

# Synthesis of Silica Aerogel with and without Glass Wool

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## Abstract

This work is focused on the simple method called the ambient pressure drying (APD) process to synthesize the silica aerogel with and without glass wool. The silica hydrogel samples were prepared from aqueous sodium silicate and HCl solution with and without glass wool in the plastic mould. The solidified hydrogel samples were passed through the solvent exchange process using ethanol and then modified using 20:30:10 ratios of TMCS (Trimethyl Monochlorosilane): n-hexane; ethanol) and the modified samples were named Type-2. The Type-2 were then dried at ambient temperature to get the final product. From the characterization, it was revealed that all Type-2 samples showed the presence of Si-C bond at peak 2964 and 844  $cm^{-1}$  to confirm the hydrophobic nature. Further, the contact angle of 146.7° was noted which is higher than 90° confirming the hydrophobic nature and electrical insulator property of the Type-2 samples was confirmed with a 4.1499 eV band gap energy value. The adsorption of organic solvents like ethanol was higher for the Type-2 sample i.e., 4.3036 g/g than 1 g of the activated carbon and also removed 15.881 ml of edible oil from 100 ml water containing 16 ml oil showing cleaning agent property. The thermal stability of the Type-2 sample was noted to be 300°C and the mechanical test was not feasible for the sample with glass wool due to cracks and improper binding between silica and glass wool. Hence, the sample made from commercially available sodium silicate resembled the properties of the silica aerogel but did not fully fulfil the criteria due to impurities in the source chemical.

## Keywords

Aerogel, Silica Aerogel, APD, TMCS

## 1. Introduction

The structure of aerogel is different from other solids in terms of density and appearance. It has the lowest density than other solids and possesses an airy appearance due to the presence of the nanopores. The appearance of the aerogel in terms of colour is yellowish or gleam-bluish and changes under the influence of sunlight and observing conditions [1]. Sol-gel process is followed to synthesize the aerogel from solid materials with porosity varying from 80 to 99.8 percent [2]. The different types of aerogels are synthesized since its invention such as starch, resorcinol formaldehyde, alginate type, etc. which are considered organic aerogel, and alumina, silica, titania, etc. are synthesized as inorganic aerogel [3]. Carbon-based aerogels are also made for different applications like supercapacitor electrodes [4], thermal insulation [5], exclusion of heavy metal ions from aqueous solution [6], etc.

The alkoxides such as sodium silicate and TMOS are the most common precursors for making aerogels through the sol-gel method. The basic steps of the sol-gel process for making aerogel are solution making, gelation, aging, drying, and densification [7]. The basic steps of the sol-gel process for making aerogel are solution making, gelation, aging, drying, and densification [7]. The silica aerogel was prepared by S.S Kistler in 1931 [8] where he replaced the water from hydrogel with alcohol (solvent exchange method) and then performed the supercritical drying to remove the alcohol which gave the final aerogel. As the Kistler process took more time to prepare the aerogel, Stanislaus Teichner introduced the new precursor TMOS (tetramethyorthosilicate) which eliminated the solvent

exchange method [9]. After that, different researchers worked on the synthesis of the aerogel. Till now, the three methods commonly used for making silica aerogel i.e., ambient pressure drying (APD) [10], supercritical drying (SCD) [11], and freeze-drying [12].

In global research endeavors, sodium silicate has been commonly employed as a precursor for silica aerogel synthesis. Notably, the weight ratio of  $Na_2O$  to  $SiO_2$  at around 1:3.3 [13] or 1:3.4 [14] has been widely adopted. These precursor formulations emphasize a higher proportion of silica ( $SiO_2$ ) by weight [15, 16, 17]. This composition facilitates the proper development of the three-dimensional silica network during the sol-gel process, ensuring an abundant yield of the wet-gel or hydrogel for subsequent synthesis stages. Sodium silicate with an elevated silica content proves advantageous for efficient aerogel production. So, there is lack of research on using the commercial grade sodium silicate for synthesis of aerogel under APD method.

Therefore, this endeavor constitutes a continuation of the preceding task, which involved the utilization of commercially available sodium silicate (commercial grade) from Nepal to produce aerogel using the APD method as a Type-1 sample. This was achieved by modifying the sample through the solution of TMCS, n-hexane, and EtOH (in a volumetric ratio of 4:30:1) and it was presented at the International Chemical Congress (ICC - 2023) within the poster presentation category [18]. In the current investigation, the modification solution's composition has been adjusted to 20:30:10, respectively, and further, commercial glass wool has been introduced as a reinforcement material for the aerogel structure.

