



Phase-Selective Gelators Based on *p*-Alkoxybenzoyl for Oil Spill Recovery and Dye Removal

Hongmei Qu¹ · Jie Zhang¹ · Yunxia Ma¹ · Zhongxuan Li¹ · Jianjun Qiao¹

Received: 2 January 2019 / Revised: 17 March 2019 / Accepted: 17 April 2019 / Published online: 17 May 2019
© Tianjin University and Springer-Verlag GmbH Germany, part of Springer Nature 2019

Abstract

Oil spills result in tremendous damage to the environment and ecosystem. In this study, several *p*-alkoxybenzoyl-based gelators (**1**, **2a**, **2b**, **2c**, **3**) synthesized from commercially available materials were designed for recovering oil from an oil–water mixture. Gels with remarkable gelation ability in various oils were characterized by nuclear magnetic resonance, Fourier transform infrared spectroscopy and X-ray diffraction to study the driving forces of self-assembly. Notably, these gelators could achieve the goal of recycling oil from the oil–water mixture at room temperature. In addition, gelator **2b** could be used to remove toxic dyes from aqueous solutions with high efficiency. Therefore, these compounds were considered promising materials for oil spill recovery and dye removal due to their practicality and high efficiency.

Keywords Organogelator · *p*-Alkoxybenzoyl · Phase selectivity · Oil spills recovery · Dye removal

Introduction

A rapid industry development has caused a tremendous environmental damage. Marine oil spills occur frequently with the exploitation and transportation of marine petroleum and introduce disasters to the marine environment [1, 2]. Dyes can also cause serious environmental pollution because of the difficulty in decomposition [3]. To date, various methods and materials have been studied for dealing with spilled oil, including bioremediation [4], in situ burning, dispersants [5] and adsorbents [6–8]. However, current methods and materials have some drawbacks. The complete conversion of oil into carbon dioxide and water under bacterial action takes a long time. The gas caused by burning is harmful to environment. And for the adsorption process, it is difficult to recover the oil due to the difficulty of desorption. In addition, the materials for adsorbing dyes [9–11] are also limited.

Therefore, researchers are still working on the study of new and effective materials for oil spill recovery or dye removal.

Low molecular weight gelators (LMWGs) that can solidify organic liquids into a solid or a viscous semi-solid via non-covalent interactions among molecules [12] have become popular over the past few decades. As a class of stimuli-responsive soft materials [13], organogels formed through the self-assembly of LMWGs exhibit reversible phase transitions under external stimuli such as temperature [14], pH [15], and ultrasound [16]. On this basis, a variety of gelators have been developed for various applications including drug delivery [17], probes [18], template reactions [19, 20] and sensors [21]. LMWGs that can phase-selectively gelate oils in the presence of water (phase-selective gelators, PSGs) have recently attracted attention and become ideal materials for oil spill recovery [22, 23]. Importantly, the recoverability of oil from organogels is superior to adsorbents because of their thermo-reversible behavior, which overcomes the shortcomings of current technologies. To date, various PSGs have been developed, including carbohydrates derivatives [24–26], modified amino acids [27, 28] and derivatives of naphthalene [29, 30]. Although these compounds are effective in recovering oil spills, there are some disadvantages, for instance, the ester or acetal in carbohydrates derivatives are unstable under acidic conditions and the synthesis of naphthalene derivatives is difficult. These

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s12209-019-00204-z>) contains supplementary material, which is available to authorized users.

✉ Hongmei Qu
ququhongmei@126.com

¹ Key Laboratory of Systems Bioengineering (Ministry of Education), Department of Pharmaceutical Engineering, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300350, China

disadvantages increase the inconvenience and cost. As a result, PSGs designed for gelation of oils must have the following characteristics: Gelators should provide strong enough intermolecular forces to drive self-assembly, they should gelate the oil in the presence of water, and compounds should be stable and easy to prepare.

Alkoxybenzoyl group is synthesized by introducing the bromoalkane into the hydroxybenzoyl group. Their derivatives can be obtained by adjusting the length and number of the alkyl chain as well as the types of carboxylic acid derivatives. The molecular structure of alkoxybenzoyl derivatives, including alkyl chains and aromatic rings, can provide self-assembly driving forces such as π - π stacking and van der Waals interactions. Some LMWGs based on (3,4,5)-trialkoxybenzoyl group and *p*-alkoxybenzoyl group have been synthesized and applied in the fields of liquid crystal materials and ion detection [14, 31, 32]. However, the use of organogelators based on *p*-alkoxybenzoyl has not been explored for oil spill recovery or dye removal.

In this study, *p*-alkoxybenzoyl group was designed as part of the structure. The hydrogen-bonding interactions were then enhanced by introducing a polyhydroxy group to the *p*-alkoxybenzoyl group via an amide bond to drive self-assembly. A series of gelators (**1**, **2a**, **2b**, **2c**, **3**) were synthesized and applied to oil spills recovery. The results of oil recovery experiments demonstrated that these gelators could successfully recover oil from the oil–water mixture quickly at room temperature. In addition, only a small number of PSGs that have been reported can also adsorb dye molecules from their aqueous solutions. Notably, efficient dye adsorption was realized by the organogel formed by **2b**. The adsorption efficiency was estimated to be 95%.

Experimental Section

Materials and Methods

Unless otherwise stated, all of the materials for synthesis and testing were purchased from commercial suppliers and used without further purification. All reagents were analytically pure. The toluene was distilled before using. The reactions were monitored using thin-layer chromatography (TLC) with energy chemical silica gel plates (GF-254) under irradiation by a UV-lamp (254 nm). Column chromatography was performed on silica gel (100–200 mesh). The NMR spectra were recorded on Bruker AVIII-400 and Varian INVOA-500 spectrometer with TMS as the internal standard. High-resolution mass spectra were measured using the Bruker micro-TOF-QII mass spectrometer. The UV–Vis absorption spectra were acquired using the 4802-UV spectrophotometer.

