

Fog Harvesting: Inspired by Spider Silk

Yijia Cen

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Jiangtao Cheng

Michael W Ellis

Xiaoyu Zheng

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ABSTRACT

The water crisis has been an increasing challenge in some places in the world. One proposed solution that has drawn lots of attention is fog harvesting. A commonly used fog collector is a vertical mesh, usually made of poly materials. Small water droplets can easily get pinned and quick evaporation is the major common challenge for vertical meshes. Coating the fog mesh with superhydrophobic chemicals is one of the solutions. However, superhydrophobicity is not durable and it may contaminate the collected water. In addition, it requires a high professional maintenance and laboratory operation standard. As a result, it is impractical to set such fog collectors in regions and countries with water crisis. Low cost, harmless, easily fabricated, higher coalesce rate and low maintenance are the five pillars for this research. This thesis topic is inspired by spider silk's ability to direct water droplets to certain locations to further enhance water collecting rate. This directional droplet movement is caused by spindle-knot and joint structure on the biomimetic silk. The spindle-knot is randomly porous, and the joint is stretched porous. In addition, the spindle-knot has a tilted angle β above the joint region. Due to these unique structures, there are three droplet movement controlling forces – surface tension force, hysteresis force, and Laplace pressure force. This thesis presents detailed equation derivations for each driving force in the introduction section. Spindle-knot is the pivot point to direct water, forming the spindle-knot structure is another focus of this thesis. Fluid coating and dip-coating with dimethylformamide (DMF), a solvent with a low evaporation rate, is the highly used methods to form the spindle-knot structures due to its simple setup and low cost. However, DMF is an extremely hazardous organic compound, and it requires high laboratory operation standards. In the second section of this thesis, DMF has been replaced with water/ethanol and

photocurable materials to construct the spindle-knots. Furthermore, Additive manufacturing (3D printing method) was adopted to synthesize bionic spider web with spindle-knot structures.

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GENERAL AUDIENCE ABSTRACT

Water shortage is one of the highest concerns all around the world and collecting fog water has drawn lots of attention recently. The focus of this thesis is to increase the fog collection rate by using less hazardous, low maintenance and low-cost methods. Commonly used fog collector is a large vertical plastic mesh. However, those large meshes suffer from water pinning and easily evaporation issues. Water repellent chemicals have been studied and used to dissolve those issues, however, the chemical coating will not last long and it will contaminate the collected water easily. Moreover, coating the water repellent chemicals requires professional operation and maintenance. To solve this issue without using chemical coating, we have learned unique water collection and directional behavior from spider silk. In a humid day, you will easily find the spider web with fully covered water droplets in an organized order. If we zoom in on single spider silk, the spider silk is composed of many puff and joint regions. Those puff regions have higher water collection ability than the joint regions, and this puff region shrinks down to form the spindle-knot shape with angle β above the joint region. This unique spindle-knot structure induces the water directional movement, and three forces- surface tension force, pinning force, and Laplace pressure force – are controlling the moving direction. Chapter 1 shows equation derivations with surface material effects, surface roughness effects and water droplet landing location effects. To form such special spindle-knot structure, commonly used formation methods are fluid coating and dip coating by using an organic polymer solvent. However, commonly used organic polymer-solvent suffer from a high level of hazardous, resulting in high laboratory requirement and operation cost. In Chapter 2 of this thesis, that commonly used organic polymer-

solvent will be replaced by water/ethanol mixture and light-sensitive materials to form the spindle-knots. Furthermore, the 3D printing method is adopted to build a spider web with spindle-knot structures.

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Table of Contents

Abstract	ii
General Audience Abstract	iv
Acknowledgements	vi
List of Figures & Tables	x
Chapter 1: Introduction.....	1
1.1Motivation: water scarcity	1
1.2 Water harvesting methods	2
1.2.1 Seawater desalination	2
1.2.2 Rainwater harvesting	3
1.2.3 Atmosphere water harvesting	4
1.3 Bio-inspired: spider silk	5
1.4 Three Forces – surface tension Force, hysteresis force, and Laplace pressure force	6
1.4.1 Surface Tension Force Due to Material Gradient	7
1.4.2 Surface Tension Force Due to Roughness Gradient	8
1.4.3 Hysteresis force caused by unsmooth and heterogeneous surface	9
1.4.4 Laplace pressure force due to spindle-knot shape	10
1.4.5 Resultant force	14
1.5 Existing spindle-knot formation methods and existing challenges	19
1.5.1 Fluid coating – PMMA powder & organic compound DMF	19

1.5.2 Dip-coating – PMMA powder & organic compound DMF	20
1.6 Thesis objective	21
Chapter 2. Methods and Results	22
2.1 Fluid coating – PMMA powder & water/ethanol	22
2.2 Dip-coating – PMMA powder & water/ethanol	26
2.3 Mold casting	29
2.4 3D printing	31
2.4.1 Fiber with spindle-knot	31
2.4.2 Spindle-knot only	33
2.5 Light Curing	34
2.5.1 Performance testing	34
2.5.2 Fluid coating with PEGDA	40
2.5.3 Manually formed spindle-knots	40
2.5.4 Dip-coating with PEGDA	43
Chapter 3. Discussion	43
Chapter 4. Conclusion	44
Appendix A. Freshwater is Abundant	47
Appendix B. Surface Tension Force Due to Material Gradient	48

<u>Appendix C. Surface Tension Force Due to Surface Roughness Gradient</u>	<u>50</u>
<u>Appendix D. Methods to Measure Receding and Advancing Angles on a Surface</u>	<u>53</u>
<u>Appendix E. Hysteresis Force Due to Unsmooth and Heterogeneous Surface</u>	<u>55</u>
<u>Appendix F. Young's relation, Wenzel's Law & Rayleigh Instability Simple Explanation</u>	<u>57</u>
<u>Appendix G. 3D Drafted Web Frame</u>	<u>60</u>
<u>Reference</u>	<u>63</u>

List of Figures & Table

Figure 1. Seawater desalination process turning saltwater into freshwater	2
Figure 2. Dry spider silk	6
Figure 3. Wet spider silk with formed spindle-knot and joint region	6
Figure 4. Graphical representation of the resultant force	7
Figure 5. Surface tension force due to material gradient	8
Figure 6. Surface tension force due to roughness gradient	9
Figure 7. Three landing positions for the water droplet	11
Figure 8. Dimension representation of each landing case	12
Figure 9. Top views of three droplet landing cases	12
Figure 10. Water directional movement on smooth and heterogenous surface	17
Figure 11. Water directional movement on unsmooth and heterogeneous surface	17
Figure 12. Water directional movement comparison on homogeneous surface	18
Figure 13. Existing Fluid Coating Method by DMF	19
Figure 14. Dip-coating by PMMA powder and DMF solvent	20
Figure 15. Fluid coating by water/ethanol solvent	23
Figure 16. Fluid coating setup	23
Figure 17. Nylon fiber fluid coating	25
Figure 18. Optical fiber fluid coating	25

Figure 19. Dip coating by U-shape holder	26
Figure 20. Water droplet collection and directional movement	27
Figure 21. Spider web holder dip coating	28
Figure 22. 3D drafted spider web frame	29
Figure 23. First mold design	30
Figure 24. Second mold design	30
Figure 25. 3D printed spindle-knot and joint regions	32
Figure 26. Spindle-knots under microscope	32
Figure 27. Water collection and movement on 3D printed spindle-knots and joints	33
Figure 28. Spindle-knot only 3D design	34
Figure 29. Smooth surface contact angles measurement	35
Figure 30. Spindle-knot formation by PEGDA	35
Figure 31. Water Collection and Movement on PEGDA knots	36
Figure 32. Water droplet dimensions by ImageJ	37
Figure 33. 3D Inventor model for spindle-knot	38
Figure 34. Water quantity comparison setup	39
Figure 35. Fluid coating with PEGDA	40
Figure 36. Manually formed spindle-knots	41

Figure 37. Water collection and movement on manually formed spindle-knot	42
Figure 38. Dip-coating with PEGDA drawback	43
Figure B1. Surface tension force due to material gradient	48
Figure C1. Surface tension force due to roughness gradient	50
Figure D1. Advancing and receding angle measurement by inflating and deflating fluid	53
Figure D2. Angle measurement by tilted surface	54
Figure E. Hysteresis force on unsmooth and heterogeneous surface	55
Figure F1. Young's relation	57
Figure F2. Wenzel's Law	58
Figure F3. Rayleigh's Instability	59
Figure G1. First spider web frame design	60
Figure G2. Second spider web frame design	61
Figure G3. Third spider web frame design	61
Figure G4. Fourth spider web frame design	62
Table 1. Four polymer materials and their surface contact angles	16
Table 2. Collected water quantity comparison	39
Table 3. Method Comparison Table	44

Chapter 1. Introduction

1.1 Motivation: water scarcity

According to The Millennium Project – Global Challenge 2, over 90% of the world has access to improved drinking water since 1990 [1]. Nevertheless, over 785 million people are still living without access to freshwater and one million people die each year due to water-related diseases [2]. From Shiklomanov and Rodda's global water content in the hydrosphere chart, earth contains only 2.53% of total freshwater volume. Among this low percentage of freshwater, there is 68.7% permanent ice, 30.1% groundwaters, 0.86% soil moisture, and 0.86% permanent ice, which are not easily accessed by human. The remaining accessible freshwater (around 0.339%) stores in lakes, wetlands, rivers, biological water, and atmosphere [3]. Even though the available freshwater percentage is low, the freshwater is plentiful for all the human beings mathematically (see Appendix A for comparison). Even with the mathematical proof that freshwater is abundant, the freshwater crisis is increasing due to uneven quantity and quality distribution. Based on the freshwater usage data on the Worldometers website [5], the lowest daily water usage is 34 liters per capita, on the opposite side, the highest rate is 16281 liters per capita. Besides, Asia takes 60% of the world's population but only has 36% water available. At the same time, North and Central America only has 8% of the world's population but has 15% water available [5][6]. The inequality not only involves the quantity but also includes the quality. Take South America for instance, it has 26% of the world's water and only takes 6% of the world's population. The water seems to be abundant only based on their quantity. However, most of the water is rainfall and less than 30% area has access to clean water. Natural factors such as geology, weather, and vegetation contribute to the uneven freshwater distribution. Furthermore, climate change and human activities – wars, agriculture, urbanization, population growth – add stress to the freshwater distribution.

1.2 Water harvesting methods

To solve the water scarcity problem, many actions and water harvesting methods have been studied and applied worldwide.

1.2.1 Seawater desalination

Turning seawater into freshwater is the most popular idea since the earth is covered by 71% sea surface and saltwater comprises about 96.5% of the earth's total water supply [3].

Figure one shows the most common and most energy-efficient desalination procedures.

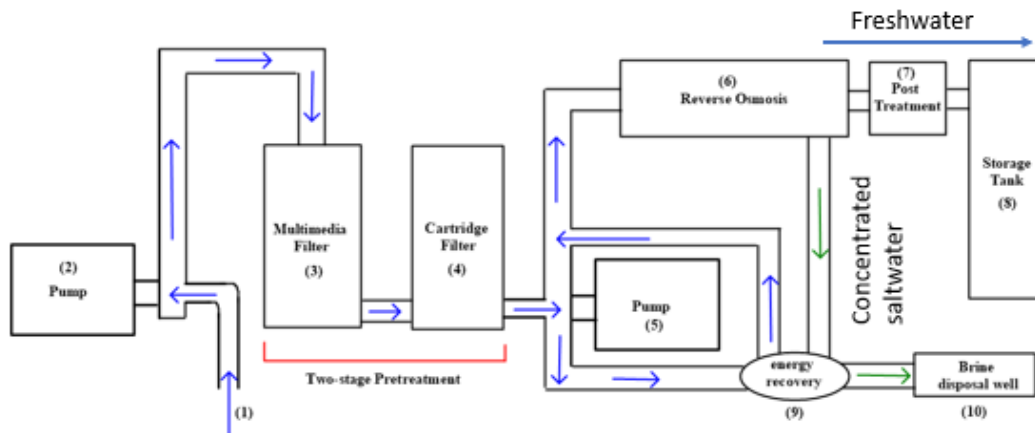


Figure 1. Seawater desalination process turning saltwater into freshwater. Devices include (1) seawater intake, (2) seawater driving pump, (3) multimedia filter, (4) cartridge filter, (5) pressurize pump, (6) reverse osmosis, (7) freshwater post-treatment, (8) storage tank, (9) energy recovery device, and (10) brine disposal well.

Generally, a seawater intake well will deliver the seawater to the two-stages pretreatment – multimedia filter and cartridge filter. The multimedia filter contains a series of layers of granular materials such as garnet, sand, pebbles, gravel. This first stage filter will remove the large particles, including sand, twigs, seaweed. After the large particles have been removed, the seawater is fed into the cartridge filter which is made of yarn-like material to remove smaller particles such as fine sand and clay. After that, the filtered seawater is pressurized and fed into the reverse osmosis semi-permeable membrane by using a high-pressure pump. The added

pressure forces the water molecule to go through the membrane and leave the salt particles behind. Then the freshwater is treated in the post-treatment by adding calcium carbonate (improving taste and neutralize pH level) and chlorine (provide disinfection properties). Meanwhile, the concentrated salt stream is flowing into the energy recovery device to provide the remaining energy to pump the new seawater into the reverse osmosis stage. After the energy has been recovered, the high concentrated salt stream will be treated to normal ocean salt level in the brine disposal well. Even though the power assumption is decreasing remarkably by almost 89% from the 1970s to 2008 [7], the reverse osmosis desalination device is not constructed into huge applications due to three major challenges and concerns. The first challenge is the higher energy usage compared to existing water harvesting methods. Most of the desalination devices contain the energy recovery device to reduce the energy cost; however, energy requirement with the energy recovery system is still relatively high. The second challenge is the cost, including valuable high-tech devices, expensive operational cost, and frequent maintenances. The most concerning challenge is environmental side-effects. The Seawater Intake device is sitting in the sea, and it might cause the death of juvenile organisms. Also, the concentrated salt stream and chemicals used in several stages might breed huge environmental risks to the ocean if they are not properly managed [7].

1.2.2 Rainwater Harvesting

Rainwater harvesting, on the other hand, is a simple, inexpensive and widely used collection method for domestic and irrigation purposes. There are two simple components – the catchment area and storage tank. The catchment area can simply be the roof of the house, and the storage tank is a safe water container. Roof area, annual rainfall, and the runoff coefficient play important roles in determining the rainwater collection efficiency [8]. Annual rainfall is the most

important and uncontrolled one among those important factors. Meanwhile, the annual rainfall (including rainfall frequency and amount) becomes the constraint to decide if the rainwater harvesting is practical. Take Egypt for instance, the average precipitation day is nearly 0 days between Jun and Sept. On other months, the precipitation days range from 1 day to 7 days per month [9]. As described, the rainwater collection is not practicable for low precipitation areas.

1.2.3 Atmosphere Water Harvesting

For those low precipitation areas with a high humidity ratio, atmosphere water harvesting-atmosphere water condensation, and fog collection- becomes the solution. Water condensation requires a temperature difference to turn the water from the vapor phase into the liquid phase. To condense more water droplets, it requires energy input to maintain the device at a lower temperature than the surrounding. On the other hand, the fog collection does not require a temperature difference and does not involve phase change; instead, the fog catch device will simply catch the water droplets in the air and collect them in a container. The major challenge of fog collection is how to collect droplets efficiently before they get evaporated. The state-of-art fog collector on the market is achieved by the collision of suspended droplets on a vertical mesh [10]. The vertical mesh is usually large and made of polyethylene or polypropylene without any chemical coatings. The mesh is a simple design and the water droplet movement is controlled by gravity and the pinning forces. Small water droplets can easily get pinned and quick evaporation is the major common challenge for vertical meshes. Coating the fog mesh with superhydrophobic chemicals is one of the solutions. However, superhydrophobicity is not durable and it may contaminate the collected water. Another solution is adding complex micro- or nanoscale topography on the surface. However, those two methods require high professional maintenance and laboratory operation standard to maintain at its desire performance. Designing

environmental-friendly, inexpensive, simple and robust meshes have attracted lots of attention. As a result, it is impractical to set such fog collectors in regions and countries with water crisis. Low cost, harmless, easily fabricated, higher coalesce rate and low maintenance are the five pillars for this research.

1.3 Bio-inspired: spider silk

Nature is the most creative designer, and our mission is to find and exploit its principles. Spider silk is one of the most inspired masterpieces. Spider silk's high level of strength and toughness have been inspired and applied to real applications. The water-absorbent and water directional abilities have attracted countless researchers. Jiang Lei's group is one of the nature followers and has conducted countless researches [11-14]. In their first published work, they found the spider silk is not just a straight line. Instead, the spider silk is composed of puff and joint regions in sequence (Figure 1), and the puff structure is rougher and larger to catch more water molecules from the air than the joint regions. With the higher water catching ability, the puff region is condensed to form a spindle knot shape (Figure 2) which has angle β in degree higher than the joint region. Moreover, the nanofibrils are randomly distributed in the spindle-knot region, and the joints have aligned nanofibrils. Hence, the spindle-knot region is more hydrophilic (water-liking) than the joint region. When the water lands on the fibers, water droplets move toward the spindle knot region to form larger water droplets [11]. Having this directional movement, unbalanced forces are acting around the water droplets.

