Amino Carbon Nanotube Modified Reduced Graphene Oxide Aerogel for Oil/Water Separation

Jingyi Cai^{1#}, Jing Tian^{1#}, Hongbo Gu^{1*} and Zhanhu Guo²

In this work, reduced graphene oxide/amino multi-walled carbon nanotube (rGO/MWCNTs-NH₂) aerogel has been successfully fabricated through the chemical reaction between carboxyl groups on graphene oxide (GO) and amino groups on multi-walled carbon nanotubes (MWCNTs-NH₂) in combination with freeze-drying and high-temperature annealing process. The optimal condition for fabrication of rGO/MWCNTs-NH₂ aerogel is mass ratio of MWCNTs-NH₂ to GO = 1:8 with GO concentration of 2 mg mL⁻¹. In addition, owing to the strong chemical bonding between amino groups on MWCNTs and carboxyl groups on GO, the improved hydrophobic property and microstructures of rGO/MWCNTs-NH₂ aerogel are obtained confirmed by scanning electron microscope (SEM), transmission electron microscopy (TEM), X-ray photoelectron spectroscopy (XPS) and contact angle measurement. After that, the adsorption tests and oil/water separation experiments prove that the rGO/MWCNTs-NH₂ aerogel possesses an excellent adsorption performance for various organic solvents and oil including ethyl acetate (152.79 g g⁻¹), cyclohexane (122.46 g g⁻¹), acetone (219.54 g g⁻¹), dichloromethane (242.31 g g⁻¹), and sesame oil (156.35 g g⁻¹), and achieves a high efficiency of oil/water separation within only 25 s. Our rGO/MWCNTs-NH₂ aerogel is expected to be a promising material for treatment of oil spills.

Keywords: Graphene oxide; Amino multi-walled carbon nanotubes; Aerogel; Oil adsorption; Oil/water separation

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1. Introduction

In recent years, oil spills and organic pollution caused by industrial accidents have brought serious environmental problems¹ and endangered people's health due to its carcinogenic content.² Adsorption,³⁻⁵ centrifugation,⁶ membrane separation,⁷ chemical demulsification,⁸ chemical oxidation,⁹ coagulation¹⁰ and electrochemical methods¹¹ are normally exploited to remove oil from polluted water. Among them, adsorption has become a research hotspot with the characteristics of high oil removal efficiency, no secondary pollution, simple processing, low energy consumption and low operation cost.¹² The adsorbents widely used in oily wastewater treatment include activated carbon,¹³ polyethylene,¹⁴ fly ash,¹⁵ bentonite,¹⁶ carbon fiber¹⁷ and super oil-adsorbing resin.¹⁸ However, they have some disadvantages, such as low oil/water separation efficiency and selectivity, difficult oil recovery and durable materials,¹⁹ which has severely limited their practical applications. Therefore, it is important to find an efficient and fast material for treating oily wastewater.

Graphene is a material, comprising a layer of carbon atoms with unique electronic characteristics.²⁰ Although it has excellent physical

properties, its poor solubility greatly hinders its processing and application.²¹ Therefore, graphene oxide (GO) is often prepared for further usage because the introduction of polar oxygen-containing groups and defects could make it to exhibit a better processability.²² Recently, aerogel has gradually attracted attentions due to its unique properties, such as high porosity, high specific surface area, low thermal conductivity and ultra-low density.^{12, 23} Among them, reduced GO aerogel has a large specific surface area and extremely hydrophobic characteristics to ensure the efficiency for the separation of oil and organic solvents from water, so it has been widely studied in the removal of oil and organic solvents from sewage.²⁴ However, due to the relatively poor mechanical properties of single rGO aerogel, the researchers commonly mix the GO with other materials to enhance its structure and properties. For example, Mi *et al.*²⁵ prepared cellulose/graphene aerogel with outstanding compression and recoverability properties that could adsorb 80-197 times its weights of oil and organic solvents. Wu *et al.*²⁶ reported a graphene-based aerogel with Cu nanoparticles and found that this aerogel exhibited an excellent catalytic performance and good capability to remove the various oils and dyes from wastewater. Unfortunately, there are few reports on the reduced GO aerogels chemical bonded with multi-walled carbon nanotubes (MWCNTs) for oil/water separation.

