



## Two-component supramolecular organogels formed by maleic N-monoalkylamides and aliphatic amines

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### ABSTRACT

The complexes of maleic N-monoalkylamides and aliphatic amines lead to the formation of sheet-like and fiber-shaped aggregates in diverse organic solvents. Their morphologies and microstructures were characterized by Fourier transform infrared spectroscopy (FT-IR), scanning electron microscope (SEM), as well as small angle X-ray diffraction (SA-XRD). The results reveal that the alkyl chain lengths of 8–12 carbons for maleic N-monoalkylamides, and 12–18 carbons for aliphatic amines, are suitable for obtaining highly efficient two-component gelators.

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### 1. Introduction

During the past two decades, more and more attention has been paid to the synthesis of supramolecular organogels due to their special properties and potential applications in fields such as drug delivery [1,2], inorganic nanomaterials [3,4], biofilm simulation [5,6], liquid crystallines [7,8], photochemistry [9,10], and electrochemistry [11,12]. It is well known that organogelators, possessing functional groups like hydroxyl, amide, linear alkyl chains, aromatic group, are able to self-assemble into aggregates with diverse morphologies such as fibers, sheets, ribbons through non-covalent interaction including hydrogen bonding, van der Waals force,  $\pi$ – $\pi$  stacking, coordination interaction, and charge transfer [13–21]. Such aggregates form a three-dimensional network through further cross-linking and immobilizing the solvents and finally result in the formation of organogels. The physical characteristics of organogels can range from those of surfactants in solution to those of polymer solutions. Gels are at the interface between “complex fluids” and phase-separated states of matter. To date, diverse categories of organogels have been prepared and studied, including steroid derivatives [22,23], cholesterol [22,24], amino acids [25,26], carbohydrates [27,28], urea [29,30], organometallics [31], and so on. Recently, some chemists begin to transfer their attention from single component organogels to two-component organogels. Compared with the single component organogels, two-component organogels can build new organogel systems, obtain more new organogels with

interesting morphologies and microstructures, and offer the potential of developing more soft materials with highly tunable microscopic and macroscopic properties. George and Weiss reported the complexes of aliphatic amines with carbon dioxide could gelate organic solvents [32,33]. Suzuki and coworkers reported a two-component gelator consisting of L-lysine derivatives and aliphatic acids [34]. The above citations provide ample evidence of the promising features of the two-component approach to achieve gel. However, most of these studies do not provide much structural insight as to how the two components interact to form the gel. It has been known that succinic N-monoalkylamides can form organogels [35], while the structurally related compounds, maleic N-monoalkylamides, do not gelatinize any organic solvents. To further enrich the two-component organogel system with desired functions and observe a well-defined structure–property relationship, herein, we report a new class of two-component organogelators comprised of maleic N-monoalkylamides and aliphatic amines. Their gelation ability, morphologies and microstructures were explored and characterized.

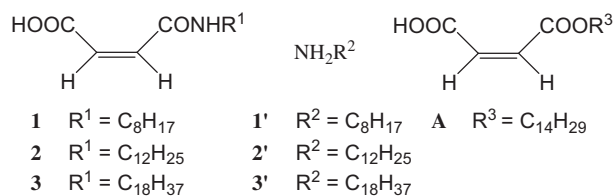
### 2. Materials and methods

#### 2.1. Synthesis

All starting materials were available from commercial sources and used without further purification. Compounds **1–3** (Scheme 1) were synthesized according to the procedure described as follows: A mixture of maleic anhydride and equivalent mol aliphatic amine was dissolved in 50 mL toluene in a round-bottomed

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**Scheme 1.** Chemical structures of maleic N-monoalkylamides, aliphatic amines and tetradecyl hydrogen maleate.

flask equipped with a condenser. After stirring for about 30 min at 80 °C, the toluene was removed by vacuumizing method, and white solid was obtained. Highly pure products of **1–3** were gotten after the white solids recrystallized using mixed solvent of ethanol/water ( $v/v = 1:1$ ) for four times, respectively. The method for preparing **A** (Scheme 1) is similar to that of **1–3**, except using 1-tetradecanol instead of aliphatic amine. Satisfactory  $^1\text{H}$  NMR, element analysis data were obtained for the corresponding maleic N-monoalkylamides (see Supporting information).

