

Reinforcing Silicone with Hemp Fiber for Additive Manufacturing

Pantea Koushki^a, Tsz-Ho Kwok^{*a}, Lucas Hof^b, Rolf Wuthrich^a

^aDepartment of Mechanical, Industrial and Aerospace Engineering, Concordia University Montreal, Canada.

^bDepartment of Mechanical Engineering, École de technologie supérieure ÉTS Montreal, QC H3C 1K3.

Abstract

This study explores the 3D printability of a new material based on silicone and hemp fibers from renewable, sustainable and non-petroleum resources with the aim of enhancing mechanical properties of silicone. To improve composites printing technology, it's required to discover desired mixing composition. At first, to determine the proper amount of fibers, samples were fabricated by molding. Incorporation of fibers improved the mechanical properties of the silicone matrix. However, fiber distribution between matrix material adversely affected the printability of silicone due to the high viscosity. Therefore, behavior of the new engineered material was analyzed using rheological study to obtain a printable material. The composition containing 15 (wt.%) hemp fibers and 20 (wt.%) solvent with enhanced mechanical properties displayed desirable printability. Moreover, the mechanical properties of the 3D printed and molded samples were studied. The results revealed that 3D printed samples outperformed the molded counterparts. Finally, a simple gripper and a honeycomb structure were fabricated to demonstrate the application of the developed material.

Keywords: 3D Printing, Silicone, Hemp fibers, Enhanced mechanical properties.

1. Introduction

Additive manufacturing offers more advantages over conventional molding methods including the ability to create complex interior structures, different degrees of hardness which makes it appropriate to be utilized in different applications.

Although natural fibers provide many advantages such as low cost, lightweight, and biodegradability over glass fiber; they have some disadvantages like low impact strength, poor thermal stability, and high moisture absorption [1]. High moisture absorbing is the major disadvantages with natural fiber decreasing their compatibility with the non-polar elastomeric matrix leading to the poor fiber-matrix adhesion. This causes low mechanical properties which is one of the drawbacks with natural fibers. Therefore, many studies have been done on modifying fiber surface to enhance interfacial adhesion between filler particles and polymer macromolecules as well as their dispersion in the polymer matrix [2, 3]. Different chemical and physical fibers surface treatment have been used to improve fiber/matrix interfacial adhesion [4].

To improve composites printing technology, it's required to find the desired mixing composition. Moreover, sustainable materials are promising to be developed to reduce material cost and environmental impact [5]. This study investigates the effect of hemp fiber loading and hemp fiber surface treatment on tensile strength, reaction force, and 3D printability behavior of hemp fiber reinforced silicone (HFRS).

Therefore, hemp fibers were incorporated with silicone to synthesize eco-friendly and high-performance 3D printing material. However, integration of hemp fibers causes major dif-

ficulties in 3D printing. This work investigates the possibility to increase the printability of natural fiber reinforced paste-like material by modification of material viscosity. Rheological tests and physical validation were performed to determine desirable solvent and hemp fiber (wt.%) in silicone composition.

2. Methodology

The components from hemp fibers and silicone were 3D printed by extruding these compositions, in a form of a paste. Hemp fibers from low-cost and sustainable sources were chosen as reinforcement component of the composite. Silicone was selected as the binder material. The main focus of this study is to determine the desirable composition of HFRS to provide enhanced mechanical properties while maintaining 3D printability. Therefore, tensile strength tests were performed on different samples to evaluate the effect of fiber loading and surface treatment. The printability of the HFRS composites with different solvent concentration was evaluated by rheological evaluation and physical validation. In addition, the mechanical properties of 3D printed composites were compared to molded composites, prepared using the same composition

2.1. Materials

All of the materials employed in this work were obtained from commercial sources. Untreated hemp fibers were provided by Ontario Hemp Materials company. The length of the fibers used in this study was ranging from 0.2 to 0.4 mm. GE 100% silicone was used in this study. 3-Aminopropyltriethoxysilane (3-APS) was purchased from Gelest Inc.

*Corresponding author. Email: tszho.kwok@concordia.ca

2.2. Hemp fibers surface modification

Different chemical treatments are applied to enhance interface interaction between the reinforcement and matrix. Many efforts have been done to modify natural fiber surface such that the interfacial adhesion between fibers and matrix are improved. Different physical and chemical treatments for natural fibers including plasma treatment, heat treatment, coupling agents (silane treatment), and mercerization are discussed in literature [6]. The two most common methods used in the fabrication of natural-fibers-reinforced elastomers are mentioned below.

2.2.1. Alkali treatment

Hemp fibers were soaked in a sodium hydroxide (NaOH) solution of concentration 5% (w/v) for 2 hrs at room temperature. Fibers were further rinsed with distilled water containing acetic acid. This procedure was done two times until the pH of the rinse water reached 7, and the water no longer indicates any alkalinity. Then the fibers were air dried for two days. At the end, fibers were dried in an oven at 80 °C for 6 hrs. NaOH treatment provides rougher surface by removing impurities and noncellulosic parts which increase the adhesive nature of natural fibers.

2.2.2. Silane treatment

Following literature [7, 8], silane solution was made by hydrolyzing 5% (wt) silane 3-APS (weight percentage regarding the fiber) in a mixture of water and methanol (40:60 w/w). The pH of the solution was adjusted to 4 with acetic acid. The fibers were immersed in this solution for 3 hrs. Then, they were washed and air-dried for two days, and further dried in an oven at 80 °C for 12 hrs.

