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Solvent-controlled Growth of Silicone Nanofilaments

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5 Silicone nanofilaments (SNFs) with different feature and hydrophobicity were grown on the surface of glass slides by simply regulating the solvent composition during hydrolysis and condensation of trichloromethylsilane (TCMS). Toluene and its homologues are ideal solvents for the growth of SNFs. The suppression of solvents and molecules containing N and/or O elements on the growth of SNFs provides a chance to directly observe the roots of SNFs, the infant SNFs and the growing process. The
10 roots of SNFs are formed by random immobilization of hydrolyzed TCMS and its oligomers (HTOs) onto the surface of glass slide. The HTOs could continuously anchor onto the exposed silanols of these roots under proper conditions, which conduces to their elongation and increase of the aspect ratio.

Introduction

Self-assembled monolayers (SAMs) made from organosilanes
15 have attracted considerable attention since 1980s in modifying surface properties owing to their excellent properties such as hydrophobicity and stability even under various harsh conditions.^{1,2} Both alkylchlorosilanes and alkylalkoxysilanes are frequently used to prepare SAMs on surfaces of silicon, glass and
20 mica, etc., for academic research and practical applications.³⁻⁵

There are many chances to tailor properties of the SAMs by choosing proper organosilanes (*e.g.*, the type and amount of reactive group and alkyl group) and reaction conditions.⁶ For a given organosilane, properties of the SAMs mainly depend on the
25 reaction conditions including water content, solvent and temperature.^{2,5,7} There were many studies related to hydrolysis of organosilanes, improvement in properties of SAMs and the self-assembly mechanism in the past decades. The influences of various factors on SAMs were gradually disclosed and a plausible
30 SAMs formation mechanism, hydrolysis-physical adsorption-surface condensation, was established.^{2,7,8-12} It is also well known that the polymerization of organosilanes should be avoided throughout the assembly process in order to prepare excellent SAMs.

35 Inspired by self-cleaning and water-repellent properties of the lotus leaf and the leg of water strider, superhydrophobic and superoleophobic surfaces develop very quickly¹³⁻¹⁹ and have potential applications in various fields including anti-fouling, oil/water separation and moisture collection.²⁰⁻²⁴ The combination
40 of proper surface roughness and materials with low surface energy is proved to be a successful way to prepare superhydrophobic surfaces.²⁵⁻²⁹ Versatile organosilanes are ideal procurers for preparing artificial superhydrophobic and superoleophobic surfaces.³⁰⁻³³ The reactive groups could easily
45 polymerize to generate certain surface roughness and the alkyl group could evidently decrease the surface tension.^{34,35} Tuteja

and Cohen et al. developed a series of excellent superoleophobic coatings based on polyhedral oligomeric silsesquioxane with perfluoro-alkyl groups.^{31,32} Sun et al. reported fabrication of very
50 interesting self-healing superhydrophobic coatings using fluoroalkylsilane.²⁸ Li et al. designed novel superhydrophobic sponges with the help of polydimethylsiloxane.^{33,36} A new group of nanostructures called “silicone nanofilaments (SNFs)” were developed since 2003 via polycondensation of
55 trichloromethylsilane (TCMS) on the surfaces of various substrates.³⁷⁻⁴³ The SNFs coatings exhibit excellent superhydrophobicity, chemical and environmental stability, and even superoleophobicity after further modification with perfluoroalkylsilanes. Also, the SNFs coatings are promising
60 materials in the fields of oil/water separation and photocatalysis, and have received much attention.^{44,45}

Obviously, the growth condition for SNFs must be different from that of SAMs. Polymerization of alkyltrichlorosilane, which should be avoided through strict control over water content in
65 SAMs preparation, is necessary for the growth of these unique SNFs. Although the SNFs have been successfully prepared³⁷⁻⁴⁵ and confirmed by other groups,⁴⁶⁻⁴⁸ the factors influencing their growth and the growth mechanism are not clear yet. The information we have got about them is just a tip of the iceberg
70 compared to the developed SAMs. The previous research about SAMs based on organosilanes provides us with abundant helpful information. With this idea in mind, we have successfully grown SNFs on various substrates via properly activating them.^{38,39} In addition, it was found that water concentration in solvent (toluene)
75 has great influences on structure of the SNFs, and then on superhydrophobicity and superoleophobicity of the coatings.⁴⁰

Here we report the solvent-controlled growth of SNFs on the basis of our previous work.³⁷⁻⁴⁵ SNFs with different structures were grown onto the surface of glass slides by simply regulating
80 the type and ratio of solvents during hydrolysis and condensation of TCMS. The structure of SNFs has great influences on the contact angle (CA) and sliding angle (SA) of water drops.

Toluene and its homologues are ideal solvents for the growth of the SNFs, whereas hydrophilic solvents and molecules containing N and/or O elements seriously hinder their growth. The combination of toluene with hydrophilic solvents and molecules could precisely tune the structure of the SNFs, which helps to understand their growing process and mechanism.

