Poly(dimethylsiloxane) Oil Absorbent with a Three-Dimensionally Interconnected Porous Structure and Swellable Skeleton

Aijuan Zhang, Mingjie Chen, Can Du, Huizhang Guo, Hua Bai, and Lei Li*

College of Materials, Xiamen University, Xiamen 361005, People's Republic of China

Supporting Information

ABSTRACT: Cleanup of oil spills is a worldwide challenge to prevent serious environmental pollution. A new kind of poly(dimethylsiloxane) (PDMS) oil absorbent with high absorption capacity and excellent reusability was prepared and used for oil/ water separation. The preparation process of PDMS oil absorbents involves direct curing of a PDMS prepolymer in a *p*-xylene solution in the presence of commercial sugar particles, which is simple and economic. PDMS oil absorbents have interconnected pores and a swellable skeleton, combining the advantages of porous



materials and gels. Absorption capacities of PDMS oil absorbents are 4-34 g/g for various oils and organic solvents, which are 3 times that reported previously. Owing to their hydrophobicity and oleophilicity, the as-obtained PDMS oil absorbents can selectively collect oils or organic solvents from water. The absorption process can be finished within tens of seconds. Furthermore, the absorbed oils or organic solvents can be recovered by compressing the oil absorbents, and after 20 absorbing/ recovering cycles, PDMS oil absorbents show little loss of their absorption capacities and own weights.

KEYWORDS: poly(dimethylsiloxane), oil absorbent, porous material, swell, reusability

INTRODUCTION

The treatment of oil spills and chemical leakage has become an important task of environmental protection to avoid pollution to water and fatal destruction to biodiversity.¹⁻³ Among technologies for cleaning up oils and organic solvents from water, absorption by an oil absorbent has distinct advantages of simple operation, low cost, and thorough cleanup without secondary pollution.^{4–6} At present, absorption materials for oils and chemicals fall into three categories: porous materials, structure-assembled fibers, and organogels.⁷⁻¹⁴ Three-dimensionally (3D) interconnected porous materials with hydrophobicity and oleophilicity are preferential candidates for oil absorption because of their outstanding selectivity for oil, high absorption capacities, fast absorption kinetics, excellent reusability, and oil recovery. Recently, aerogels based on carbon nanomaterials, such as carbon nanotube (CNT), graphene, carbon nanofiber, and carbon/inorganic hybrids, have drawn great attention because of their superhydropho-bicity and high porosity (more than 99%).^{15–19} For example, Gao et al. prepared CNT/graphene aerogels with ultrahigh oilabsorption capacities of 215-913 g/g toward a wide range of organic solvents and oils.¹⁷ However, considering the ultralow density of carbon nanomaterials, there will be various operational difficulties in practical application, such as that they can be blown up easily by a gentle breeze or breath. Meanwhile, the preparation of these carbon nanomaterials usually involves complicated processes, limiting their commercialization and industrialization on a large scale. Also, the cost of aerogels based on carbon nanomaterials is very high. Therefore, it is still necessary to develop new oil absorbents with low cost, considerable absorption capacity, simple preparation, and practical applicability.

Recently, poly(dimethylsiloxane) (PDMS) was found to be a promising candidate for oil-absorbent applications because of its oleophilicity, hydrophobicity, and commercial availability, and porous PDMS has been used to selectively separate oils or organic solvents from water.^{20,21} These porous PDMS materials can be prepared using sugar, salt, self-assembled colloidal microspheres, emulsion droplets, and CO2 gas generated by NaHCO3 as templates, among which the sacrificial sugartemplate technique has been the most popular because of its low cost and ecofriendliness.²²⁻²⁵ Choi et al. prepared a PDMS oil absorbent with high selectivity, fast absorption, and excellent reusability by a sugar-template method.²⁶ Compared with oil absorbents based on nanocarbon, PDMS oil absorbents have been easily prepared and had much more stability and practical applicability. However, its absorption capacities for oils and organic solvents were as low as 4-11 g/g. Moreover, the preparation process involves the shaping of sugar templates and the neeed for vacuum operation, inducing penetration of PDMS prepolymer into the templates. Therefore, other efforts should be made to improve the absorption capacity of PDMS oil absorbents and their preparation strategy.