2.3. Composite manufacture

2.3.1. Preparation of molded composites

Modified fibers were distributed in the silicone matrix using a vacuum mixer ARV-200. The fibers were dispersed in the matrix by mechanical stirring for 2 mins. To determine the desirable composition of hemp fiber/silicone to provide enhanced mechanical properties, dog-bone shape samples with different composition were molded. To prepare the samples, a dog-bone shape ABS mold was prepared using 3D printer according to the ASTM D412-16 standard, which is the standard test method. The tensile samples were prepared for 10%, 15%, and 20% of hemp fiber reinforced silicone composite (composites were obtained with raw fibers, alkali treated fibers, and alkali/silane treated fibers). The silicone composites were cured by exposure to atmospheric moisture at room temperature for 15 hrs to form cross-linked molecular bonds in all composites.

2.3.2. Additively manufactured composites

After determining the desired composition, the printability of the silicone/hemp fiber composites was evaluated. For 3D printing HFRS the Discov3ry paste extruder was employed. This extruder can be easily added to any existing 3D printer. The Ultimaker line is our printer of choice as it's open source, well engineered, and reliable [9]. HFRS composite was loaded into

a syringe cartridge, and the Discov3ry Extruder system forced the paste through a feed tube. The material was injected through a nozzle and deposited on platform layer by layer. This study investigated the effect of different printing parameters such as viscosity, nozzle diameter, and print speed on 3D printing of hemp fiber reinforced silicone composites.

3. Silicone/hemp fiber composites characterization

To obtain the desired composition and obtain information about fiber/matrix interaction, tensile test, Fourier transform infrared spectrometry (FT-IR), and Scanning electron microscopy (SEM) were performed.

3.1. Mechanical Characterization

To determine the tensile properties of the products, a tensile test was performed using tensile test machine from Hoskin Scientific company equipped with a 5 kN load cell. The test was developed according to requirements of ASTM D412-06a which is the standard test methods for vulcanized rubber and elastomers-tension. The testing process was carried out at a crosshead speed of 150 mm/min with a gauge length of 40 mm, and the average values of tensile strength and modulus were recorded for all experimental samples of untreated, NaOH treated, and silane treated hemp fiber silicone composites.

3.2. Fourier transform infrared spectrometry (FT-IR)

Fourier transform infrared spectrometry (FT-IR) is an analytical method for investigating the structural identification or confirmation of identified or unidentified product. An infrared spectrum allows to easily discover the presence of significant functional groups. FT-IR study of the untreated, NaOH treated, and silane treated hemp fibers were done by using a FT-IR machine (Nicolet 6700 / Smart iTR) to determine the changes in functional groups on the fiber surface. All the spectra were recorded in a range of 4000cm^{-1} to 500cm^{-1} .

3.3. Scanning electron microscopy (SEM)

To analyze the fracture surface of HFRS composite and to visualize the effect of alkali and silane treatment, Scanning electron microscopy (SEM, Hitachi, S-3400N) was utilized. The voids, microstructures, and interfacial interaction of fiber and matrix were determined using SEM at voltage 5 KV, pressure 50 Pa, and at a focusing distance of 5 to 10 mm, for magnification of up to 1000X used in this study. In order to establish effective conductivity for examination, the fracture surfaces of tensile samples were gold sputtered .

4. Printability assessment method

Printable ink must provide a number of significant requirements to be processed with DIW 3D printing method. This study investigated the desirable condition to address printability issues.

The key printing parameters such as nozzle diameter, layer height, infill density, and print speed were determined by 3D

Table 1: 3D printing parameters for fabricating HFRS composite.

Material	Layer height (mm)	Infill density (%)	Print speed (mm/s)	Travel speed (mm/s)	Nozzle diameter (mm)
Silicone	0.5	100	8	120	1.54

Table 2: Optimized 3D printing parameters for fabricating silicone

Material	Layer height (mm)	Infill density (%)	Print speed (mm/s)	Travel speed (mm/s)	Nozzle diameter (mm)
Silicone	0.3	100	10	120	0.84

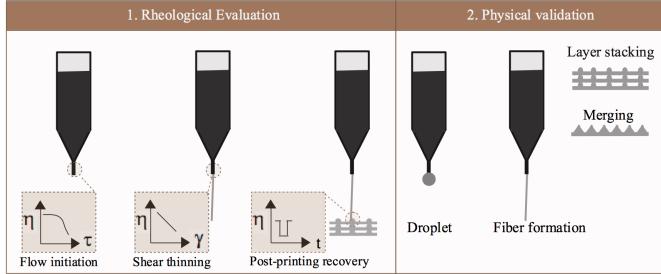


Figure 1: Schematic of the proposed research method to determine printability of the designed material.

printing and evaluating the dog-bone shape sample to acquire the best model quality. Table 1 demonstrates the best 3D printing parameters for fabricating HFRS composites.

This work proposed to characterize material printability (Figure 1).

4.1. Rheological evaluations

A MCR301 rheometer (Anton Paar) was employed to measure the rheological properties of the silicone and hemp fiber reinforced silicone. A 25 mm parallel plate with a measurement gap of 1 mm was used.

One of the goals in this study is to determine whether hemp fiber reinforced silicone could be fabricated using 3D extrusion printing. In order to understand the printability of silicone/hemp fiber, it is essential to monitor the rheological behavior of all experimental samples during processing. In this study, the following three rheological experiments at room temperature were carried out in order to find a solvent concentration which provides 3D printability without changing material characteristics: (1) For all experimental samples containing different solvent concentration, a shear stress ramp ranging from 0.01 to 100 Pa was applied, to determine the yield point, indicating the point at which the material first started to flow. (2) Rotational shear viscosity measurements were done in flow mode in a range of 0.01 to 100s^{-1} , to investigate the shear thinning properties of the materials. (3) To determine the materials recovery behavior after exposure to shear rates, the rotational recovery behavior were performed by applying a low shear rate of 0.01s^{-1} for 200s, following by high shear rate at 100s^{-1} for 100 s and then at the end a low shear rate of 0.01s^{-1} for 200

Table 3: K and N coefficients for all experimental samples.