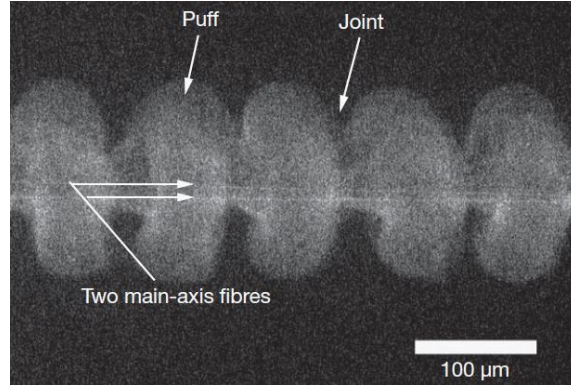


Figure 2. Dry spider silk with ordered puff and joint around the fibers. Puff regions are higher than the joint region, and each puff is connected by the joint. [11]

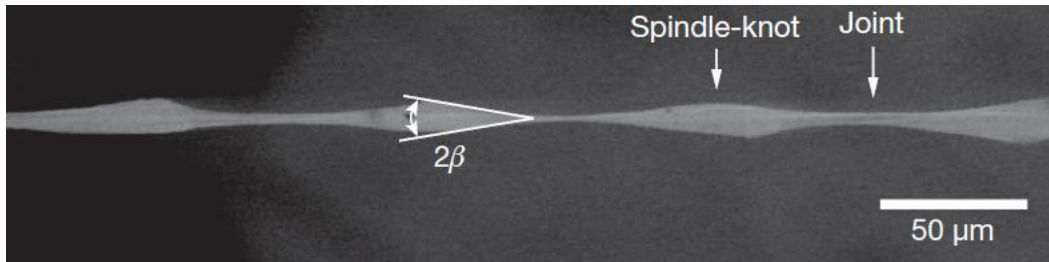


Figure 3. Wet spider silk with formed spindle-knot and joint region. The spindle-knot region forms an angle of 2β around the spider fibers. [11]

1.4 Three Forces – surface tension Force, hysteresis force, and Laplace pressure force

Those unbalanced forces are surface tension force, hysteresis force (aka pinning force), and Laplace pressure force. Surface material properties and surface roughness induce surface tension force and hysteresis force. On the other side, the spindle-knot shape influences the Laplace pressure force. Graphically, the resultant force can be represented in Figure 4. We assume the surface is ideally smooth and flat when we calculate the surface tension force due to material gradient. In a similar approach, we assume the solid surface is homogeneous and flat when we evaluate surface tension force due to the roughness gradient. Surface material difference and surface roughness gradient also affect the hysteresis force. To calculate this force, we assume the surface is flat, heterogeneous, and unsmooth. Last but not least, we consider the

spindle-knot shape to find the Laplace pressure force that acts on the droplet while keeping the surface smooth and homogeneous.

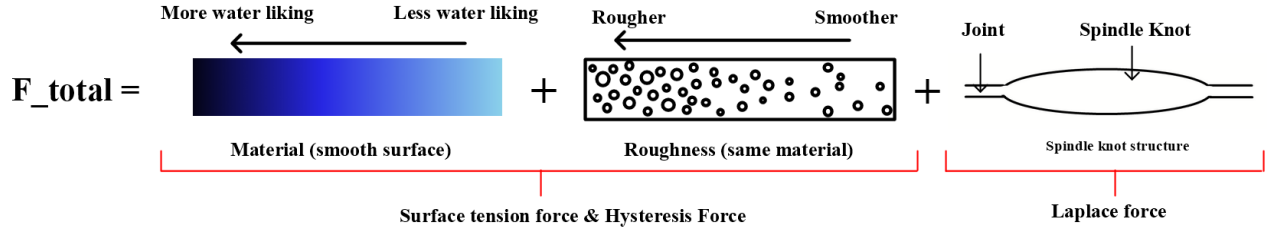


Figure 4. Graphical representation of the resultant force acting on the droplet due to material difference, surface roughness gradient, and spindle-knot structure.

Mathematically, the resultant force can be written as the summation of those three forces,

$$F_{total} = F_{\gamma,m} + F_{\gamma,r} + F_H + F_L \quad (1)$$

where sub-notation γm , γr , H , and L represent surface tension force due to material, surface tension force due to roughness, hysteresis, and Laplace pressure.

1.4.1 Surface tension force due to material gradient

The material gradient and surface roughness gradient control the surface tension force. Figure 5 shows that the water droplet lands on a smooth and flat surface. The left side is made of more hydrophilic material than the right side. Two different surface materials induce two different contact angles θ_1 and θ_2 . Due to the higher material wettability on the left side, surface contact angle θ_1 is smaller than the surface contact angle θ_2 . We assume the water droplet will move dx along the perimeter (toward the more hydrophilic side). As a result, the surface tension force due to material gradient can be calculated as, (See Appendix B for detail steps)

$$F_{\gamma,m} = \gamma_{lv}(\cos \theta_1 - \cos \theta_2)_m * 2\pi R_0 \quad (2)$$

where γ_{lv} is the liquid-vapor surface tension in newton per meter, and R_0 is the water droplet radius in meter. The surface tension force due to material gradient is always positive to direct water toward left if the left surface is made of more hydrophilic material.

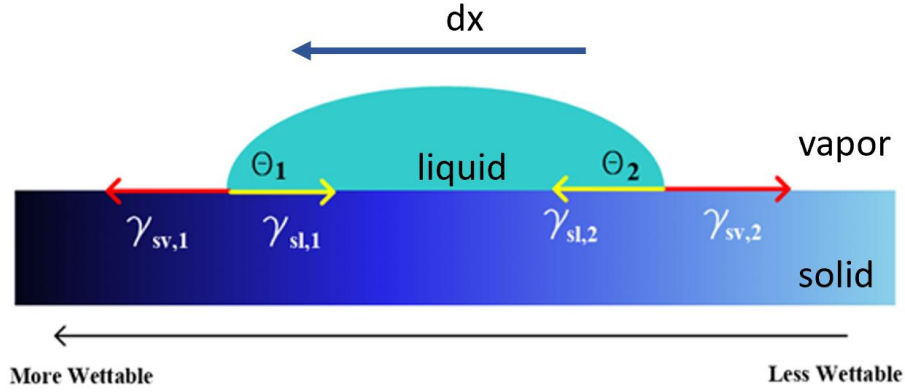


Figure 5. Surface tension force due to the material gradient. The solid surface is flat, smooth, and heterogeneous. The left side is composed of more hydrophilic material; therefore, smooth contact angle θ_1 is smaller than contact angle θ_2 in degrees.

1.4.2 Surface tension force due to roughness gradient

As we have mentioned in the previous section, the surface tension force can also be induced by surface roughness. In Figure 6, the surface is flat, homogeneous (same hydrophilic material), and unsmooth. Different roughness induces different surface contact angles; in this case, the rougher side (left side) has a smaller contact angle θ_1 . The surface tension force induced by surface roughness can be formed as, (see Appendix C for detail)

$$F_{\gamma,r} = \gamma_{lv}(\cos \theta_1 - \cos \theta_2)_r * 2\pi R_0 \quad (3)$$

where R_0 is the collected fog radius in meter on the solid surface. The direction of the surface tension force is always from right to left if we have a rougher surface on the left side than the right side. The magnitude of this force is directly proportional to the roughness gradient and water droplet size.

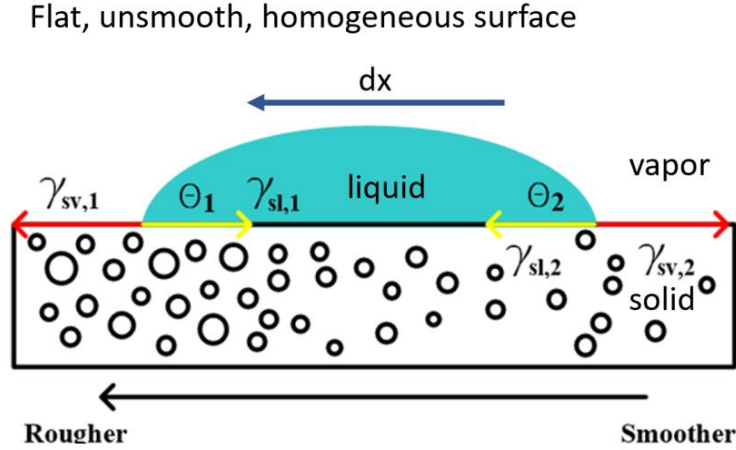


Figure 6. Surface tension force due to roughness gradient. Droplet on a homogeneous (same hydrophilic material) and rough surface. The surface tension rises due to the roughness gradient along the contact surface. The front end is rougher than the rear end. In general, the rougher surface for the same material is more water liking. As a result, the surface contact angle 1 is smaller than contact angle 2.

1.4.3 Hysteresis force caused by unsmooth and heterogeneous surface

Surface roughness and surface material properties also affect hysteresis force, which tends to prevent the movement toward more hydrophilic side. The most common method to compute the hysteresis force is measuring the receding and the advancing angles of the water droplet on the solid surface. The receding and the advancing angles are two angles when deflating and inflating a water droplet until the contact line starts to move. In other words, the water droplet contact surface with the solid will move if the contact angle is below receding angle θ_R in degrees and beyond the advancing angle θ_A in degrees. An easier way to measure those two critical angles is by tilting the solid surface, then measure the two angles right before the water droplet starts to slide (see Appendix D for methods for finding receding and advancing angles). After measuring the receding and advancing angles, the hysteresis resisting force can be evaluated as, (Appendix E for detail math derivation)

$$F_H = \pi R_0 * \gamma_{lv}[(\cos \theta_{R2} - \cos \theta_{A2}) - (\cos \theta_{R1} - \cos \theta_{A1})] \quad (4)$$

where subscript 1 and 2 indicate the spindle knot and joint contact surface. Rougher surface and more water liking chemical surface has higher advancing angle and lower receding angles [16]. Therefore, the receding angle on surface 2 (right region/rear region/joint region) is larger than the receding angle on surface 1 (left region/front region/spindle-knot region); advancing angle on surface 2 is smaller than the angle on surface 1. Mathematically, the relation can be represented as,

$$\cos \theta_{R1} > \cos \theta_{R2} \quad (5)$$

and

$$\cos \theta_{A1} < \cos \theta_{A2} \quad (6)$$

Combining equation 5 and equation 6 gives us that,

$$\cos \theta_{R1} - \cos \theta_{A1} > \cos \theta_{R2} - \cos \theta_{A2} \quad (7)$$

Because the hysteresis resistance is larger in the front end than the rear end, the hysteresis force F_H will always be negative. In other words, the hysteresis force will move the droplet away from the front end (spindle-knot region).

1.4.4 Laplace pressure force due to spindle-knot shape

The surface tension force and hysteresis force are evaluated under a flat surface; however, the spindle knot region is higher than the joint region with a β tilted angle. Those periodic high regions produce curvature contact surface, which brings us another driving force – Laplace pressure force. When water droplet lands on the fibers, there are three landing locations - fully on the spindle knot, fully on the joint region, and both joint and spindle knot - shown in Figure 7. Spindle knot and joint regions are molded by using the actual spider silk dimensions

measured from Figure 3 with a 50-micrometer scalebar. The mean diameter of water in the air is only about 10 to 15 micrometers, which is less than half of the spindle knot length.

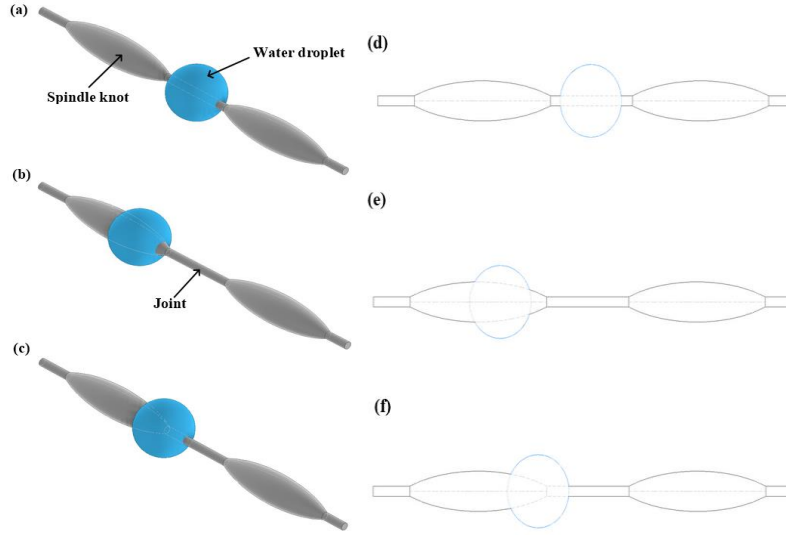


Figure 7. Three landing positions for the water droplet. (a) The water droplet fully lands on the joint region, (b) the water droplet fully lands on the spindle knot region, and (c) part of the droplet lands on the spindle knot and the remaining part will land on the joint region. Three figures on the right side are the front views of those three landing positions correspondingly.

Figure 8(a) is the enlarged image of the spindle knot and joint under the microscope and Figure 8(b) to 8(d) are simplified sketches for three cases. Furthermore, the corresponding top views can be represented in Figure 9. Figure 8(b) and Figure 9(a) represent the first case where the water droplet lands fully on the joint region. The local radius at point A and point C are equal to the fiber radius R , and the local curvature R_1' and R_2' are identical. The Laplace pressure different per unit length AC (length AC is the diameter of a water droplet) can be represented as,

$$\Delta P_{ten,1} = \gamma_{lv} \left(\frac{1}{R_2'} - \frac{1}{R_1'} \right) * \frac{1}{2R_0} \quad (8)$$

There is no Laplace pressure difference along with the water droplet. Therefore, the Laplace pressure force in case one (Figure 8b & 9a) will approximately equal to zero.

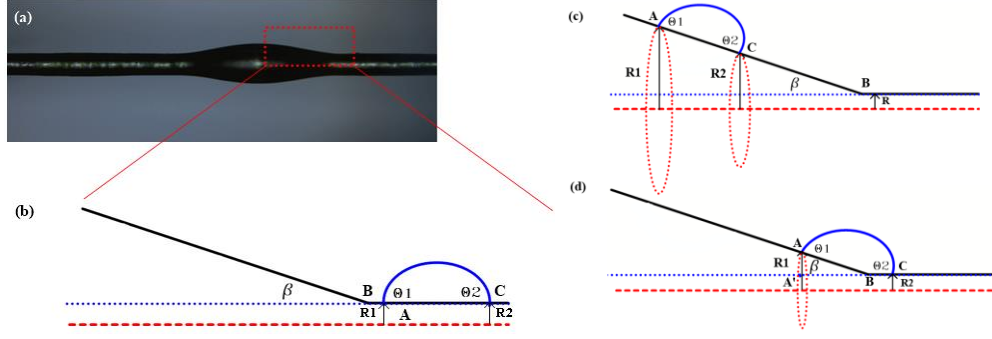


Figure 8. Dimension representation of each landing case. (a) fabricated spindle knot fiber under the microscope. (b) water droplet fully lands on the spindle knot region. (c) water droplet fully lands on the joint region. (d) water droplet lands both on the joint and spindle-knot region.

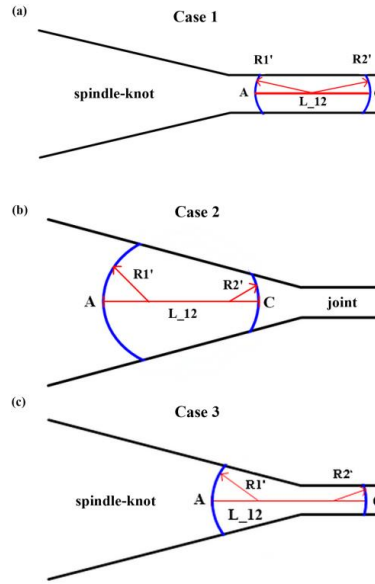


Figure 9. Top views of three droplet landing cases. a) Droplet fully lands on the joint region. b) Droplet fully lands on the spindle-knot region. c) Some of the droplet lands on the spindle-knot region, and the rest rests on the joint region.

Figure 8(c) and Figure 9(b) represent case two, where the water droplet fully lands on the spindle knot region with front contact angle θ_1 and rear contact angle θ_2 . The relation between the spindle knot angle and the radius can be represented as by using trigonometry,

$$BC = \frac{R_2 - R}{\sin \beta} \quad (9)$$

$$AB = \frac{R_1 - R}{\sin \beta} \quad (10)$$

The distance between point A and point C can be calculated as,

$$L_{AC,2} = AB - BC = \frac{R_1 - R}{\sin \beta} - \frac{R_2 - R}{\sin \beta} = \frac{R_1 - R_2}{\sin \beta} \quad (11)$$

According to Laplace's theorem, the Laplace pressure around the water droplet can be calculated as by using the local curvatures at the front and rear location R_1' and R_2' ,

$$\Delta P = \gamma_{lv} \left(\frac{1}{R_2'} - \frac{1}{R_1'} \right) \quad (12)$$

The pressure difference acts along distance L_{12} , combining equation (11) and equation (12) to get the pressure gradient along with the water droplet,

$$\Delta P_{len,2} = \gamma_{lv} \left(\frac{1}{R_2'} - \frac{1}{R_1'} \right) \frac{\sin \beta}{R_1 - R_2} \quad (13)$$

The pressure gradient is acting around the entire droplet, and under the assumption of a perfect half-sphere, we could calculate the Laplace force as [12],

$$F_{L,2} = \gamma_{lv} \left(\frac{1}{R_2'} - \frac{1}{R_1'} \right) \frac{\sin \beta}{R_1 - R_2} \frac{2}{3} \pi (R_0)^3 \quad (14)$$

The local curvature at point 2 is always smaller than the local curvature at point 1, and the local radius R_1 is greater than the local radius R_2 on the spindle knots. Therefore, the Laplace force for case two will always be positive to drive the water drop toward the higher spindle-knot region.