In this work, we have chosen the amino group functionalized MWCNTs (MWCNTs-NH₂) to fabricate the reduced GO/MWCNTs-NH₂ aerogel in order to form the strong chemical bonding between amino groups of MWCNTs and carboxyl groups on GO to boost the mechanical property and hydrophobic property of GO aerogel, which further improves its oil adsorption properties. The scanning electron microscope (SEM), transmission electron microscopy (TEM), X-ray photoelectron spectroscopy (XPS), and Fourier transform infrared spectroscopy (FTIR) have been conducted to study the microstructures

¹Shanghai Key Lab of Chemical Assessment and Sustainability, School of Chemical Science and Engineering, Tongji University, Shanghai 200092, China

²Integrated Composites Lab (ICL), Department of Chemical & Biomolecular Engineering University of Tennessee, Knoxville, TN, 37966, USA

*E-mail: hongbogu2014@tongji.edu.cn

[#]There authors are contributed equally.

and chemical structures of aerogels. Furthermore, the oil adsorption capability and oil/water separation capacity have been performed systematically.

2. Experimental

2.1 Materials

Graphite powder (~325 mesh, 99%), potassium persulfate ($K_2S_2O_8$, 99%), amino multi-walled carbon nanotubes (MWCNTs-NH₂), dichloromethane (CH_2Cl_2 , 99.5%) and Sudan Red B ($C_{24}H_{20}N_4O$, AR) were purchased from Alfa Aesar Chemical Co., Ltd., Beijing InnoChem Science and Technology Co., Ltd., Beijing Deke Daojin Science and Technology Co. Ltd., Aladdin Reagent Co., Ltd., and Maclean Biochemical Technology Co., Ltd., respectively. Phosphorus pentoxide (P_2O_5 , 99%), sulfuric acid (H_2SO_4 , 95–98%), potassium permanganate ($KMnO_4$, 99.5%), hydrogen peroxide (H_2O_2 , 30%), hydrochloric acid (HCl, 36–38%), L(+)-ascorbic acid ($C_6H_8O_6$, 99.7%), ethanol (C_2H_5O , 95%), ethyl acetate ($C_4H_{10}O$, 99.5%), and cyclohexane (C_6H_{12} , 99.5%) were purchased from Sinopharm Chemical Reagent Co., Ltd. All the chemicals were used as-received without any further treatment.

2.2 Synthesis of rGO aerogel

The GO was firstly fabricated *via* a modified Hummers method. Please see details in supplementary materials. After that, GO solution (2 mg mL⁻¹) and L(+)-ascorbic acid (LAA, mass ratio of GO : LAA = 1 : 2) were mixed for sonication of 10 min. Then the mixed solution was poured into a 25 mL of beaker with a sealing cap and put in a regular oven at temperature of 40 °C for 12 h to obtain the hydrogel. The obtained hydrogel was dialyzed in a 15% (v/v) ethanol solution for 6 h and then taken out for freeze of 12 h in a refrigerator. Thereafter, the frozen sample was placed in a freeze-dryer for 48 h to attain the aerogel. Finally, the rGO aerogel was acquired with annealing at 780 °C for 0.5 h in an infrared tube furnace (Beijing HuaCe Testing instrument Co., Ltd.).

2.3 Manufacture of rGO/MWCNTs-NH₂ aerogel

MWCNTs-NH₂ (mass ratio of MWCNTs-NH₂ to GO = 1:8) was ultrasonically dispersed in deionized water at room temperature and mechanically stirred with a speed of 150 r min⁻¹ for 1 h, and GO solution (2 mg mL⁻¹) was added for additional ultrasonic and mechanical stirring of 1 h. After that, the temperature was raised to 60 °C and kept for 2 h in order to have a chemical reaction between GO and MWCNTs-NH₂. After cooling down to room temperature, LAA (mass ratio of GO : LAA = 1 : 2) was added into above solution for sonication of 10 min. Then, the mixed solution was poured into a 25 mL beaker with a sealing cap and maintained at 40 °C for 12 h to obtain the hydrogel. The hydrogel was dialyzed in a 15% (v/v) ethanol solution for 6 h and then frozen in a refrigerator for 12 h. Afterward, the frozen sample was put in a freeze-dryer for 48 h to yield the aerogel. Finally, the aerogel was annealed at 780 °C for 0.5 h to acquire rGO/MWCNTs-NH₂ aerogel. With the purpose of obtaining the optimal preparation conditions of rGO/MWCNTs-NH₂ aerogel, the different GO concentrations (3, 4 and 5 mg mL⁻¹) and different mass ratios of MWCNTs-NH₂ to GO (1:6, 1:10, and 1:12) were applied to fabricate the rGO/MWCNTs-NH₂ aerogel with the same procedure as mentioned above.