Experimental section

Materials

Glass slides (Menzel, Braunschweig, Germany) of 24 mm × 60 mm (1#) were used as the substrates. TCMS (97%, ABCR, Germany) was handled under water free conditions and used without further purification. Toluene (99.85%, extra dry over molecular sieve) was purchased from Acros Organics. Cyclohexane, *t*-butylbenzene, dioxane, Span 80, Tween 20 and paraffin oil were purchased from Fluka. Acetonitrile, *p*-xylene, *n*-hexadecane, methyl ethyl ketone, ethyl ether and DMSO were purchased from Sigma-Aldrich. Acetone was bought from Merck and poly(dimethylsiloxane) was purchase from Suchema AG, Switzerland.

Growth of SNFs

In brief, the glass slides were ultrasonicated for 30 min in a 10% v/v solution of Deconex 11 Universal (Borer Chemie AG) at 50 °C, rinsed with deionized water and dried under a nitrogen flow. A piece of the activated glass slide was immersed in 80 mL of solvent and certain amount of co-solvent in the custom made chamber (100 mL). The water concentration in the solvents was controlled by bubbling with the mixture of dry and wet nitrogen, and then magnetically stirred for 10 min for the water concentration to be in equilibrium. The water concentration was determined using a Compact Karl Fischer Coulometer (C20, Mettler Toledo, Switzerland).⁴⁰ Subsequently 80 μL of TCMS was injected into the chamber at 25 °C to trigger the hydrolysis and condensation of TCMS. 6 h later, the coated samples were rinsed with 10.0 mL of toluene, 10.0 mL of ethanol and 10.0 mL of 50% v/v deionized water/ethanol solution successively, and then dried under a nitrogen flow.

Characterization

Measurements of CA and SA were performed with a Contact Angle System OCA20 (Stuttgart, Germany) equipped with a custom built tilting table. The syringe was positioned in a way that the water drops (10 μL) could contact surface of the samples before leaving the needle. Tilting angle of the table was adjustable (0 ~ 90°) and allowed the subsequent measurement of SA at the same position on the sample. A minimum of three readings were recorded for each sample. The micrographs of the samples were taken using a SEM (Zeiss Supra 50 VP). Before SEM observation, all samples were fixed on aluminum stubs and coated with gold (~ 7 nm).

Results and discussion

Growth of SNFs in one-component solvents

Since polymerization of organosilanes should be avoided for SAMs preparation, alkanes and toluene are frequently used as solvents because they are inert to the reaction and the water

content in them can easily be kept at a very low level.^{2, 8} Thus, toluene was often chosen as the solvent for the growth of SNFs in previous studies.^{40, 45} It was reported by Thompson et al. that the choice of the solvent has great influence on SAMs prepared by silanization of glass slides with octadecyltrichlorosilane (OTS),² which inspired us to study the role of solvent in the growth of SNFs. The effects of fourteen different solvents on the growth of SNFs on glass slides have been investigated. We have tried to control the water content in all the solvents to be around 100 ppm, however, this is not straightforward for all of them because of their different affinity to water. The water content in the solvents was kept in the reasonable range of 33 to 236 ppm because the SNFs can grow well in toluene with a water concentration of 56 to 194 ppm according to our previous study.⁴⁰ The representative SEM images are shown in Fig. 1 and the corresponding CA and SA of water drops are shown in Table 1.

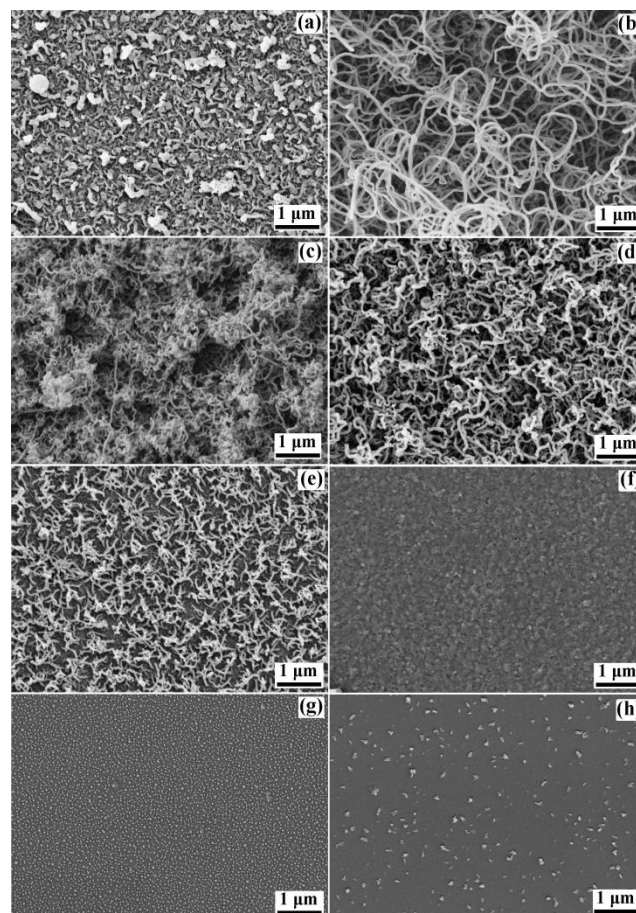


Fig. 1 SEM images of the glass slides coated with TCMS in (a) cyclohexane, (b) toluene, (c) *p*-xylene, (d) *t*-butylbenzene, (e) *n*-hexadecane, (f) poly(dimethylsiloxane), (g) paraffin oil and (h) acetonitrile.