## 2.2. Gelation test

The gelation ability of nine combinations of **1–3** with **1'–3'** was investigated by a typical test tube experiment. A weighted sample of maleic N-monoalkylamides and equivalent mol aliphatic amine was mixed in an organic solvent (1 mL) in a sealed test tube and the mixture was heated until the solid dissolved. The resulting solution slowly cooled to room temperature and then the gelation generated. Once formed, the organogel was stationary and the tube can be inverted without deformation of the gel.

The reversible gel–sol transition temperature ( $T_{\text{gel}}$ ) was measured using a classical drop-ball method as described by Takahashi [36]. A locally made steal ball with a diameter of 2 mm was gently laid on the gel surface, and the gel was heated gradually in an oil bath. The temperature at which the ball started to fall down was considered as the  $T_{\text{gel}}$ .

## 2.3. SEM measurements

Scanning electron microscope (SEM) measurements were carried out on Philips XL 30W/TMP. A drop of organogel was cast onto glass slides, which was slowly dried in air and then the remained xerogel was subjected to SEM observation.

## 2.4. FTIR measurements

Fourier transform infrared (FT-IR) spectra were recorded on Nicolet AVATAR 360. All data were collected for 128 interferograms with a resolution of  $4\text{ cm}^{-1}$ .  $\text{CaF}_2$  substrates and KBr pellet were used for transmission spectra of xerogel and crystal, respectively.

## 2.5. SA-XRD measurements

Small-angle X-ray diffraction (SA-XRD) patterns of dried organogel were measured on a Bruker D8 Focus diffractometer using the Cu K $\alpha$  radiation. X-ray was generated with a Cu anode and the Cu K $\alpha$  beam ( $\lambda = 1.5418\text{ \AA}$ ) was taken out via a graphite monochromator. The long spacing ( $D$ ) were obtained from the typical peak.

## 3. Results and discussion

### 3.1. Gelation test

The gelation test results are shown in Table 1. It was found that only **1 + 2'**, **1 + 3'**, **2 + 2'**, **2 + 3'**, **3 + 2'**, **3 + 3'** showed gelation property, while **1 + 1'**, **2 + 1'**, **3 + 1'** gave no gelation, which may be related with the alkyl chain length of both maleic N-monoalkylamides and aliphatic amines. Aliphatic amine compound **1'**, compared with **2'** and **3'**, has the shortest alkyl chain, which led to the intermolecular interactions among **1'**, maleic N-monoalkylamides and solvent molecule too weak to generate gelation. Therefore, the combinations of **1' + 1**, **1' + 2**, **1' + 3** exhibited no gelation behavior. In addition, although the rest six combinations of **1 + 2'**, **1 + 3'**, **2 + 2'**, **2 + 3'**, **3 + 2'**, **3 + 3'** exhibited gelation, their gelation performances were markedly different. **1 + 3'** and **2 + 2'** are excellent gelators, as they are able to gelate many organic liquids at very low concentrations. For instance, the minimum gel concentration (MGC  $\text{g/dm}^3$ ) of **1 + 3'** for toluene, o-xylene, and cyclohexane are  $1.3\text{ g/dm}^3$ ,  $1.3\text{ g/dm}^3$ , and  $1.4\text{ g/dm}^3$ , respectively. **1 + 2'** and **3 + 2'** also display good gelation capacity. However, the MGC of them are slightly higher than that of **1 + 3'** and **2 + 2'**. For **2 + 3'** and **3 + 3'**, although they could gelate many organic liquids, they are still considered as low efficiency gelators due to high MGC in most solvents. Comparing the molecular structures of **1–3** with **1'–3'**, it can be inferred that as a highly efficient two-component gelator, the suitable length of tail chain is 8 and 12 carbons for mono-chain maleic acid derivatives and 12 and 18 carbons for aliphatic amines. The results may be ascribed to the fact that the proper length of the carbon chain can strengthen the intermolecular interactions among maleic N-monoalkylamides, aliphatic amines, and solvent molecules.