## 2. Materials and Methods

### 2.1 Materials

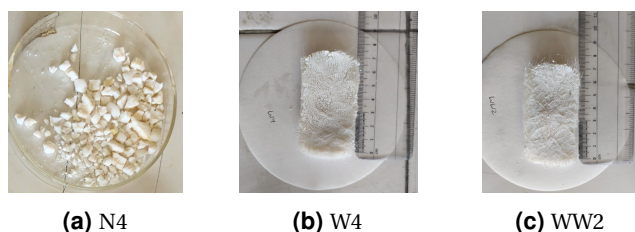
The sodium silicate (procured from NIKE Fine Chemical, India) and glass wool were bought from a local market of Nepal. Additional complementary chemicals, namely hydrochloric acid (HCl, with an acidimetric assay of 35-37 percent), ethanol (EtOH, with an assay of 99.9 percent by volume), n-hexane (with a CG assay of 99 percent), and benzene (with an assay of 99 percent), were obtained from Pulchowk Campus to facilitate the experiment. Moreover, surface modification was pursued by introducing TMCS (with a purity of 99.53 percent), sourced from Hebei Guanlang Biotechnology Co. Ltd, China.

### 2.2 Methods

The aqueous solution of the sodium silicate was made in 1:1 (w/v) with distilled water. Then, HCl of 1M concentration was added to that aqueous solution and quickly transferred the mixture into the plastic mould of size (7.3cm×3.7×1.2cm). The mixture was left for the gelation or solidification process to form a silica network forming hydro or wet-gel. To remove the sodium salt from the solidified gel, it was submerged and washed twice a day for three days. In the next stage, the water in the wet gel was replaced by EtOH by following the solvent exchange method. This process was completed in 4 cycles during which EtOH was replaced and the final sample is called alcogel. After that, the surface modification of the alcogel was done by submerging it into the solution of TMCS, n-hexane, and EtOH (20:30:10 by volume) in an airtight container for 24 hours at 50°C and it was repeated twice. The removal of the TMCS traces from the modified sample was necessary so it was immersed in n-hexane for 24 hours followed by washing and it was done three times. At last, the washed sample was dried in 4 stages; at ambient temperature for 24 hours followed by 50°C for 24 hours, 100°C for 2 hours, and 150°C for 1 hour. In this way, the two samples of the same composition were made N3 and N4. By following the same steps of N3 and N4, a sample with glass wool was prepared by placing the wool in the mould before pouring the mixture of aqueous solution and HCl into it. The W3 and W4 were made by adding 1 gm of glass wool whereas WW1 and WW2 were made by adding 1.5 gm of glass wool.

**Table 1:** Prepared samples

S.No.	Name	Glass Wool
1	N3 and N4	None
2	W3 and W4 (with glass wool)	1 gm
3	WW1 and WW2 (with glass wool)	1.5 gm



**Figure 1:** Prepared samples

In this experiment, the reason behind settling on a volumetric mixture of 20:30:10 was that the previous trials made with mixture 4:30:1 did not show the addition of  $-CH_3$  group in higher quantity in the final product with a low volume of TMCS and the presence of  $-OH$  group was high which was not desirable. So, the volume of TMCS was increased directly from 4 ml to 10 ml which also did not show an improvement so the final, volume of the TMCS was decided on 20 ml.

## 3. Analysis

### 3.1 Characterization

The existence of the functional groups in the prepared samples was determined by doing a Fourier Transform Infrared Spectrometer (FTIR) analysis. The main focus was to find the removal of the  $-OH$  group and the addition of the  $-CH_3$  group in the sample from TMCS to make it hydrophobic. The nature of the sample regarding the hydrophobic or hydrophilic was identified by performing the contact angle test. The presence of silica in the sample was examined by studying the absorbance of visible and UV light with the help of a UV-Vis spectrophotometer as well as band gap energy was measured to know the electrical conductivity. The XRD was also conducted to examine the nature of the sample either amorphous or crystalline as well as identify impurities.

### 3.2 Tests

Three organic solvents; ethanol, n-hexane, and benzene were utilized for the adsorption test by using samples; N3, and WW2. Similarly, the sample N3, WW1, and WW2 were used to test their adsorption capacity of edible oil (sunflower refined oil). The samples used in the organic solvent adsorption test were passed through the desorption test by keeping them in the oven at 30°C and noting the solvent removal at regular intervals. The thermal stability of the powder sample was approximately checked by placing it in a tube furnace in the presence of nitrogen at 300°C whereas the wool-reinforced sample was placed inside the oven at 200°C. After heating, the water was placed on both samples and the formation of droplets was checked to confirm the still presence of a hydrophobic nature.