From the viewpoint of the absorption mechanism, porous absorption materials store oils and organic solvents in pores, while gels retain them among the 3D cross-linked network of molecular chains or aggregates.²⁷ Therefore, for porous absorbents, the higher the porosity, the larger the absorption capacity.²⁸ However, considering the flexibility of the PDMS molecular chain, it is impossible to increase the porosity of

Received: July 19, 2013 Accepted: September 16, 2013 Published: September 16, 2013

ACS Applied Materials & Interfaces

PDMS without limit. High porosity may result in collapse of the porous structure. $^{\rm 29}$

Herein, to challenge the absorption-capacity limitation, we designed a novel PDMS oil absorbent by combining the absorption mechanisms of filling for porous materials and swell for organogels. These PDMS oil absorbents have interconnected pores and a swellable skeleton, both of which are able to absorb and retain oils. Without the shaping of sugar particles beforehand and vacuum operation, the preparation strategy is straightforward. Absorption capacities of PDMS oil absorbents were 4-34 g/g for various oils and organic solvents. Importantly, more than 80% of the absorbed oil or organic solvent can be recovered by squeezing the robust and flexible PDMS oil absorbents. Meanwhile, they can be used repeatedly over 20 times without decrease in their absorption capacities and loss of their own weights.

EXPERIMENTAL SECTION

Materials. Poly(dimethylsiloxane) (PDMS) prepolymer (Sylgard 184A) and thermal curing agent (Sylgard 184B) were bought from Dow Corning Corp. *p*-Xylene (PX), chloroform, cyclohexane, dichloromethane, *n*-hexane, toluene, aniline, acetone, methanol, petroleum ether (60-90 °C), and silcone oil were all chemically pure and purchased from Sinopharm Chemical Reagent Co., Ltd. (China). Gasoline and diesel were purchased from China National Petroleum Corp. Crude oil was provided by Oil Field of Tulufan-Hami (China). Saccharose particles of various sizes (coarsely granulated sugar, CGS; finely granulated sugar, FGS; soft sugar, SS) and maize oil were purchased from a local supermarket.

Preparation of PDMS Oil Absorbents. Typically, PDMS prepolymer (3.0 g) and thermal curing agent (0.3 g) were added to a vessel, and PX (6.6 g, 2 times that of the total prepolymer) was mixed. Then sugar particles [mixture of CGS and SS; 59.4 g, 1:1 (w/w)] were added to the prepared PDMS solution directly. After curing at 70 °C for 12 h, the blend was first soaked in water of 80 °C to dissolve the sugar and then in ethanol to remove PX. Consequently, after air drying, a PDMS oil absorbent was obtained.

Characterization of PDMS Oil Absorbents. The morphology of the PDMS oil absorbents prepared above was observed by a scanning electron microscope (TM3000, Hitachi, Japan) under an electron beam with an accelerating voltage of 15 keV and a working distance of 4 mm. All of the samples were coated with a thin layer of gold for better conductivity. The stress–strain curve was measured with a universal testing machine (Sun2500, Galdabini, Italy). The hydrophobicity and oleophilicity were estimated on a contact-angle measurement apparatus (DSA20, Kruss, Germany) with a 5 μ L droplet. The porosity (ϕ) was measured with a methanol saturation method²⁴ according to the following equation:

$$\phi = \frac{\gamma_{\rm sat} - \gamma_{\rm dry}}{\gamma_{\rm meth}}$$

where γ_{sat} , γ_{dry} , and γ_{meth} indicate the densities of saturated PDMS, dry PDMS oil absorbent, and methanol, respectively.

Oil and Organic Solvent Absorption by PDMS Oil Absorbents. Absorption capacities of PDMS oil absorbents for the following oils and organic solvents were measured: chloroform, cyclohexane, dichloromethane, *n*-hexane, toluene, aniline, acetone, methanol, petroleum ether, gasoline, diesel, crude oil, maize oil, and silicone oil. Their densities and solubility parameters are listed in Table S1 in the Supporting Information (SI).³⁰ PDMS oil absorbents cut into cubic shape (1 cm × 1 cm × 1 cm) were immersed in organic solvents and oils for 1 min and then picked out for weight and volume measurements. All experimental processes were carried out at room temperature (20 ± 5 °C) except for those specified. The mass absorption capacity (M_{abs}), volume absorption capacity (V_{abs}), and swell ratio were calculated by the following equation:

$$M_{abs} = \frac{m_{sat} - m_0}{m_0}$$
$$V_{abs} = \frac{m_{sat} - m_0}{\rho \nu_0}$$
swell raito = $\frac{\nu_{sat}}{\nu_0}$

m _ m

where m and ν indicate the weight and volume of the PDMS oil absorbent before (0) and after (sat) saturation by organic solvent or oil, respectively, and ρ is the density of the absorbed oil.