The microstructures of the coatings formed in various solvents are obviously different from each other. The surfaces of glass slides are coated with a lot of intertwined SNFs when toluene (Fig. 1b), *p*-xylene (Fig. 1c) and *t*-butylbenzene (Fig. 1d) are used as solvents although details of the SNFs are still different. The SNFs grown (1) in toluene are very long and thick, and stack loosely together, (2) in *p*-xylene are very thin and stack densely together, (3) in *t*-butylbenzene are similar to those grown in

toluene except for a bit shorter. All the three coatings are superhydrophobic with CA > 164° and SA < 6.5°. However, a dense layer of finger-like structure is coated onto glass slide in cyclohexane (Fig. 1a). The coating is perfectly superhydrophobic with extremely high CA and low SA. Compared with toluene and cyclohexane, a sparser coating with an intermediate structure is obtained in *n*-hexadecane (Fig. 1e). The aspect ratio is not as high as those got in toluene. Similar effect of solvent on the surface density of OTS SAMs was also reported previously.² Evidently different from the SNFs grown in the above mentioned solvents, a smooth layer with a CA of 122.6° is generated in poly(dimethylsiloxane) (Fig. 1f). A lot of uniformly dispersed nanoparticles are formed on glass slide (CA = 104.4°) in paraffin oil (Fig. 1g). The nanoparticles are in fact roots of SNFs and will be discussed in detail below. The viscosity of poly(dimethylsiloxane) and paraffin oil is very high, which hinders migration of hydrolyzed TCMS and its oligomers (HTOs) onto the surface of glass slide. For those solvents with N and/or O elements (N/O-containing solvents), such as acetonitrile and acetone, the surface morphology is very similar to each other. Only very sparse and irregular nanoparticles can be seen on glass slides (Fig. 1h and Fig. S1), and CA below 70° or even zero are recorded.

Table 1. CA and SA of water drops on the glass slides coated with TCMS in various solvents.

	H ₂ O/ppm	CA/°	SA/° ^a
Cyclohexane	88	179.8	1.3
Toluene	102	173.0	4.3
<i>p</i> -Xylene	105	171.7	5
<i>t</i> -Butylbenzene	133	164.5	6.3
<i>n</i> -Hexadecane	55	165.8	9
Poly(dimethylsiloxane)	75	122.6	90
Paraffin oil	33.3	104.4	90
Methyl ethyl ketone	213	68.1	90
Ethyl ether	182	67.2	90
Acetonitrile	86	66.1	90
Acetone	221	54.8	90
Dioxane	198	36.2	90
DMSO	173	0	-
Acetyldimethylamine	236	0	-

^a SA = 90° indicates that water drops pin stably on the surface even tilted 90°. “-” means flat water films are formed on the surfaces and it is impossible to measure SA.

It can be concluded from Fig. 1 that the SNFs grow very well in toluene, *p*-xylene and *t*-butylbenzene, whereas cyclohexane and *n*-hexadecane disturb growth of the SNFs and their growth is stopped before they grow up into long SNFs. The SNFs grow uniformly on glass slides in toluene, *p*-xylene, *t*-butylbenzene and *n*-hexadecane although details of the SNFs are different. The growth of SNFs in N/O-containing solvents is seriously hindered. The chemical and environmental stability of the SNFs is similar

to our previous SNFs coatings.⁴⁰ The mechanical stability still needs to be improved for some applications, as for other known superhydrophobic or superoleophobic coatings. The mechanical stability may be improved by finding a new approach to active the surface of substrate or by introducing some hydrophobic binder.

Growth of SNFs in mixed solvents

In order to further elucidate the effect of solvents, mixtures of toluene and N/O-containing solvents were used as the co-solvents for the growth of SNFs. The water concentration in the coating solvent was kept in the range of 150 to 160 ppm for all the experiments in this section. The difference of these N/O-containing solvents in influencing the growth of SNFs, CA and SA of water drops is obvious (Fig. 2 and Table 2).