## 4. Results

### 4.1 FTIR

From the FTIR of the N3 sample, the peak of the  $-OH$  group between the 3600 to 2700  $cm^{-1}$  frequency range is missing, and small stretching is seen at 1635  $cm^{-1}$  which indicates that the  $-OH$  group is removed in more quantity as compared to the Type-1 sample [18]. The presence of the Si-C bond is confirmed due to the peak at 2964  $cm^{-1}$  which lies between the frequency range of 2970-2010  $cm^{-1}$  [19] and 844  $cm^{-1}$  which lies between 865 to 750  $cm^{-1}$  [20]. Due to the removal of the  $-OH$  and the presence of Si-C, the N3 sample is confirmed to be hydrophobicity. Similarly, the other samples (N4, W3, W4, WW1, and WW2) also show same peaks as shown by the N3 samples.

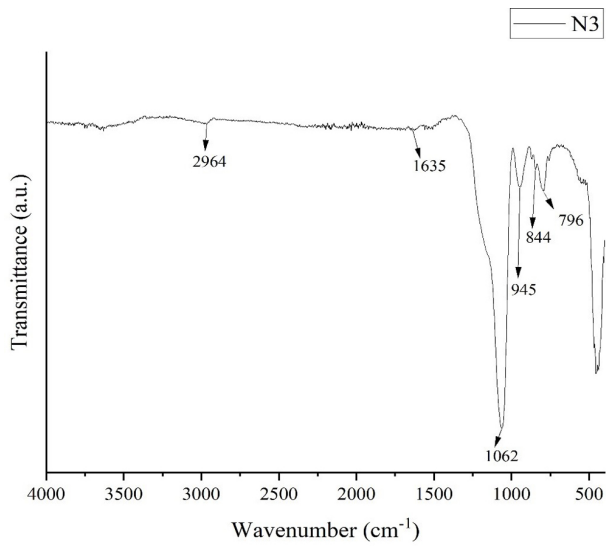


Figure 2: FTIR plot for N3

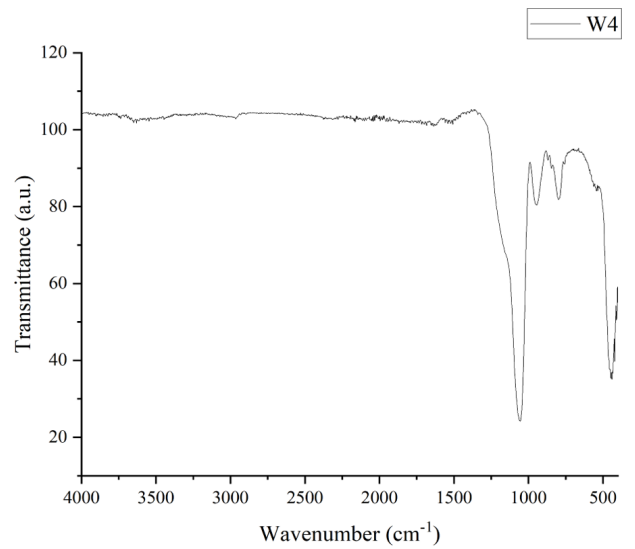


Figure 5: FTIR plot for W3

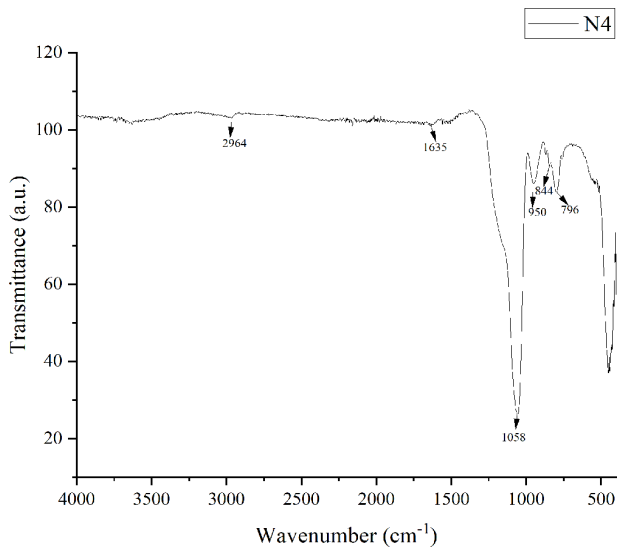


Figure 3: FTIR plot for N4

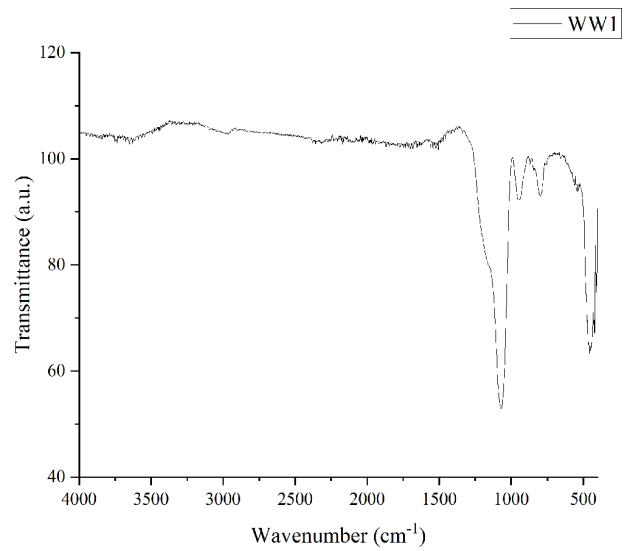


Figure 6: FTIR plot for WW1

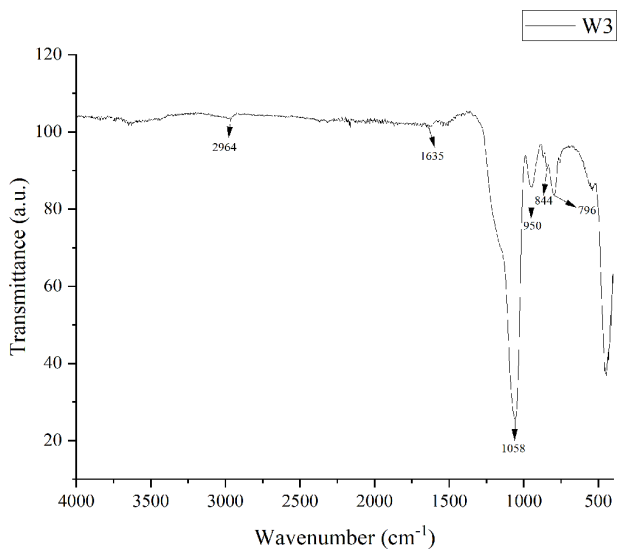


Figure 4: FTIR plot for W3

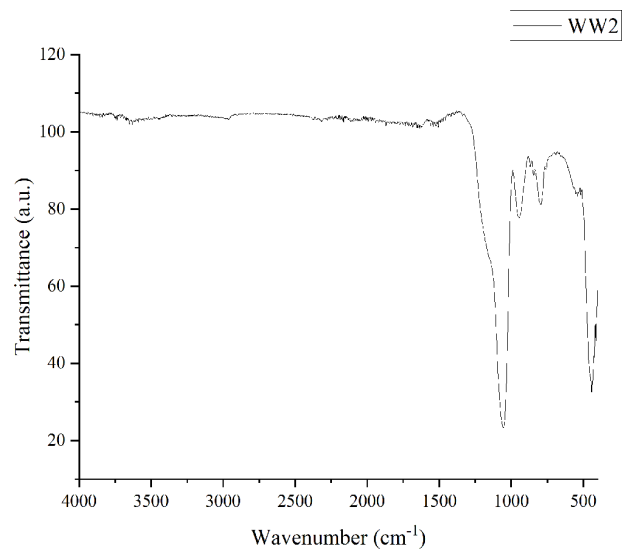


Figure 7: FTIR plot for WW2

So, all samples under Type-2 are hydrophobic in nature and contain less -OH group than Type-1 samples because the modification was twice at 50°C which shows better replacement of the -OH group with  $-CH_3$ .

### 4.2 XRD

The Type-2 samples; N3 and W3 were only examined under the XRD test where N3 represented the powder form samples and W3 represented the glass wool strengthened sample. Here, the broad peak is seen between 12.55° and 19.9° which indicates that N3 is amorphous in nature [21]. But the two sharp peaks are also noted at 31.58° and 45.32° which describes the presence of the NaCl having Miller indices (h k l) of (200) and (220) respectively. Next in W3, the broad peak is noted between 13.83° and 20.71° which explains the amorphous property. In this also, the sharp peaks are seen at 31.74°, 45.48°, 56.51°, and 75.37° which are due to the presence of NaCl with different crystal planes. The presence of NaCl in the final sample is not desired so it is considered as an impurity. Further, it confirms that NaCl was not completely washed out by doing normal washing of silica gel for several days using distilled water as mentioned in the previous section.