Figure 8(d) and Figure 9(c) show the third case where the water droplet lands on both spindle knot and joint region. To find the distance between the front end and rear end, we need to find the distance AB by using the tilted angle β and the fiber radius R,

$$AB = \frac{R_1 - R_2}{\sin \beta} = \frac{R_1 - R}{\sin \beta} \quad (15)$$

The total length between point A and point C will be the sum of length AB and BC,

$$L_{AC,3} = AB + BC = \frac{R_1 - R + BC \sin \beta}{\sin \beta} \quad (16)$$

For simplicity, we assume majority of the water droplet lands on the spindle knot region. As a result, the distance BC will approximately equal to zero and equation 16 becomes,

$$L_{AC,3} = AB = \frac{R_1 - R}{\sin \beta} \text{ or } \frac{R_1 - R_2}{\sin \beta} \quad (17)$$

A similar approach with case two, we could calculate the Laplace pressure force as,

$$F_{L,3} = \gamma_{lv} \left(\frac{1}{R_2'} - \frac{1}{R_1'} \right) \frac{\sin \beta}{R_1 - R_2} \frac{2}{3} \pi (R_0)^3 \quad (18)$$

which the expression is equivalent to the case two force expression. Similarly, the third case will have a positive Laplace force to move the water droplet from the joint region toward the spindle knot region.

1.4.5 Resultant force

Resultant force can be calculated by adding up all the forces, and the total force for all the cases can be united into single equation 19,

$$\begin{aligned} F_t = & \gamma_{lv} 2\pi R_0 [(\cos \theta_1 - \cos \theta_2)_m] \\ & + \gamma_{lv} 2\pi R_0 [(\cos \theta_1 - \cos \theta_2)_r] \\ & + \pi R_0 * \gamma_{lv} [(\cos \theta_{R2} - \cos \theta_{A2}) - (\cos \theta_{R1} - \cos \theta_{A1})] \\ & + \gamma_{lv} \left(\frac{1}{R_2'} - \frac{1}{R_1'} \right) \frac{\sin \beta}{R_1 - R_2} \frac{2}{3} \pi R_0^3 \end{aligned} \quad (19)$$

where the third term will equal to zero when the droplet lands fully on the joint region. The first term is always positive if the front side is composed of more hydrophilic material. The surface tension force due to roughness is positive if the front side has higher surface roughness than the rear side. When we have higher surface tension force on the water droplet, the water droplet is also experiencing higher hysteresis force. The resultant force is controlled by surface material, surface roughness, water droplet size, spindle-knot shape, and droplet location. For the real spider silk, it is composed of the same material with different roughness. Under this condition, the total force acts on the water droplet will be [12],

$$\begin{aligned}
F_{t,spider\ silk} = & \gamma_{lv}(\cos \theta_1 - \cos \theta_2)_r * 2\pi R_0 \\
& + \pi R_0 * \gamma_{lv}[(\cos \theta_{R2} - \cos \theta_{A2}) - (\cos \theta_{R1} - \cos \theta_{A1})] \quad (20) \\
& + \gamma_{lv} \left(\frac{1}{R_2'} - \frac{1}{R_1'} \right) \frac{\sin \beta}{R_1 - R_2} \frac{2}{3} \pi (R_0)^3
\end{aligned}$$

If the resultant force is positive, the water droplet will move from the joint region toward the spindle-knot part, vice versa. However, it is difficult to calculate the numerical values since there are lots of uncontrolled variables such as the landed water droplet size, specific landing location. Experimentally, Lei Jiang's group proved that surface roughness and surface material can control the droplet moving direction and moving speed.

They used four different polymer materials – PVA (Poly (vinyl acetate)), PMMA (polymethyl methacrylate), PS (polystyrene), and PVDF (polyvinylidene fluoride) – to form the joint and spindle knot regions. From Table 1, PVA has a smaller surface contact angle, and PS and PVDF are less hydrophilic compared to PMMA. PMMA is used to form smooth joint regions, and the rest of the materials are used to form spindle-knot sections. They began with smooth spindle-knot regions, and the results are shown in Figure 10. The first spindle-knot is

composed of PVA, and it takes over 5 seconds to direct water towards the spindle knot region. On the other hand, the PS and PVDF are less hydrophilic than the joint region (smooth PMMA), the water droplet moves away from the spindle knot region. As a result, for the same roughness surface, water droplet moves towards spindle-knot if the spindle-knot is composed of more hydrophilic materials.

Table 1. Four polymer materials and their surface contact angles. Lei Jiang’s group used PMMA to form the smooth joint section and rest of the materials to form the spindle-knot sections. PVA is more hydrophilic than PMMA, and the other two materials are less hydrophilic than PMMA. [12]

Material	Smooth Surface Contact Angle (degree)	
PVA	56.7	More Hydrophilic
PMMA	68.4	Reference (joint)
PS	93.3	Less hydrophilic
PVDF	92.7	Less hydrophilic

Then they used chemical etching methods to create roughness in the spindle-knot regions while keeping the smooth PMMA joint. With the rougher surface, PVA uses less than 5 seconds to direct the water droplet towards the spindle-knots. Instead of moving the water droplets away from knots, rough PS and PVDF knots move the water droplets towards knots. Therefore, high roughness in the spindle-knot regions increases the moving speed and changes the moving direction.

To further prove that high spindle-knot roughness can increase the moving speed, they also used PMMA to form spindle-knots. From Figure 12, rough PMMA knot takes less than 1 second to direct water droplets towards knot regions. On the other hand, with smooth PMMA

knots, it uses almost 5 seconds to direct water towards the spindle knot region. Therefore, for the same material, higher spindle knot surface roughness will increase the water droplet moving direction and collection rate.

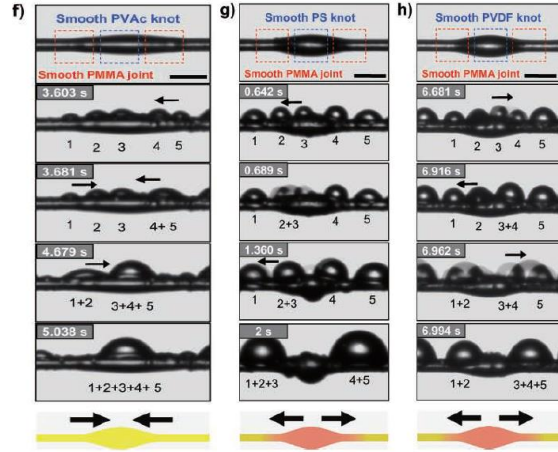


Figure 10. Water directional movement on the smooth and heterogeneous surface. Joint regions are made of smooth PMMA. (f) Smooth PVA spindle knot uses over 5 seconds to direct water towards the spindle-knot region; (g) Smooth PS spindle knot region uses 2 seconds to move the water droplet away; (h) Smooth PVDF takes almost 7 seconds to direct the water droplet away from the spindle-knot region. [12]

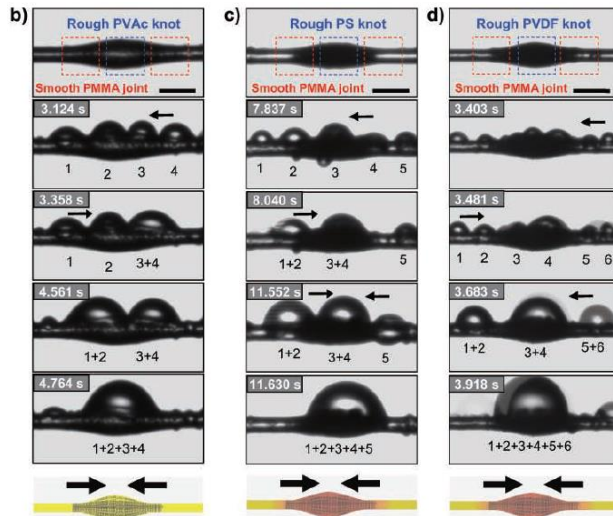


Figure 11. Water directional movement on the unsmooth and heterogeneous surface. (b) Rough PVA knots use less than 5 seconds to direct water droplet toward the knots; (c)(d) rough PS and PVDF also direct water droplet towards the knots [12]

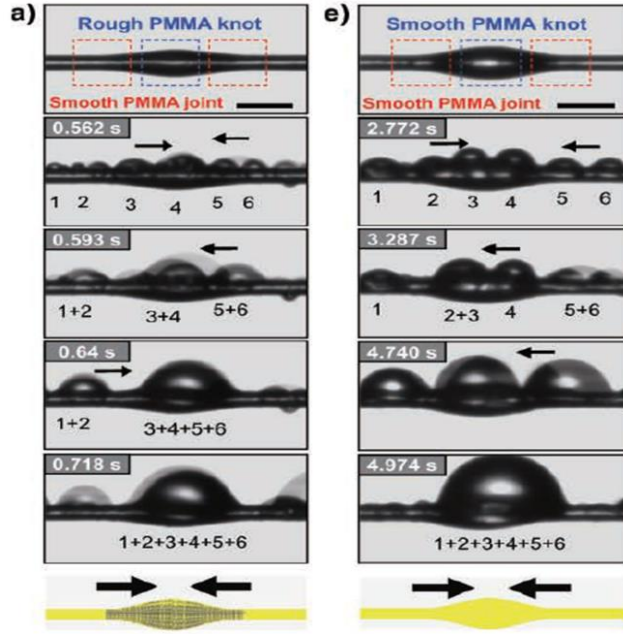


Figure 12. Water directional movement comparison on a homogeneous surface. Joint and spindle-knot regions are composed of PMMA. (a) Rough PMMA knot takes less than 1 second to direct water droplets towards knot; (b) Smooth PMMA knot uses almost 5 seconds to direct water droplets towards knots. [12]

After deriving three driving forces and seeing those experimental results, we know the spindle-knot structure is the significant fact to bring the water droplet uphill toward the spindle knot region. In the following sections, we will begin with existing spindle-knot formation methods, follows by existing challenges, and ends up with the thesis objective.

1.5 Existing spindle-knot formation methods and existing challenges

Fluid-coating and dip-coating are two standard spindle-knot formation methods. PMMA powder and organic compound DMF are combined to form the polymer solution. Then the polymer solution will be coated around the nylon fiber, and polymer solution will evaporate to form the spindle-knot shape.

1.5.1 Fluid coating – PMMA powder & organic compound DMF

The existing fluid coating method contains three sections – feeding, solution container, and receiving. According to Lei Jiang's method [17], they used 18-micrometer nylon fiber to connect those three sections. Then they filled the solution container with fully dissolved PMMA and DMF polymer solution. Then the motor in the receiving section will pull the fiber out of the solution container to form the spindle-knots. Those spindle-knots are formed automatically due to Rayleigh's instability (Appendix F1 for simple explanation) [15].

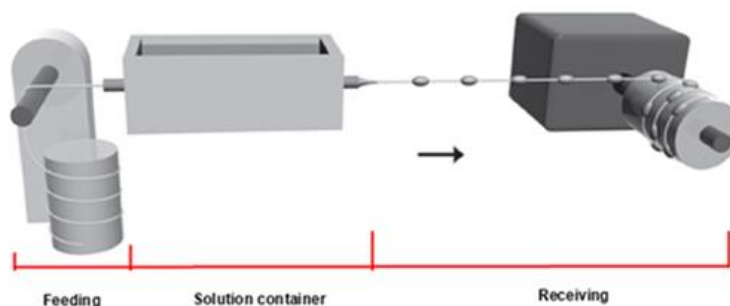


Figure 13. Existing fluid coating method. Three major sections are feeding, solution container, and receiving. PMMA powder and organic compound DMF are used to form the polymer solution.[17]

From the material side by using organic compound DMF, it dissolves PMMA powder very quickly, and the fully dissolved polymer solution evaporates quickly to form solid spindle knots. However, DMF is very hazardous and extremely dangerous to the skin and respiratory system. Due to its high rate of hazardousness, it requires professional operations and high lab operational standards. In addition to the material side, the nylon fiber is too thin and non-rigid. From the setup side, this method only requires low energy in the motor to pull the nylon fiber out of the solution container syringe to do large scale fabrication. However, centering 18-micrometer nylon fiber in the syringe is the major setup challenge.

1.5.2 Dip-coating – PMMA powder & organic compound DMF

Another standard method is dip-coating by using PMMA powder and organic compound DMF. The setup for this method is more straightforward than the fluid-coating setup, and it only requires a solution container, fiber, and polymer solution. First, the fiber is fully immersed in the polymer solution. After a couple of seconds, the fiber will be pulled out vertically with constant speed. The polymer solution is covered around the fiber, forming a thin cylindrical layer (details in Appendix F3). Due to the Rayleigh instability, the thin cylindrical layer starts to break and form the droplets in a periodic order. The formed polymer droplet will evaporate and become a solid spindle knot around the fibers.

Since this method is using the same polymer and solvent, it has the same disadvantages and advantages from the material side. From the setup side, this method requires fewer lab devices, and there is no external energy required to form the spindle-knots. However, this method can only do the mid-scale fabrication.

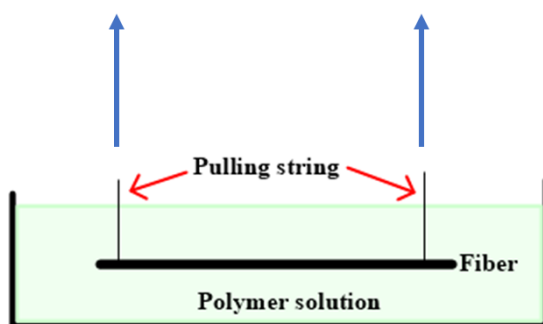


Figure 14. Dip-coating by PMMA powder and DMF solvent. Fiber is fully immersed in the polymer solution, and the spindle-knot will automatically form due to Rayleigh Instability.

1.6 Thesis objective

In section one, we begin with the freshwater percentage and how water quality and quantity are unevenly distributed (section 1.1). To alleviate the water stress, water harvesting

methods – ocean water desalination, rainwater collection, fog harvesting – have been applied in multiple countries, and each method has its cons and pros (section 1.2). Then we keep our eyes on the fog collector (section 1.2) for low precipitation areas with a high humidity ratio. However, existing non-chemical coating meshes used in fog collection have a low collection rate and low durability. Then, we introduce how spider silk has the high ability to catch water and direct the water to specific locations (This property will increase the collection rate) (section 1.3). Later, we calculated three forces acting on the water droplet and see Lei Jiang's experimental results, and we know that spindle-knot shape is the key point to control the droplet movement (section 1.4). In section 1.5, we introduced two standard existing spindle-knot formation methods: fluid-coating and dip-coating. After that, we point out the advantages and disadvantages of those two methods.

In section 2, we will introduce different methods without using hazardous DMF solvent. Those methods include fluid coating (section 2.1), dip-coating (section 2.2), mold casting (section 2.3), 3D printing (section 2.4), and light-curing (section 2.5). Besides, each section contains several sub-sections that vary in shape and procedure. Each sub-section begins with a general setup, follows by specific procedures, comes with water collection and movement, ends with advantages and drawbacks. The main purpose of this thesis is to form spindle-knots structures by simple, time saving, low cost, and unharmed methods.

In section 3, we compare each method with the existing fluid-coating method according to one constraint and six objectives. Those six objectives are polymer solution preparation time, spindle-knot solution evaporation time, less harmful material, low cost, large scale fabrication, and no professional operator requirement. At last, section 4 provides the conclusion of this thesis.

Chapter 2. Methods and Results

This section will contain four primary spindle-knot formation methods, and some method contains sub-method. For each method, we will begin with the general lab setup, follow by formation procedures, formation results, cons and pros, and some ends up with improvement proposals.

2.1 Fluid coating – PMMA powder & water/ethanol

We used a similar setup as Lei Jiang's group, as shown in Figure 15. There are three major sections – feeding, solution container, and receiving end. The motor located in the receiving section creates the driving force to pull the fiber from the feeding section to the solution container. The significant difference between those two setups is the solvent. Instead of using hazardous DMF to dissolve PMMA powder ($M_w \sim 15,000$, Sigma Aldrich), we used the mixture of water and ethanol (80% volume weight water and 20% volume weight ethanol). To fully dissolve PMMA powder, an additional heating source is required, and the heating temperature should be maintained above 80 degrees Celsius. Longer solution preparation time is another disbenefit, and it usually takes up to an hour to get the PMMA powder fully dissolved in the water/ethanol solvent.

The left figure in Figure 16 shows the general PMMA solution preparation, and the detailed setup shows at the right side in Figure 16. To begin with, we place a 20ml beaker with a magnet bar on the Cimarec Thermo device. Then we add 10 ml water/ethanol solvent and turn on the heat and stir evenly reach 80 degrees Celsius. After the solvent temperature reaches steady, we add 0.5-gram PMMA powder into the beaker, after about 10 minutes, we keep adding the solvent if the PMMA is not fully dissolved. Meanwhile, we connect the fiber holder, solution reservoir, syringe needle, blue hollow cylinder, and motor through long fiber. While connecting

those parts, we need to make sure the fiber is in the center of the syringe needle to cover the fiber fully with the polymer solution. Then we place and pre-heat the solution reservoir before we add the fully dissolved solution. After adding the fully dissolved PMMA solution, we turn on the motor and draw the fiber out of the solution reservoir. The covered solution will become solid spindle-knot after 12 hours due to Rayleigh instability [14,18].