**Table 1**

The MGC<sup>a</sup> ( $\text{g/dm}^3$ ) of each combination in different organic liquids.

Solvent	<b>1 + 2'</b>	<b>1 + 3'</b>	<b>2 + 2'</b>	<b>2 + 3'</b>	<b>3 + 2'</b>	<b>3 + 3'</b>
Carbon tetrachloride	TG 5.7	CG 3.1	TG 4.9	TG 10.8	OG 7.3	OG 15.1
1,1,2,2-Tetrachloroethane	S	P	S	P	TG 29.4	CG 10.7
Benzene	CG 4.5	CG 1.5	CG 2.1	TG 26.5	TG 13.0	TG 11.8
Toluene	CG 4.2	CG 1.3	CG 2.1	TG 21.3	TG 6.7	TG 10.4
o-Xylene	CG 8.1	CG 1.3	CG 2.2	TG 27.0	TG 7.4	TG 19.2
Mesitylene	TG 8.5	CG 2.1	TG 18.0	TG 30.8	TG 7.3	TG 12.1
Tetralin	TG 8.7	CG 1.6	TG 15.1	TG 22.2	TG 9.7	TG 7.5
Isopropyl ether	P	OG 15.3	OG 11.2	P	P	OG 18.8
Cyclohexane	OG 3.9	CG 1.4	OG 11.2	OG 16.9	OG 7.3	OG 20.0

OG: opaque gel; TG: translucent gel; CG: clear transparent gel; S: solution; P: precipitate.

<sup>a</sup> Total concentration of two components (molar ratio = 1:1) for each combination.

**Table 2**

The gelation capacities of combinations with different  $\text{CH}_2$  length.

Total $\text{CH}_2$ length	<b>1 + 2'</b>	<b>1 + 3'</b>	<b>2 + 2'</b>	<b>2 + 3'</b>	<b>3 + 2'</b>	<b>3 + 3'</b>
20	Good	–	–	–	–	–
24	–	–	Excellent	–	–	–
26	–	Excellent	–	–	–	–
30	–	–	–	Good	Low	–
36	–	–	–	–	–	Low

In addition, we found the gelation capacities of these organogels were related with the total length of the  $\text{CH}_2$  of the two components (Table 2). For  $1 + 3'$  and  $2 + 2'$ , their total length of the  $\text{CH}_2$  are 26 and 24 respectively, they exhibit excellent efficiency. However, for  $3 + 2'$  and  $3 + 3'$ , they have longer alkyl carbon chain, and show low gelation capacities. This may be reasoned that with the increasing of the total length of the  $\text{CH}_2$ , the intermolecular interactions among maleic N-monoalkylamides, aliphatic amines, and solvent molecules decreased, which was not beneficial for their gelation.

To estimate the relationship between thermal stability of gel and the concentration of combination, the  $T_{\text{gel}}$  for combinations of  $1 + 3'$ ,  $2 + 2'$ ,  $3 + 2'$  at different concentrations in toluene were measured. The plot of  $T_{\text{gel}}$  versus concentration is shown in Fig. 1. The results reveal that with the increasing of the concentration, the  $T_{\text{gel}}$  increased gradually and eventually reached a plateau of 74 °C, 66 °C, and 61 °C for  $1 + 3'$ ,  $2 + 2'$ , and for  $3 + 2'$ , respectively. The corresponding concentrations are 26.0 g/dm<sup>3</sup>, 26.0 g/dm<sup>3</sup>, 28.0 g/dm<sup>3</sup>. The results demonstrated here imply that at the beginning, the stability of each gel was gradually enhanced following the increasing of the concentration of organogelators. However, when the concentration increased to a certain value, the  $T_{\text{gel}}$  reached a constant. It should be noted that in the same concentration, the  $T_{\text{gel}}$  of  $2 + 2'$  is higher than that of  $3 + 2'$  but lower than that of  $1 + 3'$ , which is consistent with the fact that the value of MGC of  $2 + 2'$  gel in toluene is less than that of  $3 + 2'$  but more than that of  $1 + 3'$ . These result suggested that the gelation ability of  $3 + 2'$ ,  $2 + 2'$ ,  $1 + 3'$  increased in turn. This may be related to hydrophile–lypophile balance of organogelator molecules [17,35,37]. Owing to endow with suitable alkyl chain length,  $1 + 3'$  has good hydrophile–lypophile balance in toluene, leading to the best organogelator combination for toluene.

