratory refrigerator. The other chemicals were purchased from suppliers. The bacterial was cultured in the nutrient medium which was composed of 80g of sugar, 5g of Ammonium Sulphate (NH_2SO_4) and 1000ml of coconut water. The cell was pre-cultured in conical flask containing 200ml of nutrient medium and 10% of inoculum. The coconut water was filtered first to remove all the impurities before adding into the nutrient medium. The nutrient medium then was put into autoclave for 15min at 100̊ C. After the autoclave process, the nutrient medium was cooled down at room temperature. Acetic acid was added into the solution until pH 4.5 is achieved. Lastly, 10% of inoculum was transferred into clean containers that contain 200ml nutrient medium and was incubated in bench top orbital shaker for 72 hours at 150 rpm. The inoculum was carefully cultured in nutrient medium to avoid contamination with other bacterial. Then, the nutrient medium was cooled down statically at room temperature for 7 days. BC pellicle was produced in the form of fabrication layer of cellulose. The pellicle was dipped into 2.5% of NaOH to eliminate the cells and component of culture medium at room temperature for 24 hours. The pellicle then washed with distilled water repeatedly until the pH turn to 7.

2.3. Supercritical Fluid Extraction Process

This method of drying was performed on a supercritical fluid formulation apparatus SF1 (Separex, France). Hydrogel were placed on a stack which was loaded into the preheated 500 ml autoclave (40°C). The autoclave was pressurized to 10 MPa. After reaching the final pressure, the outlet valve was opened, and the autoclave was flushed with pure CO2 for 1 hour at a flow rate of 2.5 kg h-1. Ethanol was separated in a cyclone separator. After the drying period, the autoclave was depressurized slowly, and the dry aerogels were removed.

2.4. Bacterial cellulose coated with Silane

Silylated BC was coated with different concentrations of silane (0%, 5%, 10%, and 15%). The silylated sample test was immersed in the containers that have different silane concentration at room temperature for 24 hours. Then, the sample was allowed to undergo air drying for 24 hours before testing.

2.5. Characterization

In this study, there were several methods of characterization that were used to test the Silylated BC such as porosity, water absorption, thickness and swelling, tensile strength and Field Emission Scanning Electron Microscopy (FESEM).

2.5.1. Porosity

Porosity of BC was measured by using method described by Shamary et al. [4]. Dry bacterial cellulose was soaked in deionizer water for 12 hours at room temperature and then it was weighed in water. The weight was measured by immersing the sample in a device which suspend the sample in water [5]. The porosity of BC sample was calculated by using the equation below:

Porosity % = (wet weight – dry weight) / (wet weight – weight in water) x 100%

2.5.2. Water absorption

BC membrane that was coated with different silane concentrations were tested for its water absorption. The specimens were cut into 2 cm x 1 cm, then weighed on a weighing balance and recorded as initial weight. After that, the specimens were immersed into distilled water for 24 hours in room temperature. Finally, after 24 hours, the specimens are weighed again and recorded as the final weight. Water absorption of the samples were calculated using the formula below:

(Final weight (g)-Initial weight (g))/(Initial weight(g))x100% (1)

2.5.3. Thickness and swelling

The swelling percentage was determined by modifying the method used by other researchers. The sample was cut into size 2 cm length x 1 cm width. The prepared oven dried sample (OD), tray dried sample (TD) and freeze-dried sample (FD) was weighed using Electron balance model AA-200. Then, the sample was soaked in distilled water for 48 hours. The percentage of swelling was calculated as follows:

Swelling (%) = (ws-wi)/wi x 100 (2)

where Wi was the initial weight of dried sample and Ws was the weight of sample after 48 hours soaking in water.

2.5.4. Tensile strength

The tensile test was performed by using LRX, LLOYD Instruments, United Kingdom with maximum load of 2.5 kN. This test was conducted by using ASTM D 882 for thin film less than 1.0 mm in the condition of room temperature. The specimen was attached to the extension indicator and the speed of testing was set to 10 mm/min. The load-extension curve was recorded to obtain the tensile strength, elongation at break and Young's modulus.