RESULTS AND DISCUSSION

Preparation of PDMS Oil Absorbents. PDMS oil absorbents were synthesized by a modified sugar-template method. Compared with the conventional sugar-template method, the modified method avoided vacuum operation and the preparation of a shaped sugar template beforehand. Figure 1 illustrates the preparation process in detail. PDMS

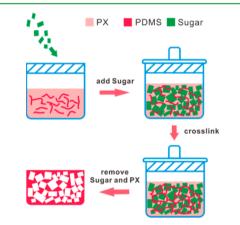


Figure 1. Schematic illustration for the preparation of PDMS oil absorbents via a sugar-particle-template method.

prepolymer was diluted with PX and then mixed with commercially available sugar particles. After thermal curing and removal of the sugar template and PX, interconnected pores were created in PDMS (Figure 2). The solvent of PX decreased the viscosity of PDMS prepolymer, increased its fluidity, and prevented the formation of bubbles, allowing the absence of vacuum operation. Hence, without a complicated procedure and expensive equipment, the advanced preparation method of porous PDMS is suitable for large-scale production. More importantly, the presence of PX caused PDMS to be in a gel state after curing, resulting in a swellable skeleton of the PDMS oil absorbent, whereas cross-linking a polymer in solution was the traditional method for preparing the gel.³¹ Hence, a simple novel method was developed for the preparation of highly swellable porous PDMS.

Performance of PDMS Oil Absorbents. 3D interconnected pores and high porosity are crucial characteristics for ideal oil absorbents. From Figure 2, it is observed that all of the obtained PDMS absorbents have interconnected pore structures, which result from rupture of the PDMS skeleton with shrinkage of curing. The porosity of PDMS oil absorbents fell into the range of 50–81%, depending on the size of the sugar-particle template (Table 1). Smaller sugar particles led to higher porosity because of their higher stacking density. However, the smallest sugar, SS, resulted in a lower porosity than that of FGS, which was attributed to the collapse of the

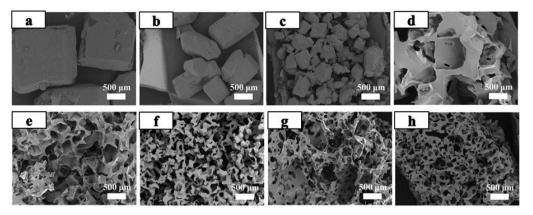


Figure 2. Scanning electron microscopy (SEM) images of sugar particles and PDMS oil absorbents. (a)-(c): CGS, FGS, and SS, respectively. (d)-(h): PDMS oil absorbents prepared by CGS, FGS, SS, a mixture of CGS and SS, and a mixture of FGS and SS, respectively.

Table 1. Preparation Conditions and Properties of PDMS Oil Absorbents

	I	preparation co	nditions	properties					
entry	PDMS (g)	PX (g)	sugar ^a	density (g/cm ³)	porosity (%)	absorption capacity ^b (g/g)	swell ratio ^b (ν/ν_0)		
1	3.3	6.6	CGS/SS, 59.4 g	0.18	81	20.8	6.0		
2	3.3	6.6	FGS/SS, 59.4 g	0.21	80	21.6	6.2		
3	3.3	6.6	CGS, 59.4 g	0.33	60	18.0	6.1		
4	3.3	6.6	FGS, 59.4 g	0.31	73	19.3	6.1		
5	3.3	6.6	SS, 59.4 g	0.36	50	14.4	6.0		
6	3.3	0	CGS/SS, 59.4 g	0.12	83	13.8	1.5		
7	3.3	3.3	CGS/SS, 59.4 g	0.16	79	15.8	4.2		
8	3.3	9.9	CGS/SS, 59.4 g	0.42	43	14.7	8.7		
9	3.3	6.6	0	1.10		1.6	3.5		
10	3.3	6.6	CGS/SS, 99.0 g	0.17	84	20.1	6.1		
^a If it is a	mixed sugar t	he weight rat	tio of the two kinds o	of sugar is 1.1 ^b The	absorbed oil is r	etroleum ether			

^aIf it is a mixed sugar, the weight ratio of the two kinds of sugar is 1:1. ^bThe absorbed oil is petroleum ether.

