Temperature and Solvent-Dependent Morphological Sol Gel Transformation: An in Situ microscopic observation

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A thermoreversible self-assemble process from gel (fiber) to sol (vesicle) state in the system alkylamine—ethylene glycol is for the first time monitored by in situ polarized optical microscopy, XRD, 1H NMR, SEM, SAXRD, FTIR and drop shape analysis. It is found that the solvent molecules are intercalated with alkylamine molecules to form the organogel and vesicle structures. A model based on structural transformation with respect to these alkylamine gelator—solvent assemblies is therefore proposed.

Introduction

Low-molecular-weight organogels (LMOGs) are currently the subject of increasing attention not only because of their numerous potential applications, but they also provide detailed structural information leading to unraveling of the principle of molecular self-assembly. The main driving forces for assembling soft molecules are believed to be primarily noncovalent such as hydrogen bonding, π−π stacking, van der Waals interactions, solvophobic interaction, etc. They can generate a wide variety of soft aggregates by incorporation of solvent molecules in their supramolecular structures giving unique morphologies.1−7 In the gelation process, LMOGs are thought to first undergo self-assemble into one-dimensional fiber-like structure, and then entangle to form a three-dimensional network to jellify the organic components. In recent years intense research has been devoted to elucidating the structural effect(s) of gelators, but the role(s) of the solvent molecule contributing in molecular assembling is far from clear. Typically, limited studies of the interaction between solvents and gelators,6,9 which could be important to explain the gelation mechanism and also account for the formation of various morphologies.10−12 Furthermore, studies on microstructures have been initiated for a number of organogel systems,10−12 but the sol–gel process of converting organogel to large aggregates, such as giant vesicles is rarely carried out particularly using optical microscopy.

We have been interested in elucidating the interaction(s) between alkylamine and polar solvent because this system has been widely employed in the preparation of mesoporous silica13 and nanoparticles synthesis as solvent/capping agent.14,15 Shinkai et al.15 argued from theoretical point of view that such system will give strong substrate-solvent interaction(s) leading to the formation of various soft assemblies. Also, the soft assemblies can form giant vesicles induced by the presence of metal ions or pH,16 which may allow a direct observation of them in micrometric scale.

Here, we employ a novel organogel system based on self-assembly of primary alkylamine in ethylene glycol (EG) which is widely used in so-called “polyol” process for the synthesis of nanoparticles. Using polarized optical microscopy the thermo-reversible transformation process from gel to vesicle state is, for the first time, recorded. Combining the in situ microscopy with 1H NMR, SEM, FTIR, SAXRD, and drop shape analysis a mechanistic model for the phase transformation with respect to gelator—solvent assemblies is hereby presented.

Experimental Section

Materials. Hexadecylamine (purity ≥99%, Aldrich), octylamine, tetradecylamine, and dodecylamine were provided by Jiangsu Feixiang Chemical Co., Ltd. Ethylene glycol was purchased from Sinopharm Chemical Reagent Co. Ltd. (SCRC). All the reagents were of analytical grade and used without further purification. Deuterated solvent was obtained from Cambridge Isotopes at the highest deuteration levels available (ethylene-d6 glycol, 98%) and was stored in a sealed container.

In a typical experiment, 0.2 g of alkylamine was added to 4.8 g of ethylene glycol with stirring under heating conditions to form a homogeneous system (sol). Then, it was cooled to a fixed temperature to form organogels.

Characterization. Optical microscopic images were taken using an Olympus BX51 optical microscope equipped with crossed polarization lenses and TP 94 heat stage. A drop of heated solution was placed on a glass plate to form a gel. The photographic images of the samples were acquired digitally. The sol–gel transformation process was recorded in repeated cycles of heating and cooling.

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Temperature-dependent $^1$H NMR spectra was recorded on a Varian Mercury VX-600 M operated at 600 MHz equipped with a variable-temperature accessory. In all NMR experiments, we used a single pulse with no echo, and the proton pulse lengths were set at 14.2 $\mu$s with the power level of 59. In a typical experiment a NMR tube containing 0.48 g of ethylene-$d_6$ glycol and 0.02 g of hexadecylamine was heated to form a homogeneous solution phase and then cooled to its gel state. Variable-temperature NMR measurements of the gel sample were recorded at designated temperatures with a minimum sample equilibration time of 30 min after next targeted temperature was reached.