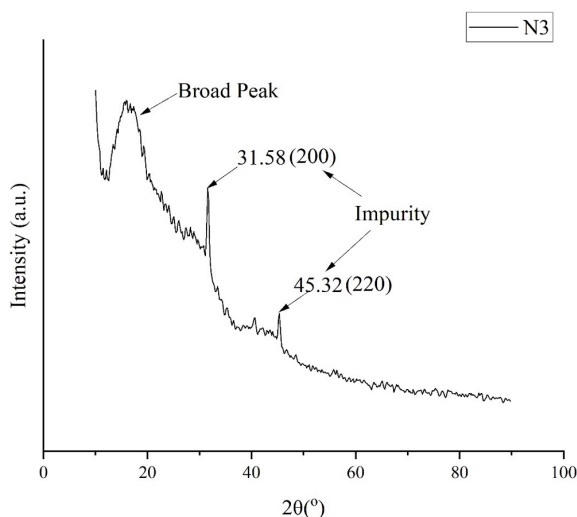


Figure 8: XRD plot of N3

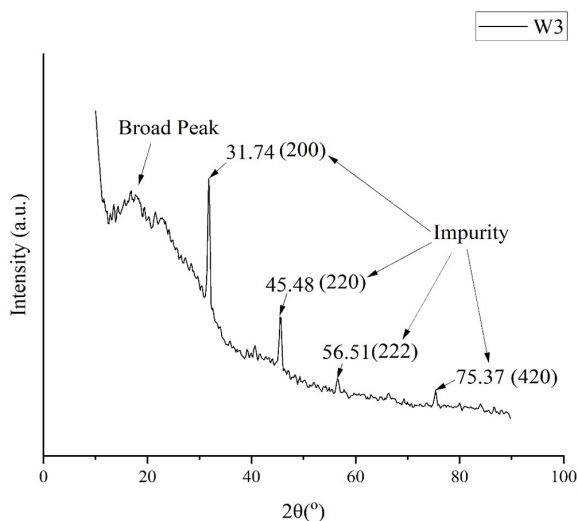


Figure 9: XRD plot of W3

### 4.3 Contact Angle

As from FTIR, all Type-2 samples show the properties of hydrophobic. Among them, the contact angle of water on the surface of sample N3 was only measured to confirm that claim. The contact angle is noted to be 146.7° which is close to the 150° [20] and greater than Type-1 samples (i.e., 124°) [18]. Thus, it is clear that the prepared sample is hydrophobic as the contact angle is higher than 90°.

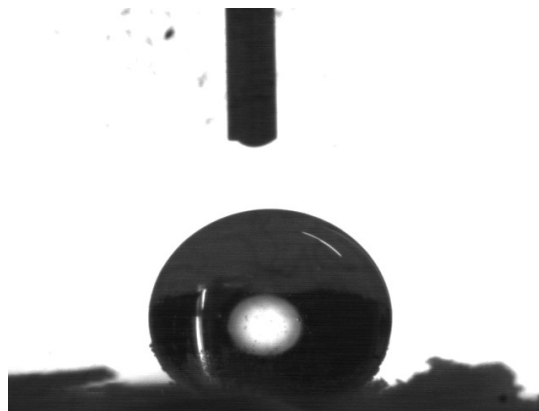


Figure 10: Contact angle measurement

### 4.4 Optical and Electrical Properties

N3 shows maximum transmittance in the visible light region and starts to decline under the wavelength below 400 nm which is the UV region. The behaviour of N3 under the UV and visible light spectrum is similar to the silica aerogel published paper [22]. Further, this shows that N3 is capable of absorbing UV light effectively. Using Kubelka Munk's relation, the  $(\alpha h\nu)^2$  was plotted against  $h\nu$ . Here, the bandgap energy of the N3 exhibits 4.1499 eV which is close to 4.25 eV [21]. Based on the band gap energy, the N3 shows the property of an electrical insulator. Here, the N3 sample was only tested as the representative of the Type 2 category because other remaining samples were synthesized in the same manner as N3 and testing the same with glass wool was difficult in the testing instrument so powder form sample N3 was tested.