Synthesis of the Gelators

The molecular structures of these gelators (**1**, **2a**, **2b**, **2c**, **3**) are shown in Fig. 1. Their synthetic routes are displayed in Scheme S1, and the specific operation of the synthesis is summarized in Supporting Information.

Gelation Test for the Compounds

Gelation Test

The gelation abilities of the gelators (**1**, **2a**, **2b**, **2c**, **3**) were examined by the “stable-to-inversion-of-a-test-vial” method. Briefly, 20 mg of gelator was added to 0.5 mL oil in a sample vial, and then, the system was heated using a hot plate heater until the solid dissolved completely. The solution was then cooled to room temperature. The sample was regarded as gel if no flow was observed when the vial was inverted after 30 min.

Critical Gelation Concentration (CGC)

CGC is an important parameter to evaluate the gelation ability of gelators. CGCs were obtained using a method of dilution [33]. Twenty milligrams of gelator was added to the oil and a gelation test was performed. If the gel was formed, this experiment was repeated by adding some oil to the vial until the gel failed to form. CGC was calculated in % (w/v,

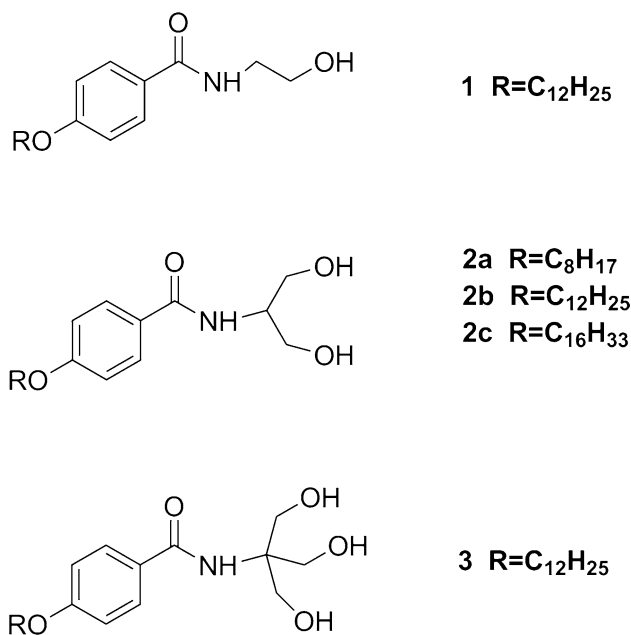


Fig. 1 Structures of gelators 1–3

g/mL) from the volume of oil that could be congealed by 20 mg of gelator.

Sol–Gel Transition Temperature (T_{gel})

T_{gel} is used to evaluate the gel stability and was determined using a MCR-301 rheometer equipped with a parallel plate (20 mm diameter). The linear viscoelasticity zone was determined from strain sweeps at constant temperature, and the frequency sweep was also performed. Temperature sweeps (heating rate 2 °C/min) were then performed with constant strain and constant frequency to obtain the storage modulus (G') and loss modulus (G''). The temperature was controlled by a circulator bath. At lower temperature, the G' dominated G'' , demonstrating a gel-like nature, while at higher temperature G'' dominated G' , demonstrating transition of the gel to sol. The T_{gel} was determined as the point where G' and G'' intersect.

Organogels Characterization

Field Emission Scanning Electron Microscopy (FE-SEM)

The microscopic morphology of the gel was observed using FE-SEM. Morphological analysis was carried out on a Hitachi S-4800 (Hitachi, Tokyo, Japan) microscope. The xerogels for SEM observation were prepared by drying toluene gels at room temperature and coating with gold using a sputtering coater.

Fourier Transform Infrared Spectroscopy (FTIR)

FTIR spectra were obtained via a Nicolet 6700 spectrometer. The solution state spectrum was obtained by placing the dimethyl sulfoxide solution of gelators on KBr pellets, and the solid-state spectrum was obtained by compressing the powder into KBr pellets.

X-ray Diffraction

The XRD spectra of compounds and xerogels were recorded using D/max-2500 (Rigaku, Japan) with $\text{CuK}\alpha$ radiation ($\lambda = 0.15418$ nm). These samples were tested from 3° to 50° at a scanning rate of 0.2 s/step. The d-spacing values were calculated by Bragg's law.

Rheology

Rheological studies were performed on a DHR-2 rheometer equipped with a parallel plate (20 mm diameter). The gels were prepared at 5% w/v and transferred to the sample plate by dropping its hot solution. The gap distance was fixed at 1 mm, and the temperature was controlled at 20 °C. The

measurements were taken in strain sweep (0.01%–100%) at a constant frequency of 1 Hz and frequency sweep (0.1–100 Hz) with 0.05% strain to obtain the storage modulus (G') and loss modulus (G'').

Determination of the Biphasic Critical Gelation Concentration (BCGC)

Twenty milligrams of gelator was added to a mixture of 0.5 mL oil and 2 mL water in a vial; then, the system was heated by a plate heater until it completely dissolved. After dissolution, the mixture was cooled to room temperature. The vial was then inverted to see whether the gel could form and support the weight of water; if it succeeded, a small amount of oil was added and this process was repeated until gel failed to form. BCGC was calculated in % (w/v) from the volume of oil that could be congealed by 20 mg of gelator.

Results and Discussion

Synthesis and Gelation Property

Both the van der Waals interactions among alkyl chains and the hydrogen bonding could promote molecular self-assembly; thus, the effect of their relative relationship on self-assembly was worth exploring. Therefore, a series of gelators (**1**, **2a**, **2b**, **2c**, **3**) containing a tail chain with different lengths and different numbers of hydroxyl groups were synthesized to investigate the effect of hydrogen bonding and van der Waals force on gelation based on self-assembly. These gelators could be synthesized in two or four steps at relatively high yields, and all materials were commercially available. In addition, these gelators were stable under slight acidic or alkaline conditions.