Scanning electron microscopic images were taken from KYKY-EM3200 (KYKY Technology Development Ltd.) operated at 30 kV. A piece of the gel was first placed on a glass and pretreated at low pressure ($> 10^{-5}$ Torr) for 3 days to remove the excess solvent and volatiles. FTIR spectra of the dried sample containing giant vesicles with the same sample pretreatments above were acquired using a Bruker Vertex 70 FTIR. It is noted that the samples were coated by gold sputtering prior to SEM examination.

Small-angle X-ray diffraction patterns were measured using Cu Ka radiation ($\lambda = 1.5406 \AA$) on D8 advance X-ray diffraction (Bruker). Diffraction data for all samples were recorded between 0.8° and 10° (2θ).

The plot of surface tension versus log c of dodecylamine in ethylene glycol was obtained using a DSA100 drop shape analysis system at 30 °C (Krüss, Germany). The area of the surfactant headgroup at the interface of the hydrophobic core–hydrophilic medium was determined from the slope before the critical micelle concentration according to Gibbs equation.

**Results and Discussion**

**Optical Microscopic Observation.** When 0.2 g of hexadecylamine was added into 4.8 g of EG with constant stirring, an organogel of wormlike structure instantly appeared at ambient temperature as seen under a heating stage optical microscope. As seen from Figure 1, the wormlike structure is comprised of interconnected fibers, and the length reaches several tenths of micrometers with about 1 $\mu$m diameter. It is interesting to find that some are aggregated as thread balls and some are well dispersed in the sample. As the temperature increased slowly from 30 to 40 °C (Figure 2b–d), vesicles of irregular shape were first formed at the expense of the fibers. The dispersed fibers also seem to condense to form smaller vesicles above the gel to sol transition temperature ($T_{gs}$) of around 40 °C. Further increasing
temperature resulted in the fusion of small vesicles into larger ones akin to the typical Ostwald ripening phenomenon (Figure 2e–g). All the giant vesicles finally appeared to be globular-like shape at 45 °C (Figure 2i), which remained unchanged even at the temperature of 70 °C (not shown here). It is evident that such a vesicle was composed of optical isotropic structure (bi- or multilayers) because of the absence of birefringence in their polarized images. Further FTIR spectra as shown in Figure 3 of the dried sample containing giant vesicles without solvent revealed that the vesicles contained characteristic peaks of 3353 cm⁻¹ (–OH stretch), 2941 cm⁻¹ (CH₂ asymmetric stretch); 2876 cm⁻¹ (CH₂ symmetric stretch); 1464–1452 cm⁻¹ (CH₂ bending modes) corresponding to pure ethylene glycol solvent (spectrum 1), this FTIR spectrum clearly demonstrates the entrapment of the solvent molecules in the giant vesicle structure.

An in situ visualization by cooling the mixture from 46 to 36 °C was also performed in order to examine the reversibility of this phase transformation process under the optical heat stage microscope (Figure 4). When a cooling rate was fixed in the range of 0.1 to 5 °C/min, a similar observation in the phase transformation was recorded with a more rapid process at higher ramp rate. As evidenced from the images in Figure 4, a fiber phase was first formed from the peripheral of the vesicle, which led to a subtle change in shape from globular to polygon upon cooling. The polarized optical microscopic images reveal that these polyhedrons gave optical anisotropy. This could be due to a surface phase segregation process when hexadecylamine molecules are crystallized from the vesicle particularly at or below 37.9 °C (corresponding well to the melting point of hexadecylamine). By further decreasing the applied temperature the fiber-phase started to grow from peripheral surface to the interior of the polygonal vesicle. By careful controlling the fixed cooling rate of 5 °C/min, the whole process was re-examined from 46 °C to room temperature (Figure 5). It is clear that each polygon vesicle indeed gave rise to a large number of rigid fibers on its surface like the flower-blossom and the created stress led to the cracking of the globular vesicle assembly. After a prolonged time (> 5 h), the flower-like structure could be reverted to the original morphology of wormlike fiber structure in organogel state depending on applied temperature. This gel–sol–gel process can be repeated many times provided that there is no severe solvent evaporation.

