RSC Advances

ROYAL SOCIETY OF CHEMISTRY

www.rsc.org/advances

RSC Advances

ARTICLE

Flexible Superhydrophobic Polysiloxane Aerogels for Oil-water Separation *via* One-pot Synthesis in Supercritical CO_2

Received 00th January 20xx,
Accepted 00th January 20xx

DOI: 10.1039/x0xx00000x

www.rsc.org/

Fangxin Zou,[a] Li Peng,[b] Wenxin Fu,*[a] Jianling Zhang*[b] and Zhibo Li*[c]

Porous aerogel is an inspiring absorbent for oil-water separation and water purification. In particular, hydrophobic silica aerogels from abundant and cheap sources offer both economical and environmental benefits. Conventional silica aerogels from alkoxy siloxane precursors involve time-consuming and laboursome multistep processes. Herein, we demonstrate the formation of flexible superhydrophobic polysiloxane aerogels through a facile hydrosilylation from functionalized polydimethylsiloxane in supercritical carbon dioxide. This robust aerogel exhibits high oil absorption capacity, superior recyclability and extraordinary mechanical property even under harsh heating and cooling cycles. These characters favor this polysiloxane aerogel more competitive than common alkoxy silica ones for oily water treatment, oil spill clean-up and oil recovery.

Introduction

The fast oil-water separation is becoming a global tickler for solving the water pollution and conserving water resources.[1,2] The frequent oil spill accidents and the oil-contaminated industrial wastewater caused the severely bad effects to environment and ecological system.[3-5] It is extremely urgent to develop effective and inexpensive oil-water separating materials for the oily pollution in water system. Hydrophobic aerogels, as a class of highly porous material filled with air, demonstrated higher oil absorption capacity, longer retention time, better oleophilicity and reusability than normal absorbents (e.g. inorganic mineral clays,[6] synthetic polymeric materials,[7,8] natural cellulose-based materials[9,10]. Recently, a variety of hydrophobic aerogels with different chemical compositions have been reported as good candidates for oil-water separation, including ultralight carbon-based aerogels,[11-14]nanostructured metal oxide aerogels[15,16] and cellulose fibers from natural resources and wastes.[17-19]

Among the diverse kinds of hydrophobic aerogels, silica aerogels, as the first kind of aerogel from abundant and cheap sources, could be the ideal alternative for oil absorption in practical and industrial applications. Compared with inorganic silica ones, the organic silicon gels own the superhydrophobic surfaces and a certain degree of compression property, due to the rich methyl groups directly bonded onto silicon atoms and the flexible network backbones from Si-O/Si-C bond. Several kinds of hydrophobic silica aerogels have been prepared, such as the silica aerogel microspheres,[20] magnetic silica sponges,[21] marshmallow-like porous silica gels[22-25] and the polymer reinforced silica composite aerogels.[26-28] All of these silica aerogels were prepared from alkoxy siloxane precursors and needed to go through multistep processes, including hydrolysis, pre-condensation, aging, solvent exchange, drying and/or further surface modification.[25,29] To develop novel kinds of hydrophobic silica aerogels through facile route is interesting, but still remains challenging.

In this study, we present a facile method to prepare the flexible superhydrophobic polysiloxane aerogels directly from functionalized polydimethylsiloxanes (PDMS) by a one-pot hydrosilylation reaction in supercritical CO_2. Compared to previous sol-gel process, the efficient and controllable hydrosilylation reaction was applied to prepare aerogels, which can allow modulatoin of crosslinking degree of polysiloxane aerogels. Besides, the aerogels directly from PDMSs were inherently superhydrophobic without the tedious surface post-modifications. Furthermore, the hydrosilylation in supercritical CO_2 integrated the gelation and drying technique, avoiding the time-consuming multi-steps, and demonstrated a robust and flexible porous skeleton with characterized microstructures. The resultant polysiloxane aerogels were used as oil absorbents for oil-water separation, which exhibited high oil absorption capacity, superior recyclability and extraordinary mechanical property even under harsh heating and cooling cycles.