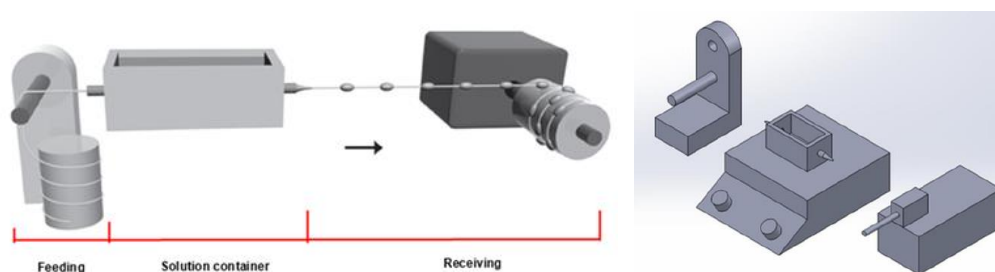


Figure 15. Fluid coating by water/ethanol solvent - Left: Jiang Lei's setup by using PMMA and DMF. Right: Lab setup with ethanol/water mixture and additional heating source.



Figure 16. Fluid Coating Setup. Left: Preparing the PMMA solution. Right: Fiber drawing to form Spindle-Knot. The motor drives the blue hollow cylinder clockwise to draw the fiber out of the solution reservoir. The collecting speed can be controlled by replacing different hollow cylinder size. The drawing speed can control the fluid coating layer thickness.

We begin with the 22-micrometer nylon fiber, and it is separated from the nylon fiber twine. Then, we connect the fiber holder, solution reservoir, and collector. Since the nylon fiber is in the micrometer scale, we place the 5X magnifying glass on top of the syringe needle to

make sure the nylon fiber is not touching the syringe needle edges. Less than 10% of the nylon fiber drawings can successfully form the spindle knot, and few of them are shown in Figure 17. Those three successful trials form different spindle-knot sizes, space, and thickness. The bottom figure has the longest length and smallest spindle-knot spacing, and the central figure has the largest spacing and highest thickness. Those formed spindle-knots are not consistent even under the same drawing speed and similar polymer concentration. This low success rate and low consistency are due to three primary reasons. First, the 22-micrometer nylon fiber is not centered, which causes the fluid not uniformly cover the fiber fully. The second reason is that the nylon fibers have been collected without fully evaporated and dried. Last but not least, the thin nylon fiber is not durable, and it can easily break into parts before the spindle-knot formation.

From the material side by using water/ethanol solvent, it is harmless, and it does not require high lab operation standards as DMF does. However, polymer solution preparation time and evaporation time are much longer than using DMF solvent. Besides, 22-micrometer nylon fibers are still too thin and nonrigid. From the setup point, additional heating source and centering thin nylon fibers are two significant challenges.

To solve the slight and nonrigid nylon fiber issue, we replace the nylon fiber with 0.25-millimeter optical fiber. This optical fiber saves fiber preparation time and connection time. In addition, it is much easier to place the fiber in the center of the syringe. Under a similar procedure, Figure 18 shows that this method can successfully form uniform spindle-knot by using the water/ethanol solvent.



Figure 17. Nylon fiber fluid coating. Less than 15% of the trials form the spindle-knot, and they all form different spindle knot sizes and thicknesses.

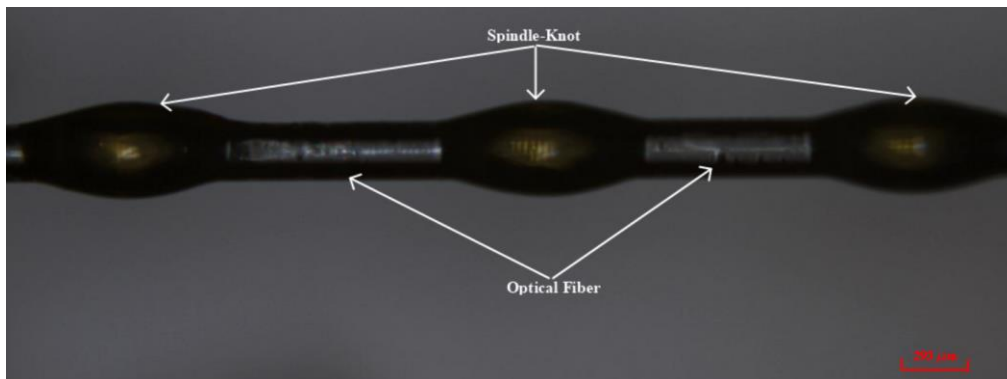


Figure 18. Optical fiber fluid coating. The spindle knot is uniformly and periodically around the 0.22 mm optical fiber. By replacing the dangerous DMF solvent, we can still form the spindle-knot structure by using a similar setup.

However, additional energy cost and long PMMA solution preparation time are still two significant challenges by replacing the DMF with water/ethanol solvent.

2.2 Dip-coating – PMMA powder & water/ethanol

We also tried the dip-coating method with the water/ethanol solvent. This method has the same PMMA preparation steps, but this method has a simpler spindle-knot formation setup. First, we attach several optical fibers on top of the U-shape holder (by VT 3D Design Studio) and hold them on the holders by double side tape and duct tape. Duct tape also serves as the interspace to ensure the PMMA solution fully covers the fiber. After placing the optical fibers on the holder, we flip the holder and place it inside the solution container, as showing in Figure 19 (b). Then we add the PMMA solution along the container edge and then pull the holder vertically after 10 seconds. From Figure 19 (c), the spindle knots are all around the optical fiber with quite uniform space and dimensions. Roughly overnight, the polymer solution evaporate, and solid spindle-knots are formed around the fibers.

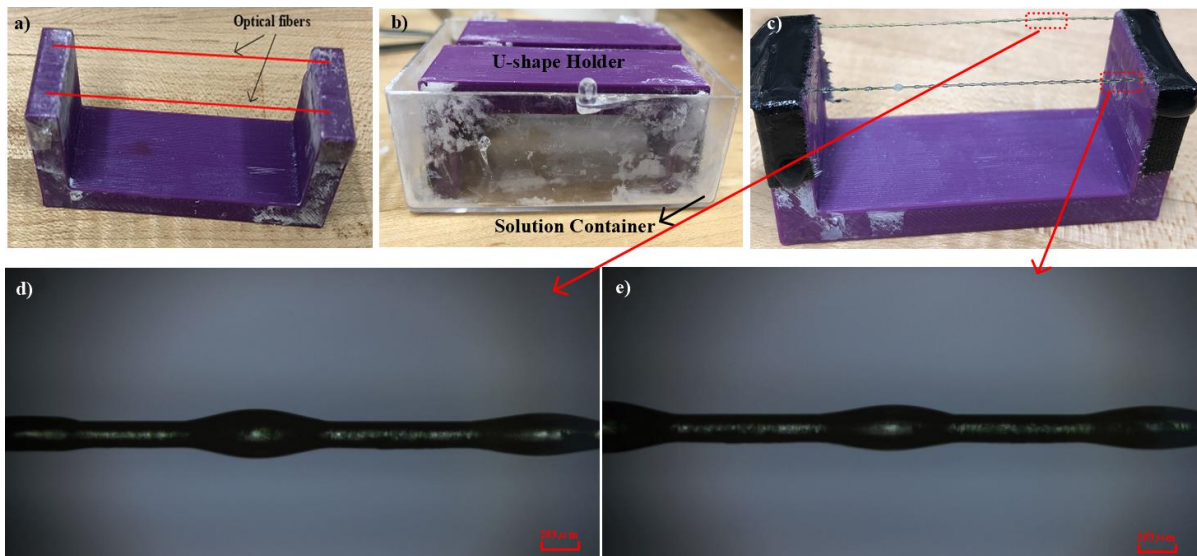


Figure 19. Dip coating by U-shape holder. **a)** Multiple optical fibers are taped on the top of the U-shape holder. **b):** Flip and place the U-shape Holder with the optical fibers inside the solution container. Then pour fully dissolved PMMA solution in the container to cover the optical fiber

fully. (c) – (e) Wait about 10 seconds, we manually pull up the holder vertically with uniform speed and place it on the flat table. Figure c was taken right after that, and the last two figures are sections of evaporated optical fiber roughly after 12 hours. Spindle knots are uniformly spaced, and most of them have identical thickness and width.

After forming the solid spindle-knots around the optical fiber by using dipping and lifting, the next step is to observe water condensation and water droplet movement. Setting the fibers under the microscope and adjusting the light to have a clear view. Then we place the ultrasonic humidifier in front of the spindle-knots and turn on the humidifier at the highest speed. At 26 seconds, water starts to land on the spindle-knots (water droplets 1, 3, 5) and joint regions (water droplets 2, 4). After 2 seconds, water droplet 4 starts to move toward the spindle-knot in the center and forms a larger water droplet (droplet 3+4). At 43 seconds, water droplet 2 starts to move toward the spindle-knot region and forms a bigger droplet. The last figure shows that large water droplets are collected on the spindle knot regions.

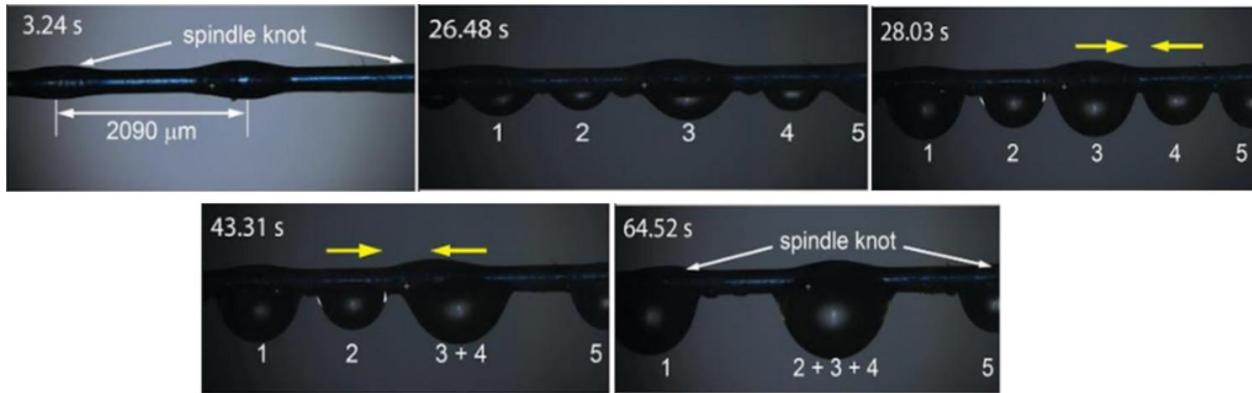


Figure 20. Water droplet collection and directional movement. The ultrasonic humidifier is constantly supplying fog onto the spindle-knots and fibers. At 26.48 s, water droplets attach to the joints and spindle knots. In less than 3 seconds, the water droplet 4 starts to move toward droplet 3 and form a larger water droplet (3+4). At 43.31 second, the water droplet 2 starts to move toward the right and become larger water droplet (2+3+4). Meanwhile, droplets 1 and 5, sitting on other spindle-knots, are forming to larger droplets.

This coating method shows good water collection and water droplet movement results. However, continuously producing a long fiber with spindle-knot structure is the major challenge of this method. To increase the fabrication scale, we will replace the U-shape holder with a spider web shape holder. Straight optical fiber is circled and attached to a spider web holder. Figure 21 shows the general setup for this approach. The holder is connecting to the rolling plate by a fixed pulley, and the rolling plate is connected to the multi-speed motor. Adjusting the lifting speed will affect the spindle-knot dimensions. Instead of using straight optical fiber directly, knitting the optical fiber into web shape or using 3D drafted spider web frames is required. Four drafted spider web frames are shown in Figure 22, and detail designs are listed in Appendix F.

Long polymer solution preparation time, long evaporation time, and additional heating source are still three significant challenges by using water/ethanol solvent. To solve those challenges, we will show the 3D printing method and the light-curing method.

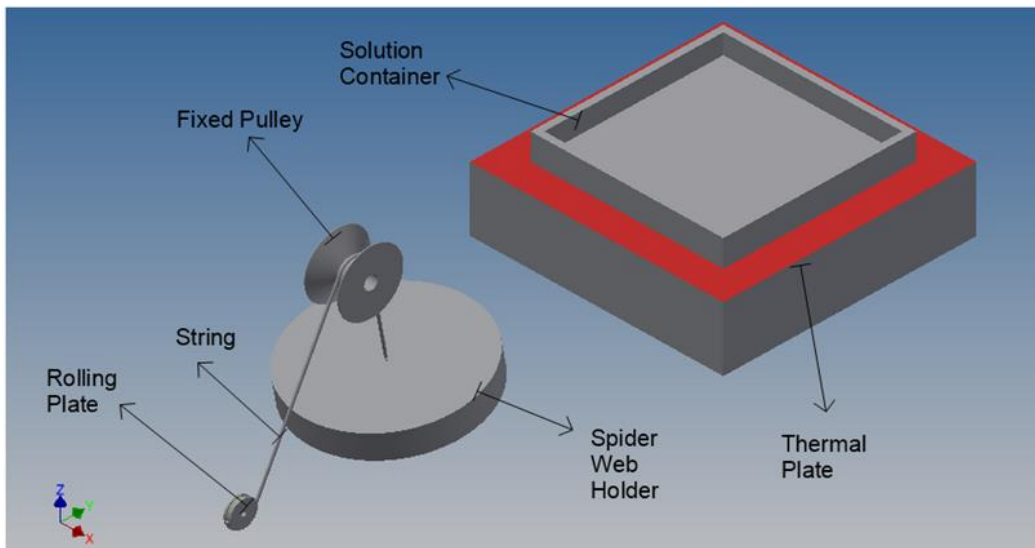


Figure 21. Spider web holder dip coating. By using this approach, the produced optical fiber with spindle knot structure is longer and lifting speed can be controlled by machine instead of human measurement.

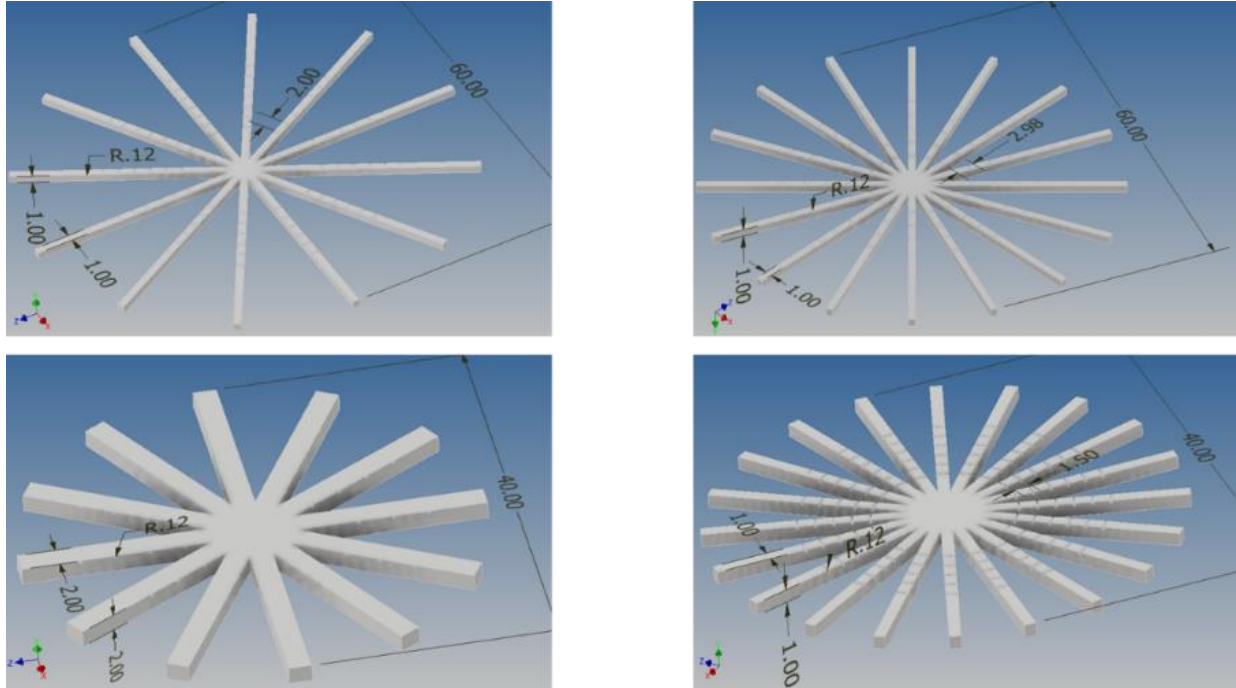


Figure 22. 3D drafted spider web frame. Here are four designs. Each frame design has grooves space to hold the optical fibers, and frame side views are presented in Appendix G.

2.3 Mold casting

Mold casting has been highly applied in many categories from art to industry. Mold casting contains two processes – molding and casting. Molding is manufacturing the mold to have a desired hollow cavity. On the other hand, casting is pouring the liquid into the mold and forming solid shapes.