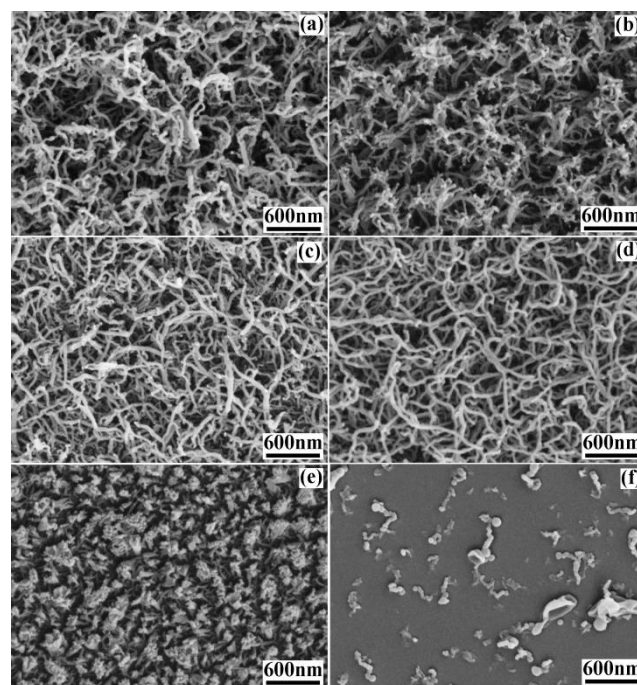


Fig. 2 SEM images of the glass slides coated with TCMS in the mixture of toluene (80 mL) and various N/O-containing solvents (1 mL). (a) acetonitrile, (b) methyl ethyl ketone, (c) ethyl ether, (d) dioxane, (e) acetone and (f) DMSO.

Table 2. CA and SA of water drops on the glass slides coated with TCMS in the mixture of toluene (80 mL) and various N/O-containing solvents (1 mL).

	CA/°	SA/°
Acetonitrile	177.6	2.0
Methyl ethyl ketone	170.2	3.5
Ethyl ether	169.5	14.0
Dioxane	164.2	13.7
Acetone	159.5	35.3
DMSO	103.7	90.0

The SNFs still are synthesized in a very similar shape when 1

mL (1.23% v/v) of acetonitrile, methyl ethyl ketone or ethyl ether is used as the co-solvent as shown in Fig. 2a-c and S2, but interestingly branched at the end compared with those grown in pure toluene. The superhydrophobicity of the coatings is kept when acetonitrile or methyl ethyl ketone is used as the co-solvent, however, an increase in SA to 14.0° was recorded in the case of ethyl ether. The SNFs grown in the dioxane/toluene mixture are the same as in pure toluene except that the coating becomes more flat, resulting in an increased SA of 13.7°. As can be seen from Fig. S1, the surface morphology is almost identical using pure acetone and methyl ethyl ketone, respectively. However, when the mixture of 1 mL of these solvents and 80 mL of toluene is used as the medium for SNFs growth, obvious differences can be seen from Fig. 2b and e. The SNFs are formed at the beginning of the formation process, however, subsequently aggregates on top of the coating are formed when an acetone/toluene mixture is used (Fig. 2e). Accordingly, CA decreases to 159.5° and SA evidently increases to 35.3°. Only a few wormlike products composed of nanoparticles can be observed on the surface of glass slide when 1 mL of DMSO is introduced into toluene (Fig. 2f). The CA is only 103.7° and water drops pin stably on the glass slide. The great impact of DMSO on the growth of SNFs, CA and SA is in accordance with the data in Table 1, in which a flat water film is formed on the surface and it is impossible to measure the SA when pure DMSO is used as the solvent.

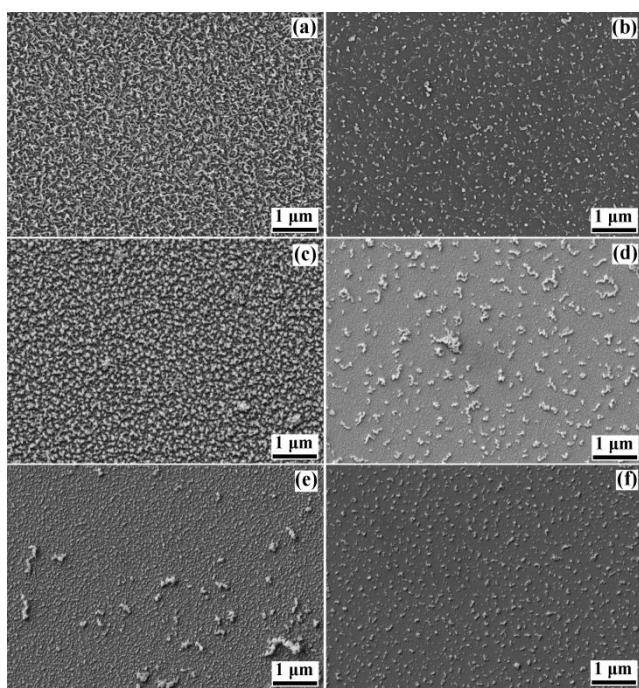


Fig. 3 Representative SEM images of the glass slides coated with TCMS in the mixture of toluene (80 mL) and various N/O-containing solvents. (a) 3 mL of acetonitrile, (b) 7 mL of acetonitrile, (c) 3 mL of methyl ethyl ketone, (d) 7 mL of methyl ethyl ketone, (e) 3 mL of acetone and (f) 5 mL of acetone.