### 3.2. SEM characterization

In order to gain an insight into the aggregate morphology, xerogels of different combinations were studied by scanning electron microscopy (SEM). As shown in Fig. 2, the xerogels of  $1 + 2'$  from toluene,  $1 + 3'$  from carbon tetrachloride and  $2 + 3'$  from carbon

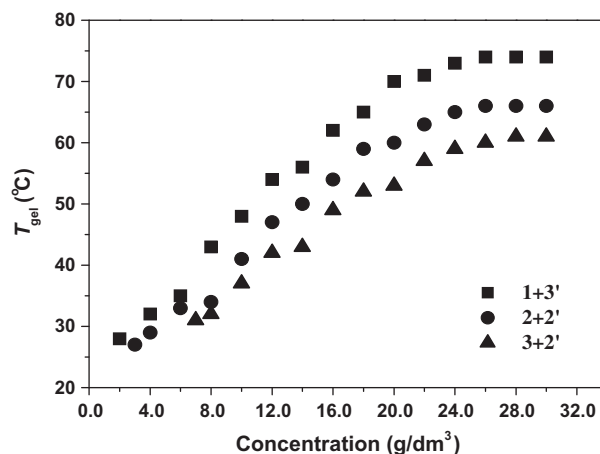


Fig. 1. Plot of  $T_{\text{gel}}$  versus organogelator concentration in toluene (g/dm<sup>3</sup>).

tetrachloride have a similar sheet-like structure with the width of 5–15  $\mu\text{m}$  (Fig. 2a, b and d). The xerogel of  $2 + 2'$  from cyclohexane displays a intertwined fiber structure with a diameter of about 0.5  $\mu\text{m}$  (Fig. 2c). Organogelator molecules created complex three-dimensional networks by entangling numerous tiny fibers and entrapped abundant organic liquid in the interspace of the networks by surface tension and capillary forces, leading to the formation of organogels [38].

### 3.3. FT-IR spectroscopy

The FT-IR spectra of **1** and  $1 + 3'$  gel from  $\text{CCl}_4$  are shown in Fig. 3. The spectrum of **1** shows a intense peak at 3240  $\text{cm}^{-1}$  can be assigned to the stretching of the amide N–H bonds. The peaks at 1708  $\text{cm}^{-1}$  and 1642  $\text{cm}^{-1}$  can be attributed to the characteristic adsorption of the carboxylic group, and the peak at 1591  $\text{cm}^{-1}$  is corresponding to the adsorption of C=C bonds stretching. After combined with **3'**, the peak at 1708  $\text{cm}^{-1}$  disappeared and two