Using the 3D printer to print out the mold with spindle-knot and nylon fiber cavities with desired spacing and dimensions is the first step. Figure 23 and Figure 24 are two mold designs. The next step is to assemble the two pieces and hold them tight by using clamps on all sides. After the mold is assembled tight, injecting the pre-made PMMA solution from the top by using syringes through each hole on the mold.

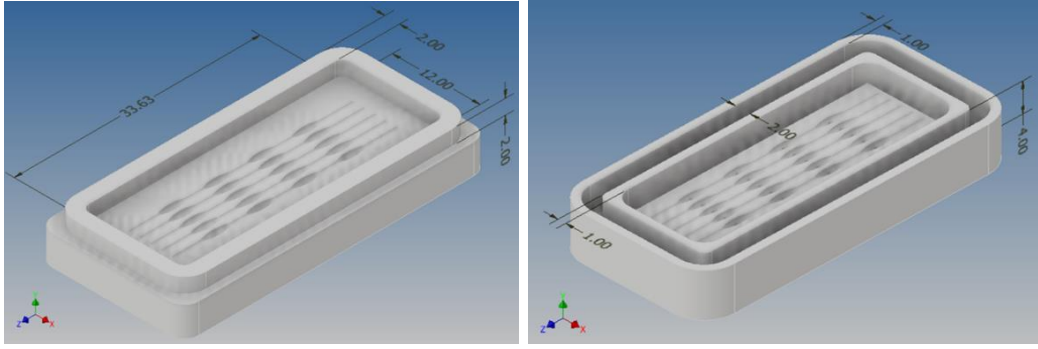


Figure 23. First mold design. All the numbers show in the figure are in millimeter. LEFT: Mold base; The optical fiber will be placed in each of the cavity, and the mold design can adjust the spindle-knot dimensions. RIGHT: Mold top will have identical design as the mold base. Besides, each spindle-knot cavity will have holes for liquid injection.

In Figure 24, the right figure captures the formed spindle-knot structure around the optical fiber. However, the formed spindle-knot is not rigid, and it can easily break into PMMA powder. This result has been the best result by using this method. In many other trials, the spindle knots are still in liquid form even after 24 hours of waiting. Another downside of this method is that the injected solution run all over the mold, and the mold cannot be completely sealed.

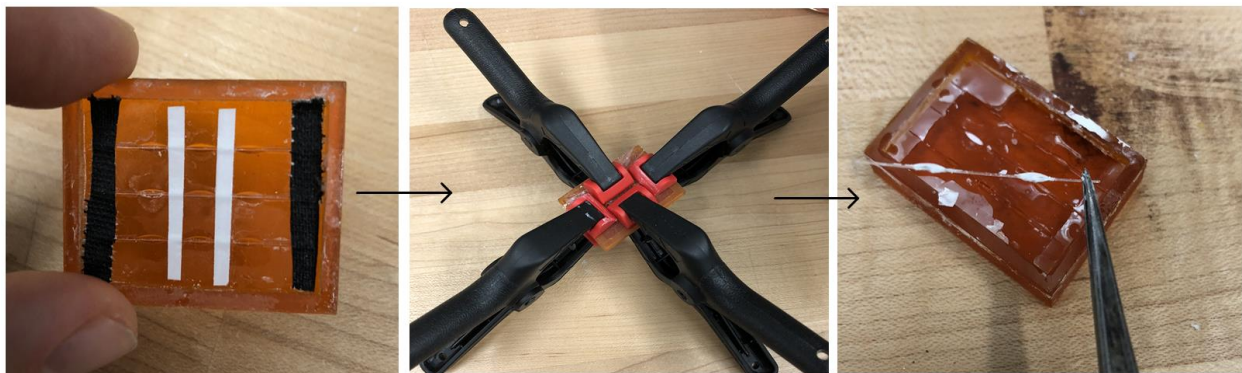


Figure 24. Second mold design. This design is similar to the first design with less material. It also has a top and bottom plate, and the top plate has the projection holes. The right figure is one of the formed spindle-knot.

2.4 3D printing

3D printing has been the hottest subject for a long period, and it is keeping growing its influences all over the places such as vessels [18], house, shoes. Using 3D printing to form spindle knots structure give us more control over the size and spacing. In this method, there are two sub-methods – fiber with spindle-knots and spindle-knot only. Those two sub-methods use the same UV curable resin – PEGDA. Pure PEGDA is transparent, and 0.05% of Sudan is added to appear as an orange color.

2.4.1 Fiber with spindle-knot

The fourth method uses a 3D printer to print out fibers and spindle-knots, refer to Figure 25. In the same printing platform, there are two spindle-knots sizes with the same spacing and same fiber diameter. The top layer shows good printing results, and we pick and observe four printed spindle-knots under the microscope shown in Figure 26. Spindle-knots are well-printed, and dimensions are consistent. Then we place the humidifier at the front to observe the water collection and water droplet directional movement. Those movements are shown in Figure 27. At 38 seconds, water droplets start to land on joint and spindle-knot regions. Water droplets keep growing, and at 80 seconds, water droplet 1 starts to move toward the spindle-knot region. After 8 seconds, water droplet 1 fully lands on the spindle-knot region and forms a larger water droplet with water droplet 2. Since the fiber dimension is in the micrometer, it cannot provide enough support for the second layer. From Figure 26, the top section has better spindle-knot printing quality and quantity than the bottom layer. The bottom spindle-knot is misaligned, and adjacent spindle-knots are all clustered due to low connection support. As a result, 3D printing can only do small scale fabrication.

From the material side, we save the solution preparation time and spindle-knot evaporation time. In addition, the printing material is less harmful compared to DMF. By using this method, no additional optical fibers need to be purchased. From the setup point, the spindle-knot dimensions and spacing between each spindle-knot can be fully controlled. The major downside is its high requirements for professional printing device and printing skills. As we have mentioned before, this method can only do small scale fabrication due to low fiber support for different layers.

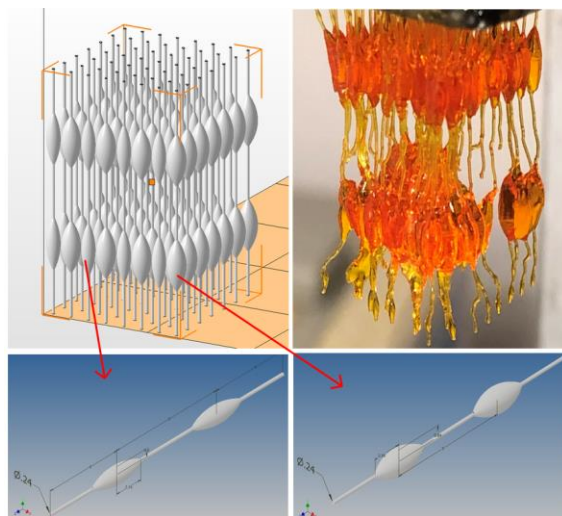


Figure 25. 3D printed spindle-knot and joint regions. Printed direction from bottom to the top. (by Zhenpeng Xu) There are two different sizes with the same fiber size. The bottom left spindle-knots are smaller than the bottom right spindle-knots.

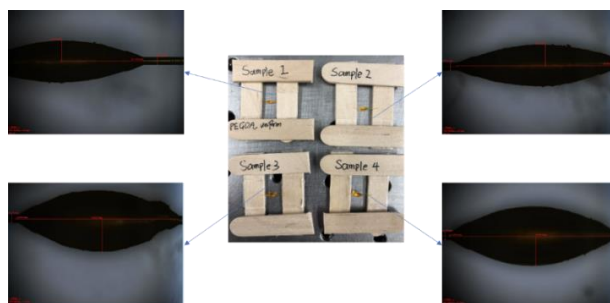


Figure 26. Spindle-knots under the microscope. Samples 1 and 2 have the same dimensions, and samples 3 and 4 have similar dimensions.

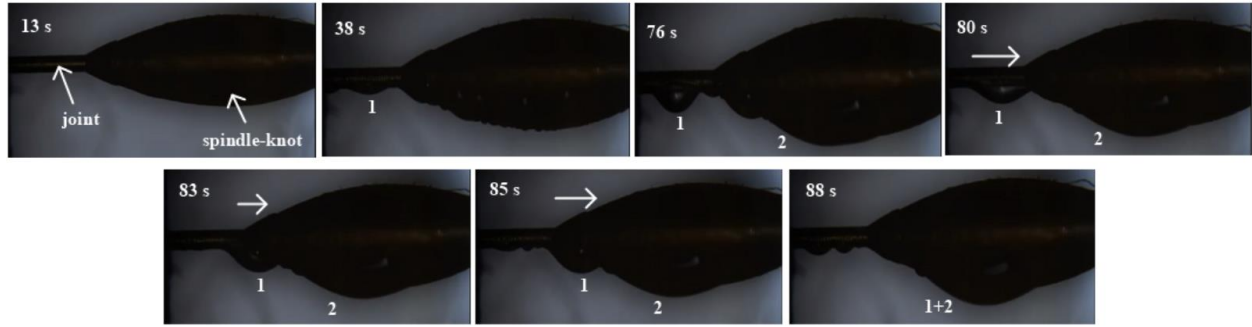


Figure 27. Water collection and movement on 3D printed spindle-knots and joints. The humidifier is on at 13 s, after 15 seconds, the water droplets start to form on the joints and spindle-knots. Water droplet 1 forms on joints region, and water droplet 2 forms on the spindle-knot region. At 80 seconds, the water droplet 1 starts to move uphill toward water droplet 2. After 8 seconds, water droplet 1 and water droplet 2 form a larger water droplet.

2.4.2 Spindle-knot only

The previous method directly prints spindle-knot and joint regions, but the joint regions are misaligned due to weak fiber support. To improve the printing quantity, we decide to print the spindle-knot region with inner opening, and then assemble the spindle-knots with optical fibers. Spindle-knot design is shown in Figure 28, from the first image, the spindle-knots are printed from bottom to the top with a thick supporting layer.

The spindle-knots are well designed and well printed; however, the resolution for the inner opening is too small. Due to the small, aspheric, and rough surface, the optical fiber will not be able to penetrate through the printed spindle-knots. Increasing the spindle-knot inner opening diameter will make the optical fiber easier to penetrate. However, fixating the spindle knot in a specific location will be a new challenge. If the spindle-knots are not well attached to the optical fiber, collected water might go into the openings instead of moving on top of the spindle-knots.

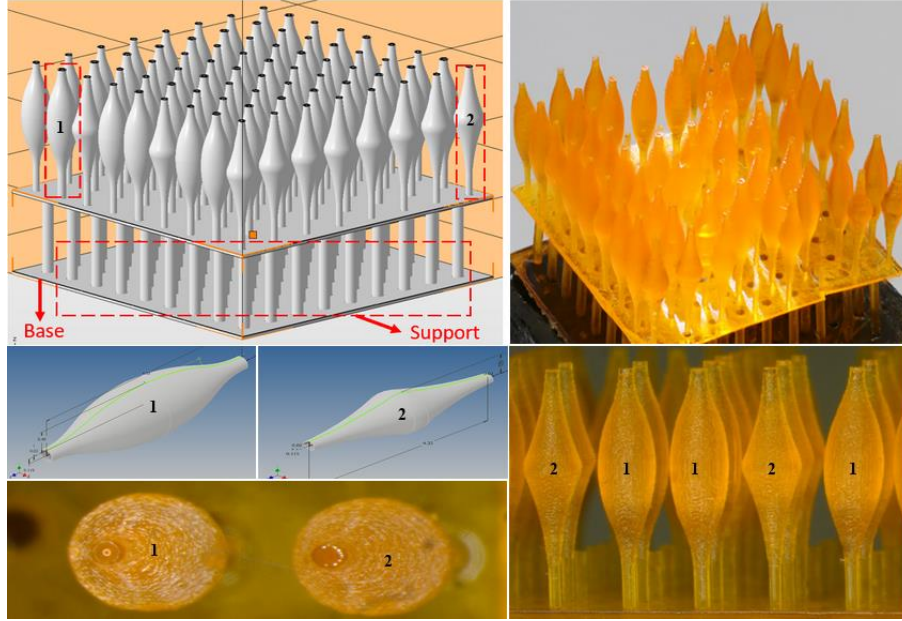


Figure 28. Spindle-knot only 3D design. This method prints two different hollow spindle-knots (denoted by 1 and 2). It has two printing parts – support region and spindle-knots regions, and it is printed from the bottom to the top layer by layer. Since our joint regions are in micrometer, the supporting region will provide additional force to hold the spindle-knots.

2.5 Light curing

PEGDA is UV curable resin, and the PEGDA liquid will become solid in a few seconds under UV light. By this unique feature, replacing the PMMA solution with PEGDA solution will save preparation time and evaporation time. In addition, the heating device is not required by using photocurable material.

2.5.1 Performance testing

To verify this method will form the spindle-knots and direct water droplets toward the spindle-knot region, we first measure and compare the surface contact angles. In Figure 29, the PEGDA surface forms a smaller surface contact angle than the PMMA surface does. In other words, the PEGDA surface is more hydrophilic than the PMMA surface. Then we used a simplified fluid-coating method to build the spindle-knots. We simply add two drops of PEGDA

in the wire cutter. Then we use the wire cutter to remove the optical fiber coat. As soon as the spindle-knots are formed due to Rayleigh's Instability, we use UV light to form solid spindle-knots around the optical fibers. In Figure 30, multiple spindle knots are formed around the optical fiber, and most of them have an identical size.

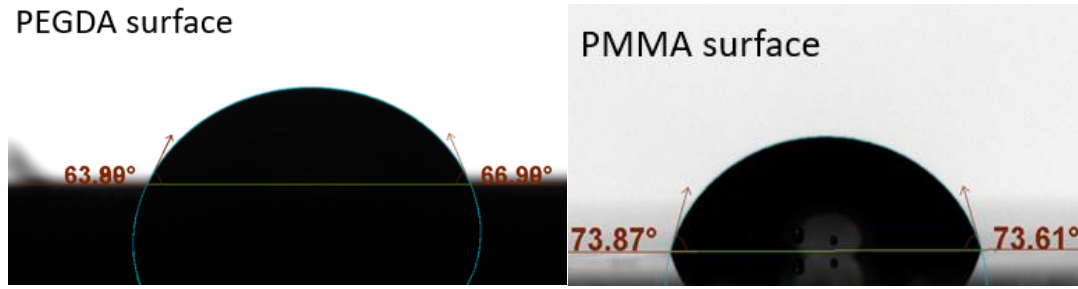


Figure 29. Smooth surface contact angles measurement. One water droplet land on PEGDA and PMMA surface and forms two different angles. PEGDA surface has smaller contact angles, and it is more hydrophilic than the PMMA surface.

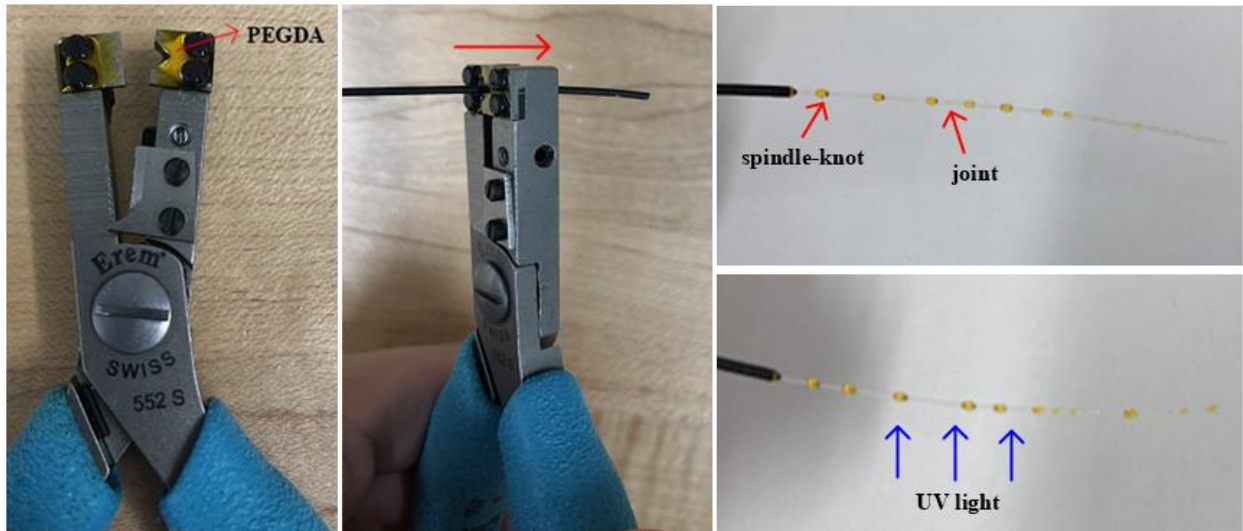


Figure 30. Spindle-knot formation by PEGDA. Two drops of PEGDA solution is placed on the wire clip. The wire clip cut depth can be adjusted by turning the screw. Prepare a short piece of coated optical fiber, then cut the coat off by the adjusted wire clip. Hold the optical fiber steady and move the clip to the right. Right after that, use UV light to form solid spindle-knots.

After forming the spindle-knots, we place a small section of fiber on the holder and do the water droplet collection and movement under the microscope. This sample contains multiple

spindle-knots, and Figure 31 shows only two of the spindle-knots. Base on the figure below, water starts to form at 0 seconds. After 4 seconds, water droplet 2 moves uphill and form a larger droplet with water droplet 3. Meanwhile, water droplet 5 moves toward water droplet 4 and form a larger water droplet. New water droplets start to form, and those droplets grow over time. When we move the fiber to the right, two spindle-knot regions have larger water droplets than the joint regions.