---

[a] Beijing National Laboratory of Molecular Sciences (BNLMS), Institute of Chemistry, Chinese Academy of Sciences, Zhongguancun North First Street 2, Beijing 100190, China.

[b] CAS Key Laboratory of Colloid, Interface and Chemical Thermodynamics, Institute of Chemistry, Chinese Academy of Sciences, Zhongguancun North First Street 2, Beijing 100190, China.

[c] School of Polymer Science and Engineering, Qingdao University of Science and Technology, Qingdao 266042, China.

E-mail: fuwenxin@iccas.ac.cn, zhangjl@iccas.ac.cn and zbli@qust.edu.cn

† Electronic Supplementary Information (ESI) available. See DOI: 10.1039/x0xx00000x

Experimental

Materials

Tetramethyldisiloxane (M^HM^H), octamethylcyclotetrasiloxane (D_4), 1,3,5,7-tetramethylcyclotetrasiloxane (D_4^H), 1,1,3,3-, 1,1,3,3-tetramethyl-1,3-divinyldisiloxane ($M^{Vi}M^{Vi}$) were purchased from Beijing HWRK Chem Co. Ltd. Toluene, H_2SO_4 (98%) and $CuCl_2•2H_2O$ were purchased from Sinopharm Chemical Reagent Beijing Co., Ltd. CO_2 (>99.95%) was provided by Beijing Analysis Instrument Factory. Karstedt's catalyst solution (Platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane complex solution in xylene, Pt 2%) was purchased from Aladdin Industrial Corporation. All chemicals were used without further purification. Oil Red O was purchased from Sigma-Aldrich Shanghai Trading Co. Ltd.

Synthesis of the methyl hydrogen polysiloxane (A)

Different molar ratios of D_4^H, D_4 and M^HM^H were placed in a glass flask, and the initiating amount of H_2SO_4 (98%) was added under stirred. Then the reaction continued for 5 hours at room temperature. After that, toluene and water were added. The toluene layer was separated and washed with water three times. The organic layer was dried over anhydrous magnesium sulfate and filtered. The solvent was removed by rotary evaporator and a colorless liquid product was obtained. The characterizations of chemical structures for precursors A are shown in Supplementary Information (ESI) and Fig. S1-4.

Synthesis of the vinyl-terminated polydimethylsiloxane (B)

D_4 (36.91 g, 0.124 mol) and $M^{Vi}M^{Vi}$ (3.09 g, 0.0166 mol) were placed in a glass flask, and the initiating amount of H_2SO_4 (98%) (2 g, 0.02 mol) was added under stirred. Then the reaction continued for 5 hours at room temperature. After that, toluene and water were added. The toluene layer was separated and washed with water three times. The organic layer was dried over anhydrous magnesium sulfate and filtered. The solvent was removed by rotary evaporator and a colorless liquid product B (36.2 g) was obtained with 90.5% yield. The characterizations of chemical structures for precursors B are shown in ESI and Fig. S1-4.

Preparation of aerogel

The equal molar weight of Si-H and vinyl from methyl hydrogen polysiloxane (A) and vinyl-terminated PDMS (B), respectively, were added into a 50 mL high-pressure reactor equipped with a magnetic stirrer. Then, CO_2 was charged into the reactor at 40 ^oC until the whole system turned to be homogeneous. Then 100 ppm of total weight Karstedt's catalyst was injected into the cell and mixed well quickly. After the gelation for 4 hours without agitation, CO_2 was released (Fig. 1).

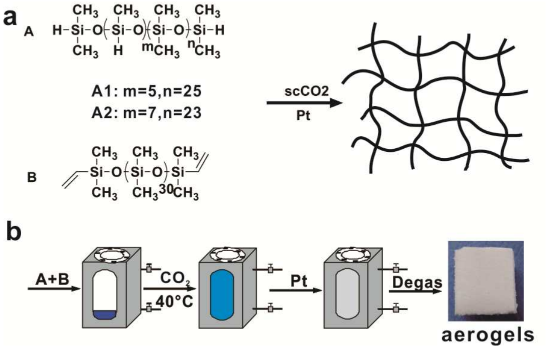


Fig. 1 a) Synthesis route of polysiloxane aerogel in $scCO_2$. b) Schematic description of the one-pot preparation for the target polysiloxane aerogel in $scCO_2$.