Sample	K	N
Silicone	564.74	0.2842
Silicone/Hemp fiber	1526.2	0.2923
Silicone/Hemp fiber/20(wt.%) solvent	458.43	0.3126
Silicone/Hemp fiber/30(wt.%) solvent	51.351	0.3647
Silicone/Hemp fiber/40(wt.%) solvent	25.39	0.3872

s. All these measurements were used to characterize materials and select a suitable candidate for 3D printing.

4.1.1. Mathematical model for shear rate at nozzle

For some rheological measurements to simulate the condition inside the nozzle, it is required to know the shear rate inside the nozzle.

The shear rate for shear thinning material was calculated via the following equation [10, 11]:

$$\gamma = \frac{3n + 1}{4n} \frac{32\dot{Q}}{\pi D^3}, \quad (1)$$

where D is the nozzle diameter, \dot{Q} is the flow rate, and n is the coefficient derived from the Power Law equation [12]:

$$\sigma = k\gamma^n \quad (2)$$

The behavior of each sample was characterized by fitting the Power Law equation to the shear stress-shear rate rheology plot for each material. Figure 3 demonstrates the shear stress-shear rate graphs.

The calculated coefficients n and k are shown in Table 3. These coefficients were employed to predict and analyze the condition as present in the needle. The calculations revealed that the shear rate inside the nozzle is around 100s^{-1} for all experimental samples.

4.2. Physical validation

To ensure printability of the designed material, and confirm rheological measurements results, the a physical test was performed. All the experimental samples containing different solvent concentrations were separately loaded into the syringes' barrel and pressure was applied to the syringes' plunger. Then, the material flow was observed to determine the composition which had the capability to form a continuous flow. Moreover, honeycomb structures were 3D printed to study material capability to keep its shape after being deposited on the surface.

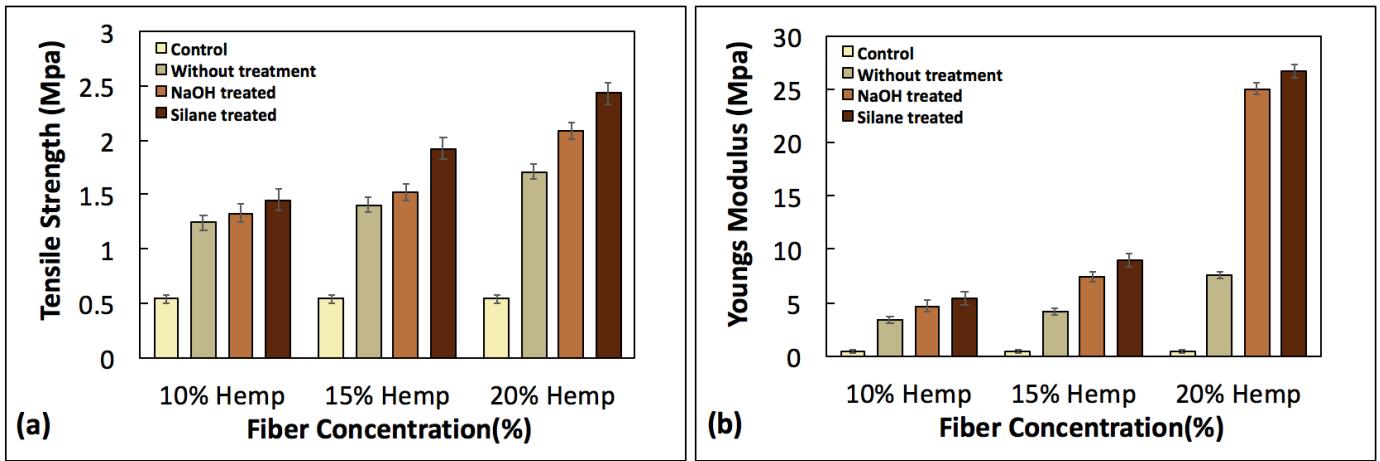


Figure 2: Mechanical properties of molded hemp fibers reinforced silicone composites; (a) Tensile strength,(b) Young's modulus.

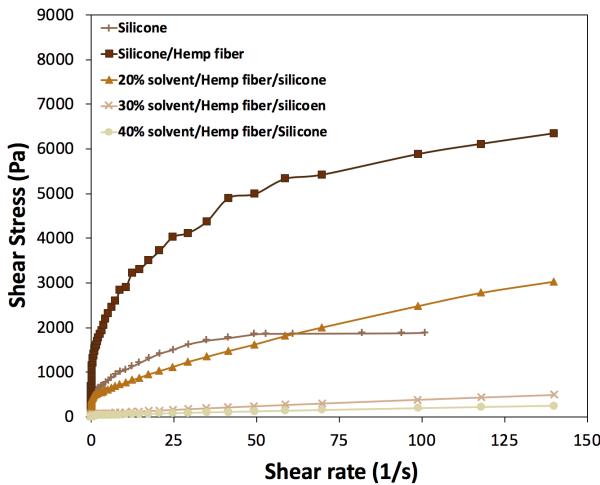


Figure 3: Shear stress-shear rate results for all experimental samples.