2.4 Adsorption test

2.4.1 Oil adsorption test

Firstly, the weighing bottle filled with ethyl acetate was weighed on an electronic balance. Then, the aerogel was put into this weighing bottle

filled with ethyl acetate for adsorption of 3 min and removed quickly with tweezers. The weighing bottle was weighed again on the electronic balance. The weight difference was the adsorbed ethyl acetate. The adsorption tests of aerogel to cyclohexane, acetone, dichloromethane and sesame oil were done with the same procedure of ethyl acetate. The reported value was the average of three measurements with a deviation of 5%.

2.4.2 Oil/water separation test

The cyclohexane and Sudan Red B were poured into a small beaker and magnetically stirred for complete dissolution. Afterward, this solution was added into a beaker filled with deionized water. Then, the rGO/MWCNTs-NH₂ aerogel was used to do oil/water separation test.

2.5 Characterizations

The surface morphology of rGO/MWCNTs-NH₂ aerogel was observed on a field emission scanning electron microscope (FE-SEM, Hitachi S-4800 system) and a high resolution TEM (Tecnai G2 F20 S-TWIN). The chemical composition of the products was acquired by XPS (Kratos AXIS Ultra DLD spectrometer), FT-IR (Thermo Nicolet NEXUS spectrometer) and Raman spectra (Renishaw in Via Reflex Raman spectrometer). Adsorption-desorption isotherm curves of rGO aerogel and rGO/MWCNTs-NH₂ aerogel were measured by Tristar3020 specific surface area and porosity analyzer with nitrogen as the adsorbent at liquid nitrogen temperature (-196 °C), and then combined with the BET (Brunauer-Emmett-Teller) equation and the BJH (Barrett-Joyner-Halenda) model to analyze the specific surface area and pore size distribution of samples. Water contact angles of rGO aerogel and rGO/MWCNTs-NH₂ aerogel were measured on a contact angle measuring instrument (JC2000DS1, Shanghai Zhongchen Digital Technology Equipment Co., Ltd.).

3. Results and discussion

3.1 Optimal condition exploration for preparation of rGO/MWCNTs-NH₂ aerogel

The rGO/MWCNTs-NH₂ aerogels with different mass ratios of MWCNTs-NH₂ to GO (including 1:6, 1:8, 1:10 and 1:12) and different GO concentrations (2, 3, 4 and 5 mg L⁻¹) have been synthesized. The porous structure is observed in the rGO/MWCNTs-NH₂ aerogel fabricated with different mass ratios of MWCNTs-NH₂ to GO as depicted in the SEM images of Fig. S1. These samples were used to do the ethyl acetate adsorption test and the results are shown in Fig. S3(a). As the mass ratio of MWCNTs-NH₂ to GO is 1:6, 1:8, 1:10, 1:12, ethyl acetate adsorption capacity of aerogels is 137, 152.8, 116.7, 88.0 g g⁻¹, respectively, revealing that the best mass ratio of MWCNTs-NH₂ to GO for producing rGO/MWCNTs-NH₂ aerogel is 1:8.