Characterization

^1H NMR and ^{29}Si NMR measurements were carried out on Bruker AV-400 and AV-300 NMR instruments, respectively, using the residual protonated solvent as an internal standard. Mass spectra (MALDI-TOF-MS) were determined on a Bruker BIFLEX III Mass spectrometer. Transmission electron microscopy (TEM) samples were examined with a JEM2200FS (200 keV). Before measurement, the samples were grinded into pieces and then dispersed in dry ethanol and sonicated for 1-2 min. After that, the sample solution was casted on a carbon-coated copper grid. After one minute, the excess sample solution was carefully removed using a piece of filter paper. The surface microstructure was examined by Scanning Electron Microscope (SEM) (6700F) instrument. Contact angle data were obtained on Krüss Drop Shape Analysis System-100 (DSA 100) by a sessile water drop method with 5 μL liquid drops. Thermogravimetric analyses (TGA) was made using a 7 Series thermal analysis system (Perkin-Elmer). The sample was heated from 30 to 800 °C at a rate of 5 °C min^{-1} in a dynamic nitrogen atmosphere with a flow rate of 70 mL min^{-1}. Bulk density was obtained from the weight/volume ratio of specimens. The skeletal density was measured by UltraPYC 1200e automatic density analyzer from Quantachrome Instruments. The porosity (%) of each sample was calculated as $(1-\rho_b/\rho_s) \times 100\%$, where ρ_b and ρ_s refer to the bulk and skeletal densities, respectively. Specific surface area and pore size were determined by PoreMaster-60-17 from Quantachrome Instruments.

Results and discussion

In this work, a series of aerogels were prepared with methyl hydrogen polysiloxane (A) and vinyl-terminated PDMS (B) through hydrosilylation reaction in supercritical CO_2 (Table S1). This method is simple, time-saving, and environmental friendly, avoiding the troublesome solvent exchange, complicated drying process, surface modification and the use of organic solvents.

Structure, hydrophobicities and mechanical properties of obtained aerogels

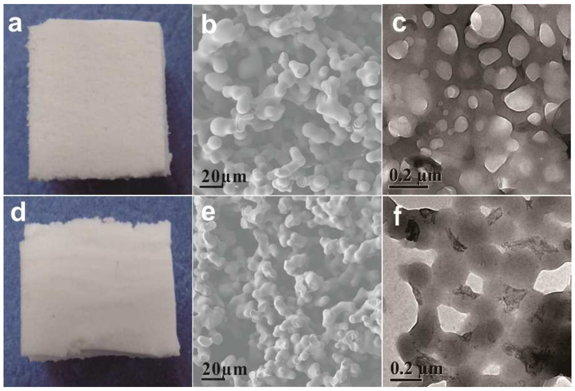


Fig. 2 Photographs, SEM and TEM images of A1-B (a, b, c) and A2-B (d, e, f) aerogels synthesized at 22 MPa and the concentration of 136 mg ml^{-1}.