5. Results and discussion

5.1. Mechanical testing results

Tensile strength and modulus for different compositions of untreated, alkali treated, and silane treated hemp fibers reinforced silicone composites are presented in Figure 2 (a),(b). It is illustrated that with increasing treated hemp fiber content in the silicone matrix the tensile strength and modulus are also increasing. The incorporation of 10%, 15%, and 20% of hemp fibers increased tensile strength by 51%, 61%, and 68% respectively and increased tensile modulus by 86%, 89%, and 94% respectively compared with pure silicone. This is explained by the fact that the applied stress is distributed to the hemp fibers by the silicone matrix. The effective and uniform stress distribution is also dependent on fiber loading. Hence, the composite with higher fiber loading tolerates higher load before failure in comparison with lower fiber loading and pure silicone [13, 14]. In other words, in silicone/hemp fiber composites, hemp fibers act as a carrier of load and stress which results in uniform force

distribution, therefore, the higher the fiber loading the more enhanced the mechanical properties. The incorporation of 10%, 15%, and 20% of silane treated hemp fiber in silicone composites improved tensile strength by 14%, 27 %, and 29% respectively, and tensile modulus by 37%, 54%, and 71% respectively, compared with untreated hemp fibers reinforced silicone composite.

As untreated fibers are covered by wax and contamination, there would be poor adhesion between hemp fiber and the silicone matrix. Consequently, the stress cannot be transferred effectively from matrix to fiber. In contrast, the significant increase in tensile strength of silane (3-AMPS) treated hemp fiber reinforced composites is due to a better interfacial interaction between fiber and matrix. The improvement in fiber/matrix interaction is ascribed by enriched roughness on the surface of the fibers and chemical bonding between functional groups of polysiloxane and functional groups of silicone. Incorporation of more than 15% hemp fibers in the silicone matrix presented a challenge in 3D printing. Therefore, 15% fiber loading was fixed as an upper limit, and all the experiments were conducted accordingly.

5.2. Composites analysis

5.2.1. Interfacial interaction between fiber/matrix (FT-IR)

FT-IR was performed to explain enhancement in chemical and mechanical interaction between fibers and matrix. The existence of modifications in the chemical bonding in the NaOH treated, Silane treated and untreated hemp fibers were determined by FT-IR spectroscopy. Figure 4 demonstrates spectra of treated and untreated hemp fibers. The peaks around 3280 and 3380cm^{-1} confirm the presence of hydrogen bonded O-H stretching present in aliphatic or aromatic alcohols in the fiber components. The increase in the absorption Of O-H region after NaOH could be due to the possible withdrawal of hemicellulose and lignin which results in increasing number of exposed O-H groups from the surface of the fibers. The peak around 2859 and 2920cm^{-1} corresponds to C-H stretching vibration from aliphatic saturated compounds, like aliphatic

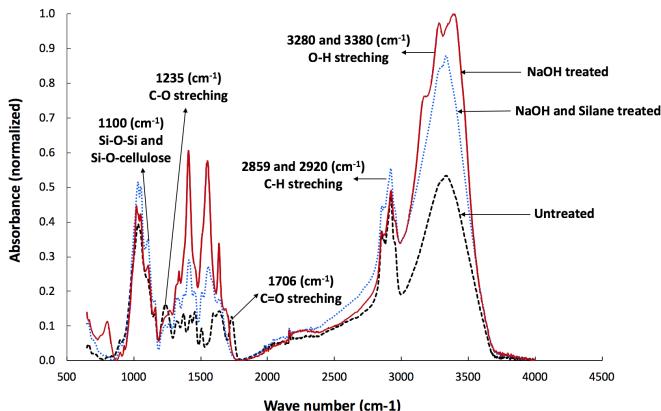


Figure 4: Fourier transform infrared (FT-IR) spectrum for treated and untreated hemp fiber.

moieties in cellulose and hemicellulose. The peak 1706cm^{-1} , results from vibrational stretching of carbonyl groups ($\text{C}=\text{O}$) which is related to hemicelluloses and lignin. As it is illustrated, the reduction in the peak intensity around 1706cm^{-1} , is due to the removal of lignin and hemicelluloses after alkali treatment. Moreover, the peak intensity around 1235cm^{-1} ascribed C-O stretching of acetyl groups in the hemicellulose diminished by alkali treatment. The appearance of the peak around 1100cm^{-1} is corresponding to asymmetric stretching of Si-O-Si, and Si-O-cellulose. The intensity of the peak around 1100cm^{-1} increased after silane treatment.

These adjustments are required for natural fibers used as reinforcement for polymer matrix. The polysiloxane has higher functional groups compared with fiber surface, which can easily react with the functional group of polymer matrix and established stable bonds. It can be concluded from the results that the silane coupling agent was grafted on the fiber surface.

5.2.2. Morphology of the hemp fibers (SEM)

Figure 5 demonstrates the surface micrographs of untreated and alkali treated hemp fiber. It is clearly evident that the surface of raw hemp fibers differs from NaOH treated fibers in roughness . .

SEM images demonstrate that the surface of untreated hemp fiber was smoother than the surface of alkali treated fibers, leading to weak interfacial bonding with polymer matrix, however, the alkali treatment increased the roughness of fiber surface (R_z). In fact, alkali treatment made fiber surface rougher due to removal of noncellulosic parts like pectin, wax, and lignin. This rough surface facilitated mechanical interlocking between fiber and silicone matrix. Therefore, alkali treatment increased the effective surface area available of contact with the matrix and improved the possibility of load transfer from matrix to fibers.