In view of the similar unapparent effects of 1 mL of acetonitrile and methyl ethyl ketone on SNFs growth, and the significant effect of 1 mL of acetone, the influences of the amount of these three co-solvents on SNFs growth, CA and SA of water were further studied as shown in Fig. 3, S3 and 4. With increasing acetonitrile content from 0 to 7 mL, the SNFs

gradually become shorter and the surface roughness decreases (Fig. 3a, b and Fig. S3a, b). Only sparse wormlike products and nanoparticles can be seen in Fig. 3b. Accordingly, a decrease in CA and an increase in SA were observed, especially when the acetonitrile content is over 5 mL. Similar trends were observed in the case of methyl ethyl ketone and acetone. The difference between acetonitrile and methyl ethyl ketone in influencing SNFs growth is clear by increasing their amount (Fig. 3a and c). The SNFs are even shorter when the same amount of methyl ethyl ketone was used instead of acetonitrile. These three co-solvents are in the order of acetonitrile > methyl ethyl ketone > acetone in influencing SNFs growth, CA and SA of water, which is in accordance with the data in Table 2. No SNFs can be seen even just 3 mL of acetone was introduced.

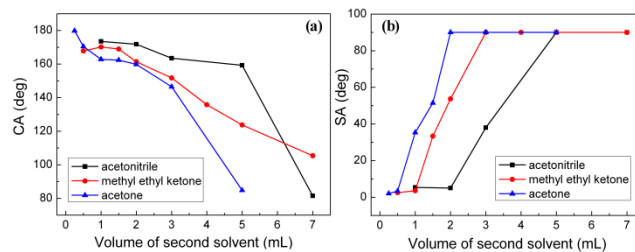


Fig. 4 Variation of CA and SA of water on the TCMS coated glass slides with volume of acetonitrile, methyl ethyl ketone and acetone in toluene (80 mL).

The results in this section prove that the choice of the solvent plays an important role for the growth of SNFs and the surface hydrophobicity. The growth of SNFs can be tuned simply by controlling the solvent composition. In addition, the SNFs are gradually shortened by increasing the content of these co-solvents, which gives us a chance to see the SNFs/glass slide interface. The introduced co-solvent hinders the growth of SNFs. Only randomly distributed irregular nanoparticles or wormlike products can be seen under the existence of proper amount of the co-solvents (Fig. 1g, Fig. 2b, d-f). There are in fact roots of the SNFs, which are most likely produced by random immobilization of HTOs onto the glass slide via bonding between their hydroxyl groups. The methyl groups tend to be directed toward the outside to decrease the surface tension^{49, 50} and effectively limit further attachment of the reactive components (HTOs) around them. Consequently, the HTOs could only continuously condense onto the remained exposed silanols, which results in elongation and asymmetric growth of the roots and finally in infant SNFs as shown in Fig. 3b. These infant SNFs can continue to grow into long SNFs with high aspect ratio under proper conditions.

Growth of SNFs in toluene containing Tween 20 and Span 80

To further support our discussion in the above section, Tween 20 and Span 80 with long alkyl groups and abundant O-containing groups (-OH, C=O and C-O) were used instead of the N/O-containing co-solvents to impact the growing process of SNFs. We found that the growth of SNFs can even be better controlled by using Tween 20 instead of using co-solvents. The effect of Tween 20 on CA and SA is more evident because it has a lot of O-containing groups. The CA decreases linearly to 91.2° with increasing Tween 20 content to 0.277 mmol/L (Fig. 5). The SA increases quickly and water drops pin stably on the surface when

the Tween 20 content is over 0.042 mmol/L. From the SEM images in Fig. 6 and S4, we can clearly see how the nanoparticles (roots of SNFs) gradually grow into SNFs by decreasing the Tween 20 content.

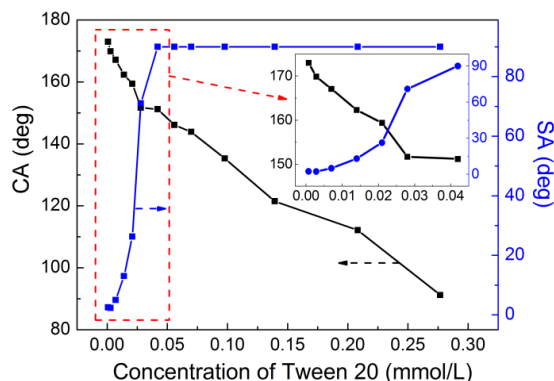


Fig. 5 Variation of CA and SA of water drops on the TCMS coated glass slides with concentration of Tween 20 in toluene (80 mL).