The results of the gelation ability test are summarized in Table S1. The gelators could gelate various oils including crude oil and edible oil with CGC from 0.36 to 4.5% (Fig. S1). Notably, the CGCs of **2b** were less than the other gelators, which may be caused by the difference in the structure. Compared with **2a**, the longer alkyl chain of **2b** provided stronger van der Waals interactions in the process of gelation, but for **2c**, there was no enhancement in the gelation ability, or even a slight decrease with increasing tail chain length on the basis of **2b**. Additionally, the more hydroxyl groups of **2b** compared with **1** strengthened the hydrogen-bonding interactions, while **3** displayed poor gelation behavior in aliphatic hydrocarbon, because its polarity increased with increasing hydroxyl groups, resulting in poor solubility. Therefore, the appropriate length of the tail chain and suitable number of hydroxyl groups were favorable for the gelator to

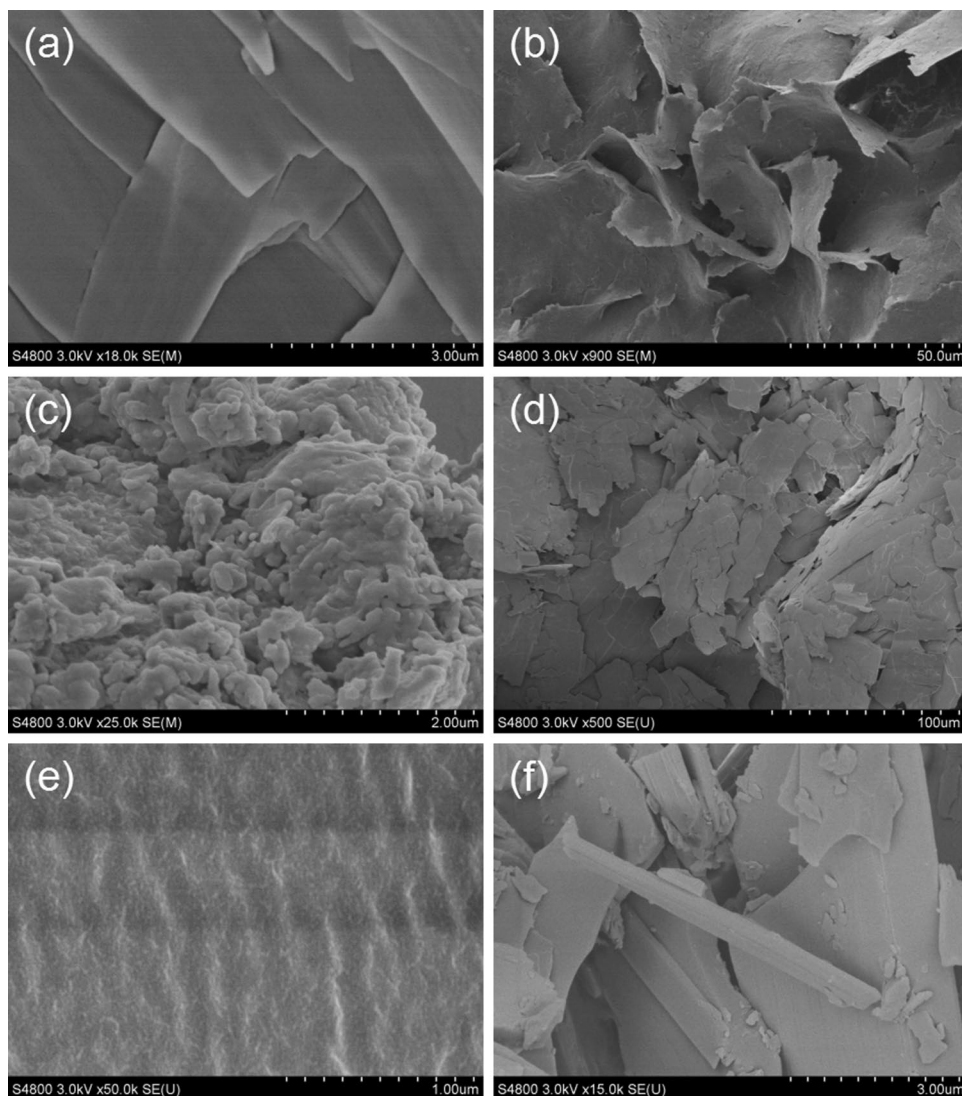
reach a dissolution–precipitation equilibrium. Therefore, a delicate balance of various non-covalent interactions was always crucial to obtain a good gelator.

In addition, the T_{gel} of different gelators in various oils, as well as different dosages of gelator in the same oil, were tested (Fig. S2). The results are summarized in Table S2 and Fig. S3, demonstrating that the interactions between the gelator molecules and the oils being solidified had a significant influence on the T_{gel} . The T_{gel} increased with the increase in the alkyl chain length and the number of hydroxyl group, suggesting higher thermal stability of the gel formed by stronger intermolecular interactions. Alternatively, the T_{gel} value of gelators increased with increasing concentration, showing enhancement in the gel stability at high concentration.

Gel Characterization

Compound **2b** was chosen as a representative of the gel structural characterization due to its low CGCs and good thermal stability. To gain a visual insight into the gel microstructures, some typical SEM images of the xerogels are exhibited in Fig. 2. It should be noted that the gel morphology was affected by the solvents. The xerogel of **2b** in toluene exhibited a ribbon-like structure, the xerogel of **2b** in ethylene glycol and gasoline adopted sheet-like structure and the xerogel in cyclohexane showed flower-like microstructures. Alternatively, the xerogel of **1** in toluene showed a flowing stream-like structure and **3** in toluene showed a sheet-like structure. The sheet structure of **3** was narrower than the others, which may be caused by its poor solubility. So, these results indicated that a small change in the structures of gelators could result in the difference of gel morphology.