5.2.3. Effect of chemical treatment on interaction between fiber and matrix

To investigate the interaction between fibers and matrix, scanning image analysis was performed giving essential infor-

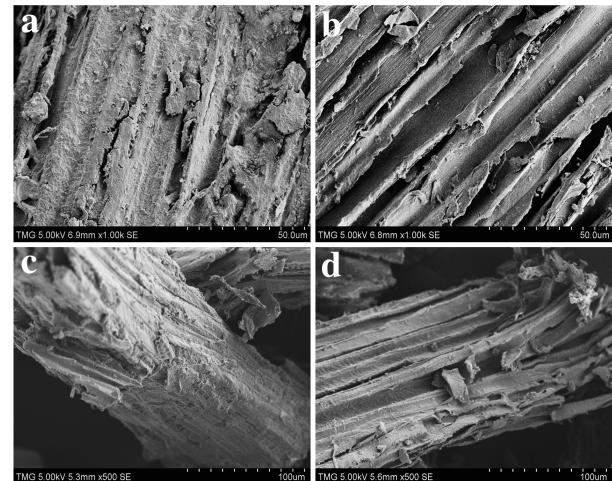


Figure 5: SEM images of (a),(c) untreated (b),(d) alkali treated hemp fibers.

mation about fibers distribution in matrix, and the adhesion between two phases. Figure 7 represents SEM images of the impact fracture surface of the composites. This figure illustrates, that untreated fibers were presented in the form of multi-fibers instead of distributing uniformly. It's demonstrated that there is poor adhesion between untreated fiber and matrix which led to appearance of pull-out holes in composites. Indeed, there is no physical contact between both components in the interfacial region.

Figure 8 demonstrates the effect of NaOH/silane treatment on interface interaction between hemp fiber and silicone matrix. As it's demonstrated that fibers are totally covered by the matrix and there is no void between the two phases. In fact, the interfacial adhesion is affected by the changes of surface topography, because the higher the roughness of alkali treated fiber the better mechanical interlocking between fiber and matrix. Therefore, chemical treatment improved the adhesion at the interface leading to higher mechanical properties.

6. Printability assessment

Extrusion of material with continuous flow and the ability of the fabricated sample to keep its shape without flowing are the two essential requirements for 3D printing. Silicone is a printable material. However, incorporation of hemp fibers in silicone will increase its viscosity which leads to unprintability behavior. Different solvent concentrations were used to reduce the viscosity of the composition, and bring back printability behavior. Therefore, rheological evaluation and physical validation were performed to determine the printable composition without changing material characteristics.

6.1. Rheological evaluation

6.1.1. Shear thinning characterisation

Figure 6 (a) demonstrates the shear viscosity profiles of all the samples which were measured by rheology measurements. All experimental samples displayed shear thinning behavior in which the viscosity decreases over increasing shear rate. The 30

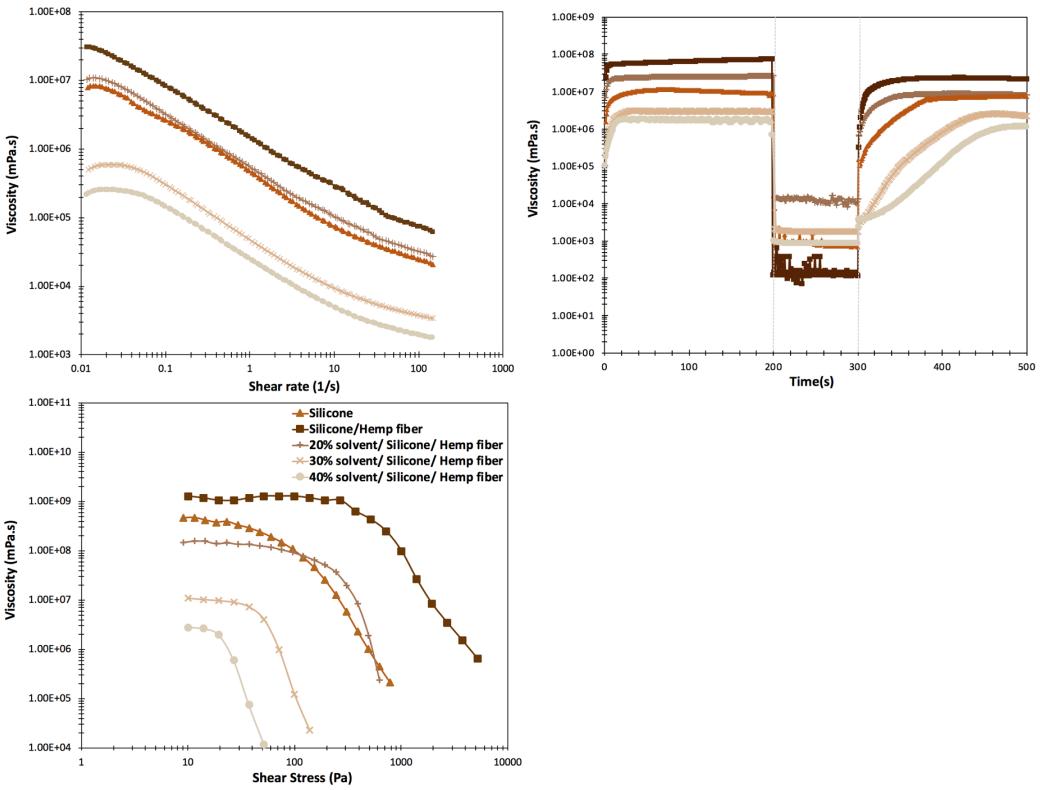


Figure 6: Rheological evaluation; (a) Shear-viscosity results for all experimental samples,(b) Recovery behavior of experimental samples to determine material behavior under a shear rate (100s^{-1}) to near-zero shear to simulate the environment experienced by the materials during extrusion printing, (c) Shear stress ramp for all experimental samples.