Fig. 2a and 2d show the photographs of two polysiloxane aerogels, A1-B and A2-B, obtained from the reaction of B with precursor A1 and A2, respectively. They display a much soft and elastic appearance. The bulk densities of A1-B and A2-B aerogels were 227 and 258 mg cm^{-3}, respectively (Table S1). Their microstructures were further characterized by SEM and TEM. The SEM images (Fig. 2b, e) show that both of A1-B and A2-B aerogels have a three-dimensional interconnected network, a highly porous structure , the Pt particles were uniformly distributed in the aerogel (Fig. S5) and the characteristic crosslinked bulb structures with diameters of about 8 μm. After pounded in a mortar to break the bulb structures, both A1-B and A2-B aerogels also demonstrate nanoporous structures with the pore size in range of 30-180 nm (Fig. 2c, f) and . Because the highly porous structure is conducive to the rapid transport of gas and liquid in the aerogel, they can be used for oil absorption. Notably, A1-B shows more inter-connective nanopores and less regional aggregations than A2-B, which is beneficial to enhance the porosity and specific surface area (Table S1 and Fig. S6). The difference in microstructures of the two aerogels can be attributed to their specific chemical compositions. As shown in Fig.1, A2 has high mole percentage of Si-H, so even at the same reaction concentration, A2-B aerogel will has more crosslinkings. During the gelation, more crosslinkings in A2-B will lead to faster precipitation and more aggregates in scCO$_2$ than those of A1-B, resulting in the less porosity and specific surface area of A2-B aerogel than that of A1-B aerogel.

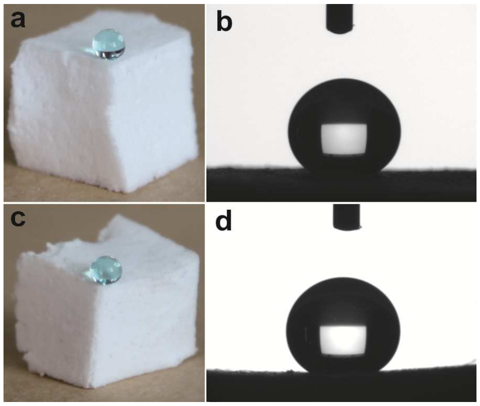


Fig. 3 Optical images of one water droplet deposited on A1-B (a) and A2-B surface (c). Water is colored by CuCl$_2$. The water contact angles for A1-B (b) and A2-B (d) are 154° and 150°, respectively.

One of the most important features of polysiloxane aerogel is its superhydrophobicity (Fig. 3a, c). The surface hydrophobicities of A1-B and A2-B aerogels were investigated by using water-contact angle measurement. Both of them are superhydrophobic with a contact angle over 150° (Fig. 3b, d) with neither chemical nor physical surface treatment. The hydrophobic feature of the aerogel is due to the microscopic rough surface from the intrinsic porous structures and the presence of plenty of methyl groups.

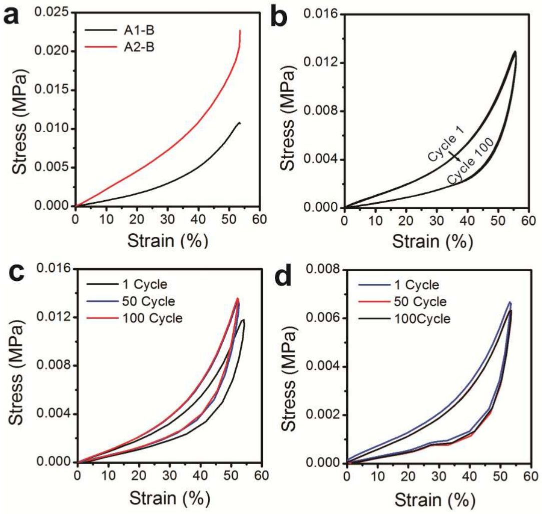


Fig. 4 a) Stress-strain curves of a uniaxial compression test on A1-B and A2-B aerogels. 100 cycles repeated compression of A1-B aerogel at room temperature (b), 150 °C (c) and -50 °C (d).