Fig. 2 SEM images of xerogels. **a** **2b** in toluene at 2%, **b** **2b** in ethylene glycol at 2%, **c** **2b** in cyclohexane at 2%, **d** **2b** in gasoline at 1%, **e** **1** in toluene at 4% and **f** **3** in toluene at 2%



FTIR spectra can provide useful information to confirm the driving forces of self-assembly. The FTIR spectra of **2b** were measured in the dissolved state (dimethyl sulfoxide), as a net solid and xerogel state (Fig. 3). Generally, the non-hydrogen-bonding stretching vibrations of the N–H (O–H) and C=O appeared at 3600–3400 cm^{-1} and around 1660 cm^{-1} [34]. Therefore, the absorption bands of dimethyl sulfoxide solution at 3423 and 1666 cm^{-1} were attributed to the vibrations of νOH (νNH) and $\nu\text{C=O}$ groups, but the absorption bands appeared at 3292, 1633 cm^{-1} and 3292, 1632 cm^{-1} for the net solid and xerogel obtained from toluene gel, respectively. The red shift of these absorption bands indicated that amide groups were involved in the conformation of hydrogen bonding, so hydrogen-bonding interactions were one of the driving forces. Furthermore, the asymmetric ($\nu_{\text{as}}\text{CH}_2$) and symmetric ($\nu_{\text{s}}\text{CH}_2$) stretching vibrations of the long alkyl chains usually appeared at 2917–2920 and 2848–2851 cm^{-1} [35]. For the net solid and xerogel, the absorption bands of CH_2 in **2b** appeared at 2917, 2850 cm^{-1}

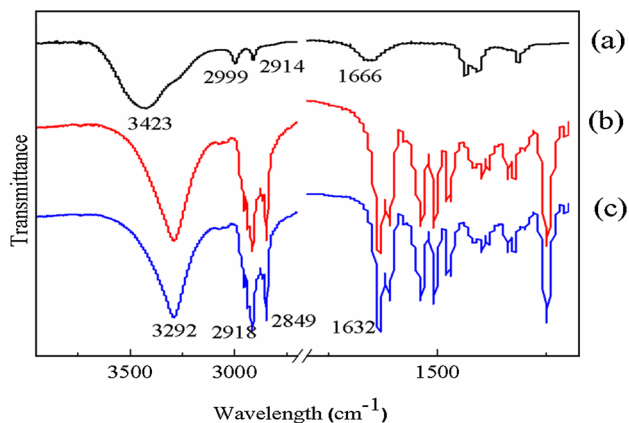


Fig. 3 FTIR spectra of **2b**. **a** DMSO solution, **b** net solid, **c** xerogel from toluene (1%)

and 2918, 2849 cm^{-1} , respectively. For comparison, the corresponding peaks of **2b** in dimethyl sulfoxide solution appeared at 2999 and 2914 cm^{-1} . These results indicated that the van der Waals interaction between the alkyl chains plays an important role in the self-assembly.

^1H NMR spectroscopy is another powerful technique to study the non-covalent interaction in the gel state. Concentration-dependent ^1H NMR experiments of **2b** were recorded in CDCl_3 at 298 K. The NH signal and OH signals were marked by symbols H_1 and H_2 , respectively. As shown in Fig. S4, the signal of the NH proton of **2b** appeared at 6.86×10^{-6} at low concentration, but it gradually shifted to 6.91×10^{-6} at a near-gel concentration. Additionally, the sharp signals of OH protons gradually broadened with the increase in concentration. The results indicated the involvement of hydroxyl and amide N–H protons in the self-assembly [27]. In addition, the aromatic proton signals corresponding to the phenyl rings showed a slight shift in high concentration compared with low concentration, which provided the support for the π – π stacking.

Molecular arrangements and intermolecular interactions are believed to be responsible for the structural characteristics of self-assembled gels. To further investigate the molecular packing and confirm the driving forces of self-assembly during gelation, the XRD experiments of the net solid and xerogel obtained from toluene were conducted. As shown in Fig. 4, the X-ray diffraction patterns of the net solid and the xerogel are indistinguishable, which indicated that gelator **2b** and **1** had similar molecular packing arrangement in both states [36]. The X-ray diffraction patterns of the xerogel **2b** showed periodical reflection peaks, and the corresponding spacings (d) were 1.97, 0.99, 0.66, 0.49 and 0.39 nm, which were exactly in the ratio of 1: 1/2: 1/3: 1/4: 1/5, thus indicating a layered organization within the aggregates of gel **2b** (Fig. 4a). The result was in good agreement with the FE-SEM data. The net solid of **2b** also showed periodical