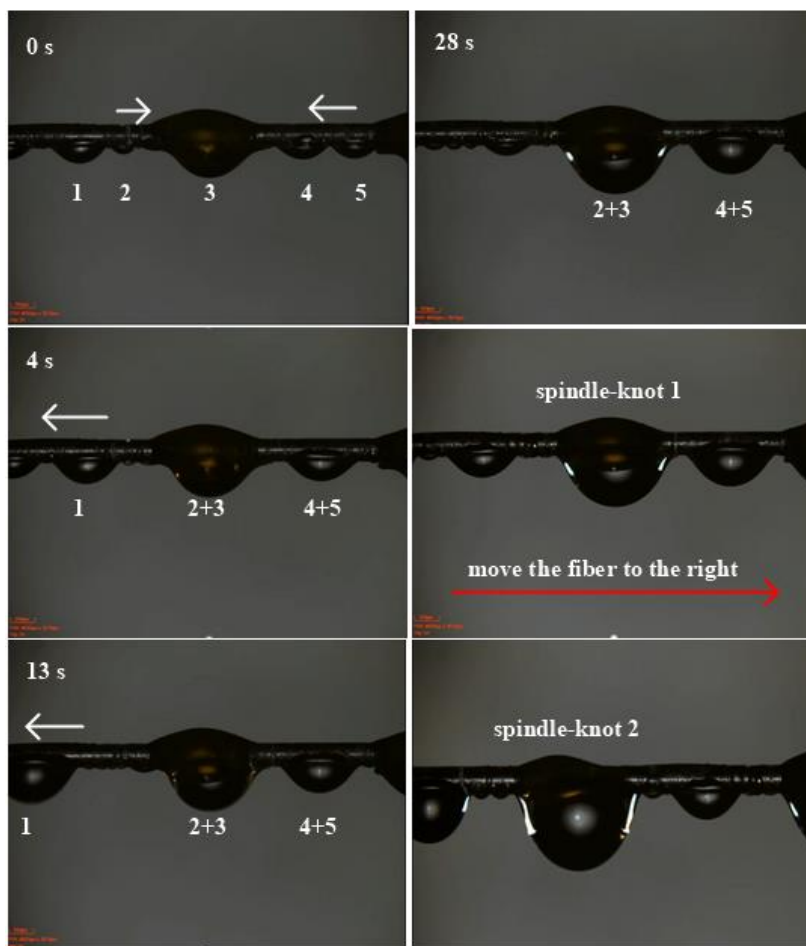


Figure 31. Water Collection and Movement on PEGDA knots. The sample has a couple of spindle-knots, and two spindle-knots are showing in this figure. Water droplet 5 moves toward left and merge with water droplet 4 to form a larger water droplet. Meanwhile, water droplet 2 is moving uphill toward the spindle knot. Water droplet 1 moves toward the left spindle-knot. Water droplets grow over time, and the spindle-knot regions collect more water than the joint region.

We use ImageJ to measure the water droplet and spindle knot dimensions. With the measured water droplet dimensions, we can calculate two water droplet volumes as,

$$V_{w_sk} = \frac{2}{3}\pi \left(\frac{1445.89 \text{ } \mu\text{m}}{2} * \frac{1 \text{ mm}}{1000 \text{ } \mu\text{m}} \right)^3 = 0.791 \text{ mm}^3 \quad (21)$$

$$V_{w_j} = \frac{2}{3}\pi \left(\frac{1020.544 \text{ } \mu\text{m}}{2} * \frac{1 \text{ mm}}{1000 \text{ } \mu\text{m}} \right)^3 = 0.278 \text{ mm}^3 \quad (22)$$

where w_sk and w_j stands for water on spindle-knot and joint. Since the water droplet on the spindle-knot region covers half of the spindle-knot, then we use Inventor to calculate the spindle-knot volume (see Figure 33). We subtract the half spindle-knot volume from the spindle-knot water droplet volume, and get the new water droplet volume,

$$V_{w_sk} = 0.791 \text{ mm}^3 - \frac{0.067}{2} \text{ mm}^3 = 0.7575 \text{ mm}^3 \quad (23)$$

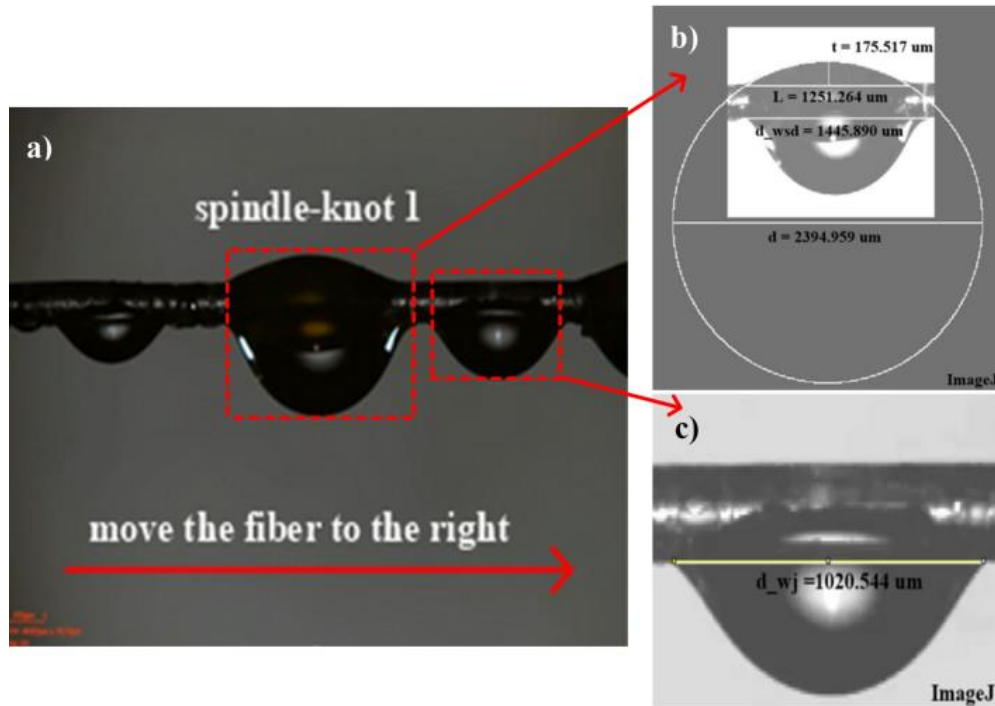


Figure 32. Water droplet dimensions by ImageJ. Two water droplets – one on the joint region and another on the spindle-knot region- are picked from the figure a. Figure b and c shows measured dimensions of water droplets on picked regions.

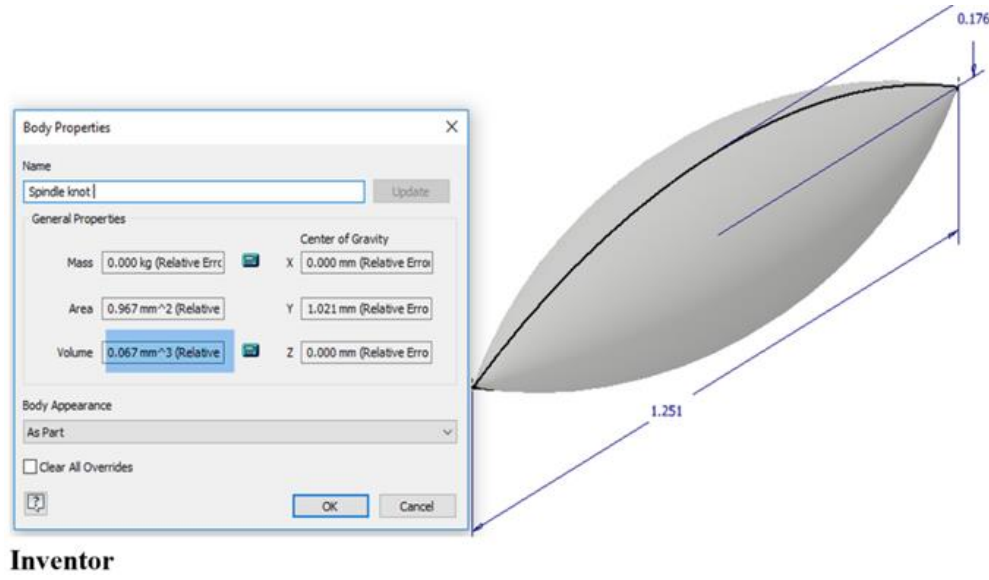


Figure 33. 3D Inventor model for spindle-knot. Measured dimensions are used in Inventor to build the spindle-knot structure. The volume of the modeled spindle-knot is 0.067 mm³.

Therefore, we compare the water droplet volumes between spindle-knot location and joint location. In equation 24, water quantity on the spindle knot region is about 2.72 times greater than the quantity on the joint region.

$$\frac{V_{w-sk}}{V_{w-j}} = 0.7575:0.278 \approx 2.72 \quad (24)$$

To be more accurate about the water collection quantity, we prepare two fibers – one with spindle-knot and another without spindle-knot. Figure 34 shows the general setup for collecting and comparing water quantity. The optical fiber is attached to the U-shape holder, while the ultrasonic humidifier is placed at the front. The water collector is set right below the fiber to catch water droplets. Dry-wipe is attached at the back of the U-shape holder to catch water droplets that land on the holder. Lab room temperature is 25 degrees Celsius, and humidity ratio is around 16%, under this same operational condition, we applied fog to each fiber for 10 minutes, and repeat the same procedures for five times. After each testing, we scale weight

collected water quantity and record the measured value in Table 2. Using optical fiber with spindle-knots increases the water collection quantity by 16 percent.

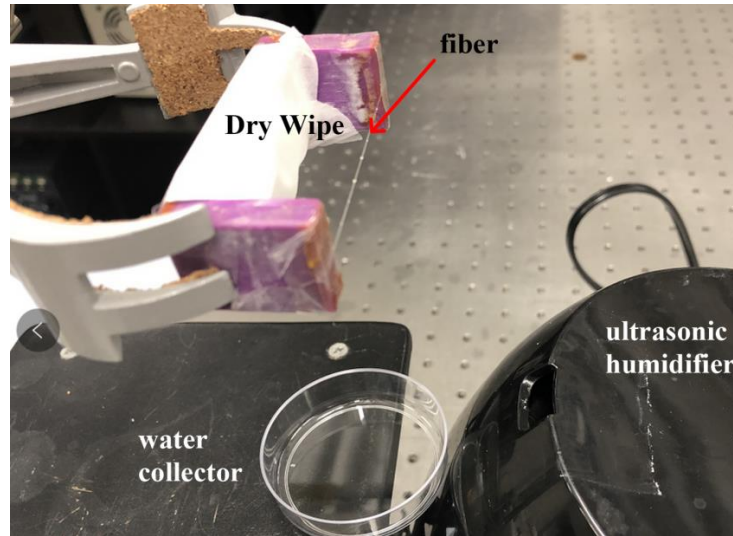


Figure 34. Water quantity comparison setup. Optical fiber is placed on the U-shape holder, and an ultrasonic humidifier is placed in front of the fiber. Dry wipe is used to catch water droplets that land on the holder.

Table 2. Collected water quantity comparison between an ordinary optical fiber and spindle-knot fiber. Fiber with spindle knot can increase the water collection quantity by 16%.

	Fiber without spindle-knots(grams)	Fiber with spindle-knots (grams)
1	0.1501	0.1666
2	0.1528	0.1724
3	0.1526	0.1765
4	0.152	0.1833
5	0.1513	0.1835
Mean	0.15176	0.17646
Percentage	$(0.17646 - 0.15176)/0.15176 * 100\% \approx 16.28 \%$	

According to the formed spindle-knot shape and water collection and movements, replacing the PMMA solution with PEGDA is a feasible, more straightforward, and time-saving method. In the following sections, we will introduce updated methods by using PEGDA.

2.5.2 Fluid coating with PEGDA

We will use the existing fluid-coating setup with two minor changes. In the solution container section, the pre-prepared PMMA solution is replaced by UV curable resin PEGDA. The last section will integrate a UV light to solidify spindle-knots. By using the PEGDA, we further eliminate the PMMA solution preparation time and spindle-knot evaporating time. Likewise, it further saves the heating energy used during PMMA solution preparation. From the safety perspective, PEGDA does not require high standard operating conditions as DMF does.

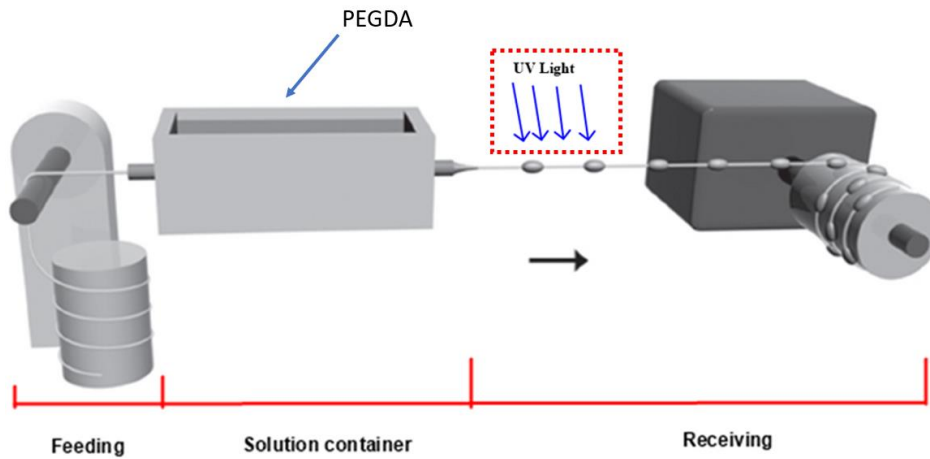


Figure 35. Fluid coating with PEGDA. PEGDA replaces the PMMA polymer solution in the solution container. In addition, UV light is added in the receiving section to turn the liquid into solid.

2.5.3 Manually formed spindle-knots

Another method we have tried is manually dripping. This method has the most straightforward setup materials— optical fiber, PEGDA, UV light, double tapes, scotch tape,

pipette (Fisherbrand Elite), and a holding frame. First, plastic frames are attached to double tapes. Then, manually circle the optical fiber around the frame. After that, add one more layer of scotch tape to hold the optical fiber in place. The following step is using a pipette to drip the PEGDA solution around the optical fiber. The pipette can adjust its liquid volume from 0.2 to 2 in microliter. The size of the spindle knot is fully controlled by the pipette volume while dripping the PEGDA. Right after attaching the liquid PEGDA on the optical fiber, use UV light to get solid spindle-knots, as shown in Figure 36.

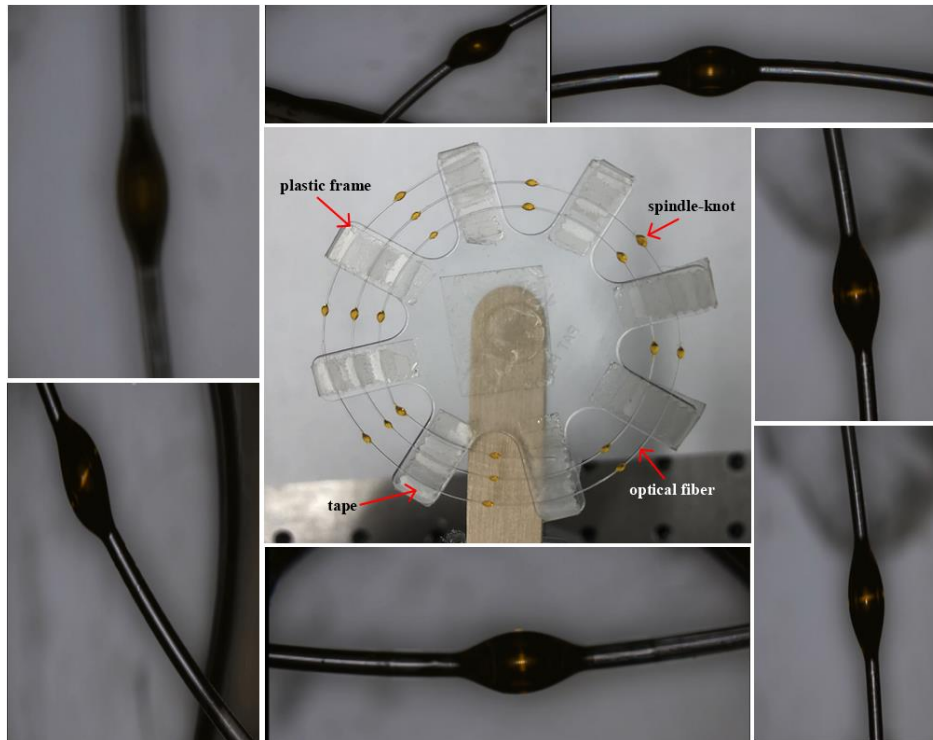


Figure 36. Manually formed spindle-knots. Spindle-knots are formed manually by dripping the PEGDA solution around the optical fiber and use UV light to obtain solid spindle shape.

In Figure 37, the spindle knot is located at the lowest point of the optical fiber. At 5 seconds, water droplet lands and grows on the joint and spindle-knot regions. Small water droplets formed right next to the spindle-knot regions continuously move towards the knot. Water droplet 3 starts to move to its left after 7 seconds, and it fully lands on the knot region at

18 seconds. Meanwhile, new water droplets land on the joint location, and the next directional movement cycle begins. We also move around the manually formed web to compare the water quantity. In the second column of Figure 36, all the spindle-knots contain larger water quantity.