PDMS skeleton because it was too thin and soft to support its own weight. Therefore, it was difficult to achieve porosity higher than 73% using sugar particles of single size as the template. Mixed sugar particles with various sizes offered an alternative to give high porosity because of its unique hierarchical porous structure, which contained big voids with some windows on the wall (Figure 2g,h). The thick pore walls provided mechanical support to the 3D structure, while the small open windows on the wall gave an additional porosity. Experimentally, mixed sugars gave the highest porosity of more than 84%, higher than the previously reported value of 64%.³² However, the amount of sugar particles had a slight effect on the porosity of the obtained PDMS oil absorbents. Under the same preparation conditions except for the size of the sugar particles, absorption capacities of PDMS oil absorbents for petroleum ether increased from 14.4 to 21.6 g/g with their porosity increasing from 50% to 80%.

Unlike other porous oil absorbents, the obtained PDMS oil absorbents are swellable, which can increase their absorption capacities further. As listed in Table 1, with the amount of PX increasing from 0 to 3 times that of PDMS, the swell ratio of PDMS oil absorbents increased from 1.5 to 8.7 when saturated by petroleum ether. The main reason for the swell performance of PDMS oil absorbents may be that the presence of PX decreased the cross-linking degree of PDMS. Also the more PX used, the less is the cross-linking degree of PDMS. With the amount of PX increasing from 0 to 2 times that of PDMS, the petroleum ether absorbency of the obtained PDMS oil absorbents increased from 13.8 to 20.8 g/g. However, a further

increase of PX led to a decrease of the absorption capacity, although the swell ratio still increased. This can be explained by a decrease of the porosity. When the amount of PX was over 2 times that of PDMS prepolymer, the cross-linking degree of PDMS was so low that the skeleton could not support its own weight and collapsed (Figure S1 in the SI). With collapse, the porosity decreased from 81% to 43%.

Importantly, the as-obtained PDMS oil absorbents have intrinsic hydrophobicity and oleophilicity. The water contact angle (CA) of the PDMS oil absorbent was as high as 144° (Figure 3a and the SI, movie 1), and when it was pressed into water, its surface was surrounded by air bubbles, demonstrating its hydrophobicity. On the contrary, the CAs of PDMS oil absorbents for organic solvents and oils were almost 0°. Once droplets of organic solvent or oil were in contact with the surface of the PDMS oil absorbent, they spread and penetrated

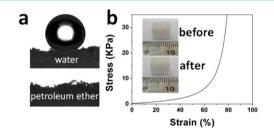


Figure 3. (a) High hydrophobicity and oleophilicity of PDMS oil absorbents. (b) Stress-strain curve of the PDMS oil absorbent. Insets: digital photographs before and after compression.

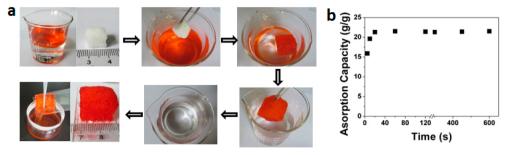


Figure 4. (a) Removal and collection of petroleum ether (dyed with Sudan III for observation) from the surface of water by a piece of PDMS oil absorbent. (b) Absorption rate curve of the PDMS oil absorbent for petroleum ether.

oil absorbent	oil	density of the oil absorbent (mg/cm ³)	$M_{ m abs} \ ({ m g/g})$	V_{abs}^{a} (cm ³ /cm ³)	desorption methods	ref
swellable porous PDMS	gasoline	180	22	5.35	squeeze or wash	this worl
	diesel		12	2.57		
	crude oil		9.0	2.19		
	chloroform		34	4.14		
	toluene		18.7	3.9		
PDMS sponge	chloroform	180	11	1.34	squeeze	26
	toluene		5.0	1.03		
	transformer oil		4.3	0.89		
TiO ₂ /nanocellulose aerogel	paraffin	30	27	0.88	wash	8
Me-CNT sponges	diesel	15	56	1.00	heat treatment	15
graphite/isobutylene—isoprene aerogel	diesel	154	22	3.96	squeeze or centrifuge	7
carbon nanofiber aerogel	diesel	6.0	170	1.21	burn	18
CNT/GGO aerogel	crude oil	0.45	350	0.18	burn	17
graphene-coated sponge	chloroform		165	1.26	squeeze or distill	38
CNT solids	hexane	24	26	0.96	burn or squeeze	39
graphene/CNT foam	toluene	6.9	126	1.00	wash or heat treatment	40
graphene/polypyrrole foams	diesel	10	108	1.29	wash	41
conjugated microporous polymer	octane	27	23	0.60	wash	42
CNT sponge	diesel	7.5	143	1.28		19
cotton towel	diesel	240	5	1.43		19
PU sponge	diesel	30	42	1.50		19