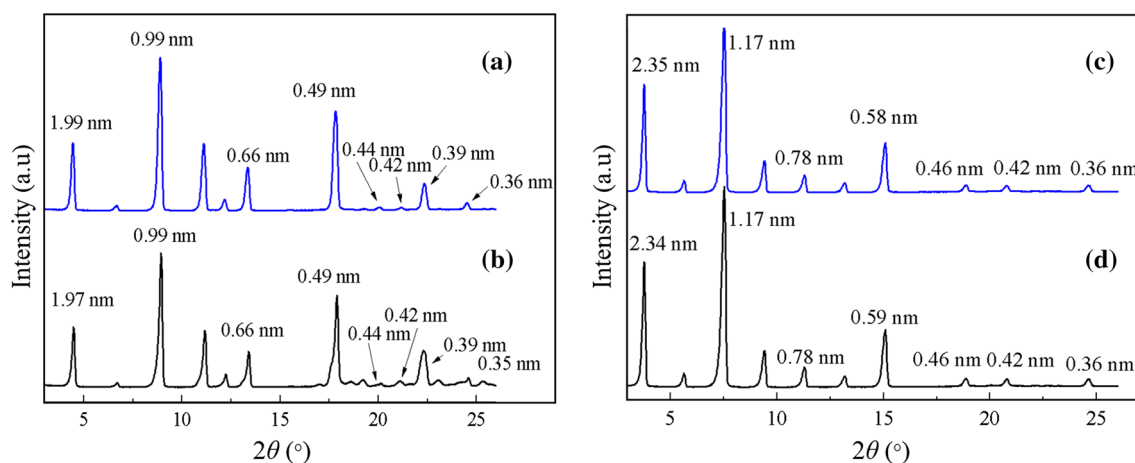
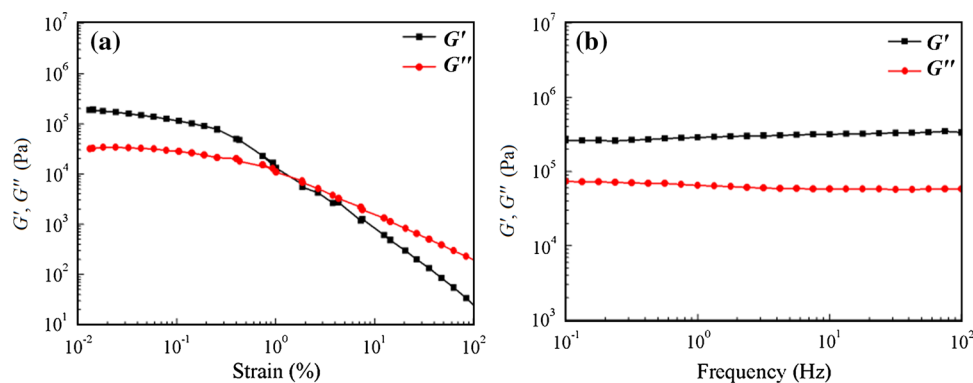


Fig. 4 X-ray diffraction patterns. **a** Net solid of **2b**, **b** xerogel of **2b** from toluene, **c** net solid of **1**, **d** xerogel of **1** from toluene

reflection peaks following the ratio of about 1: 1/2: 1/3: 1/4: 1/5, suggesting the existence of crystal-like ordered structure in the net solid [24, 37]. In addition, the diffraction pattern of the xerogel demonstrated several reflection peaks in the wide-angle region. The peak at $2\theta = 19.24^\circ$ ($d = 0.44$ nm) could correspond to hydrogen-bonding distance of gelator molecules in the gel state [28]. The peaks at $2\theta = 21.16^\circ$ ($d = 0.42$ nm) and $2\theta = 25.36^\circ$ ($d = 0.35$ nm) could correspond to packing distance of the alkyl chains and π - π stacking distance of the aromatic rings, respectively [38]. Therefore, three kinds of non-covalent interactions participated in the self-assembly of gelators. Similar results were also observed from the diffraction pattern of **2b**. The attempts to obtain single crystals of **2b** were failed, so that the actual packing of the net solid is still unknown. The XRD of gelator **1** and its xerogel also exhibited several sharp reflection peaks following the ratio of approximately 1: 1/2: 1/3: 1/4, thus indicating a layered organization within the aggregates. The other reflection peaks of xerogel **1** were obtained at $2\theta = 18.92^\circ$ ($d = 0.46$ nm), $2\theta = 21.26^\circ$ ($d = 0.42$ nm) and $2\theta = 24.70^\circ$ ($d = 0.36$ nm), respectively, which were consistent with the presence of H-bonding interactions, van der Waals and π - π stacking interactions in the xerogel state.

The mechanical strength of the organogel is an extremely important property for its applications and was studied by rheology. Strain sweep measurements of the toluene gel of **2b** are shown in Fig. 5a, indicating that the value of G' was more than one order magnitude greater than the G'' value over a wide range of strains, which was in agreement with the characteristic expected from a true gel [37]. The G' then decreased rapidly and became smaller than the G'' after the critical strain region, suggesting a transition from the gel state to the sol state. Frequency sweep measurements of the gel were taken to further investigate its mechanical property. Figure 5b shows that both G' and G'' were roughly invariant with frequency throughout the experimental region, and what's more the G' remained higher than G'' . This observation confirmed the formation of viscoelastic gel materials [39]. The value of G' was on the order of 10^5 , indicating higher stiffness and strength of the gel.

Fig. 5 Rheological studies for the toluene gel of **2b** at 5%. **a** Strain sweep and **b** frequency sweep



Oil Spills Recovery

Having detected the gelation of diesel, gasoline and crude oil, we then turned our attention to the phase-selective gelation ability of these gelators in the presence of water. In the beginning, the phase-selective gelation of crude oil, gasoline and olive oil was tested using the heating-cooling method; gelators **1**, **2b** and **2c** demonstrated impressive results. As shown in Figs. S5 and S6, the resulting gels of these gelators remained at the bottom and support the weight of water when the vial or tube was inverted, while **2a** distributed in two phases when the system was heated and it failed to gelate the oil after cooling, which might attribute to its weak hydrophobicity. Additionally, the BCGCs of gelators were detected in some of the representative oils and the values are summarized in Table S3. The gelators were slightly higher than CGCs, indicating that the presence of water had some effect on gelation. The BCGCs of **3** were greater than the others, just like its CGCs, while the BCGCs of **2b** were relatively lower than the BCGCs of **1** and **2c**. **2b** was selected as the representative of these PSGs for oil spill recovery due to its low BCGCs.