2.5.5. Field Emission Scanning Electron Microscopy (FESEM)

Field Emission Scanning Electron Mircrosopy (FESEM) was used to determine the morphology of the surface and cross section of the silylated aerogels. BC sample was sputter coated with gold and observed under a Zeiss Supra-40VP (FESEM) at accelerating voltage of 5kV and 10K and 25K time's magnification.

3. Results and Discussion

3.1. Porosity

Figure 1 shows the porosity for the BC samples coated with different concentrations of silane. It shows that the porosity of pure BC recorded 91.83% while the lowest porosity is the one with 5% concentration of silane marked 88.59% porosity. The BC with 10% silane concentration showed 93.46% of porosity followed by BC samples coated with 15% silane concentration recorded 94.68%, highest porosity.

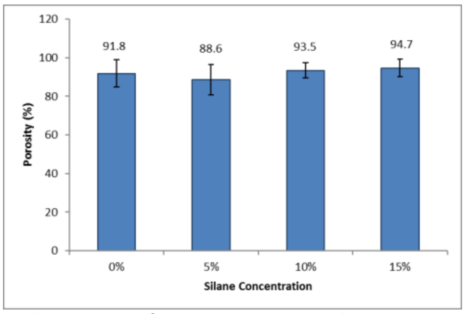


Fig. 1: The porosity of BC samples versus silane concentration.

Shamary et al. [4] found that the porosity of BC silylated aerogels was approximately 80% when used 2% to 8% inoculum. The porosity of BC silylated aerogels did not show much difference when the inoculum volume of BC used was 2%. The cell growth increased with increasing size of inoculums and that led to the increasing BC production. The large number production of micro fibrils often led to a compact structure and lower porosity for the BC membrane. Statistical analysis of one-way ANOVA also confirmed that p-value is 0.3609 which are larger than 0.05 and this infers, no significant correlation between porosity and concentration of silane. The porosity of BC aerogel samples was found not to be affected by silane concentration.

3.2. Water absorption

The water absorption of BC aerogel samples decreased from 0% to 15% of silane concentration (Fig. 2). The BC silylated aerogels absorbed less water compared to pure BC. Pure BC without silane coating recorded a higher value with 93.31%. The result for bacterial cellulose coated with 5%, 10% and 15% concentration of silane were 46.82%, 26.57% and 12.34%. Based on the graph, the amount of water absorption decreased as the concentration of silane increased.

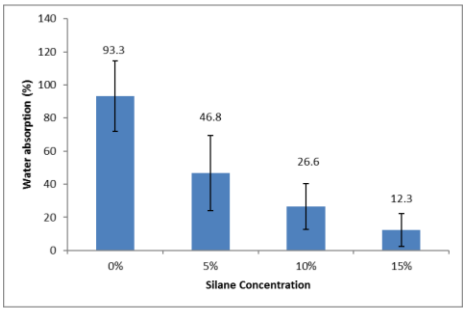


Fig. 2: Water absorption properties of BC aerogel coated with different concentrations of silane

Pure BC without silane marked the highest BC water absorption due to the hydrophilic nature of the pure BC. According to the Zhang et al. [6], water content can be up to 98% in BC pellicle, about 50 times the weight of the fibres. The capacity absorption of the gel is reduced as the concentration of silane increased because of the strong bonding between hydrophobic membrane and cellulose produced when silane was applied. BC silylated membrane coated with a stronger adhesion produced a greater hydrophobicity and less absorption [7].

3.3. Thickness and swelling

Fig. 3 shows that the thickness swelling of BC aerogels samples decreased from 0% to 5% concentration of silane. Pure BC without silane exhibited highest value of absorption recorded 122.86%. Meanwhile, BC silylated aerogels coated with silane of 5%, 10% and 15% concentration marked 23.69%, 17.07% and 7.25% respectively. The BC silylated aerogels swelled less compared to the pure BC.