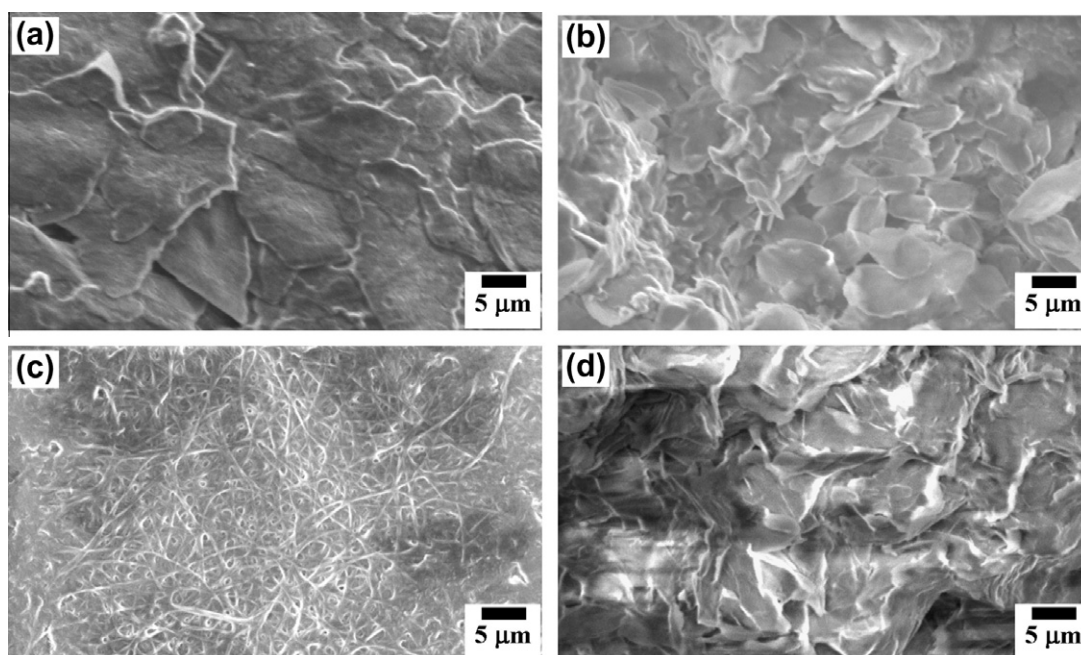


Fig. 2. SEM images of xerogels: (a)  $1 + 2'$  from toluene, 8.0 g/dm<sup>3</sup>; (b)  $1 + 3'$  from  $\text{CCl}_4$ , 8.0 g/dm<sup>3</sup>; (c)  $2 + 2'$  from cyclohexane, 15.0 g/dm<sup>3</sup> and (d)  $2 + 3'$  from  $\text{CCl}_4$ , 15.0 g/dm<sup>3</sup>.

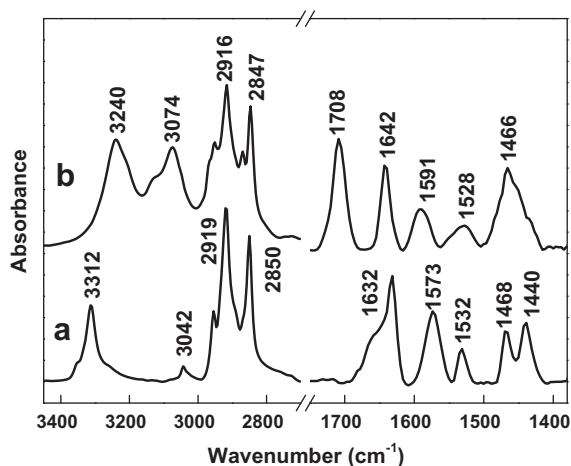


Fig. 3. FT-IR spectra of **1** + **3'** gel from CCl<sub>4</sub> at 8 mg/mL (a) and **1** (b), respectively.

Table 3

The long spacing (*D*) obtained by XRD method and the molecular length evaluated by optimized CPK model.

	Long spacing ( <i>D</i> <sub>n</sub> ) (by XRD, nm)	Evaluated molecular length (by CPK model, nm)
<b>1</b> + <b>2'</b>	3.42	3.44
<b>1</b> + <b>3'</b>	4.14	4.19
<b>2</b> + <b>2'</b>	3.93	3.94
<b>2</b> + <b>3'</b>	4.67	4.69
<b>3</b> + <b>2'</b>	4.68	4.69

new peaks at 1573 cm<sup>-1</sup> and 1440 cm<sup>-1</sup> appeared, indicating that **1** reacted with **3'** to form **1** + **3'** gel [34]. In addition, the peak at 3440 (ν<sub>N-H</sub> of amide), 1680 cm<sup>-1</sup> (ν<sub>C=O</sub> of amide) for a free amide group red-shifted to 3312, 1632 cm<sup>-1</sup> for amide group in gel state, respectively, implying the hydrogen bonds had formed between neighboring amides [34,39]. Furthermore, two strong peaks at 2919 cm<sup>-1</sup> and 2850 cm<sup>-1</sup> of **1** + **3'** gel, arising from antisymmetric and symmetric CH<sub>2</sub> stretching vibration, suggested that hydrocarbon chains of organogelator molecules adopted trans-zigzag conformation in organogels [40,41].