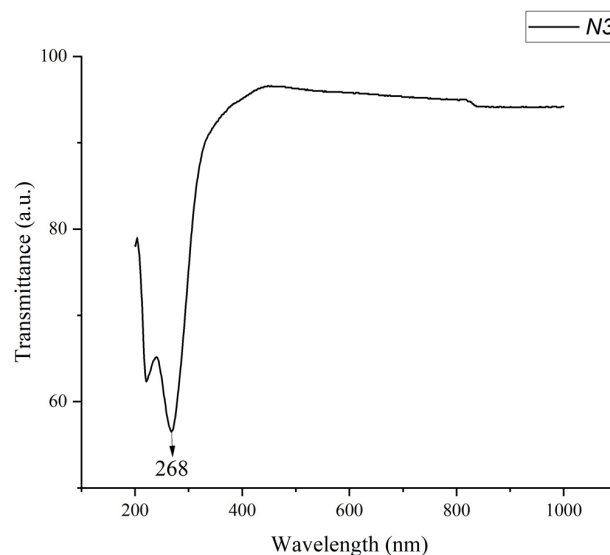


Figure 11: Optical property of N3



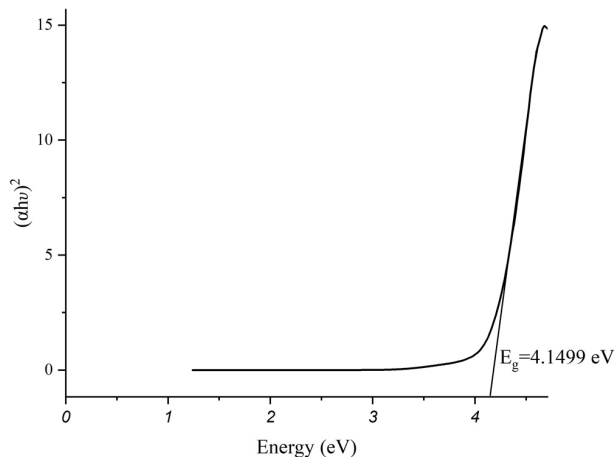


Figure 12: Bandgap of N3

4.5 Adsorption

First, the powder form sample was tested to find how much 1 g of the sample could adsorb organic solvent. The 1 g of N3 sample was able to adsorb 4.9421 g of benzene, 4.3036 g of ethanol, and 3.7831 g of n-hexane which is less than the standard value of 7 g/g adsorption capacity of the silica aerogel but found to be higher than activated carbon (i.e., less than 1 g/g for an organic solvent) [23] and Type-1 samples which were made in previous work [18]. From this test, it is also confirmed that the adsorption of the prepared sample increased with the higher surface tension of the solvent. Next, the sample with the glass wool was tested where 3.455 g of WW2 was soaked in ethanol, it adsorbed 15.1104 g ethanol which gives 4.3735 g of ethanol adsorbed by 1 g of WW2. By comparing both powder form (N3) and solid strengthen form (WW2), the adsorption of organic solvent was slightly higher in WW2 which indicated that the strengthened form was better in handling and adsorbing the organic solvent. Further, the comparison was made between powder form (N3) and strengthened form (WW1) for adsorbing the sunflower oil and

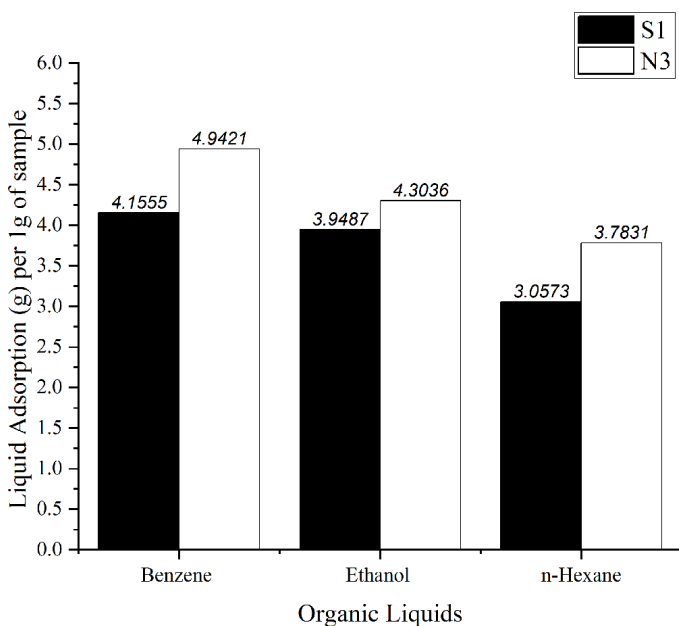


Figure 13: Adsorption comparison between Type-1(S1) [18] and Type-2 (N3)

found 1 g N3 and WW1 were able to adsorb 4.325 g and 6.3481 g of edible oil respectively. This also confirmed that the strengthened form was better than the powder form. In the next test for examining the water cleaning agent property, the mixture was prepared of oil and water (16:100 ml ratio) and 3.349 g of WW2 strengthen sample was able to remove 15.881 ml of edible oil after placing it in the solution for 1 minute.