For the phase-selective gelation experiments to remediate the spilled oil, the practical dispersion method was crucial to the application of PSG. It was obviously impractical to promote the dispersion of gelators in oil floating on the sea by the heating method mentioned above. Therefore, the wetting powder strategy was adopted for promoting dispersion (Fig. 6). A typical procedure obtaining wetting powder was as follows: PSG was heated to dissolve in a chosen solvent and cooled to produce a solvent-containing gelator in the form of sticky powder; the solvent chosen should have a relatively high solubility for PSG. In this work, crude oil (0.8 g) was added to the water (100 mL) in a beaker to simulate actual situation of oil spills. The wetting powder was then produced by mixing 0.1 g of gelator **2b** and 0.2 g of ethyl acetate. The powder was sprinkled over the oil layer, and the crude oil was solidified into semi-solid state after 3 h (Fig. 6b). Compared with the heating method, the wetting method was feasible but it took a long time. Given the

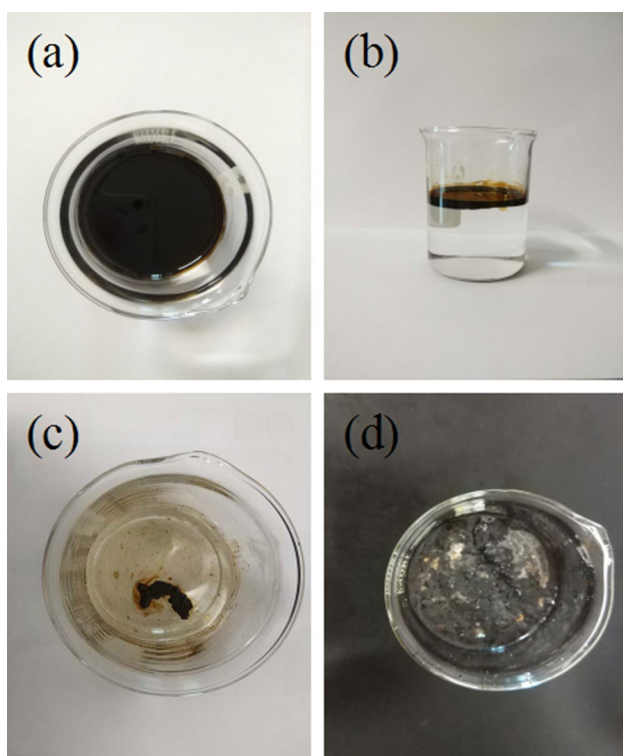


Fig. 6 Phase-selective gelation of crude oil from a oil/water mixture. **a** Top view of oil/water mixture, **b** front view of oil/water mixture, **c** treating with wetting powder, **d** treating with carrier solvent

spilled oil is harmful to the marine ecological environment, it should be separated as quickly as possible. Therefore, the carrier solvent was used to promote the dispersion of the gelator, and toluene was chosen as a carrier solvent due to its high boiling point and immiscibility with water. In addition, the use of toluene could effectively reduce secondary marine pollution because it could also be congealed. A typical procedure was performed by adding a hot solution of **2b** (0.1 g) in toluene (0.5 mL) to the beaker containing crude oil (0.8 g) and water (100 mL). Remarkably, crude oil was congealed into a semi-solid state within 30 s, and it could be removed by a small scoop. Furthermore, the hot solution of **2b** (0.4 g)

and toluene (1.5 mL) was added to the beaker containing crude oil (5 g) and water (100 mL). Although the thickness of the oil layer increased, the crude oil was congealed into a semi-solid within 2 min. The results demonstrated that the use of a carrier solvent could effectively promote the dispersion and reduce the time to gelation. In addition, the gelled gasoline could be recovered by distillation. In actual situations of the treatment of crude oil spills, the recovered gelled crude oil could be fractionally distilled to produce consumable petroleum products.

Dye Removal

Organic synthetic dyes are another class of pollutants that pollute the environment. An important issue in the current research is the adsorption of organic dyes from wastewater. Several PSGs have been applied in the field of removing dyes [40, 41]. Therefore, gelator **2b** was used tentatively to adsorb dye from its aqueous solution. For the dye adsorption experiment (Fig. 7), 20 mg of **2b** was added to toluene (1 mL) to obtain toluene gel, and 1 mL of crystal violet solution (0.1 mmol/L) was then added to the vial containing the above gel. In order to investigate the adsorption process, the mixture was sampled at intervals, and the concentration of dye in water was detected by an ultraviolet–visible spectrophotometer. The absorption spectroscopy showed that 94% of the dye could be adsorbed by toluene gel after 24 h (Fig. 8a). Additionally, although toluene could extract the crystal violet from water to the organic layer, the efficiency was much lower according to the contrast test (Fig. 8b). The absorption spectrum showed that only 25% of the crystal violet was removed by the extraction of toluene. Therefore, **2b** promotes the adsorption for the dyes. The effect of gelator concentration on dye removal was studied (Fig. S7). The results showed that up to 95% of the dye could be removed with increasing concentration. A similar removal effect was also observed from methylene blue (Fig. S8). Furthermore, **2b** could be recovered by eluting the dye-adsorbed gel with methanol and reused; the recovery efficiency was nearly up to 90%. It was speculated that the existence of both the hydrophilic hydroxyl head and hydrophobic