After that, different GO concentrations (including 2, 3, 4 and 5 mg L⁻¹) have been selected to produce the rGO/MWCNTs-NH₂ aerogel under the MWCNTs-NH₂ to GO mass ratio of 1:8. And the obtained results are listed in Fig. S2 and S3(b). The adsorption capacity for ethyl acetate of aerogels is 152.8, 88.4, 78.7, 61.8 g g⁻¹, separately, for rGO/MWCNTs-NH₂ aerogel synthesized with GO concentration of 2, 3, 4 and 5 mg L⁻¹, Fig. S3(b), resulting from the decreased porous structure with the increasing GO concentration (as displayed in Fig. S2). This confirms that the best GO concentration for preparation of rGO/MWCNTs-NH₂ aerogel is 2 mg L⁻¹. In a word, the optimal GO concentration for synthesis of rGO/MWCNTs-NH₂ aerogel is MWCNTs-NH₂ to GO mass ratio of 1:8 with GO concentration of 2 mg L⁻¹.

3.2 Structure and morphology of rGO/MWCNTs-NH₂ aerogel

As shown in Fig. 1(a) and (b), it's found that the gained rGO/MWCNTs-NH₂ aerogel has a good cylindrical shape. Its diameter and height are respectively measured to be 1.95 and 2.37 cm. By weighing its weight on an electronic balance (42.9000 mg), its density is calculated to be 6.06 mg cm⁻³. It's seen that this aerogel could be placed on a flower, Fig. 1(c) and (d), with no obvious deformation, indicating that the aerogel possesses a very low density.

Fig. 2 displays the SEM images of (a) GO aerogel, (b) rGO aerogel, (c) MWCNTs-NH₂, (d) and (e) rGO/MWCNTs-NH₂ aerogel and (f) TEM image of rGO/MWCNTs-NH₂ aerogel. In Fig. 2(a), it's noted that the graphene sheet is completely peeled off, proving that the

graphite has been completely oxidized. There are many wrinkles on the rGO sheet after adding the reducing agent LAA and high temperature annealing, Fig. 1(b). In Fig. 1(c), the MWCNTs-NH₂ presents an irregular entangled network structure. As depicted in Fig. 1(d) and (e), the MWCNTs-NH₂ is noticed to be evenly dispersed within graphene sheet. Especially, in TEM image, Fig. 1(f), the MWCNTs-NH₂ was partially embedded inside the graphene layer, demonstrating that the MWCNTs-NH₂ is covalently bonded with GO because of chemical bonding between oxygen-containing functional groups on the GO and amino-group of MWCNTs-NH₂, leading to the uniform distribution of MWCNTs-NH₂ within graphene layer. From the elemental mapping

Fig. 1 (a) Diameter and (b) height measurements of rGO/MWCNTs-NH₂ aerogel; (c) and (d) digital photographs of rGO/MWCNTs-NH₂ aerogel, showing that this aerogel could be placed on a flower.

Fig. 2 SEM images of (a) GO aerogel; (b) rGO aerogel; (c) MWCNTs-NH₂ (d) and (e) rGO/MWCNTs-NH₂ aerogel, and (f) TEM image of rGO/MWCNTs-NH₂ aerogel.

image of rGO/MWCNTs-NH₂ aerogel (Fig. S4), the existence of N element (red) is attributed to the addition of MWCNTs-NH₂. The uniform distribution of N element further demonstrates the possible chemical reaction between MWCNTs-NH₂ and GO.

In the Raman spectra, Fig. 3(a), GO aerogel mainly has two characteristic peaks, containing G peak caused by the E_{2g} vibration model of sp^2 hybrid carbon atoms (near 1591 cm⁻¹) and D peak (1354 cm⁻¹) resulted from the defect structure of GO edge and amorphous structure.²⁷ In comparison, the D peak and G peak of rGO aerogel respectively appear at 1348 and 1577 cm⁻¹. The presence of peaks at 2686 and 2911 cm⁻¹ attributed to the two phonon lattice vibrations of

graphene, revealing that structural defects of rGO aerogel are reduced relative to GO aerogel. The shift in the D peak (1344 cm⁻¹) of the rGO/MWCNTs-NH₂ aerogel with its G peak at 1589 cm⁻¹ compared with GO aerogel manifest that there is an interaction between GO and MWCNTs-NH₂.

The FTIR spectrum (Fig. 3(b)) of GO aerogel reveals the existence of a large number of oxygen-containing groups in the chemical structure of GO aerogel, such as the stretching vibration peaks of C-O-C and C=O at 1045^{28, 29} and 1732 cm^{-1, 30, 31} respectively. These peaks are significantly reduced or disappear in FTIR spectrum of rGO aerogel, indicating that GO aerogel has been successfully reduced. The

Fig. 3 (a) Raman spectra; (b) FT-IR spectra; (c) high resolution XPS spectrum of C1s; and (d) nitrogen adsorption-desorption isotherms and pore size distributions of rGO/MWCNTs-NH₂ aerogel.