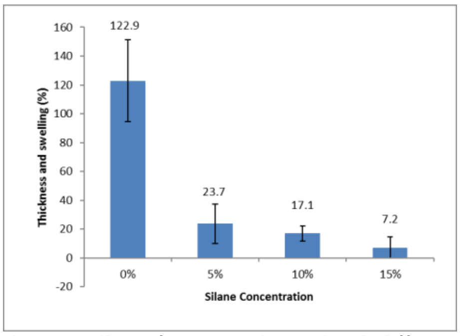


Fig. 3: Thickness swelling of BC aerogel coated with different concentration of silane.

Pure BC without silane coating demonstrated highest value of thickness swelling due to high water holding capacity and hydrophilic nature of pure BC. Whereas, according to Zakaria et al. [8], BC coated with silane possesses much compact structure with smaller porosity which reduced the water uptake and swelling. Therefore, as the concentration of silane increased, the thickness and swelling of the BC aerogels decreased. This is because of strong bonding between cellulose and hydrophobic membrane produced when the silane was applied. According to Gang Li et al. [9], BC silylated membrane gets masked with a stronger adhesion and produces a greater hydrophobicity and less absorption.

3.4. Tensile strength

Fig. 4 shows that the pure BC aerogels recorded lowest MOR which is 9.66 MPa. Based on different concentrations of silane coating, it was found that BC coated with 15% of silane exhibited higher strength recorded 12.27 MPa. The BC silylated aerogels showed that the strength and modulus of aerogels increase as the concentration of silane increase. It showed that the pure BC aerogels attained lowest MOE marked 153.34 MPa, with the highest recorded 284.15 MPa which for those made of 15% concentration of silane.

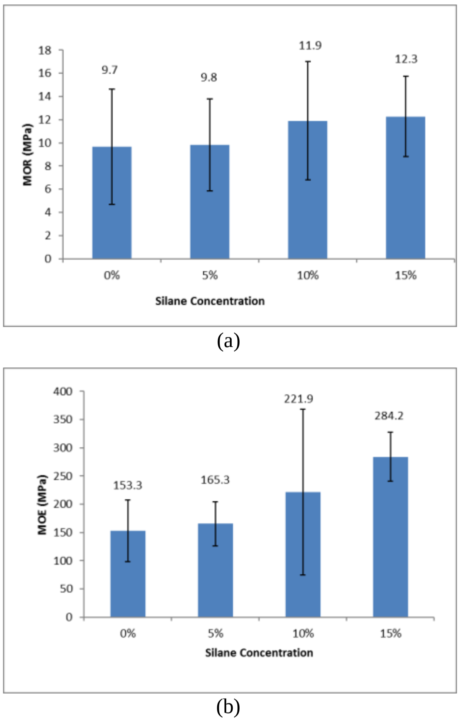


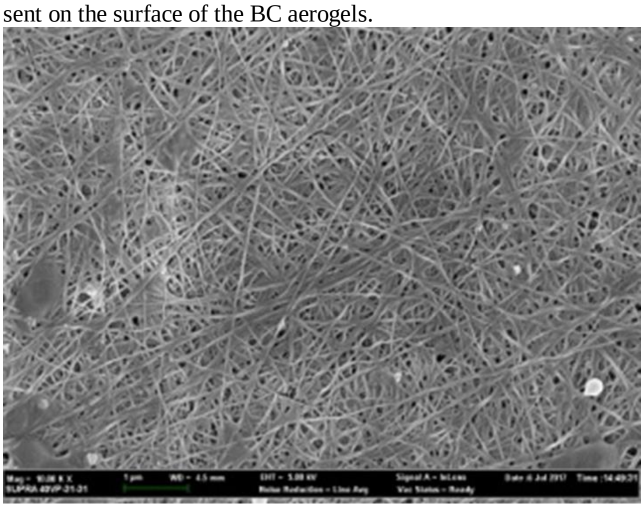
Fig. 4: (a) Tensile strength (MOR) and (b) Tensile modulus (MOE) of BC aerogels coated with different concentrations of silane

The pure BC aerogel without silane attained lowest value of MOE because of weak bonding of the cellulose compared to the cellulose that coated with silane. According to Andresen et al. [10], silane can form a durable bond between organic and inorganic materials. The tensile strength of aerogel is more sensitive to the