To further determine whether acid–base interaction and hydrogen bonding play important roles in the formation of organogels, we designed a new compound **A** (Scheme 1), which has ester group instead of amide group. The gelation test were carried out by mixing **A** and aliphatic amine using the similar procedure described above. The result illustrated that no gelation was observed for **A** + **1'**, **A** + **2'** and **A** + **3'**, this may be reasoned that the hydrogen bonds cannot be formed between combined molecules.

### 3.4. X-ray diffraction

To testify the order structures of the organogelator molecules, we measured the X-ray diffraction patterns (XRD) of xerogels of **1** + **2'**, **1** + **3'**, **2** + **2'**, **2** + **3'** and **3** + **2'** from CCl<sub>4</sub> gel. As shown in Fig. S1, each pattern assumes more than three periodical diffraction peaks, indicating the organogelator molecule indeed assembled into an ordered lamellar structure.

The long spacing (*D*) obtained by XRD method and the molecular length evaluated by optimized CPK model are listed in Table 3. It can be known that the value of *D* is almost equal to the summation of molecular lengths of corresponding combination. For instance, the molecular lengths for **1** and **3'** are 1.77 nm and 2.42 nm, respectively. The summation is 4.19 nm and is greatly close to the value of *D* for **1** + **3'** (4.14 nm) obtained from the XRD result. Therefore, combined with the FT-IR results, it can be

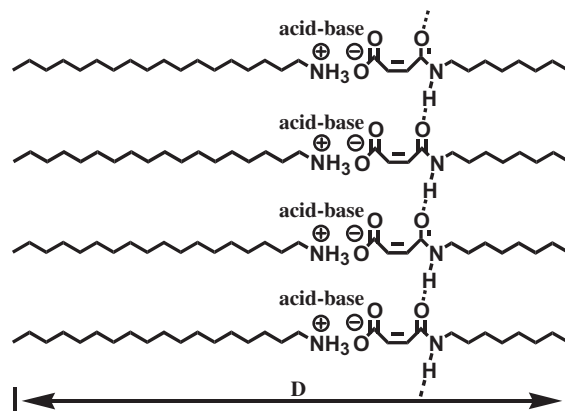


Fig. 4. Local microstructure of aggregates in organogel formed by **1** + **3'** in solvents.

deduced that the gel aggregates consist of a lamellar structure, which is stabilized by an acid–base interaction between carboxyl of **1** and amino of **3'**. The adjacent line-shaped molecules are further connected by intermolecular hydrogen bonds to form the superstructure as illustrated in Fig. 4. This orientation is also preferable for alkyl chains to arrange in parallel by hydrophobic attractive interaction.

## 4. Summary

In summary, we have identified an extremely simple but novel two-component organogelation system using the principle of organic salt formation involving commercially available maleic N-monoalkylamides and aliphatic amines of suitable length. These organic salt organogelators have good organogelation abilities for some organic solvents. The FTIR, XRD studies reveal that the organic salt organogelators self-assemble into ordered lamellar aggregates in organic solvents, in which hydrogen bonding, intermolecular acid–base interaction and van der Waals interaction play important roles. The alkyl chain length of 8–12 C atoms for maleic N-monoalkylamides and 12–18 C atoms for aliphatic amines are suitable for obtaining highly efficient two-component gelators. Maleic N-monoalkylamides do not gelatize any organic solvents, while the structurally related compounds, succinic N-monoalkylamides, have good organogelation abilities [35]. These results imply that we can construct various two-component organogelation systems with different function by organogelators structurally related compounds and appropriate counterparts. Such organogelators may be applied to biomimetics and soft materials in the future.

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## Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jcis.2011.06.016.

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