Fig. 4 Water contact angle between (a) rGO aerogel and (b) rGO/MWCNTs-NH₂ aerogel.

reduction of these hydrophilic oxygen-containing functional groups might improve the hydrophobic properties of rGO aerogel.

Fig. 3© shows the high resolution C 1s XPS spectrum of rGO/MWCNTs-NH₂ aerogel, in which the C 1s XPS spectrum could be deconvoluted into four peaks, i.e. 284.6, 286.4, 287.5 and 288.3 eV, corresponding to four functional groups on rGO/MWCNTs-NH₂ aerogel including C=C, C-O, C=O and N-C=O.³² In addition, the appearance of N-C=O around at 288.3 eV might be from the chemical reaction between the amino group of MWCNTs-NH₂ and the carboxyl group of GO by covalent bonding, which is consistent with SEM, TEM, and Raman results.

In the nitrogen adsorption-desorption isotherm curves, Fig. 3(d), the BET specific surface areas of rGO aerogel and rGO/MWCNTs-NH₂ aerogel are estimated to be 261.05 and 233.44 m² g⁻¹, respectively. The average pore diameter of rGO/MWCNTs-NH₂ aerogel centered at 2.1 nm indicates that its internal structure owns a uniform pore size and belongs to mesoporous materials, whereas the pore size distribution curve of rGO aerogel has no obvious peak, meaning that its internal structure is non-uniform pore size.

3.3 Adsorption performance of rGO/MWCNTs-NH₂ aerogel

Before doing adsorption test, the contact angle between aerogels and deionized water is measured and the results are displayed in Fig. 4. The water contact angle for rGO aerogel and rGO/MWCNTs-NH₂ aerogel is 83.54° (Fig. 4(a)) and 115.99° (Fig. 4(b)), respectively. This verifies that the addition of MWCNTs-NH₂ could improve the hydrophobicity of rGO aerogel, which might be beneficial for the selective adsorption of grease. The adsorption capacity of rGO/MWCNTs-NH₂ aerogel for different oil and organic solvents, including ethyl acetate, cyclohexane, acetone, dichloromethane, and sesame oil, has been studied and the results are 152.79, 122.46, 219.52, 242.31 and 156.35 g g⁻¹, respectively, as shown in Fig. 5. In comparison with the results in literature as listed in Table 1, it's seen that the rGO/MWCNTs-NH₂ aerogel exhibits excellent adsorption capacity for various organic solvents, so that it has a prospective application in treatment of organic pollution or oil leakage. Subsequently, an oil/water separation experiment by rGO/MWCNTs-NH₂ aerogel is performed in a beaker with application of Sudan Red B to dye cyclohexane. The obtained results are listed in Fig. 6. It turns out that the rGO/MWCNTs-NH₂ aerogel could adsorb cyclohexane within 15 s and reach an oil-water separation completely within only 25 s. It could conclude that this material with fast oil/water separation capability and superior oil adsorption capacity is expected to

Fig. 5 Adsorption capacity of rGO/MWCNTs-NH₂ aerogel for ethyl acetate, cyclohexane, acetone, dichloromethane and sesame oil.

Fig. 6 Oil/water separation experimental results of rGO/MWCNTs-NH₂ aerogel for cyclohexane.

be a potential candidate for large-scale oil removal from water.