For many silica aerogels, the low mechanical strength limited their applications. But in this work, this problem can be solved by the one-pot hydrosilylation reaction of methyl hydrogen polysiloxane (A) and vinyl-terminated PDMS (B) in supercritical CO$_2$. Stress-strain curves of a uniaxial compression tests on A1-B and A2-B aerogels demonstrate their high compressibility and remarkable flexibility (Fig. 4a). The A1-B aerogel could recover to the original shape under the 55%

deformation after 100 repetitions at room temperature, 150 and -50 °C, respectively (Fig. 4b-d and Fig. S7a). The shape recovery and persistence indicate that the polysiloxane aerogels possess integrated structures and good mechanical stability. However, silica aerogel without further chemical or physical surface treatment will be broken and collapse with small stress, because it has many silanol groups on the surface and the silanol groups close together will further react to form siloxane bonds during a compression test. But for the obtained polysiloxane aerogels without silanol groups, the elastic PDMS-like backbones, highly porous gel networks and repulsive interactions between methyl groups endow the areogel high compressibility, remarkable flexibility and no collapse during compression test. Compared with traditional crumbly silica aerogels (Table S2),[20, 30-33] the excellent toughness of A1-B aerogel is especially significant for reusable oil absorbents in industrial applications. It is noteworthy that A1-B owns better flexibility than A2-B under the same uniaxial compression (Fig. 4a), which can be attributed to the fact that A1-B owns less chemical crosslinkings than A2-B.

As a result of the excellent mechanical stability of the polysiloxane aerogels, the superhydrophobic aerogels also exhibit considerable robustness against organic solvents (Fig. S4b). Meanwhile, the polysiloxane aerogels are stable up to ca. 400 °C from the thermogravimetry analysis (Fig. S8 and Table S1). The thermal stability of the as-synthesized polysiloxane aerogels is much better than that of the common oil-water separation materials from carbon-based polymers, for which the initial decomposition temperature is ca. 200 °C (Table S3).[17, 34]

Absorption capacities of obtained aerogels

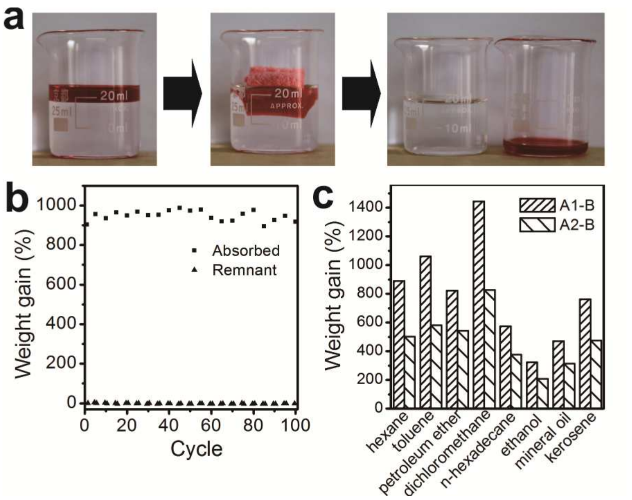


Fig. 5 a) Separation of n-hexane from water by A1-B aerogel and hexane is colored by Oil Red O. b) Weight gain of A1-B aerogel during 100 cycles of n-hexane absorption. c) Absorption capacities of the A1-B and A2-B aerogels for various organic solvents, as indicated by weight gain.

With high porosity, superhydrophobicity, high compressibility and remarkable flexibility, the polysiloxane aerogels can be considered as an ideal candidate for oil-water separation. To further verify the feasibility for oil-water separating applications, n-hexane was chosen as a model absorbate to investigate the separation performance of the A1-B aerogel. As shown in Fig. 5a, n-hexane is completely separated from water quickly by A1-B aerogel and also can be all squeezed out by hand. Even after 100 cycles of absorption and squeezing-out, not only the A1-B aerogel's absorption performance still keeps stable and has no significantly decrease (Fig. 5b), but also its morphology has no damages, indicating its excellent recyclability (Fig. S7b). The reusability of the polysiloxane aerogel as oil absorbent can be attributed to its robust skeleton. Furthermore, the superhydrophobic A1-B aerogel displays excellent absorption capacities towards a wide range of organic solvents, up to 4.7-14.5 times of its dry weight depending on the density of the organic liquid (Fig. 5c and S9). The aerogel could be recycled and recovered by facile squeezing-out, even for the high-density organic solvents (e.g., chloroform) and viscous oils (e.g., mineral oil with a comparable kinetic viscosity of ca. 44.6 mm s^{-1} at 40 °C to medium crude oil[35]). Therefore, this kind of polysiloxane aerogel can be used for the regeneration of the oil. Particularly, A1-B demonstrates better absorption ability than A2-B, probably due to its lower crosslinking density and larger specific surface area than A2-B (Table S1). It is worth noting that the oil absorbency of A1-B polysiloxane aerogel is much more superior to that of many other reported silica ones (Table S4).[31, 32] So the excellent absorption capacity of the polysiloxane aerogel is not only due to the highly porous framework, oleophilic nature, capillary action and robust skeleton, but also attributed to the sufficient swelling of cross-linked flexible polysiloxane network in organic solvents.