Fig. 7 Removal of the crystal violet using toluene gel and recovery of **2b**

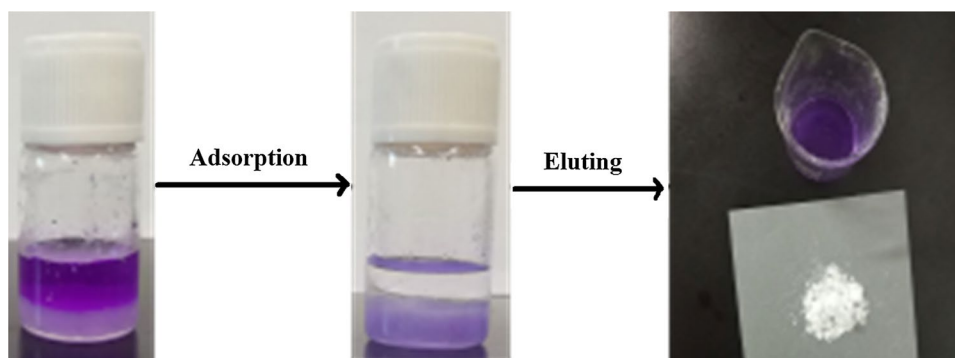
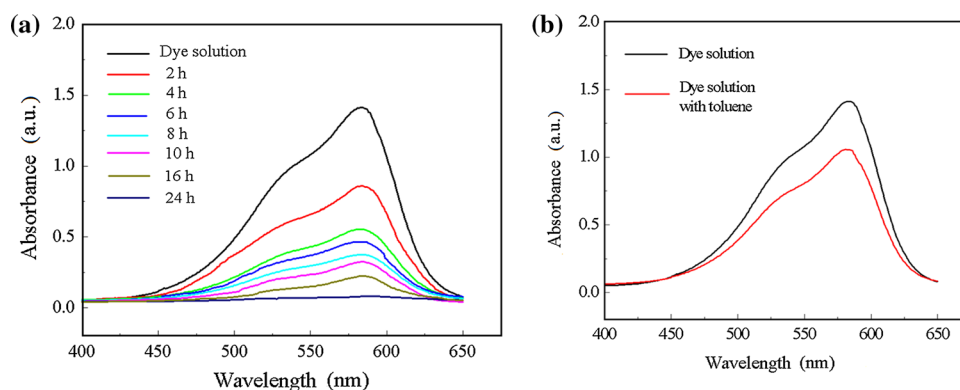


Fig. 8 Absorption spectra of crystal violet in an aqueous solution: **a** removal of the crystal violet using organogel and **b** removal of the crystal violet using toluene



alkyl chain in the **2b** structure was highly advantageous for the adsorption of organic dyes. The hydrophilic hydroxyl head was a benefit for materials to contact with water, and the hydrophobic regions could accommodate and retain organic substances [42, 43]. Furthermore, the large surface area of the self-assembled structure was a critical factor for adsorption of the dye molecule with a relatively high efficiency. These findings indicated that **2b** could be regarded as an excellent water purification agent due to its good performance in the adsorption of these dyes.

Conclusions

In summary, several compounds based on the *p*-alkoxybenzoyl group were designed and synthesized for the first time. The structures were stable and the synthesis was simple. The gelation ability could be changed by adjusting the length of the tail chain and the number of hydroxyl groups; some of these compounds demonstrated excellent gelation ability in a variety of oils including crude oil and edible oil with low CGCs. The results of the gel structural characterization confirmed that the interactions of hydrogen bonding, van der Waals force and π - π stacking were the driving forces of the self-assembly. In addition, compounds **1**, **2b** and **2c** had a good performance on phase-selective gelation of oil from the oil–water mixture at room temperature, and **2b** could gelate oil spills rapidly and recover oil spills efficiently in the experiments that simulating oil spills recovery. The gel formed by **2b** could also adsorb crystal violet and methylene blue from the solution with a relatively high adsorption efficiency, and **2b** could be regenerated by simple eluting. These results indicate that the PSGs are promising materials for oil spill recovery and dye removal.

References

- Al-Majed AA, Adebayo AR, Hossain ME (2012) A sustainable approach to controlling oil spills. *J Environ Manag* 113(1):213–227
- Beyer J, Trannum HC, Bakke T et al (2016) Environmental effects of the Deepwater Horizon oil spill: a review. *Mar Pollut Bull* 110(1):28–51
- Le TXH, Esmilaire R, Drobek M et al (2017) Nitrogen-doped graphitized carbon electrodes for biorefractory pollutant removal. *J Phys Chem C* 121(28):15188–15197
- Swannell RP, Lee K, McDonagh M (1996) Field evaluations of marine oil spill bioremediation. *Microbiol Rev* 60(2):342–365
- Nyankson E, Demir M, Gonen M et al (2016) Interfacially active hydroxylated soybean lecithin dispersant for crude oil spill remediation. *ACS Sustain Chem Eng* 4(4):2056–2067
- Ding X, Wang R, Zhang X et al (2014) A new magnetic expanded graphite for removal of oil leakage. *Mar Pollut Bull* 81(1):185–190
- Wang F, Lei S, Li C et al (2014) Superhydrophobic Cu mesh combined with a superoleophilic polyurethane sponge for oil spill adsorption and collection. *Ind Eng Chem Res* 53(17):7141–7148
- Ge J, Zhao HY, Zhu HW et al (2016) Advanced sorbents for oil-spill cleanup: recent advances and future perspectives. *Adv Mater* 28(47):10459–10490
- Low LW, Teng TT, Alkarkhi AFM et al (2011) Optimization of the adsorption conditions for the decolorization and COD reduction of methylene blue aqueous solution using low-cost adsorbent. *Water Air Soil Pollut* 214(1–4):185–195
- He Y, Li H, Zhou L et al (2019) Removal of methyl orange from aqueous solutions by a novel hyper-cross-linked aromatic triazine porous polymer. *Acta Phys-Chim Sin* 35(3):299–306
- Ahmad A, Mohd-Setapar SH, Chuong CS et al (2015) Recent advances in new generation dye removal technologies: novel search for approaches to reprocess wastewater. *RSC Adv* 5(39):30801–30818
- Wang Y, Wang Y, Yan X et al (2016) Toluene diisocyanate based phase-selective supramolecular oil gelator for effective removal of oil spills from polluted water. *Chemosphere* 153:485–493
- Liu M, Ouyang G, Niu D et al (2018) Supramolecular gelators: towards the design of molecular gels. *Org Chem Front* 5(19):2885–2900
- Cao H, Wang F, Zeng H et al (2014) Morphology tunable organogels based on benzoylhydrazine derivatives. *J Mol Liq* 196:94–97
- Kar T, Debnath S, Das D et al (2009) Organogelation and hydrogelation of low-molecular-weight amphiphilic dipeptides: pH responsiveness in phase-selective gelation and dye removal. *Langmuir* 25(15):8639–8648
- Xu J, Wang Y, Shan H et al (2016) Ultrasound-induced organogel formation followed by thin film fabrication via simple doctor blading technique for field-effect transistor applications. *ACS Appl Mater Interfaces* 8(29):18991–18997