4. Conclusions

In this work, we provide a method to synthesize the reduced graphene oxide/amino multi-walled carbon nanotube (rGO/MWCNTs-NH₂) aerogel by chemical reaction between graphene oxide and amino multi-walled carbon nanotube in combination with freeze-drying and high-temperature annealing process. The SEM, TEM and XPS affirm the presence of strong chemical bonding between the amino groups of MWCNTs and carboxyl groups on GO, enhancing the hydrophobic property and microstructures of rGO/MWCNTs-NH₂ aerogel. The MWCNTs-NH₂ is observed to be uniformly distributed within graphene layer verified by SEM and TEM. The optimal condition of rGO/MWCNTs-NH₂ aerogel is the mass ratio of MWCNTs-NH₂ to GO of 1:8 with GO concentration of 2 mg mL⁻¹. Subsequently, the oil adsorption capacity of rGO/MWCNTs-NH₂ aerogel to ethyl acetate, cyclohexane, acetone, dichloromethane, and sesame oil is 152.79, 122.46, 219.52, 242.31 and 156.35 g g⁻¹, accordingly. This rGO/MWCNTs-NH₂ aerogel could separate oil and water within 25 s, exhibiting a high oil/water separation efficiency.

Supporting Information

Additional Figs. S1-S4.

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References

1. Z. Li, B. Wang, X. Qin, Y. Wang, C. Liu, Q. Shao, N. Wang, J. Zhang, Z. Wang, C. Shen and Z. Guo, *ACS Sustainable Chem. Eng.*, 2018, **6**, 13747-13755.
2. J. Xiao, W. Lv, Y. Song and Q. Zheng, *Chem. Eng. J.*, 2018, **338**, 202-210.
3. S. F. Chin, A. N. B. Romainor and S. C. Pang, *Mater. Lett.*, 2014, **115**, 241-243.
4. D. N. Tran, S. Kabiri, T. R. Sim and D. Losic, *Environ. Sci. Water Res. Technol.*, 2015, **1**, 298-305.
5. Y. Yang, H. Yi and C. Wang, *ACS Sustainable Chem. Eng.*, 2015, **3**, 3012-3018.
6. L. Guerrini, O. Luca Pantani and A. Parenti, *J. Food Process Eng.*, 2017, **40**, e12489.
7. X. Jiang, S. Li, Y. Bai and L. Shao, *J. Mater. Chem. A*, 2019, **7**, 10898-10904.
8. Y. H. Yau, V. Rudolph, K. Ho, C. C. Lo and K. Wu, *J. Water Process Eng.*, 2017, **17**, 40-49.
9. M. Usman, P. Faure, K. Hanna, M. Abdelmoula and C. Ruby, *Fuel*, 2012, **96**, 270-276.

Table 1 Comparison of properties for rGO-MWCNTs-NH₂ with literature.

Materials	Density (g cm ⁻³)	Adsorption capacity (g g ⁻¹)
GO-Coated Polyurethane Sponge ³³	-	Gasoline 46 Chloroform 34 Acetone 32
Graphene -based foam ³⁴	-	Heptane 50 Acetone 68 Chloroform 120 Phenixin 130
Graphene aerogel ³⁵	5.0	Acetone 140 Dioxane 170 Chloroform 230 Hexane 110 Paraffin oil 130
Cellulose/graphene aerogel ²⁵	5.9	Hexane 80 Gasoline 100 Toluene 140 Dichloromethane 190
Amphipathic graphene aerogel ³⁶	4.9 - 10	Chloroform 250 Toluene 150 Soybean oil 140
This work	6.3	Ethylacetate 152.79 Cyclohexane 122.46 Acetone 219.52 Dichloromethane 242.31 Sesame oil 156.35