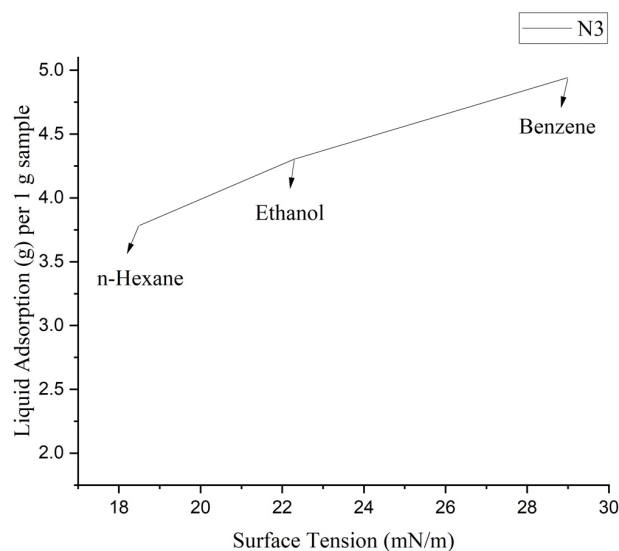


Figure 14: Adsorption variation with surface tension

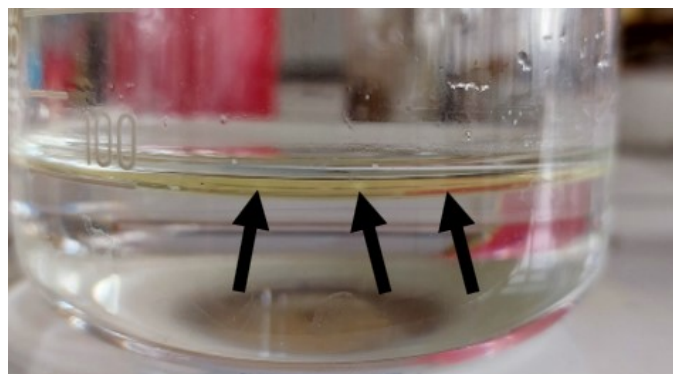


Figure 15: Edible oil and water mixture (16:100 ratio by vol)



Figure 16: Oil adsorbed by WW2

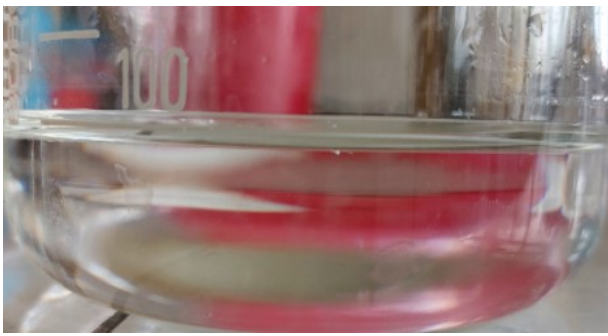


Figure 17: Oil removed from water

#### 4.6 Desorption

The soaked N3 sample was then dried in an oven at 30°C during which ethanol took 30 minutes to evaporate, benzene took 15 minutes, and n-hexane took 5 minutes. Likewise, the completely soaked WW2 took around 3 hours to lose all ethanol from it at the same temperature and it suggests that the prepared sample can be used again for adsorbing the solvent over and over.

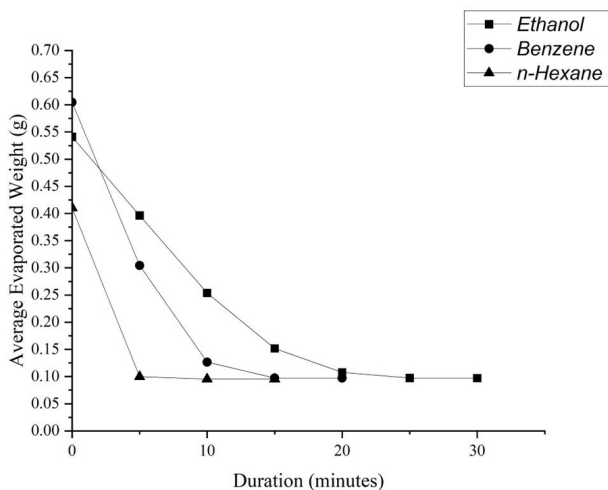


Figure 18: Average evaporate weight of soaked N3 over time

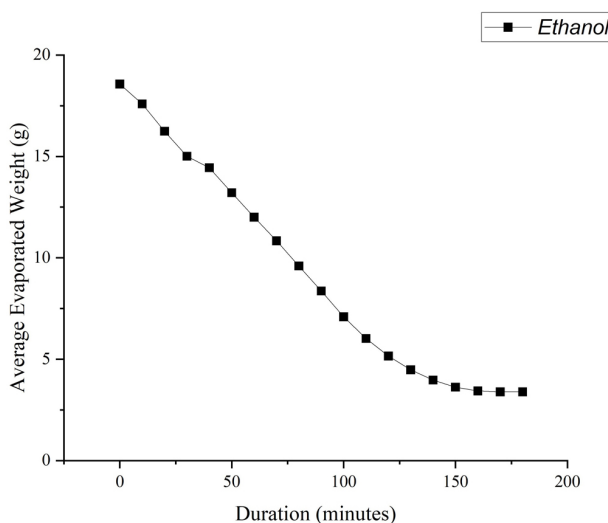


Figure 19: Average evaporate weight of soaked WW2 over time

#### 4.7 Thermal Stability

The thermal stability of the sample is mainly done using TG-DTA but due to unavailability, the approximate stability was performed using a box oven and tube furnace. The WW2 sample was placed in a box oven for 1 hr at 200°C. N3 powder was placed in a glass tube and heated at 300°C in the presence of inert gas i.e., Nitrogen for 1 hr. After heating, the hydrophobicity of both samples was checked by placing the water droplet on them. The image of the spherical water droplets on the surface of the sample is shown below which indicates that they still possess hydrophobic nature. Based on this, the maximum heat resistance of the Type-2 sample is considered to be 300°C but the heat resistance of the sample may be more because the critical temperature at which hydrophobicity will be lost is not determined.