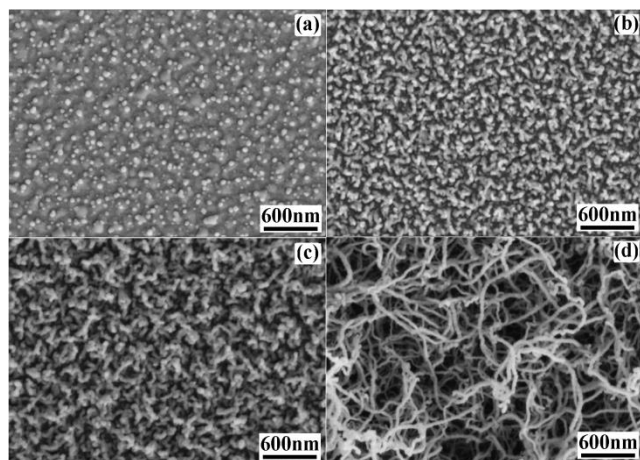


Fig. 6 Representative SEM images of the TCMS coated glass slides in toluene with a Tween 20 concentration of (a) 0.277, (b) 0.042, (c) 0.021 and (d) 0.001 mmol/L.

Similar variation of SNFs growth, CA and SA of water also can be observed in the case of Span 80 as shown in Fig. 7, S5 and 8. One of the differences is that a higher concentration of Span 80 is needed for an evident impact on SNFs growth and wettability. The roots of SNFs (green circles in Fig. 7a) and infant SNFs (red squares in Fig. 7a) could only be observed when the Span 80 concentration is more than 7.13 mmol/L, which is much higher than that of Tween 20 (0.277 mmol/L). This is probably due to the lower number of O-containing groups of the Span 80 molecule (Fig. S6). The asymmetric growth of SNFs roots and infant SNFs can be seen clearly with decreasing the Span 80 content to 3.77 mmol/L (Fig. 7b). In addition, the surface of the SNFs is not as smooth as those generated in other system and becomes very rough with decreasing the Span 80 concentration to 1.18 mmol/L (Fig. S5a and 7c). This difference can be seen clearly in Fig. 9. There are plenty of nanoparticles on the surface of the SNFs and the SNFs seem to be an assembling of nanoparticles. The surface of the SNFs becomes smooth again and no nanoparticles can be seen with further decreasing the Span 80 concentration to 0.02 mmol/L (Fig. S5b-d and 7d).

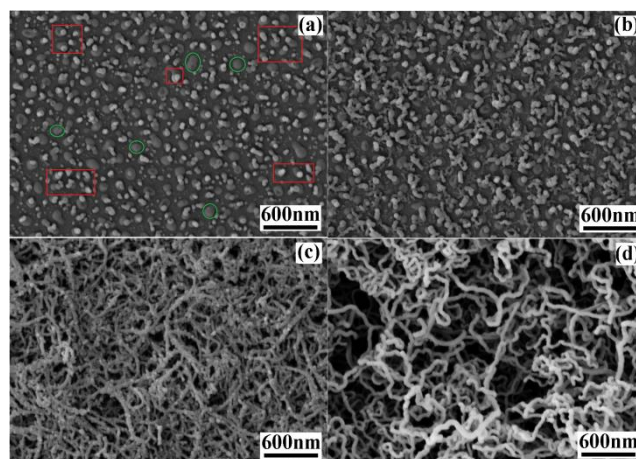


Fig. 7 SEM images of the TCMS coated glass slides in toluene with a Span 80 concentration of (a) 7.13, (b) 3.77, (c) 1.18 and (d) 0.02 mmol/L.

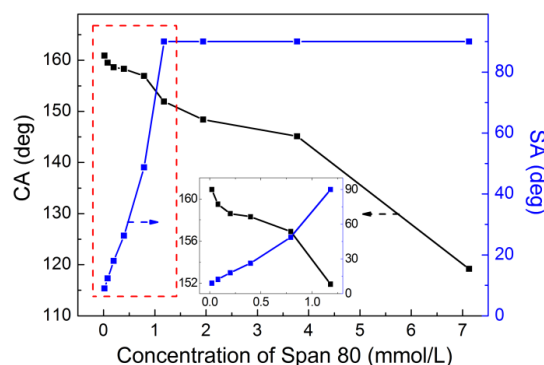


Fig. 8 Variation of CA and SA of water drops on the TCMS coated glass slides with concentration of Span 80 in toluene (80 mL).

Based on the results described above, we have concluded that the SNFs are formed by random immobilization of HTOs onto a glass surface. However, we still have not got the direct evidence for the immobilization of HTOs onto SNFs until we got the SEM image as shown in Fig. 9b. This is because the size of HTOs composed of the SNFs is very small in toluene and other systems studied above. So, the SNFs are formed perfectly with very smooth surface and we cannot see the details. Differently, in the case of Span 80 with a narrow range of concentration (1.18 ~ 1.94 mmol/L), the HTOs have the chance to grow big enough before taking part in forming the SNFs owing to the interaction among Span 80 molecules, water, HTOs and glass slide. Consequently, the SNFs with very rough surface were observed.