17. Hu B, Sun W, Li H et al (2018) Systematic modifications of amino acid-based organogelators for the investigation of structure-property correlations in drug delivery system. *Int J Pharm* 547(1–2):637–647
18. Li X, Zhang Y, Chen A et al (2017) A ferrocene-based organogel with multi-stimuli properties and applications in naked-eye recognition of F⁻ and Al³⁺. *RSC Adv* 7(59):37105–37111
19. Rajkamal Chatterjee D, Paul A et al (2014) Enantiomeric organogelators from *D*-/*L*- arabinose for phase selective gelation of crude oil and their gel as a photochemical micro reactor. *Chem Commun* 50(81):12131–12134
20. Xue P, Lu R, Li D et al (2004) Novel CuS nanofibers using organogel as a template: controlled by binding sites. *Langmuir* 20(25):11234–11239
21. Kishimura A, Yamashita T, Aida T (2005) Phosphorescent organogels via “metallophilic” interactions for reversible RGB-color switching. *J Am Chem Soc* 127(1):179–183
22. Bhattacharya S, Krishnan-Ghosh Y (2001) First report of phase selective gelation of oil from oil/water mixtures. Possible implications toward containing oil spills. *Chem Commun* 2:185–186
23. Okesola BO, Smith DK (2016) Applying low-molecular weight supramolecular gelators in an environmental setting—self-assembled gels as smart materials for pollutant removal. *Chem Soc Rev* 45(15):4226–4251
24. Jadhav SR, Vemula PK, Kumar R et al (2010) Sugar-derived phase-selective molecular gelators as model solidifiers for oil spills. *Angew Chem Int Ed* 49(42):7695–7698
25. Kesava Raju CS, Pramanik B, Ravishankar R et al (2017) Xylitol based phase selective organogelators for potential oil spillage recovery. *RSC Adv* 7(59):37175–37180
26. Mukherjee S, Mukhopadhyay B (2012) Phase selective carbohydrate gelator. *RSC Adv* 2(6):2270–2273
27. Mukherjee S, Shang C, Chen X et al (2014) N-Acetylglucosamine-based efficient, phase-selective organogelators for oil spill remediation. *Chem Commun* 50(90):13940–13943
28. Basak S, Nanda J, Banerjee A (2012) A new aromatic amino acid based organogel for oil spill recovery. *J Mater Chem* 22(23):11658–11664
29. Yao H, Yang LP, He Z et al (2017) A phase-selective, bis-urea organogelator with a curved bis-naphthalene core. *Chin Chem Lett* 28(4):782–786
30. Datta S, Samanta S, Chaudhuri D (2018) Near instantaneous gelation of crude oil using naphthalene diimide based powder gelator. *J Mater Chem A* 6(7):2922–2926
31. Zhai Y, Chai W, Cao W et al (2015) Organogelators based on *p*-alkoxybenzamide and their self- assembling properties. *Front Chem Sci and Eng* 9(4):488–493
32. Wan JH, Mao LY, Li YB et al (2010) Self-assembly of novel fluorescent silole derivatives into different supramolecular aggregates: fibre, liquid crystal and monolayer. *Soft Matter* 6(14):3195–3201
33. Vibhute AM, Muvvala V, Sureshan KM (2016) A sugar-based gelator for marine oil-spill recovery. *Angew Chem Int Ed* 128(27):7913–7916
34. Hanabusa K, Koto C, Kimura M et al (1997) Remarkable viscoelasticity of organic solvents containing trialkyl-1, 3, 5-benzenetricarboxamides and their intermolecular hydrogen bonding. *Chem Lett* 26(5):429–430
35. Masuda M, Vill V, Shimizu T (2000) Conformational and thermal phase behavior of oligomethylene chains constrained by carbohydrate hydrogen-bond networks. *J Am Chem Soc* 122(49):12327–12333
36. Xu H, Song J, Tian T et al (2012) Estimation of organogel formation and influence of solvent viscosity and molecular size on gel properties and aggregate structures. *Soft Matter* 8(12):3478–3486
37. Ran X, Li Y, Gao Q et al (2017) A smart phase-selective gelator for recycling aromatic solvents, the removal of toxic dyes, and molecular delivery. *Asian J Org Chem* 6(1):95–101
38. Yu H, Liu B, Wang Y et al (2011) Gallic ester-based phase-selective gelators. *Soft Matter* 7(11):5113–5115
39. Zhuan C, Li Y, Yuan X et al (2019) A sorbitol-based phase-selective organogelator for crude oil spills treatment. *J Appl Polym Sci* 136(6):47052
40. Zhang X, Song J, Ji W et al (2015) Phase-selective gelators based on closed-chain glucose derivatives: their applications in the removal of dissolved aniline/nitrobenzene, and toxic dyes from contaminated water. *J Mater Chem A* 3(37):18953–18962
41. Narayana C, Upadhyay RK, Chaturvedi R et al (2017) A versatile carbohydrate based gelator for oil water separation, nanoparticle synthesis and dye removal. *New J Chem* 41(6):2261–2267
42. Bekiari V, Lianos P (2006) Ureasil gels as a highly efficient adsorbent for water purification. *Chem Mater* 18(17):4142–4146
43. Cho EJ, Jeong IY, Lee SJ et al (2008) Terpyridine-based smart organic–inorganic hybrid gel as potential dye-adsorbing agent for water purification. *Tetrahedron Lett* 49(6):1076–1079