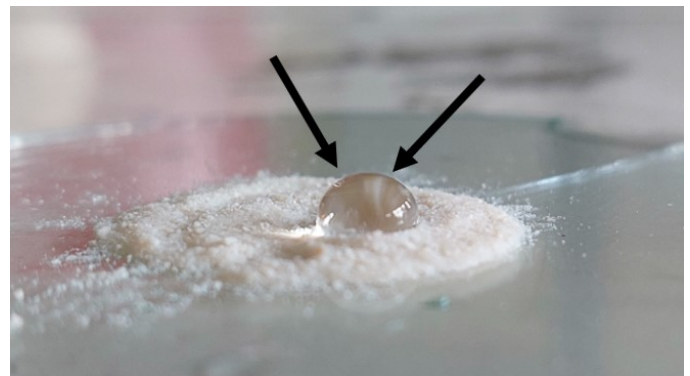


Figure 20: Visual inspection of N3 for hydrophobicity after heating at 300°C



Figure 21: Visual inspection of WW2 for hydrophobicity after heating at 200°C

#### 4.8 Mechanical Strength

Mechanical tests like flexural and tensile are not feasible for the samples prepared in this research. The two samples (N3 and N4) of Type-2 were in powder form. Further, samples W3, W4, WW1, and WW2 containing glass wool contains flaw like the silica network is not properly bonded with the glass wool as seen in the photos and the powder from the sample kept falling while handling them. The glass wool samples already have cracks and gaps which will not allow to give the exact load at which the initial crack will be seen. Although, it is seen that

the sample with glass wool was able to maintain the moulded structure as compared to the non-wool samples. So, it is clear that glass wool was able to provide the reinforcement structure upto some extent.



Figure 22: Presence of cracks

## 5. Conclusion

FTIR analysis confirmed the presence of Si-C at  $2964\text{ cm}^{-1}$  and  $844\text{ cm}^{-1}$  peak, indicating the replacement of -OH groups with  $-CH_3$  groups during modification, confirming the Type-2 samples are hydrophobic. XRD analysis of the powder sample (N3) showed broad peaks between  $12.55^\circ$  and  $19.9^\circ$  and the strengthened sample (W3) showed between  $13.83^\circ$  and  $20.71^\circ$  which confirmed the amorphous nature of the samples but also revealed the presence of NaCl impurities that did not wash away completely. The contact angle measurement of N3 showed  $146.7^\circ$  confirming the hydrophobic nature of the sample and also exhibited the band gap energy of  $4.1499\text{ eV}$  showing the electrical insulator property. From the adsorption test, 1 of the powder form N3 adsorbed  $4.9421\text{ g}$  of benzene,  $4.3036\text{ g}$  of ethanol, and  $3.7831\text{ g}$  of n-hexane which was higher than activated carbon.  $1\text{ g}$  of the strengthened sample WW2 adsorbed  $4.3735\text{ g}$  of ethanol which was higher than N3. The strengthened sample WW1 adsorbed more amount of sunflower oil ( $6.3481\text{ g/g}$ ) than powder form N3 ( $4.325\text{ g/g}$ ). Based on outcomes from the adsorption test, it is clear that the strengthened sample showed better adsorption of organic solvent and edible oil. The strengthened sample WW2 was able of removing  $15.881\text{ ml}$  of edible oil from water containing  $16\text{ ml}$  of oil in  $100\text{ ml}$  of water in  $1\text{ minute}$  which indicated the water cleaning agent for removing the edible oil efficiently. Desorption data suggested that increasing temperature could reduce the evaporation time of soaked organic solvent for reusing the samples. The powder sample (N3) remained stable up to  $300^\circ\text{C}$  in a tube furnace which suggested the Type-2 samples have thermal stability upto  $300^\circ\text{C}$ . The samples with glass wool (W3, W4, WW1, and WW2) exhibited better structural integrity due to the presence of glass wool in it. In summary, Type-2 samples exhibited properties resembling silica aerogel, with the study attributing inefficiencies in silica aerogel synthesis because of impurities in commercial sodium silicate and the presence of NaCl in the final product.

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