The spindle-knots are fully formed manually; however, those procedures can be quickly learned by machine learning. Then those procedures can be operated automatically to output more precise location and size. In addition, lots of uncertainties and errors caused by human operating can be further eliminated.

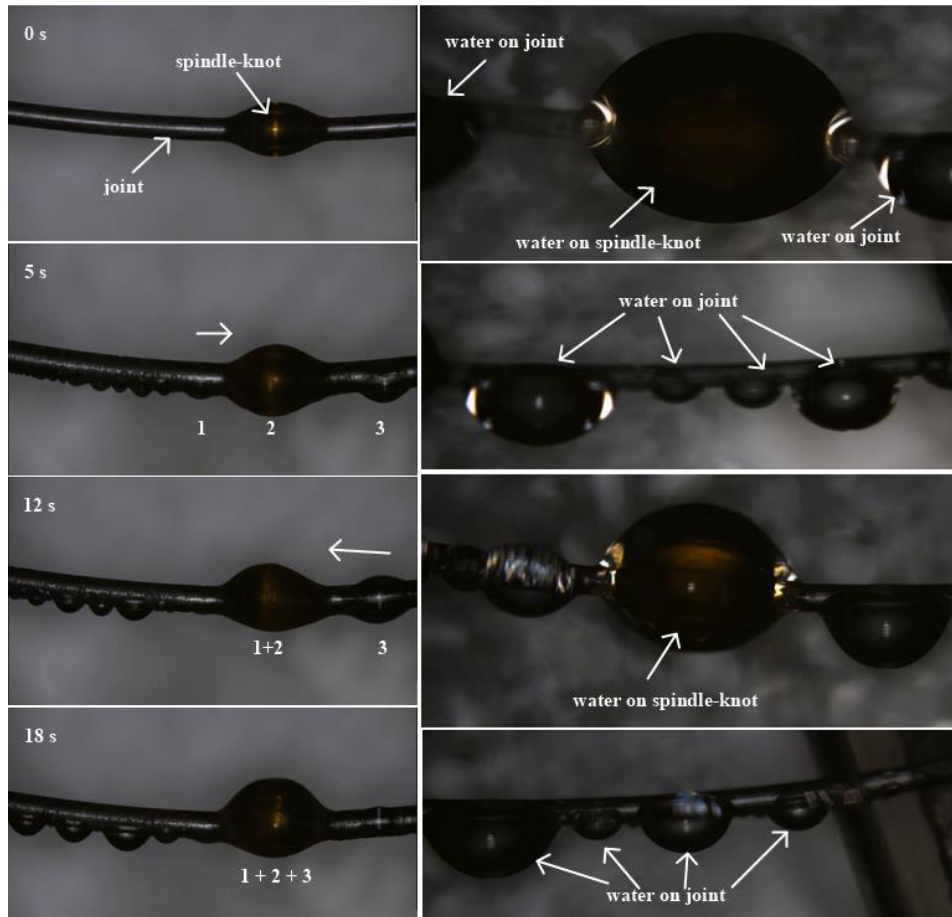


Figure 37. Water collection and movement on manually formed spindle-knot. Droplets 1 and 2 on the joint move toward the spindle-knot and this movement takes about 18 seconds. Meanwhile, new droplets start to form on the joints and move uphill. The second column of pictures shows the spindle-knot can collect more water than the joint region. It further approves spindle-knot structure can attract water from the side.

2.5.4 Dip-coating with PEGDA

The last method we have tried is dip-coating with PEGDA. We prepare a container with a thin layer of PEGDA solution (it can be any photocurable solution) and dip the optical fiber frame into the container. Wait for about 10 seconds, we lift the frame with constant speed, without flip the frame, we shine the UV light directly on the formed spindle-knots. Dip-coating with PEGDA can form a large number of spindle-knots. However, curing the PEGDA creates a thick layer of PEGDA on the frame, refer to Figure 38. The right figure in Figure 38 shows the comparison between dip-coating and manually dripping. The dip-coating forms PEGDA on all the frames, and it looks less neat. On the other hand, the manually dripping side is clean and more organized.



Figure 38. Dip-coating with PEGDA drawback. LEFT: Dip-coating creates a thick layer of PEGDA all over the plastic frame. RIGHT: Compare dip coating (top half) with manually dripping (bottom half), the bottom half is neater than the top half.

Chapter 3. Discussion

Fluid coating with DMF is the reference method, and we value each method by one constraint and six objectives. Those six objectives are solution preparation time, spindle-knot formation time, less harmful material, low cost, large scale fabrication, and no professional

operator requirement. Moreover, the only constraint is the successful spindle-knot formation rate. According to the table, molding casting and spindle-knot only methods do not meet the constraint. The remaining methods can successfully form the solid spindle-knot around fiber and direct water droplet toward the knot regions. Methods with water/ethanol solvent are the least harmful method; however, they require longer solution preparation and spindle-knot evaporation time. Methods with PEGDA solution are the top three methods since PEGDA is less harmful, and it can be cured by UV light just in few seconds. Among those top three methods, fluid-coating with PEGDA and manually dripping method have the highest score due to their large-scale fabrication.

Table 3. Method Comparison Table. Fluid coating with DMF is the reference, and each method is scored based on one constraint and six objectives.

Methods	Success	Solution Preparation Time	Spindle-knot formation time	Less Harmful material	Low Cost	Large scale fabrication	No professional operator required	Total
FC with DMF	YES	0	0	0	0	0	0	0
DC with DMF	YES	0	0	0	0	-1	0	-1
FC with water/ethanol	YES	-1	-1	1	0	0	1	0
DC with water/ethanol	YES	-1	-1	1	0	-1	1	-1
FC with PEGDA	YES	0	1	1	0	0	1	3
DC with PEGDA	YES	0	1	1	0	-1	1	2
Mold Casting	NO	-1	-1	1	0	-1	1	NO
3D printing	YES	0	0	1	0	-1	0	0
Spindle-knot only	NO	0	0	1	0	1	0	NO
Manually dripping	YES	0	1	1	0	0	1	3

Chapter 4. Conclusion

Water scarcity has been an increasing challenge, and the fog collection method has been widely used in low precipitation areas. Existing fog collection meshes are inexpensive and straightforward; however, water droplets usually get pinned on the mesh. To reduce the pinning issue and to increase the water collection quantity, the water droplet directional movement

inspired by spider silk has been learned and tested. Surface tension force, hysteresis force, and Laplace pressure force are three controlling forces that act around the water droplets to cause the movement. With the support of experimental results from Lei Jiang's group, surface material and surface material are controlling the water droplet moving speed and moving direction. For a smooth and flat surface, water droplet moves toward spindle-knot regions if spindle-knot is composed of more hydrophilic material. Similarly, for homogeneous and flat surface, water droplet moves toward knot regions if the spindle-knot has higher surface roughness. Spindle-knot regions should be more hydrophilic than the joint region to increase the water collection rate. Using more hydrophilic material or creating higher surface roughness are two common approaches to increase the hydrophilicity. Forming the spindle-knot structure is critical, and existing spindle-knot formation methods are fluid-coating and dip-coating by PMMA powder and organic compound DMF. Hazardous DMF can be replaced by 80% water/20% ethanol solvent to dissolve PMMA powder fully. By using water/ethanol solvent, it usually takes 20 to 30 minutes to dissolve PMMA powder, and 12 hours to form solid spindle-knots. Besides, a heating source is required, and the heating temperature should be maintained above 80 degrees Celsius. To solve those three major downsides, photocurable material PEGDA has been used and tested to form the spindle-knot regions. The PEGDA surface contact angle is measured first, and it is more hydrophilic than the PMMA surface. Spindle-knots can successfully and quickly be formed around the 250-micrometer optical fibers by using PEGDA. Then we run some water quantity tests on those formed knots. The water droplet on the spindle-knot region is 2.72 times greater than the water droplet volume on the joint. In addition, an optical fiber with spindle-knots can collect 16% more water than the optical fiber without spindle-knot. We further compare each method according to one constraint and six objectives with the existing fluid-coating method.

Based on the method comparison table, using photocurable material is the winning method. However, it is cured with UV light, and it will remain sensitive to UV light while exposed outside. Reducing the UV light sensitivity of cured photocurable material will be a new challenge to increase spindle-knot performance further.

Appendix A. Freshwater is Abundant [4]

From Hoekstra and Chapagain's water footprints of nations (between 1997 - 2001), the global water footprint average is 1240 m^3 per capital per year. World population is around 7,000,000,000, and then the total water usage could be calculated as,

$$W_{need} = (7 * 10^9 \text{ capita}) * \left(1240 \frac{\text{m}^3}{\text{capita} * \text{year}}\right) = 8.68 * 10^{12} \frac{\text{m}^3}{\text{year}} \quad (\text{A1})$$

During the hydrological cycle, water will evaporate into the sky and then become precipitation to get back on land and sea. The global annual average precipitation is around 0.99 meters, with the total land area $1.48 * 10^{14}$ in unit of square meter, the global annual precipitation volume is,

$$W_{prec} = \left(0.99 \frac{\text{m}}{\text{year}}\right) * (1.48 * 10^{14} \text{m}^2) = 146 * 10^{12} \frac{\text{m}^3}{\text{year}} \quad (\text{A2})$$

The ratio between the annual precipitation and the required freshwater will be,

$$W_{prec} : W_{need} = 146 : 8.68 = 16.82 : 1 \quad (\text{A3})$$

The ratio can tell that the freshwater is abundant for human needs.

Appendix B. Surface Tension Force Due to Material Gradient

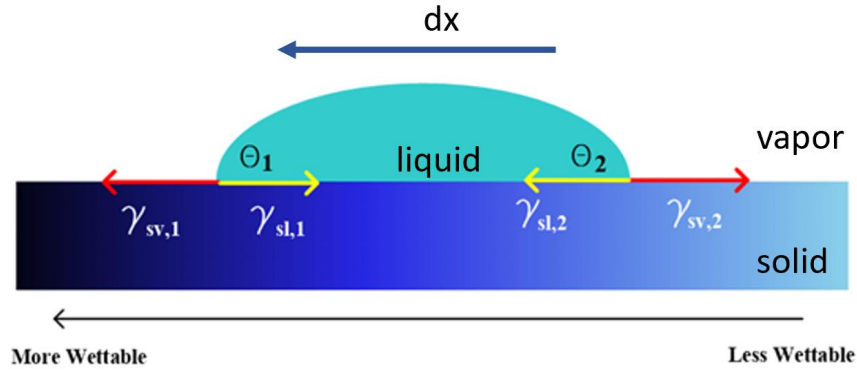


Figure B1. Surface tension force due to material gradient. The droplet sits on a smooth and material varying solid surface. The surface tension raises due to the different material wetting properties. The smooth surface contact angles at the front end and rear end are denoted by θ_1 and θ_2 .

Based on the figure above, the surface tension force on each contact surface can be written as,

$$dF_{sv,1} = \gamma_{sv,1}(dx) \quad \text{B1}$$

$$dF_{sl,1} = -\gamma_{sl,1}(dx) \quad \text{B2}$$

$$dF_{sv,2} = -\gamma_{sv,2}(dx) \quad \text{B3}$$

$$dF_{sl,2} = \gamma_{sl,2}(dx) \quad \text{B4}$$

Adding up the forces together from equation A1 to equation A4,

$$dF_{\gamma,m} = (\gamma_{sv,1} - \gamma_{sl,1})dx - (\gamma_{sv,2} - \gamma_{sl,2})dx \quad \text{B5}$$

Young's equation (Appendix F1 for simple explanation) states the following relation between the surface tensions and surface contact angles,

$$\gamma_{lv} \cos \theta = \gamma_{sv} - \gamma_{sl}$$

Substitute the Young's equation into equation A5,

$$dF_{\gamma,m} = \gamma_{lv}dx(\cos \theta_1 - \cos \theta_2) \quad \text{B6}$$

Take integral on both side from point x_1 to point x_2 where the fog landed on the spindle knot and joint accordingly,

$$F_{\gamma,m} = \gamma_{lv} \int_{x_1}^{x_2} (\cos \theta_1 - \cos \theta_2) dx \quad \text{B7}$$

The contact surface is a circular, so we can derive the equation B7 into equation B8,

$$F_{\gamma,m} = \gamma_{lv} (\cos \theta_1 - \cos \theta_2)_m * 2\pi R_0 \quad \text{B8}$$

Where R_0 is fog droplet radius in millimeter.

Appendix C. Surface Tension Force Due to Surface Roughness Gradient

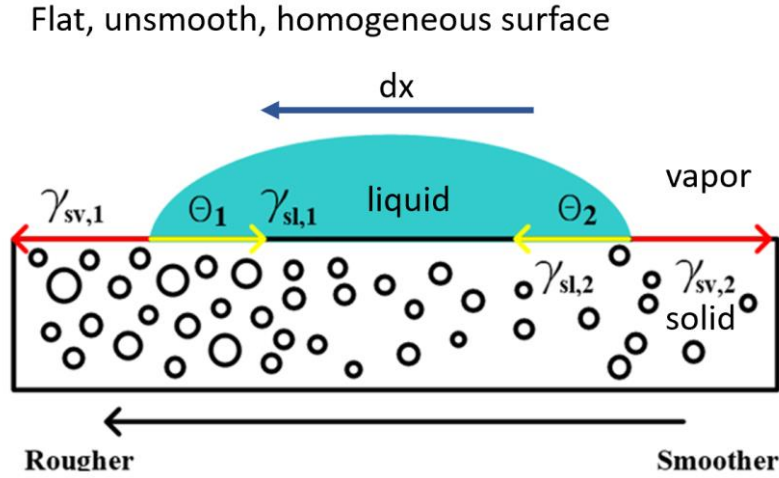


Figure C1. Surface tension force due to roughness gradient. Droplet on a homogeneous (same hydrophilic material) and rough surface. The surface tension rises due to roughness gradient along the contact surface. The front end is rougher than the rear end. In general, rougher surface for the same material is more water liking. As a result, the surface contact angle 1 is smaller than contact angle 2.

It is reasonable to assume the droplet will move dx along the perimeter toward the more hydrophilic region – the front region in Figure C1. Then the surface tension forces for each contact surface can be written as following,

$$dF_{sv,1} = \gamma_{sv}(r_1 dx) \quad C1$$

$$dF_{sl,1} = -\gamma_{sl}(r_1 dx) \quad C2$$

$$dF_{sv,2} = -\gamma_{sv}(r_2 dx) \quad C3$$

$$dF_{sl,2} = \gamma_{sl}(r_2 dx) \quad C4$$

where r_1 and r_2 represent the dimensionless surface roughness on front and rear half (surface roughness = total length/projected length), γ_{sv} is the solid air surface tension coefficient in unit of Nm^{-1} , and γ_{sl} is the solid liquid surface tension coefficient in unit of Nm^{-1} . The next step is to sum the forces from equation C1 to C4 and rearrange them in equation C5 form,

$$dF_y = (\gamma_{sv} - \gamma_{sl})(r_1 dx - r_2 dx) \quad C5$$

From Young's relation, the relationship between liquid air surface tension coefficient, solid liquid surface tension coefficient, and solid air surface tension coefficient can be represented as,

$$\gamma_{lv} \cos \theta = \gamma_{sv} - \gamma_{sl}$$

where the angle θ is the contact angle on ideal smooth surface. Plug in the young's relation in equation C5, and the new total force will be as,

$$dF_{y,r} = \gamma_{lv} \cos \theta (r_1 dx - r_2 dx) \quad C7$$

Multiply each term, and take out dx term, the equation can also be in the following form,

$$dF_{y,r} = (\gamma_{lv} r_1 \cos \theta - \gamma_{lv} r_2 \cos \theta) dx \quad C8$$

Wenzel's Law (Appendix F2 for simple explanation) describe the relationship between contact angle on a rough surface, ideal smooth contact angle and surface roughness. In our case above, θ_1 and θ_2 are two contact angles on rough surfaces with surface roughness r_1 and r_2 respectively. Based on Wenzel's law, we can show the following expressions,

$$\cos \theta_1 = r_1 \cos \theta \quad C9$$

$$\cos \theta_2 = r_2 \cos \theta \quad C10$$

The next step is to substitute those two terms in equation C8,

$$dF_{y,r} = (\gamma_{lv} \cos \theta_1 - \gamma_{lv} \cos \theta_2) dx \quad C11$$

Take integral on both side from point x_1 to point x_2 where the fog landed on the spindle knot and joint accordingly,

$$F_{\gamma,r} = \gamma_{lv} \int_{x_1}^{x_2} (\cos \theta_1 - \cos \theta_2) dx \quad \text{C12}$$

Since the fog droplet is not two single points, instead, the fog droplet will form a circular contact surface with the solid. Based on the circular surface, we need to sum up the surface tension force around the fog droplet edges, and equation C12 becomes,

$$F_{\gamma,r} = \gamma_{lv} (\cos \theta_1 - \cos \theta_2) * 2\pi R_0 \quad \text{C13}$$

where R_0 is fog droplet radius in meter.

Appendix D. Methods to Measure Receding and Advancing Angles on a Surface

Method 1: Inflate and Deflate

The first step is to place the water droplet on the surface of interest. Then carefully adding more water in the droplet until the water droplet starts to move. The angle right before the movement is the advancing angle. Similar approach to estimate the receding angle, instead of adding water into the droplet, we will carefully remove the water from the droplet until we see the water droplet movement. The angle right before the movement will be the receding angles.