into the 3D porous structure immediately (Figure 3a and the SI, movie 1). The combined hydrophobicity and oleophilicity of the PDMS oil absorbent are attributed to the low surface free energy of the PDMS material and the porous structure.^{10,33,34} PDMS was reported to have a low surface free energy of 20-30 mN/m, and a smooth surface showed excellent hydrophobicity with a water CA of 105° .³³ According to the Cassie–Baxter wetting model, porous morphology can increase the water CA of the hydrophobic surface by air entrapment.³⁵ Thus, the water CA of the PDMS oil absorbent is larger than that of smooth PDMS. In contrast to water, organic solvents and oils have surface free tensions (20-30 mN/m) comparable to that of PDMS, so they can spread on PDMS and penetrate into the pores of oil absorbents under wicking. These properties enable the oil absorbents to separate oil from water.

The obtained PDMS oil absorbent, similar to a sponge, has a prominent elasticity. Its stress—strain curve under compression is plotted in Figure 3b, in which the modulus increases slowly until a strain of about 60%, a typical performance of soft foam. It can be compressed to 20% of its original volume and recover its original state immediately after decompression, which favors recycle of PDMS oil absorbents and collection of the absorbed oil. The elasticity is due to the flexible chemical chain of PDMS (-Si-O-). Also, the repeatable compressibility is mainly attributed to the high content of pores filled with air. In addition, soft PDMS oil absorbents can be readily tailor-made into any customized shape and size by scissors or knives, convenient for practical application and competitive with commercial-scale production.

Selective Absorption of Oils and Organic Solvents from Water. With 3D interconnected porous structure, combined hydrophobicity and oleophilicity, and flexibility, the as-obtained porous PDMS may serve as an ideal candidate as an oil absorbent for oil/water separation purposes. As expected, the obtained PDMS oil absorbents had excellent selective absorption for oil from water. When placed on the surface of an oil/water mixture, PDMS oil absorbents absorbed oil completely in tens of seconds, and all water was left (Figure 4a and the SI, movie 2). The absorption rate curve of the PDMS oil absorbent toward petroleum ether is plotted in Figure 4b, in which the maximum absorbency of 22 g/g is observed in 20 s. The absorption rate is much higher than that of gel oil absorbents, which reached a maximum absorbency in 2 h or even longer.^{12,36,37} Moreover, PDMS oil absorbents

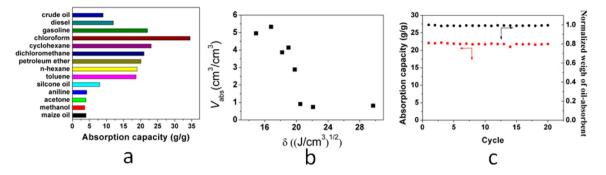


Figure 5. (a) Mass absorption capacities of PDMS oil absorbents for various organic solvents and oils. (b) Volume absorption capacities of PDMS oil absorbents for organic solvents with various solubility parameters. (c) Stability of PDMS oil absorbents with multirecycles.

saturated by oil can float on the surface of water and be taken out by tweezers without leakage of oil, exhibiting excellent oilkeeping performance, which will be crucial for oil collection in practical application.

PDMS oil absorbents prepared from various conditions had different absorption capacities for oil, as listed in Table 1, which can be attributed to their various porosities and cross-linking degrees. Entry 2 gave the PDMS oil absorbent the highest absorption capacity. Table 2 compares the absorption capacities of several typical oil absorbents reported in the literature. The swellable porous PDMS oil absorbent can absorb chloroform for 34 g/g, more than 3 times that of a PDMS sponge, reported by Choi et al. (11 g/g).²⁶ The high oil absorbency is mainly attributed to the swellable pore walls, which can absorb additional oil. However, $M_{\rm abs}$ of the PDMS oil absorbent is not as high as that of carbon aerogels, which possess superlow density and high porosity. Although $M_{\rm abs}$ is widely used in the literature, $V_{\rm abs}$ can fully characterize the oil absorbent because it is not influenced by the density of oils and oil absorbents.^{8,15} If compared by V_{abst} the absorbency of the PDMS oil absorbent is higher than those of other porous oil absorbents, including aerogels based on carbon nanomaterials.