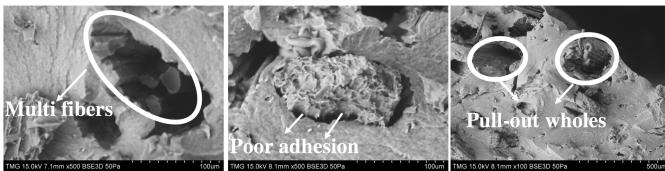


Figure 7: Fractured surface of untreated hemp fiber reinforced silicone.

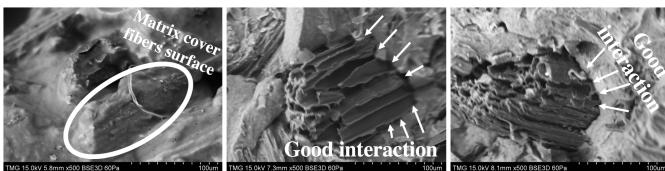


Figure 8: Fractured surface of NaOH/silane treated hemp fiber reinforced silicone.

(wt.%) and 40 (wt.%) solvent, demonstrate significantly lower viscosities compared to silicone. The composition containing 20 (wt.%) solvent, aligned very closely in shear thinning characteristics to printable pure silicone. In other words, the shear thinning profile for the 20 (wt.%) solvent is similar to pure silicone. Material slipping out between plates at higher shear rate led to difficulties in the shear-viscosity measurements and

caused inconsistencies at higher shear rate. Therefore, high shear rates (more than 100s^{-1}) were excluded from these experiments.

6.1.2. Recovery behaviour

Recovery testing was performed to analyze shape fidelity after 3D printing for all specimens. As the rheometer cannot measure the viscosity at true zero-shear, shear rate of 0.01s^{-1} was chosen to mimic at-rest condition during printing. To simulate the condition in the nozzle tip a shear force of 100s^{-1} was applied to the samples for 100s (calculation for shear rate inside nozzle was explained in experimental procedures chapter, section 3.4.2). Finally, to measure the recovery of the materials a low shear rate of 0.01s^{-1} was again applied. Figure 6 (b) demonstrates the recovery results for different solvent concentrations. Here, the pure silicone show rapid recovery (50 s) after exposing under high shear rate. This behavior makes the material to quickly increase its viscosity after extruding and keep its shape after depositing. It can be observed that it takes 140 s for the sample containing 30 (wt.%), and 40 (wt.%) solvent to get steady in viscosity which causes difficulties in formation of continuous flow and shape fidelity. This is another evidence that samples with 30 and 40 (wt.%) solvent are showing unprintability characteristics. In contrast, the composition containing 20 (wt.%) solvent exhibits shorter recovery time to its initial viscosity in less than 75 s. Therefore, 20 (wt.%) solvent concentration exhibits closer printability characteristics to pure

Table 4: Yield point of the different samples.

	Yield stress(Pa)
Silicone	85.40
Silicone/Hemp fiber	440.76
Silicone/Hemp fiber/20(wt.%) solvent	152.64
Silicone/Hemp fiber/30(wt.%) solvent	45.79
Silicone/Hemp fiber/40(wt.%) solvent	21.82

silicone.

6.1.3. Yield stress measurements

Yield stress is an important parameter in determining printability. Shear stress ramp test was performed to measure the yield stress of the hemp fiber/silicone with different solvent composition. Figure 6 (c) demonstrates the viscosity-shear stress curves which give information about yield stress. Yield stress was determined using the intersection of the plateau region in which the material is deformed elastically, and the region that the viscosity drops and the material starts to flow. Table 4 summarizes the results for different composition containing different solvent concentration. By comparison, the hemp fiber/ reinforced silicone sample without solvent displayed highest yield stress which justified material capability to keep its shape after printing. However, this composition exhibited high viscosity which makes it unprintable. The composition containing 20 (wt.%) solvent also displayed higher yield stress compared with silicone. In other words, the composition containing 20 (wt.%) solvent has higher ability to keep its shape after fabrication compared with silicone due to its high yield stress.

6.2. Physical validation

To confirm the rheological results, material flow was also monitored during manual extrusion. Figure 9 demonstrates the material flow for different samples with different composition. 40 (wt.%) (Figure 9 (a)) demonstrated droplet formation

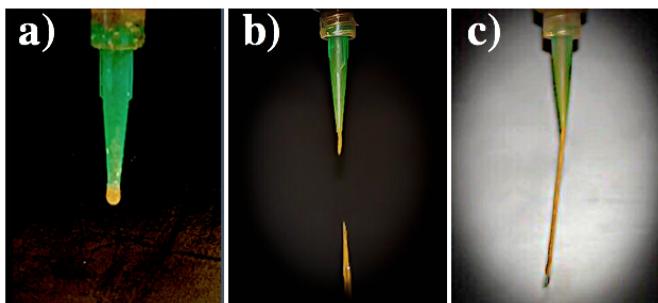


Figure 9: Effect of solvent concentration on material flow; a) 40 (%) solvent b) 30 (%) solvent c) 20 (%) solvent.

and could not form consistent material flow. 30 (wt.%) (Figure 9 (b)) displayed partial fiber formation, however material flow was inconsistent and short. Therefore, both 30 and 40

(wt.%) also exhibited unprintable behavior in line with rheological results. On the other hand, 20 (wt.%) solvent (Figure 9 (c)) showed continuous and consistent material flow which are the required characteristics for 3D printability.