Mechanism for the formation of polysiloxane aerogels in $scCO_2$

To figure out the hydrosilylation in $scCO_2$, the effects of different experimental conditions on the formation of polysiloxane aerogels were investigated. Firstly, upon the decrease of reaction concentration, the resulting A2-B aerogels have more unconsolidated structures. Their microscopic structures reveal some regional small bulb aggregates and unevenly distributed bigger pores over hundreds of micrometers (Fig. S10). The porosities of A2-B aerogels synthesized at concentrations of 68, 95 and 136 mg mL^{-1} are 86.8%, 86.0% and 76.5%, respectively, which suggests that the porosity decreases with the increasing of sample concentration. Under high reaction concentration, the skeleton density is constant, but the increased bulk density could result in the decrease of pore size, which is consistent with SEM results (Fig. 2e and S10). Moreover, with the decrease of concentration, the A2-B's contact angle increases (Table S1), indicating the enhanced hydrophobicity. It probably can be due to the more porous surface with lower concentration. It is worth noting that A1-B aerogel could not be formed at the concentration lower than 136 mg mL^{-1} (Table S1) because of the less crosslinkings for A1-B. So it could not form stable three-dimensional interconnected networks but the precipitations. Secondly, the polysiloxane aerogels could not be formed under 16 MPa. Although the precursors, A1 and

B, can be well solubilized in $scCO_2$ at 16 MPa, during the gelation, the crosslinked A1-B slowly precipitated and finally the hard and fragile monoliths was formed, which displays as an amorphous closely packed bulk aggregate (Fig. S11). It suggests that $scCO_2$ can not only work as solvent to dissolve the precursors and reaction medium for gelation, but also favor the gel skeleton formation at an enough high pressure. For comparison, we prepared the A1-B gel in dioxane by freeze-drying. The gel appears as a shrunk plastic and the network collapses (Fig. S12), that is similar to the reported silicon aerogels formed in organic solvents.[36, 37] It further proves that $scCO_2$ is critical for the aerogel formation.

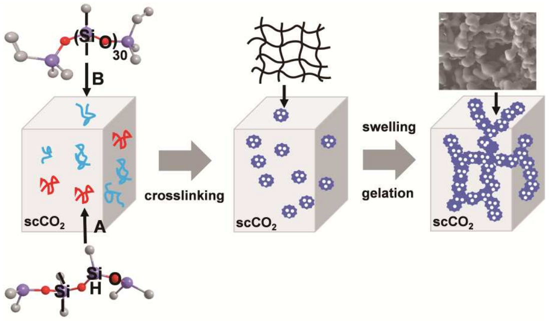


Fig. 6 Schemetic illustration for the formation of polysiloxane aerogels in $scCO_2$.

Based on the above experimental results, a mechanism for the formation of polysiloxane aerogels is proposed in Fig. 6. The two precursors (A and B) are first dissolved in $scCO_2$, forming a homogeneous solution. During the hydrosilylation process, the crosslinked polysiloxanes own worse solubility than monomers in $scCO_2$ and could partially precipitate as the micron-sized aggregates shown in Fig. 2. Meanwhile, the $scCO_2$ could suffuse, swell and support the microstructures in that aggregates and the whole skeleton of the final aerogel. Therefore, after degasing and drying procedure the polysiloxane aerogel preserving the skeleton structure are obtained.