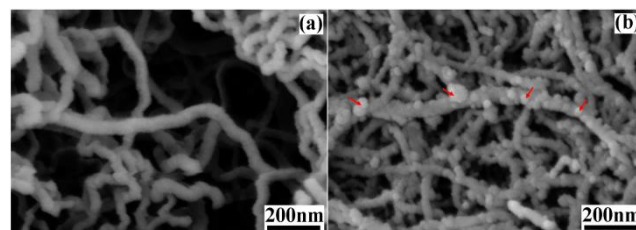


Fig. 9 SEM images of the TCMS coated glass slides in toluene (a) without and (b) with Span 80 (1.18 mmol/L).

Conclusions

We show that the choice of the solvent has great influences on the

growth of SNFs, the CA and SA of water drops. By simply regulating the solvent composition during hydrolysis and condensation of TCMS, SNFs with various features were obtained on the surface of glass slides. The morphology of SNFs has great influence on CA and SA of water drops. Toluene and its homologues are ideal solvents for the growth of the SNFs, whereas N/O-containing solvents and molecules could seriously hinder their growth. The variation of N/O-containing solvents and molecules provides a good opportunity to influence the growing process of SNFs. The roots of SNFs are formed by random immobilization of HTOs onto the surface of glass slides. Subsequently, the HTOs anchor onto the exposed silanols of these roots under proper conditions, which results in their elongation and increase of the aspect ratio.