Absorbent capacities of PDMS oil absorbents for various kinds of oils and organic solvents were investigated. $M_{\rm abs}$ values of PDMS oil absorbents fell in the range of 4-34 g/g (Figure 5a). Also, for gasoline, diesel, and crude oil, $M_{\rm abs}$ values were 22, 12, and 9.0 g/g, respectively. Unlike other porous oil absorbents, $M_{\rm abs}$ did not increase with an increase in the oil density,⁸ which was attributed to the swellable performance of PDMS oil absorbents. As discussed above, the swell ratios of PDMS oil absorbents have an important effect on their absorption capacities. However, the swell ratio determines not only the cross-linking degree of PDMS oil absorbents but also the dissolving capacity of oils to PDMS.43 The solubility parameter (δ) is universal to estimate the dissolving capacity of solvent to polymer.³⁰ Therefore, the correlation between the absorption capacity and δ of oils and organic solvents was investigated. In order to take out the effect of the oil density, $V_{\rm abs}$ was used to describe the absorption capacity. As shown in Figure 5b, V_{abs} had a peak value of 5.3 cm³/cm³ near δ = 16.8 $(J/cm^3)^{1/2}$ (cyclohexane), and larger or smaller δ caused a decrease in V_{abs} . When δ was increased to 29.7 $(J/cm^3)^{1/2}$ (methanol), V_{abs} decreased to 0.81 cm³/cm³. At the same time, the swell ratios of PDMS oil absorbents after saturated by oils and organic solvents decreased from 5.5 to 1.0 (Table S1 in the SI). This can be explained by the solubility parameter theory. One substance is soluble in another if the internal energy

change, ΔE , is small enough.⁴⁴ For interaction between the solvent and polymer,

$$\Delta E = \varphi_1 \varphi_2 (\delta_1 - \delta_2)^2$$

where δ and φ are the solubility parameter and volume fraction of the solvent (1) and polymer (2), respectively. Therefore, if the oil has a δ value close to that of PDMS, the swelling process of PDMS by the oil will become thermodynamically favorable because of small ΔE . Therefore, those oils and organic solvents whose δ values are close to that of PDMS [14.9–15.5 (J/ cm³)^{1/2}], such as petroleum ether, gasoline, cyclohexane, and *n*hexane, have large $V_{\rm abs}$. The above correlation between $V_{\rm abs}$ and δ may be useful for estimating the absorption capacity of the absorbents for other organic solvents and oils and will be helpful for designing new oil absorbents.

More importantly, PDMS oil absorbents can be recycled by a simple treatment of compression, which is a crucial requirement in practical application to reduce cost (Figure 4a and the SI, movie 2). At the same time, more than 80% of the absorbed oil can be gathered and reused by compression. To evaluate the recyclability and durability of PDMS oil absorbents, they were immersed in petroleum ether repeatedly, followed by compression to recover the oil and drying. After 20 recycles, a decrease in neither the absorption capacity nor the weight of the PDMS oil absorbent was observed (Figure 5c), indicating their excellent stability and durability. The excellent recyclability of the PDMS oil absorbent can attributed to the excellent flexibility and 3D interconnected porous structure. In addition, compared with other porous oil absorbents modified by hydrophobic materials, such as graphene,³⁸ CNT,¹⁵ or polysiloxane,¹⁰ the PDMS oil absorbent has inherent properties of hydrophobicity and oleophilicity. The PDMS material is resistant to bacteria and heat,^{45,46} which helps for long-time storage and use. Therefore, the PDMS oil absorbent is competitive in stability and durability. Also its absorption capacity for oils had no obvious change in a wide range of temperatures (Figure S2 in the SI), allowing their applications under extreme conditions.

CONCLUSIONS

In conclusion, a new kind of PDMS oil absorbent with high absorption capacity and excellent reusability was reported. The preparation process via the sugar-particle-template method was simple and economic without demanding a shaped template and vacuum operation, feasible for large-scale production. PDMS oil absorbents possessed 3D interconnected porous structures with a swellable skeleton. Hence, with the synergistic effect of filling and swelling, absorption capacities of PDMS oil

ACS Applied Materials & Interfaces

absorbents are 4-34 g/g for oils and organic solvents, depending on their solubility parameter and density, which are 3 times that reported previously. Owing to their hydrophobicity and oleophilicity, the obtained PDMS oil absorbents can selectively collect oils and organic solvents from water, and the separation process can be finished in tens of seconds. Furthermore, the absorbed oils and organic solvents can be recovered by a simple treatment of compression, and PDMS oil absorbents can be recycled more than 20 times without loss of their absorption capacities and own weights. Because of easy preparation, good commercial availability of raw materials, high absorption capacity, and excellent reusability, the PDMS oil absorbent reported here might be a promising candidate for cleanup of oil spills and chemical leaks. More importantly, a new strategy was proposed to improve the absorption capacity of a polymer-based absorbent, which may pave a new way for designing novel oil absorbents.