Figure 3. Optical microscopic image (left) and FT-IR (right) analysis of dried sample containing giant vesicles (spectrum 2): the characteristic peaks of 3353 cm⁻¹ (–OH stretch); 2941 cm⁻¹ (CH₂ asymmetric stretch); 2876 cm⁻¹ (CH₂ symmetric stretch); 1464–1452 cm⁻¹ (CH₂ bending modes) corresponding to pure ethylene glycol solvent (spectrum 1), this FTIR spectrum clearly demonstrates the entrapment of the solvent molecules in the giant vesicle structure.

Figure 4. Morphological transformation of vesicle to polygon of fibers (hexadecylamine in ethylene glycol) from 46 to 36 °C at various cooling rates. Parts A–D correspond to the images obtained at different cooling rates: (A) 0.1 °C/min, (B) 0.5 °C/min, (C) 2 °C/min, and (D) 5 °C/min. In the series shown in part A, the first three figures contain optical polarized images captured in different zones; in the series in parts B–D, the last images are the optical polarized patterns of the corresponding former ones. The scale bar is 100 μm.
As a result of in situ microscopic investigation of heating and cooling processes, molecular assembles in forms of vesicle at elevated temperature and organogel with worm-like fiber structure at low temperature are clearly structurally related and can be interconverted to each other. To further examine the formation of giant vesicles in sol state and their reversible transformation to the gel structure, a similar microscopic observation was also conducted for other primary alkylamine of chain length from 12 to 18. It was found that all the tested alkylamines can form giant vesicles in ethylene glycol under heating conditions. Figure 6 presents the photographs of the gel—sol—gel reversible transformation in the system of tetradecylamine-EG. It is clear that the giant vesicles formed at elevated temperatures and their transformation to the fiber structure is totally thermoreversible. In the cooling process, an anisotropic phase was also observed, similar to the case of the hexadecylamine—EG system. The transition temperature for the formation of anisotropic structure resembled closely to the melting point of the tetradecylamine but independent of its concentration change. By visual examination the gelation process was gradually developed from the bottom to the top of the vial: the white opaque phase emerged first in the bottom and then expanded to the whole vial. These results demonstrate that these anisotropic structures during the gelation process are clearly initiated by the crystallization of alkylamines, the phase segregation of which may be governed by crystal mismatching as proposed by Liu et al.17

1H NMR and Small-Angle XRD Characterization. To reveal the internal structures of these soft assembles variable temperature 1H NMR and small-angle XRD were conducted. The 1H NMR spectral changes of hexadecylamine in deuterated ethylene-d6 glycol are shown in Figure 7A. For comparison, the 1H NMR examination was also performed for the pure ethylene-d6 glycol (Figure 7A) and hexadecylamine in CDCl3 (Figure 7D). It is remarkable to note from the Figure 7A that despite the weak intensity the NMR peaks of hexadecylamine they do not disappear and remain very sharp in the gel state as comparable to soluble counterpart in CDCl3 (Figure 7D). In most of the cases reported in the literature, the gelator NMR signals are completely disappeared in the gel state while they are only present in the liquid state.18 This can be explained by the fact that the gel fiber can be considered as a crystal of the gelator in which the molecular motion is very limited and the solvent molecules are excluded from the fiber. Therefore, disappearance of the 1H NMR peaks has been described to be one criterion for “dry gels” state. Our result clearly suggests that the amine gelator molecules still keep a good thermal motion in the fibers (only slightly broaden peak compared to soluble counterpart); this feature is consistent, with a “wet gel” for which the solvent molecules (EG) are incorporated into the gel fibers. From the enlarged spectra of the gel at various

Figure 5. Morphological transformation of vesicle to wormlike fiber structure (hexadecylamine in ethylene glycol) upon cooling from 46 °C to room temperature kept at 5 °C/min; in the part D series, the magnification of the latest image is twice that of the third one, and its scale bar is 50 μm; otherwise, all scale bars are 100 μm.