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Notes and references

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- † Electronic Supplementary Information (ESI) available: [Characteristic SEM images of the TCMS coated glass slides in other solvents besides Fig. 1. Other SEM images of the TCMS coated glass slides in the mixture of toluene and various N/O-containing solvents besides those shown in Fig. 3. Other SEM images of the TCMS coated glass slides in toluene with different Tween 20 concentration besides those shown in Fig. 6. Other SEM images of the TCMS coated glass slides in toluene with different Span 80 concentration besides those shown in Fig. 7. Molecular structure of Tween 20 and Span 80.]. See DOI: 10.1039/b000000x/
- J. D. Swalen, D. L. Allara, J. D. Andrade, E. A. Chandross, S. Garoff, J. Israelachvili, T. J. McCarthy, R. Murray, R. F. Pease, J. F. Rabolt, K. J. Wynne and H. Yu, *Langmuir*, 1987, **3**, 932.
 - M. E. McGovern, K. M. R. Kallury and M. Thompson, *Langmuir*, 1994, **10**, 3607.
 - L. W. Moore, K. N. Sprjnger, J. X. Shi, X. G. Yang, B. I. Swanson, D. Q. Li, *Adv. Mater.*, 1995, **7**, 729.
 - M. C. Howland, A. R. Sapuri-Butti, S. S. Dixit, A. M. Dattelbaum, A. P. Shreve and A. N. Parikh, *J. Am. Chem. Soc.*, 2005, **127**, 6752–6765.
 - A. N. Parikh and D. L. Allara, *J. Phys. Chem.*, 1994, **98**, 7577–7590.
 - B. L. V. Prasad, S. I. Stoeva, C. M. Sorensen, V. Zaiikovski and K. J. Klabunde, *J. Am. Chem. Soc.*, 2003, **125**, 10488.
 - B. C. Bunker, R. W. Carpick, R. A. Assink, M. L. Thomas, M. G. Hankins, J. A. Voigt, D. Sipola, M. P. de Boer and G. L. Gully, *Langmuir*, 2000, **16**, 7742.
 - R. R. Rye, G. C. Nelson and M. T. Dugger, *Langmuir*, 1997, **13**, 2965.
 - A. Y. Fadeev and T. J. McCarthy, *Langmuir*, 2000, **16**, 7268.
 - C. P. Tripp and M. L. Hair, *Langmuir*, 1995, **11**, 149.
 - P. Silberza, L. Løger, D. Ausserré and J. Benattar, *J. Langmuir*, 1991, **7**, 1647.
 - J. Sagiv, *J. Am. Chem. Soc.*, 1980, **102**, 92.
 - J. L. Wang, A. Raza, Y. Si, L. X. Cui, J. F. Ge, B. Ding and J. Y. Yu, *Nanoscale*, 2012, **4**, 7549.
 - K. S. Liu, X. Yao and L. Jiang, *Chem. Soc. Rev.*, 2010, **39**, 3240.
 - J. F. Gao, J. S. P. Wong, M. J. Hu, W. Li, R. K. Y. Li, *Nanoscale*, 2013, **6**, 1056.
 - J. K. Yuan, X. G. Liu, O. Akbulut, J. Q. Hu, S. L. Suib, J. Kong and F. Stellacci, *Nat. Nanotechnol.*, 2008, **3**, 332.
 - M. J. Liu, Y. M. Zheng, J. Zhai and L. Jiang, *Accounts Chem. Res.*, 2010, **43**, 368.
 - W. Barthlott and C. Neinhuis, *Planta*, 1997, **202**, 1.
 - H. X. Wang, Y.H. Xue, J. Ding, L. F. Feng, X. G. Wang and T. Lin, *Angew. Chem. Int. Ed.*, 2011, **50**, 11433.
 - W. Ren, S. J. Guo, S. J. Dong and E. K. Wang, *Nanoscale*, 2011, **3**, 2241.
 - M. Guix, J. Orozco, M. Garc á, W. Gao, S. Sattayasamitsathit, A. Merkoç, A. Escarpa and J. Wang, *ACS Nano*, 2012, **6**, 4445.
 - D. Yoon, C. Lee, J. Yun, W. Jeon, B. J. Cha and S. Baik, *ACS Nano*, 2012, **6**, 5980.
 - N. Miljkovic, R. Enright and E. N. Wang, *ACS Nano*, 2012, **6**, 1776.
 - A. Li, H. X. Sun, D. Z. Tan, W. J. Fan, S. H. Wen, X. J. Qing, G. X. Li, S. Y. Li and W. Q. Deng, *Energy Environ. Sci.*, 2011, **4**, 2062.
 - T. Verho, C. Bower, P. Andrew, S. Franssila, O. Ikkala and R. H. A. Ras, *Adv. Mater.*, 2011, **23**, 673.
 - E. Hosono, S. Fujihara, I. Honma and H. S. Zhou, *J. Am. Chem. Soc.*, 2005, **127**, 13458.
 - K. S. Liu, Y. Tian and L. Jiang, *Prog. Mater. Sci.*, 2013, **58**, 503–564.
 - Y. Li, L. Li and J. Q. Sun, *Angew. Chem. Int. Ed.*, 2010, **49**, 6129.
 - S. M. Kang, I. You, W. K. Cho, H. K. Shon, T. G. Lee, I. S. Choi, J. M. Karp and H. Lee, *Angew. Chem. Int. Ed.*, 2010, **49**, 9401.
 - X. Deng, L. Mammen, H. J. Butt and D. Vollmer, *Science*, 2012, **335**, 67.
 - A. Tuteja, W. Choi, M. Ma, J. M. Mabry, S.A. Mazzella, G. C. Rutledge, G. H. McKinley and R. E. Cohen, *Science*, 2007, **318**, 1618.
 - S. P. R. Kobaku, A. K. Kota, D. H. Lee, J. M. Mabry and A. Tuteja, *Angew. Chem. Int. Ed.*, 2012, **51**, 10109.
 - H. X. Su, A. Li, Z. Q. Zhu, W. D. Liang, X. H. Zhao, P. Q. La, W. Q. La, *ChemSusChem*, 2013, **6**, 1057.
 - J. P. Zhang, B. C. Li, L. Wu and A. Q. Wang, *Chem. Commun.*, 2013, **49**, 11509.
 - L. Wu, J. P. Zhang, B. C. Li and A. Q. Wang, *J. Mater. Chem. B*, 2013, **1**, 4756.
 - H. Sun, A. Li, X. Qin, Z. Zhu, W. Liang, J. An, P. La and W. Deng, *ChemSusChem*, 2013, **6**, 2377.
 - J. Zimmermann, S. Jung and S. Seeger, EP 1644450, 2004.
 - G. R. J. Artus, S. Jung, J. Zimmermann, H. P. Gautschi, K. Marquardt and S. Seeger, *Adv. Mater.*, 2006, **18**, 2758.
 - J. Zimmermann, F. A. Reifler, G. Fortunato, L. C. Gerhardt and S. Seeger, *Adv. Funct. Mater.*, 2008, **18**, 3662–3669.
 - J. P. Zhang and S. Seeger, *Angew. Chem. Int. Ed.*, 2011, **50**, 6652.
 - A. Stojanovic, S. Olveira, M. Fischer and S. Seeger, *Chem. Mater.*, 2013, **25**, 2787.
 - J. P. Zhang, A.Q. Wang and S. Seeger, *Adv Funct Mater.*, 2014, **24**, 1074.
 - J. P. Zhang, A.Q. Wang and S. Seeger, *Polym. Chem.*, 2014, **5**, 1132.
 - H. X. Su, A. Li, Z. Q. Zhu, W. D. Liang, X. H. Zhao, P. Q. La, W. Q. La, *ChemSusChem*, 2013, **6**, 1057.
 - G. R. Meseck, R. Kontic, G. R. Patzke and S. Seeger, *Adv. Funct. Mater.*, 2012, **22**, 4433.
 - L.C. Gao and T. J. McCarthy, *J. Am. Chem. Soc.*, 2006, **128**, 9052.
 - D. E. Rollings and J.G.C. Veinot, *Langmuir*, 2008, **24**, 13653.
 - R.G. Chen, X.G. Zhang, Z.H. Su, R. Gong, X. Ge, H.J. Zhang and C. Wang, *J. Phys. Chem. C*, 2009, **113**, 8350.
 - M. A. Raza, E. S. Kooij, A. Silfhout and B. Poelsema, *Langmuir*, 2010, **26**, 12962.
 - M. J. Pellerite, E. J. Wood and V. W. Jones, *J. Phys. Chem. B*, 2002, **106**, 4746.

Graphical Abstract

Silicone nanofilaments (SNFs) with different feature and hydrophobicity were prepared by regulating solvent composition during hydrolysis and condensation of trichloromethylsilane.