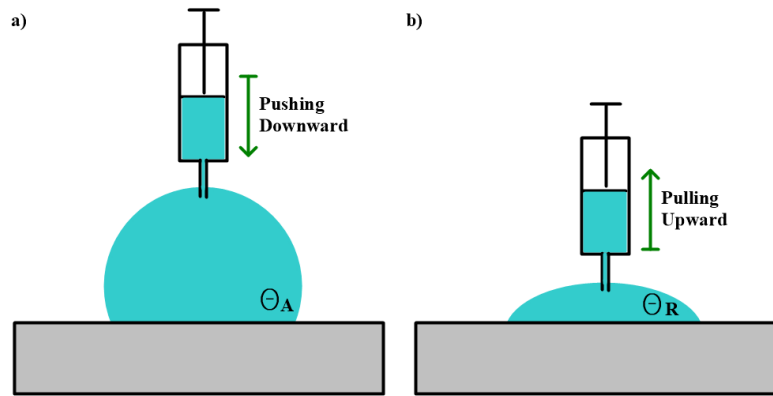


Figure D1. Advancing and receding angle measurement by inflating and deflating fluid. First method to find the advancing and receding angles by inflate and deflate the water droplet on the surface of interest. a) Inflating the water droplet by adding water and measure the advancing angle right before we see the droplet movement. b) Deflating the water droplet by removing water by syringe and measure the receding angle right before the droplet movement.

Method 2. Rising Surface

Instead of measuring the receding and advancing angles separately, we could rise the surface up to measure those two angles at the same time. First step is to place the droplet on a horizontal surface, then, we will keep rise one end of the surface until we see the droplet movement. We will keep recording the process, so it allows us to go back and measure receding and advancing angles right before the movement.

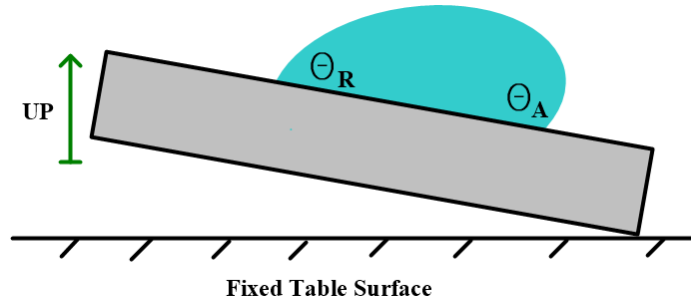


Figure D2. Angle measurement by tilted surface. Second method to measure the receding and advancing angles at the same time by rising one end of the surface until the water droplet starts to slide. Advancing angle is always larger than the receding angle.

Appendix E. Hysteresis Force Due to Unsmooth and Heterogeneous Surface

Before we jump right into the force derivation, two assumptions need to be announced and declared. The front half is experiencing the same contact surface 1, and the rear half is experiencing the same contact surface 2. The front half and rear half are two equally divided semi-circular.

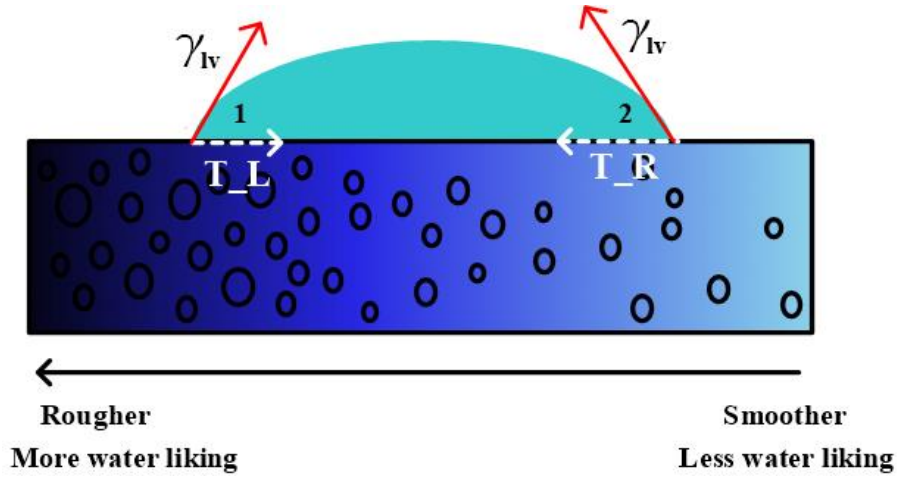


Figure E. Hysteresis force on unsmooth and heterogeneous surface. Different surface roughness and surface material (all hydrophilic) also induce hysteresis force on the contact surface. Left side is composed of more hydrophilic material and it has higher surface roughness.

The hysteresis force can be calculated as the left force subtract the force pulling toward right,

$$F_H = F_R - F_L \quad \text{E1}$$

If the resultant force is positive, the droplet will move from right to the left. On the other hand, the water droplet will move to the right if the force is negative. The hysteresis contact angle difference (HCAD) for front and rear end can be written as

$$HCAD_1 = \cos \theta_{R1} - \cos \theta_{A1} \quad \text{E2}$$

$$HCAD_2 = \cos \theta_{R2} - \cos \theta_{A2} \quad \text{E3}$$

Knowing the hysteresis angle difference, we are able to calculate the tensions on the left and right,

$$T_L = \gamma_{lv} * (\cos \theta_{R1} - \cos \theta_{A1}) \quad E4$$

$$T_R = \gamma_{lv} * (\cos \theta_{R2} - \cos \theta_{A2}) \quad E5$$

Since each tension acts on half of the circle, we could calculate the force by multiplying the perimeter,

$$F_L = \pi R_0 * \gamma_{lv} (\cos \theta_{R1} - \cos \theta_{A1}) \quad E6$$

$$F_R = \pi R_0 * \gamma_{lv} (\cos \theta_{R2} - \cos \theta_{A2}) \quad E7$$

Then the hysteresis force can be evaluated by adding up equation E6 and E7,

$$F_H = \pi R_0 * \gamma_{lv} [(\cos \theta_{R2} - \cos \theta_{A2}) - (\cos \theta_{R1} - \cos \theta_{A1})] \quad E8$$

Hysteresis force will always be negative and try to move the droplet away from the spindle knot.

Appendix F. Young's relation, Wenzel's Law & Rayleigh Instability Simple Explanation

F1. Young's Relation

Young's relation is valid for a static droplet at equilibrium or for a perfectly smooth surface. It shows the surface tension between the solid, liquid and vapor phase. Figure F1 shows a static water droplet lands on the smooth solid surface. Sum the equilibrium forces along the x direction, we can derive the equation below,

$$\Sigma F_x = \gamma_{sv} - \gamma_{sl} - \gamma_{lv} \cos \theta = 0 \quad (F1)$$

where s, v, l represents solid, vapor and liquid phase.

Rearrange the terms in equation F1, we will get the Young's relation as following,

$$\gamma_{lv} \cos \theta = \gamma_{sv} - \gamma_{sl} \quad (F2)$$

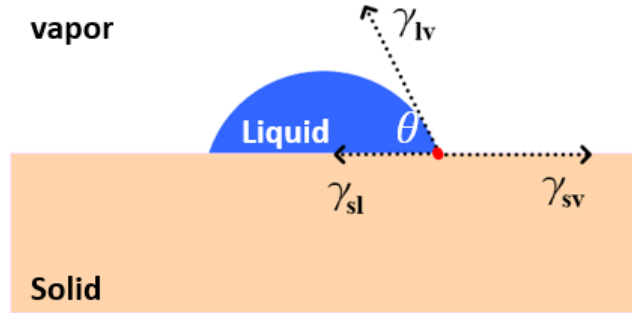


Figure F1. Young's relation. Static water droplet lands on a perfectly smooth surface. It relates the surface tension between three phases – solid, liquid and vapor.

F2. Wenzel's Law

Wenzel's law is valid only for rough homogeneous surface. In Wenzel's Law, the water droplet penetrates through the rough surface as shown in left figure in Figure F2. Wenzel's law equation can be derived by energy balance.

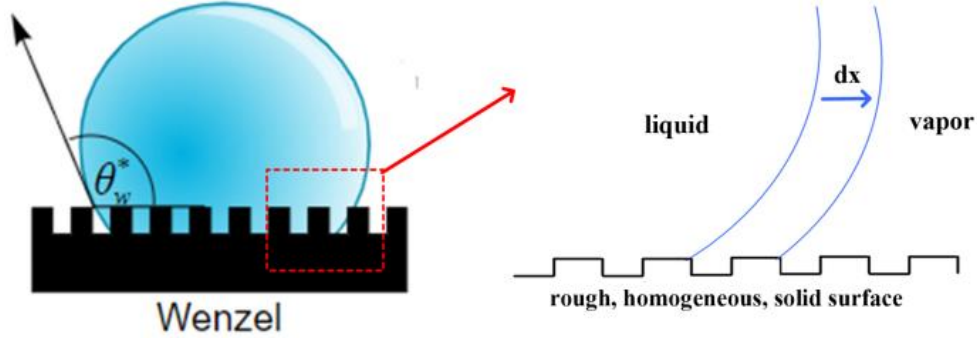


Figure F2. Wenzel's Law. In Wenzel's law, water droplet penetrates through the rough surface layer and form new surface contact angle θ_w^* (left figure courtesy by Wikipedia). The right figure shows water droplet moves dx along the perimeter, and this figure will be used to derive the Wenzel's law equation.

From the right figure in Figure F2, we assume the water droplet move dx along the perimeter towards right. Solid vapor interface is replaced by the solid liquid interface, and the energy balance equation can be represented as,

$$dE = (\gamma_{sl} - \gamma_{sv})r dx + \gamma_{lv} \cos \theta_w^* dx = 0 \quad (F3)$$

where r is the roughness ratio between total surface area and projected area. Rearrange equation and apply Young's relation, we will get the relation between smooth surface contact angle, rough surface contact angle and surface roughness,

$$\cos \theta_w^* = r \cos \theta \quad (F4)$$

F3. Rayleigh Instability

Rayleigh's instability is the balance between water surface tension and gravity. When a thin layer of fluid covers the fiber, gravity has the tendency to pull down the fluid and surface tension pulls up the fluid [15].

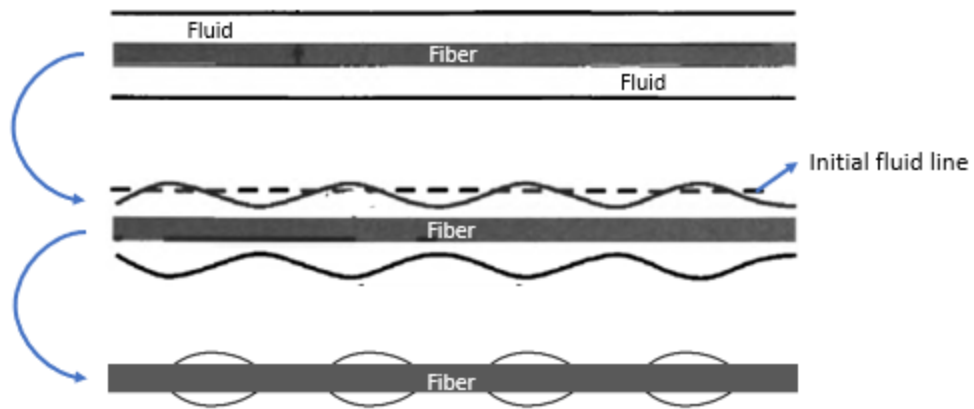


Figure F3. Rayleigh's Instability. Layer of fluid coated around the fiber will form periodic droplet along the fiber due to gravity and surface tension. Gravity will pull down the fluid, and the surface tension pull up the fluid to form droplet form fluid. [15]

Appendix G. 3D Drafted Web Frame

Frame 1:

In this frame design, half of the optical fiber can be placed in the frame. All the dimensions in the left figure is in millimeter, and each groove is 2 mm apart. This design allows us to easily remove the optical fiber after being fully evaporated. However, keeping the optical fiber in the designed location is the challenge. While placing the optical fiber along the groove spaces, making sure the space between the adjacent optical fibers should be large enough to prevent forming the liquid film.

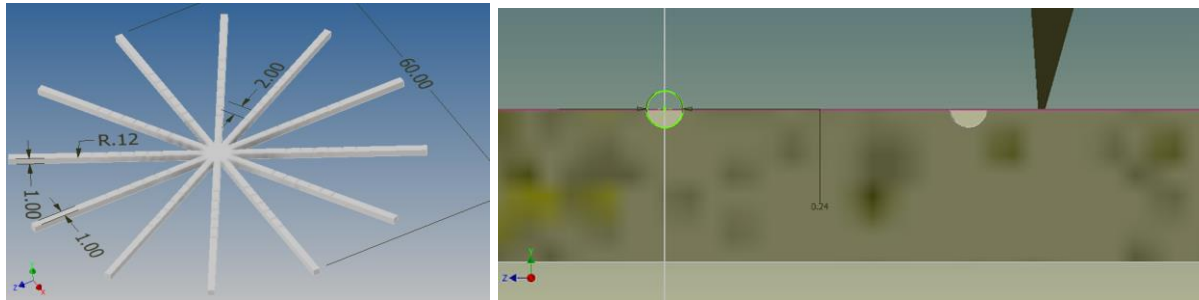


Figure G1. First spider web frame design.

Frame 2.

All the dimensions are in millimeter, and R in the left figure stands for radius. In this design, we keep the same web diameters (60 mm), but increase the adjacent groove space to 2.38 mm. In addition, the supporting frame is increased from 6 to 8 to provide support to the optical fiber. Another significant change is the groove shape. The groove depth is increased, so the optical fiber can secure in the specified space without additional forces. The downside by increasing the groove depth will give a hard removing the frame after the spindle knot is completely dried and

formed. Using flexible material like silicone gel to print the 3D frame might solve previous issue.

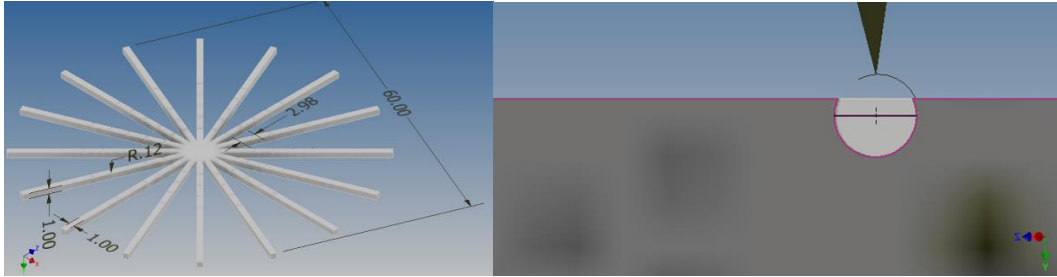


Figure G2. Second spider web frame design.

Frame 3.

This design has more supporting frames, and the frame diameter is reduced from 60 mm to 42 mm to save more material and make the structure more robust. Moreover, the groove distance is decreased down to 1.5 mm. The groove depth is further increase to secure the optical fiber in place.

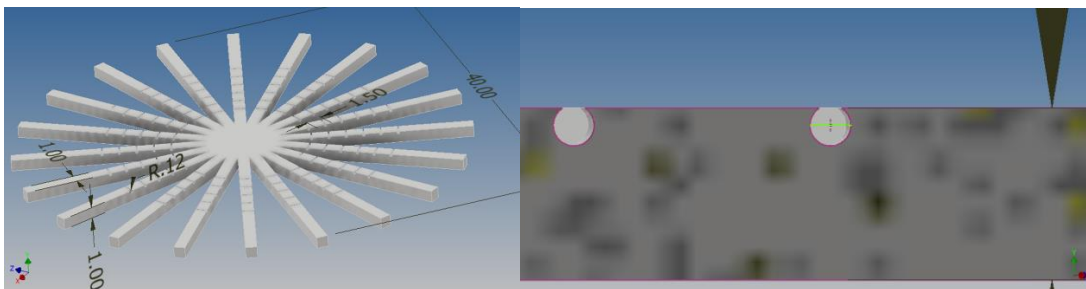


Figure G3. Third spider web frame design.

Frame 4.

Single frame is thicker from 1 mm to 2 mm to further increase the strength of the support, and each frame is shorted by 20 mm to save more materials. More importantly, the grooves are inside the frame to provide support to the optical fiber from all the sides.

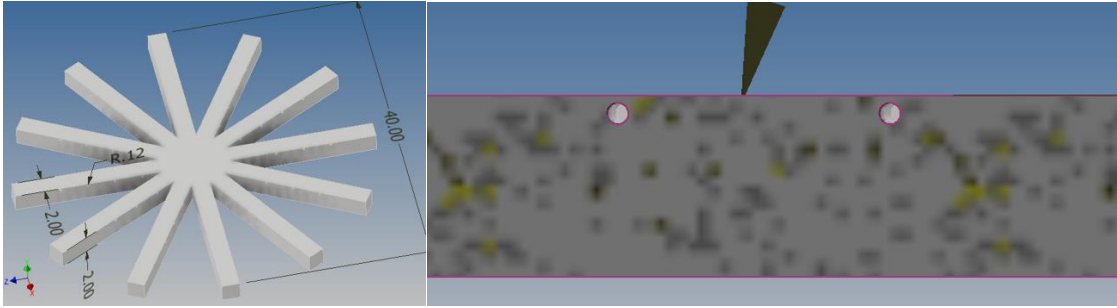


Figure G4. Fourth spider web frame design.

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