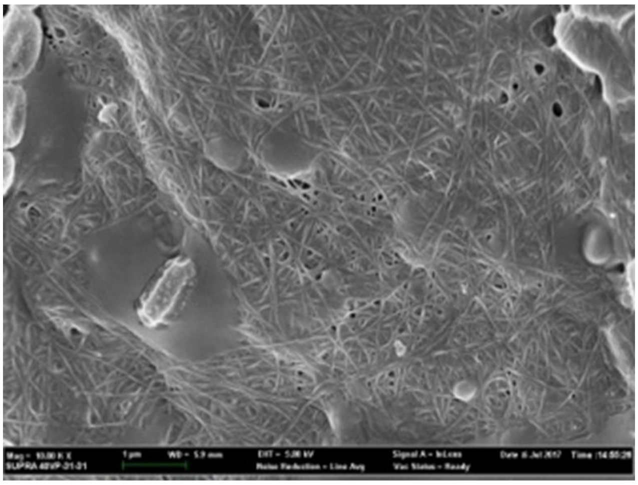
silane interfacial properties which can improve tensile strength and a strong interface of membrane [11].
Based on the factorial test using ANOVA, there are no significant different between silane concentration and the tensile strength and modulus for the BC silylated aerogels.

3.5. Field Emission Scanning Electron Microscopy (FESEM)

The morphology of BC silylated aerogels was examined by using FESEM. It is expected that the surface morphology of coated BC with silane and uncoated BC aerogels should be different in term of their porosity. Fig. 5 and 6 show the FESEM images of the pure BC aerogel without silane coating and those coated with 5%, 10% and 15% concentration of silane. There are porous structures present on the surface of the BC aerogels.



(a)



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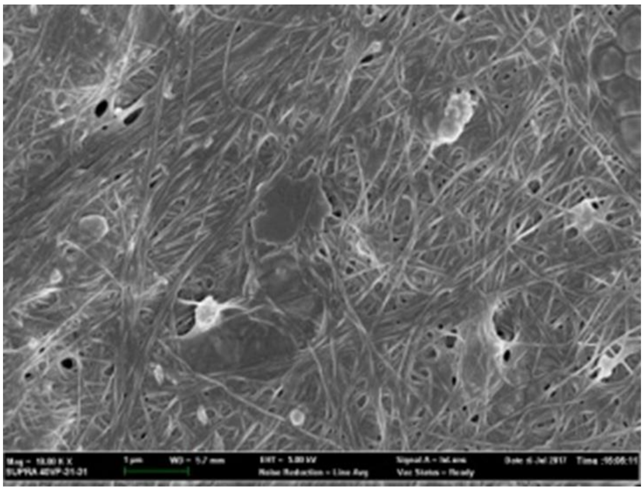