Conclusions

In conclusion, an excellent superhydrophobic polysiloxane aerogel has been prepared through a simple and green one-pot hydrosilylation cross-linking in $scCO_2$. The polysiloxane aerogel exhibits high oil absorption capacity, superior recyclability, extraordinary mechanical property and robust stability regardless of the harsh external conditions, such as various organic solvents, heating and cooling tests. It is believed that the distinct chemical compositions from polydimethylsiloxane and highly porous microstructures afford the resulting aerogel superhydrophobicity, outstanding elasticity and reusability. The robust polysiloxane aerogel has great potentials in the industrial applications of oily water treatment, oil spill clean-up and oil recovery.

Acknowledgements

We are grateful for the support by the National Basic Research Program of Ministry of Science and Technology of China (2012CB933201) and National Natural Science Foundation of China (51303187 and 51225306).

References

1 R. W. Field, *Nature*, 2012, **489**, 41-42.
2 K. Li, J. Ju, Z. X. Xue, J. Ma, L. Feng, S. Gao and L. Jiang, *Nat. Commun.*, 2013, **4**, 2276.
3 M. Schrope, *Nature*, **2011, 472**, 152-154.
4 W. Lei, D. Portehault, D. Liu, S. Qin and Y. Chen, *Nat. Commun.*, 2013, **4**, 1777.
5 J. Yuan, X. Liu, O. Akbulut, J. Hu, S. L. Suib, J. Kong and F. Stellacci, *Nat. Nanotechnol.*, 2008**, 3**, 332-336.
6 M. O. Adebajo, R. L. Frost, J. T. Kloprogge, O. Carmody and S. *J.* Kokot, *Porous Mat.*, 2003, **10**, 159-170.
7 L. Feng, Z. Y. Zhang, Z. H. Mai, Y. M. Ma, B. Q. Liu, L. Jiang and D. B. Zhu, *Angew. Chem. Int. Ed.*, 2004, **43**, 2012-2014.
8 W. B. Zhang, Y. Z. Zhu, X. Liu, D. Wang, J. Y. Li, L. Jiang and J. Jin, *Angew. Chem. Int. Ed.*, 2014, **53**, 856-860.
9 H. J. Li, Y. M. Cao, J. J. Qin, X. M. Jie, T. H. Wang, J. H. Liu and Q. Yuan, *J.Membr. Sci.*, 2006, **279**, 328-335.
10 C. B. Spricigo, A. Bolzan, R. A. F. Machado, L. H. C. Carlson and J. C. C. Petrus, *J. Membr. Sci.*, 2001, **188**, 173-179.
11 A. G. Sadekar, S. S. Mahadik, A. N .Bang, Z. J. Larimore; C. A. Wisner, M. F. Bertino, A. K. Kalkan, J. T. Mang, C. Sotiriou-Leventis and N. Leventis, *Chem. Mater.*, 2012, **24**, 26-47.
12 M. A. Worsley, S. O. Kucheyev, J. D. Kuntz, T. Y. Olson, T. Y. J. Han, A. V. Hamza, J. H. Jr. Satcher and T. F. Baumann, *Chem. Mater.*, 2011, **23**, 3054-3061.
13 H. Sun, Z. Xu and C. Gao, *Adv. Mater.*, 2013, **25**, 2554-2560.
14 Z. Y. Wu, C. Li, H. W. Liang, J. F. Chen and S. H. Yu, *Angew. Chem. Int. Ed.*, 2013, **52**, 2925-2929.
15 L. Li, S. Xiang, S. Cao, J. Zhang, G. Ouyang, L. Chen and C. Y. Su, *Nat. Commun.*, 2013, **4**, 1774.
16 G. Zu, J. Shen, W. Wang, L. Zou, Y. Lian, Z. Zhang, B. Liu and F. Zhang, *Chem. Mater.*, 2014, **26**, 5761-5772.
17 Q. Zheng, Z. Cai and S. Gong, *J. Mater. Chem. A*, 2014, **2**, 3110-3118.
18 S. T. Nguyen, J. Feng, N. T. Le, A. T. T. Le, H. Nguyen, V. B. C. Tan and H. M. Duong, *Ind. Eng. Chem. Res.*, 2013, **52**, 18386-18391.
19 J. T. Korhonen, M. Kettunen, R. H. A. Ras and O. Ikkala, *ACS Appl. Mater. Interfaces*, 2011, **3**, 1813-1816.
20 S. Yun, H. Luo and Y. Gao, *RSC Adv.*, 2014, **4**, 4535-4542.
21 L. X. Li, B. C. Li, L. Wu, X. Zhao and J. P. Zhang, *Chem. Comm.*, 2014, **50**, 7831-7833.
22 G. Hayase, K. Kanamori, G. Hasegawa, A. Maeno, H. Kaji and K. Nakanishi, *Angew. Chem. Int. Ed.*, 2013, **52**, 10788-10791.
23 G. Hayase, K. Kanamori, M. Fukuchi, H. Kaji and K. Nakanishi, *Angew. Chem. Int. Ed.*, 2013, **52**, 1986-1989.
24 Z. Wang, Z. Dai, J. J. Wu, N. Zhao and J. Xu, *Adv. Mater.*, 2013, **25**, 4494-4497.
25 H. Maleki, L. Duraes and A. *J.* Portugal, *Non-Cryst. Solids*, 2014, **385**, 55-74.
26 D. P. Mohite, Z. J. Larimore, H. Lu, J. T. Mang, C. Sotiriou-Leventis and N. Leventis, *Chem. Mater.*, 2012, **24**, 3434-3448.
27 J. P. Randall, M. A. B. Meador and S. C. Jana, *J. Mater. Chem. A*, 2013, **1**, 6642-6652.
28 A. Fidalgo, J. P. S. Farinha, J. M. G. Martinho, M. E. Rosa and L. M. Ilharco, *Chem. Mater.*, 2007, **19**, 2603-2609.
29 A. Du, B. Zhou, Z. Zhang and J. Shen, *Materials*, 2013, **6**, 941-968.
30 Y. Duan, S. C. Jana, B. Lama and M. P. Espe, *Langmuir*, 2013, **29**, 6156-6165.