In addition, hexagon honeycomb structures were 3D printed to determine shape fidelity characteristics of different compositions. Figure 11 shows 3D printed hexagon honeycomb structures with 40, 30, 20 (wt.%) solvent concentrations over curing process. The images clearly show that structures containing 40 and 30 (wt.%) solvent concentrations did not keep the hexagon shape over the curing process. On the other hand, the honeycomb structure containing 20 (wt.%) solvent maintained the hexagon shape. These observations, are in agreement with the rheological results.

From the rheological evaluation and the physical validation, it is concluded that the composition containing 15 (wt.%) hemp fiber and 20 (wt.%) solvent had the capability to form continuous flow and maintain its shape which are required for 3D printing.

7. Effect of 3D printing on mechanical properties of hemp fiber reinforced silicone

In order to determine impact of 3D printing on mechanical properties, dog bone shape samples were 3D printed (parts printed at 0° angle). Figure 12 demonstrates the tensile strength and young's modulus of 3D printed hemp fiber reinforced silicone composites. It is evident that the printed samples have higher stiffness and tensile strength in comparison with non-printed samples. Indeed, 3D printed samples possess higher mechanical properties due to a densification of the paste as it's extruded through the nozzle.

Figure 10 shows the surface fractured of 3D printed and molded samples after tensile test. SEM images demonstrate the extent of void inclusion in molded samples. Indeed, there are large voids within the molded samples compared to 3D printed samples with the same composition. It shows that the bubbles were removed as the material passed through the nozzle. Therefore, the 3D printed product has a denser structure leading to improved mechanical properties.

In addition, The enhanced mechanical properties could be explained by the fact that hemp fibers were aligned during 3D printing process. SEM images show the directional alignment of hemp fibers along printing direction. In contrast, for molded samples, there is no fiber alignment. Direct ink writing (DIW) is an extrusion-based 3D printing in which fibers are distributed randomly before they reached to nozzle tip and aligned under the shear and extensional flow field that applied by nozzle during 3D printing. In fact, under the simple shear flow fibers tend to align in the shear direction. Figure 13 shows directional alignment of hemp fibres along the scanning/printing direction. This is leading to higher mechanical properties compare to those fabricated by mold.

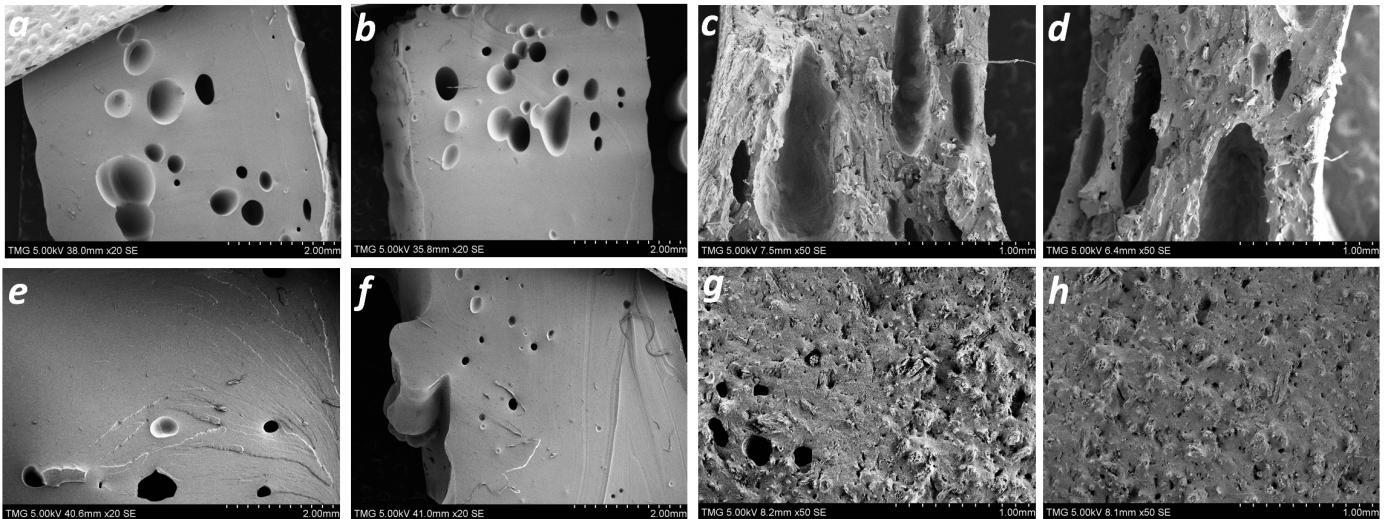


Figure 10: Cross-section of 3D printed and molded samples a,b) Cross-section of molded silicone c,d) Cross-section of 3D printed silicone e,f) Cross-section of molded hemp fiber reinforced silicone g,h) Cross-section of 3D printed hemp fiber reinforced silicone.

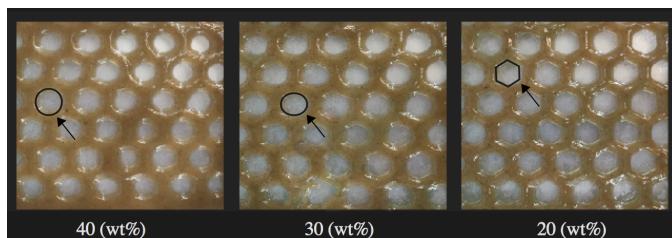


Figure 11: Effect of solvent concentration on 3D printing shape fidelity.