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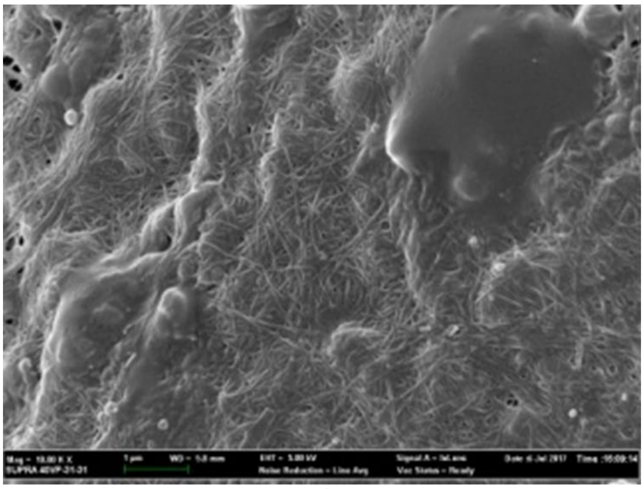
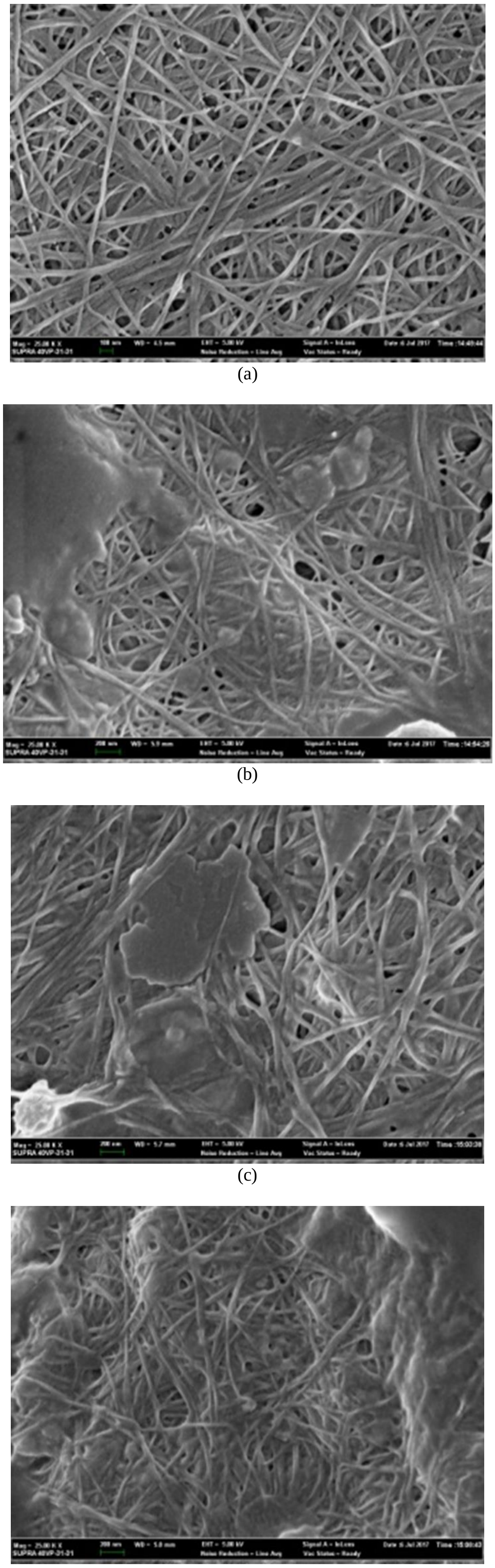


(d)

Fig 5: FESEM micrographs for (a) pure BC, (b) BC with 5% silane concentration , (c) BC with 10% silane concentration and (d) BC with 15% silane concentration at 10K magnification.

Fig. 5(a) and 6(a) show the image of pure BC and the porosity is the highest among the others. From Fig. 5(b) and 6(b) it indicates that there is still porous structure remains on the BC aerogels due to lower silane concentration. Fig. 5(c) and 6 (c) represent the image of BC coated with 10% concentration. It shows that the surface of BC silylated was lower in porosity compared to those as captured in Fig. 5(a,b) and 6(a,b). Fig. 5(d) and 6 (d) display the image of BC coated with 15% concentration of silane. The structure captured is compact and the porous structure is absent due to high concentration of silane coated on the surface of BC. Singha et al. [12] stated that the surface smoothness of BC samples has increased with the increase of silane concentration coating.



(a) (b) (c) (d)

Fig. 6: FESEM micrographs for (a) pure BC, (b) BC with 5% silane concentration, (c) BC with 10% silane concentration and (d) BC with 15% silane concentration at 25K magnification.

4. Conclusion

In this paper, the effect of different silane concentration on the physical and mechanical properties of BC silylated aerogels was studied as the water absorption and swelling capacity become lower as the concentration of silane increased due to strong bonding between hydrophobic membrane and cellulose produced when silane was applied. Based on different concentration of silane, BC coated with 15% of silane exhibited higher strength of 12.27 MPa. The BC silylated aerogels showed that the strength and modulus of the BC aerogels increased as the concentration of silane increased. It is deduced the surface of BC silylated was highly porous compared to pure BC and less silylated BC. The compact and porous structure is absent due to high concentration of silane coating on the surface of the BC aerogels.

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