ASSOCIATED CONTENT

S Supporting Information

Density and solubility parameter of organic solvents and oils and M_{abs} , V_{abs} , and swell ratio of PDMS oil absorbents for various organic solvents and oils, SEM images of PDMS oil absorbents prepared with various amounts of PX, absorption capacities of a PDMS oil absorbent at various temperatures, the hydrophobicity and oleophilicity, and the oil cleaning capability of PDMS oil absorbents. This material is available free of charge via the Internet at http://pubs.acs.org.

AUTHOR INFORMATION

Corresponding Author

*E-mail: lilei@xmu.edu.cn. Tel: +86-592-2186296. Fax: +86-592-2183937.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

L.L. gratefully acknowledges the National Natural Science Foundation of China (Grants 51035002, 21174116, and 20974089) and Key Laboratory for Ultrafine Materials of Ministry of Education.

REFERENCES

- (1) Jernelov, A. Ambio 2010, 39, 353-366.
- (2) Kingston, P. F. Spill Sci. Technol. Bull. 2002, 7, 53-61.
- (3) Shannon, M. A.; Bohn, P. W.; Elimelech, M.; Georgiadis, J. G.; Marinas, B. J.; Mayes, A. M. *Nature* **2008**, 452, 301–310.
- (4) Gaaseidnes, K.; Turbeville, J. Pure Appl. Chem. 1999, 71, 95–101.
 (5) Nordvik, A. B. Spill Sci. Technol. Bull. 1995, 2, 17–46.
- (6) Wayment, E. C.; Wagstaff, B. Pure Appl. Chem. **1999**, 71, 203–208.
- (7) Hu, Y.; Liu, X.; Zou, J.; Gu, T.; Chai, W.; Li, H. ACS Appl. Mater. Interfaces **2013**, *5*, 7737–7742.
- (8) Korhonen, J. T.; Kettunen, M.; Ras, R. H. A.; Ikkala, O. ACS Appl. Mater. Interfaces 2011, 3, 1813–1816.
- (9) Wu, J.; Wang, N.; Wang, L.; Dong, H.; Zhao, Y.; Jiang, L. ACS Appl. Mater. Interfaces **2012**, *4*, 3207–3212.
- (10) Zhu, Q.; Chu, Y.; Wang, Z.; Chen, N.; Lin, L.; Liu, F.; Pan, Q. J. Mater. Chem. A 2013, 1, 5386–5393.
- (11) Wang, J. T.; Zheng, Y. A.; Wang, A. Q. Chem. Eng. J. 2012, 213, 1–7.
- (12) Wrede, M.; Ganza, V.; Bucher, J.; Straub, B. F. ACS Appl. Mater. Interfaces **2012**, *4*, 3453–3458.