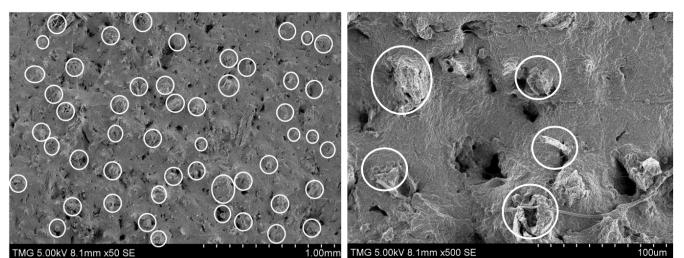


Figure 13: Hemp fibers were aligned during extrusion.

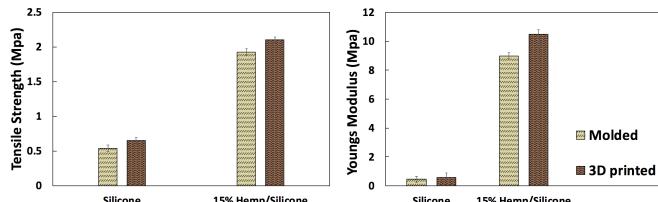


Figure 12: Mechanical properties of 3D printed products.

8. Conclusions

This study presented the design of a new printable material developed by the incorporation of hemp fiber in silicone elastomers with enhanced mechanical properties. In other words, the idea of this work was to design a new printable material with high functionalities. To enhance hemp fiber/silicone composites mechanical properties and to ensure its printability, it was required to determine suitable mixing composition. Therefore, mechanical tests and microscopic examinations were used to figure out the hemp fibers loading which provided desirable mechanical properties. Incorporation of 15 (wt.%) fiber increased tensile strength and modulus of hemp fiber/silicone composites

by 61% and 89% respectively, compared to silicone. Hence, 15% fiber loading was chosen as a suitable candidate.

One of the main drawbacks of natural fibers polymer composites is the poor compatibility between the fibers and matrix. Therefore, alkali and silane treatments were used to modify the surface of hemp fibers. Alkali and silane treatments improved tensile strength and modulus of hemp fiber/silicone composites by 27% and 54% respectively, compared to the composition containing untreated hemp fibers. Alkali treatment increased the surface roughness of the hemp fiber and exposed more reactive OH groups on the fiber surface to facilitate chemical bonding. On the other hand, silane coupling agent acts as a bridge between hemp fiber and silicon. Therefore, silane treatment increased the fiber-matrix interfacial bonding strength resulted in enhanced mechanical properties.

In this study, hemp fiber silicone composite was 3D printed using hemp fiber as the reinforcement and silicone as the matrix. The challenge with 3D printing this material was the high viscosity which made them unprintable. Different solvent concentrations were added to the composition to determine the composition with printable behavior. Two-step printability assessment was performed to obtain printable material; rheologi-



Figure 14: Simple gripper fabricated using DIW 3D printing method.

cal evaluation and physical validation. Rheological evaluations provided a clear understanding of the material behavior during the entire 3D printing process, i.e., during extrusion, material deposition and curing. While manual extrusion was performed to obtain information about flow formation at the tip of the nozzle during the extrusion. The results revealed that hemp fiber/silicone composition containing 20 (%wt) solvent exhibited 3D printable behavior. Based on rheological studies and physical validation, this composition was capable to form continuous flow and keep its shape after deposition. Mechanical testing demonstrated that 3D printed products have improved mechanical properties compared to the molded samples. Improved mechanical properties were explained by densification of the paste as it was extruded through the nozzle, and fibers alignment made by the printing process.

Finally, by employing Direct ink writing 3D printing method, a simple gripper was fabricated based on silicone and compared to its silicone hemp fiber counterpart. The results demonstrated that the silicone hemp fiber gripper applied higher force than silicone gripper. Indeed, incorporation of fibers makes the gripper more strong and reliable.

9. Future work

In this study advanced manufacturing 3D printing technology was employed to fabricate a simple soft gripper based on silicone/hemp fiber and it was compared with its silicone counterparts (Figure 14).

In order to make a comparison between the strength of the two grippers, a micro force sensor was used. To prevent unwanted movements the soft grippers were held on a flat plate. An adjustable angle clamp was used to provide the same situation for each soft gripper. In other words, the clamps were fixed during measurement and the grippers were inserted between them. Figure 15 shows that there is a significant difference in measured micro sensor voltage between the silicone gripper and silicone reinforced by hemp fiber gripper. The results show that the silicone/hemp fiber gripper is capable of lifting around 130 grams, however, the silicone gripper can lift around 30 grams. Therefore, incorporation of hemp fibers significantly affected silicone properties and made it stronger. The silicone/hemp base gripper can efficiently grip heavier objects, while its silicone counterpart cannot grasp objects with the weight above 30 grams.

Therefore, fabricating functional soft robots like a four channel tentacle, a pneunet actuator, and a quadruped using this

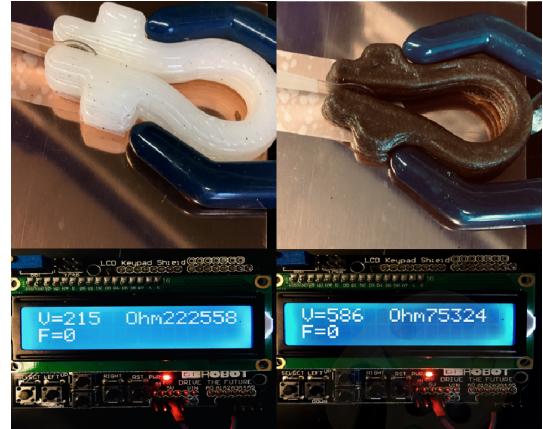


Figure 15: Using micro force sensor to find the differences between two grippers.

new material with high reaction force is recommended for future works.

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