10. L. Shamaei, B. Khorshidi, B. Perdicakis and M. Sadrzadeh, *Sci. Total Environ.*, 2018, **645**, 560-572.
11. Y. Yavuz, H. M. A. El-Lateef, A. M. Abu-Dief and M. A. Mohamed, *J. Mol. Struct.*, 2017, **1130**, 522-542.
12. H. Zhang, S. Lyu, X. Zhou, H. Gu, C. Ma, C. Wang, T. Ding, Q. Shao, H. Liu and Z. Guo, *J. Colloid Interface Sci.*, 2019, **536**, 245-251.
13. K. G. Raj and P. A. Joy, *J. Environ. Chem. Eng.*, 2015, **3**, 2068-2075.
14. X. Zhang, X. Wang, X. Liu, C. Lv, Y. Wang, G. Zheng, H. Liu, C. Liu, Z. Guo and C. Shen, *ACS Sustainable Chem. Eng.*, 2018, **6**, 12580-12585.
15. T. Sakthivel, D. L. Reid, I. Goldstein, L. Hench and S. Seal, *Environ. Sci. Technol.*, 2013, **47**, 5843-5850.
16. Z. Huang, Y. Li, W. Chen, J. Shi, N. Zhang, X. Wang, Z. Li, L. Gao and Y. Zhang, *Mater. Chem. Phys.*, 2017, **202**, 266-276.
17. H. Bi, Z. Yin, X. Cao, X. Xie, C. Tan, X. Huang, B. Chen, F. Chen, Q. Yang and X. Bu, *Adv. Mater.*, 2013, **25**, 5916-5921.
18. J. Chen, P. Fang, Y. Du and X. Hou, *Colloid Polym. Sci.*, 2016, **294**, 119-125.
19. Y. Liao, M. Wang and D. Chen, *Ind. Eng. Chem. Res.*, 2018, **57**, 8472-8483.
20. H. I. Rasool, C. Ophus and A. Zettl, *Adv. Mater.*, 2015, **27**, 5771-5777.
21. J. X. Zhang, Y. X. Liang, X. Wang, H. J. Zhou, S. Y. Li, J. Zhang, Y. Feng, N. Lu, Q. Wang and Z. Guo, *Adv. Compos. Hybrid Mater.*, 2018, **1**, 300-309.
22. X. Gong, Y. Liu, Y. Wang, Z. Xie, Q. Dong, M. Dong, H. Liu, Q. Shao, N. Lu, V. Murugadoss, T. Ding and Z. Guo, *Polymer*, 2019, **168**, 131-137.
23. H. Gu, X. Zhou, S. Lyu, D. Pan, M. Dong, S. Wu, T. Ding, X. Wei, I. Seok, S. Wei and Z. Guo, *J. Colloid Interface Sci.*, 2020, **560**, 849-856.
24. D. D. Nguyen, N.-H. Tai, S.-B. Lee and W.-S. Kuo, *Energy Environ. Sci.*, 2012, **5**, 7908-7912.
25. H. Y. Mi, X. Jing, A. L. Politowicz, E. Chen, H. X. Huang and L. S. Turng, *Carbon*, 2018, **132**, 199-209.
26. T. Wu, M. Chen, L. Zhang, X. Xu, Y. Liu, J. Yan, W. Wang and J. Gao, *J. Mater. Chem. A*, 2013, **1**, 7612-7621.
27. H. Gu, X. Xu, M. Dong, P. Xie, Q. Shao, R. Fan, C. Liu, S. Wu, R. Wei and Z. Guo, *Carbon*, 2019, **147**, 550-558.
28. Y. Zhang, H. Sun, H. Sadam, Y. Liu and L. Shao, *Chem. Eng. J.*, 2019, **371**, 535-543.
29. H. Gu, H. Zhang, J. Lin, Q. Shao, D. P. Young, L. Sun, T. D. Shen and Z. Guo, *Polymer*, 2018, **143**, 324-330.
30. X. Jiang, S. He, S. Li, Y. Bai and L. Shao, *J. Mater. Chem. A*, 2019, **7**, 16704-16711.
31. H. Gu, X. Xu, J. Cai, S. Wei, H. Wei, H. Liu, D. P. Young, Q. Shao, S. Wu, T. Ding and Z. Guo, *Chem. Commun.*, 2019, **55**, 10068-10071.
32. X. Jiang, S. Li, S. He, Y. Bai and L. Shao, *J. Mater. Chem. A*, 2018, **6**, 15064-15073.
33. R. Tjandra, G. Lui, A. Veilleux, J. Broughton, G. Chiu and A. Yu, *Ind. Eng. Chem. Res.*, 2015, **54**, 3657-3663.
34. C. Ji, K. Zhang, L. Li, X. Chen, J. Hu, D. Yan, G. Xiao and X. He, *J. Mater. Chem. A*, 2017, **5**, 11263-11270.
35. L. Xu, G. Xiao, C. Chen, R. Li, Y. Mai, G. Sun and D. Yan, *J. Mater. Chem. A*, 2015, **3**, 7498-7504.
36. J. Cao, Z. Wang, X. Yang, J. Tu, R. Wu and W. Wang, *Appl. Surf. Sci.*, 2018, **444**, 399-406.

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