31 J. L. Gurav, A. V. Rao, D. Y. Nadargi and H. H. Park, *J. Mater. Sci.*, 2010, **45**, 503-510.

32 M. Sachithanadam and S. C. Joshi, *J. Mater. Sci.*, 2014, **49**, 163-179.

33 J. C. H. Wong, H. Kaymak, S. Brunner and M. M. Koebel, *Microporous Mesoporous Mat.*, 2014, **183**, 23-29.

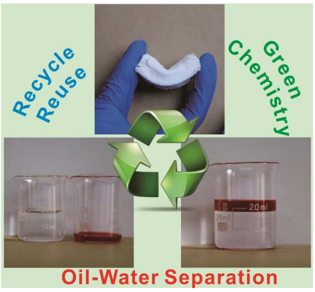
34 A. Javadi, Q. F. Zheng, F. Payen, A. Javadi, Y. Altin, Z. Y. Cai, R. Sabo and S. Q. Gong, *ACS Appl. Mater. Interfaces*, 2013, **5**, 5969-5975.

35 J. M. Al-Besharah, O. A. Salman and S. A. Akashah, *Ind. Eng. Chem. Res.*, 1987, **26**, 2445-2449.

36 L. Durães, M. Ochoa, N. Rocha, R. Patrício, N. Duarte, V. Redondo and A. Portugal, *J. Nanosci. Nanotechnol.*, 2012, **12**, 6828-6834.

37 C. Alié, N. Tcherkassova, F. Ferauche, S. Lambert, B. Heinrichs, R. Pirard and J. P. Pirard, *J. Non-Cryst. Solids*, 2006, **352**, 2763-2771.

For Table of Contents Only



A green one-pot synthesized flexible polysiloxane aerogel possesses extraordinary mechanical property, robust stability, and exhibits great potentials in oil-water separation.