Figure 6. Optical microscopic images of structure transformation process in the system of tetradecylamine and ethylene glycol under heating and cooling conditions. (A) images acquired on the heating process; (B and C) images obtained on the cooling process. The latter three images in series B and the first three ones in series C were acquired with polarization. The scale bar in part A was 100 μm and was 50 μm for parts B and C.

temperature and the pure ethylene-d₆ glycol (Figure 7B), it was found that a significant and progressive upfield change occurs in the chemical shift of free O–H in EG (a in Figure 7B) as compared to pure ethylene-d₆ glycol because of their extensive exchange in solution with the N–H over the whole temperature. The protons of O–H in EG are shifted upfield to 5.32 ppm in the gel state as compared to 5.37 ppm in the pure solvent and are further up-shifted as the temperature increase (in the sol state), implying a stronger intermolecular hydrogen interactions in the network structures. To examine clearly the amine molecule interactions with the solvent molecules, the peaks of hexadecylamine are magnified 15 times of their original intensity as shown in Figure 7C. Comparing to soluble hexadecylamine in CDCl₃ (Figure 7D) the chemical shifts of the C–H neighboring to N–H move to the downfield direction: C–H in α position has changed from 2.67 ppm in the soluble amine to 2.76 ppm in the gel state. These results clearly demonstrate the existence of hydrogen bonding of the terminal amine NH₂ group with O–D group of ethylene-d₆ glycol in the gel state. It can also be clearly seen that the proton signals of the N–H and the neighboring α C–H in the amine head region show an obvious shift with the temperature increase. It is also noted that peak h exists in both the spectra of pure EG-d₆ (Figure 7A) and amine–ethylene glycol gel state, which remains unchanged during the whole heating process. This peak is derived from the impurity of EG-d₆ which can be used a marker in the NMR characterization.

Characterization of the organogels was carried out using small-angle XRD. Initially, for comparative purpose, solid hexadecylamine was studied and the patterns are shown in Figure 8. It is noted that, the solid hexadecylamine exhibits a large number of Bragg peaks in our diffractogram which appears to be very different from the pattern of pure hexadecylamine reported by Belman et al. Considering the fact that this solid hexadecylamine had been exposed in air for prolonged time prior XRD examination (in contrast to the short acquisition time in the NMR experiments), the difference in pattern could probably be attributed to the formation of alkylammonium–alkylcarbamate molecular pairs when the amine molecules bind carbon dioxide from air. Belman et al. have also reported a similar artifact when their primary alkylamine crystal was exposed to ambient air for a period of time. Thus, the tested hexadecylamine could be composed of a mixture of hexadecylamine–hexadecylcarbamate (HAHC). According to our diffraction pattern (Figure 8) two groups of peaks: one appears at 1.88°, 5.64°, and 9.36° with the d spacing corresponding to 1:1/3:1/5; the other at 2.2°, 4.4°, 6.6°, and 8.8°, with the d spacing of 1:2/1:3/1:4 are indeed observed. Thus, the derived interlayer separations are 4.70 nm (higher quantity according to their peak intensities) and 4.01 nm,

![Figure 7](https://example.com/figure7.png)

**Figure 7.** (A) Temperature-dependent ¹HNMR spectra of the gel at various temperature and pure ethylene glycol-d₆ with TMS as standard at 20 °C. (B) Enlarged spectra of part A in the range 3.0–6.0 ppm; (C) enlarged spectra of the gel at different temperature in the range 0–3.0 ppm. (D) ¹HNMR of hexadecylamine in CDCl₃. It is noted that peaks labeled h are the impurities from the ethylene-d₆ glycol, which does not exist in hexadecylamine (h position is marked).