- (13) Prathap, A.; Sureshan, K. M. Chem. Commun. 2012, 48, 5250–5252.
- (14) Lin, J.; Tian, F.; Shang, Y.; Wang, F.; Ding, B.; Yu, J. Nanoscale **2012**, *4*, 5316–5320.
- (15) Gui, X.; Zeng, Z.; Lin, Z.; Gan, Q.; Xiang, R.; Zhu, Y.; Cao, A.; Tang, Z. ACS Appl. Mater. Interfaces 2013, 5, 5845–5850.
- (16) Chu, Y.; Pan, Q. M. ACS Appl. Mater. Interfaces 2012, 4, 2420–2425.
- (17) Sun, H.; Xu, Z.; Gao, C. Adv. Mater. 2013, 25, 2554-2560.
- (18) Wu, Z.; Li, C.; Liang, H.; Chen, J.; Yu, S. Angew. Chem., Int. Ed. 2013, 52, 2925–2929.
- (19) Gui, X.; Wei, J.; Wang, K.; Cao, A.; Zhu, H.; Jia, Y.; Shu, Q.; Wu, D. Adv. Mater. **2010**, 22, 617–621.
- (20) Park, I.; Efimenko, K.; Sjoblom, J.; Genzer, J. J. Dispersion Sci. Technol. 2009, 30, 318-327.
- (21) Uragami, T. Polym. J. 2008, 40, 485-494.
- (22) Jiao, K.; Graham, C. L.; Wolff, J.; Iyer, R. G.; Kohli, P. J. Membr. Sci. 2012, 401, 25–32.
- (23) Li, J.; Zhang, Y. Chem. Mater. 2007, 19, 2581-2584.
- (24) Peng, S.; Hartley, P. G.; Hughes, T. C.; Guo, Q. Soft Matter 2012, 8, 10493-10501.
- (25) Han, J. W.; Kim, B.; Li, J.; Meyyappan, M. Appl. Phys. Lett. 2013, 102, 051903.
- (26) Choi, S. J.; Kwon, T. H.; Im, H.; Moon, D. I.; Baek, D. J.; Seol, M. L.; Duarte, J. P.; Choi, Y. K. ACS Appl. Mater. Interfaces 2011, 3, 4552–4556.
- (27) Yang, J. China Synth. Resin Plast. 1991, 39-43.
- (28) Tan, D. Z.; Fan, W. J.; Xiong, W. N.; Sun, H. X.; Li, A.; Deng, W. Q.; Meng, C. G. Eur. Polym. J. 2012, 48, 705-711.
- (29) You, L.; Temiyasathit, S.; Tao, E.; Prinz, F.; Jacobs, C. R. Cell. Mol. Bioeng. 2008, 1, 103–107.
- (30) Hansen, C. M. Ind. Eng. Chem. Prod. Res. Dev. 1969, 8, 2-11.
- (31) Deng, S. J.; Xu, H. J.; Jiang, X. S.; Yin, J. Macromolecules 2013, 46, 2399–2406.
- (32) Cha, K. J.; Kim, D. S. Biomed. Microdevices 2011, 13, 877-883.
- (33) Khorasani, M. T.; Mirzadeh, H.; Kermani, Z. Appl. Surf. Sci. 2005, 242, 339–345.
- (34) Zhang, X.; Li, Z.; Liu, K.; Jiang, L. Adv. Funct. Mater. 2013, 23, 2881–2886.
- (35) Cassie, A.; Baxter, S. Trans. Faraday Soc. 1944, 40, 546-551.
- (36) Song, C.; Ding, L.; Yao, F.; Deng, J. P.; Yang, W. T. Carbohydr. Polym. 2013, 91, 217–223.
- (37) Ono, T.; Sada, K. J. Mater. Chem. 2012, 22, 20962-20967.
- (38) Nguyen, D. D.; Tai, N. H.; Lee, S. B.; Kuo, W. S. *Energy Environ. Sci.* **2012**, *5*, 7908–7912.
- (39) Hashim, D. P.; Narayanan, N. T.; Romo-Herrera, J. M.; Cullen, D. A.; Hahm, M. G.; Lezzi, P.; Suttle, J. R.; Kelkhoff, D.; Munoz-Sandoval, E.; Ganguli, S.; Roy, A. K.; Smith, D. J.; Vajtai, R.; Sumpter, B. G.; Meunier, V.; Terrones, H.; Terrones, M.; Ajayan, P. M. *Sci. Rep.* **2012**, *2*, 363.
- (40) Dong, X.; Chen, J.; Ma, Y.; Wang, J.; Chan-Park, M. B.; Liu, X.; Wang, L.; Huang, W.; Chen, P. *Chem. Commun.* **2012**, *48*, 10660–10662.
- (41) Li, H.; Liu, L.; Yang, F. J. Mater. Chem. A 2013, 1, 3446-3453.
- (42) Li, A.; Sun, H.; Tan, D.; Fan, W.; Wen, S.; Qing, X.; Li, G.; Li,
- S.; Deng, W. Energy Environ. Sci. 2011, 4, 2062-2065.
- (43) Rao, K. V.; Mohapatra, S.; Maji, T. K.; George, S. J. Chem.—Eur. J. 2012, 18, 4505–4509.
- (44) Barton, A. F. M. CRC Handbook of solubility parameters and other cohesion parameters, 1983 ed.; CRC Press: Boca Raton, FL, 1991.
- (45) Baud-Grasset, F.; Palla, J. C. U.S. Patent 6,020,184, 2000.
- (46) Camino, G.; Lomakinb, S. M.; Lazzari, M. Polymer 2001, 42, 2395-2402.