respectively. The 4.01 nm matches well with the Belman quoted lattice value derived from pure hexadecylamine. For comparison, solid hexadecylamine with short air exposure was also characterized, as shown in Figure 8B, which shows a similar XRD pattern of hexadecylamine from Belman et al. However, this solid amine sample still contained a small quantity of HAHC despite the short air exposure suggesting its high propensity for CO₂ capture in solid form. Nevertheless, when amine was in solution form in our NMR experiments there was no evidence for the formation of carbamate. Thus, the pure solid hexadecylamine contained an ordered layer packing with a separation of 4.01 nm. Comparing the molecular size of hexadecylamine of ~2.11 nm, tail to tail packing of hexadecylamine with tilted carbon chains to account for an interlayer spacing of 4.01 nm seems reasonable. This bended packing is expected to be thermodynamically more favorable for more efficient molecular assembly, which has been commonly observed in stacking molecules with long hydrophobic tails. As our hexadecylamine molecules were in the gel state in EG, an apparent structural difference was clearly detected: only two new peaks appeared at 1.98° and 5.72° (Figure 8 C, D), and the peaks detected in solid state were all absent. These SAXRD patterns seem corresponding well to the worm-like structure reported in hexagonal mesoporous silica (HMS) prepared using hexadecylamine. In the gel state, the derived interlayer separation is 4.46 nm which is 0.45 nm longer than the bended packing of 4.01 nm in solid. It is very interesting to reveal that the difference in the interlayer distances matches well with the molecular length of ethylene glycol. Combining with the results obtained from ¹H NMR, the polar solvent molecule, ethylene glycol is likely incorporated in between the two polar amine head groups. This can maximize solute-solvent interaction by trapping the solvent molecules into the amine aggregates as a gel state. It should be noted that a strong thermodynamic driving force is expected to play a key role in assembly of elongated molecules of polar head and nonpolar tail with polar solvent molecules. That is to reduce the undesirable interface between polar segments with nonpolar one in the final assembly. Thus, the EG is envisaged to link between the polar amine groups. In contrast, attempts to produce this soft assemble by predissolving hexadecylamine in DMF, 1,3-propanediol, and 1,4-butanediol upon heating conditions were failed. As shown in

Figure 8. Small-angle X-ray diffraction patterns of solid hexadecylamine powder and aggregates in various solvents: (A) solid hexadecylamine (exposed to ambient air for a period of time), (B) fresh solid hexadecylamine (C) hexadecylamine organogel in ethylene glycol, (D) 10 times magnification of part C, and (E) aggregates formed in DMF, 1,3-propanediol, and 1,4-butanediol.

Figure 9. (A) SAXRD profiles of the organogels of the alkylamines: (1) octyldecylamine, (2) hexadecylamine, (3) tetradecylamine, (4) dodecylamine. (B) Bilayer thickness ($D_n$) values as functions of the tail length ($n$).

Figure 10. $\gamma$–log $c$ plot of dodecylamine in ethylene glycol at 30 °C.
Figure 8E, the molecular aggregates formed in these solvents display identical diffraction patterns of two low angle peaks. The maximum interlayer separation (0.15 nm) between the d spacing (4.01 nm) in solid comparing to that (4.16 nm) in aggregates state is virtually the same, indicating that the amine molecules are in relatively disordered packing but with no entrapment of solvent molecule. Thus, it is clear that the relatively nonpolar solvent molecules with longer hydrocarbon linkers will destabilize the packing if they are entrapped between the polar amine headgroup segment hence no soft amine gelator—solvent assemble can be formed.

To gain further understanding of the amine gelator—solvent soft assembles and their intertransformation, further SAXRD characterization of four organogels systems by using alkylamines of different carbon chain lengths of 12 to 18 in ethylene glycol was conducted. As seen from Figure 9A, they exhibit similar diffraction pattern with corresponding diffraction peaks shifted to the lower angles with the increase in carbon chain lengths from C12 to C18. Figure 9B shows undisputedly that the measured spacing (D_n) from the SAXRD shows a linear dependent on the carbon chain length (n). This clearly indicates a similar packing mechanism of the organogels at room temperature irrespective of the alkyl chain length. The linear relationship between D_n and n of the samples can be written as:

\[ D_n = 0.23n + 0.73 \]

(1)

It is accepted that the slope and intercept of the eq 1 can reflect the contributions of alkyl chain and headgroup toward the interseparation spacings in the bilayer assembles (D_n), respectively.\(^{22}\) According to the multilayer model of packing aliphatic alky surfactant molecules with polar head groups, the chain orientation angle with respect to the layered structure can be calculated from the slope (25° is commonly reported in packing alkyl chains in literature, which is close to our 0.23 obtained in our measurements).\(^{23}\) Apart from the contribution from the small size amine group, the large intercept may suggest an involvement of ethylene glycol in the headgroup. Thus, basing on the packing likeness with our system the solvent molecules are thought to be intercalated into the layered structure of alkylamine molecules through hydrogen bonding to form the lamella gel in ethylene glycol.

**Drop Shape Analysis.** The concept of molecular packing parameter P was originally developed by Israelachvili et al.,\(^{24}\) which can be used to predict the morphology of molecular self-assembled structure in surfactant systems in polar solvent. Thus, the packing parameter was calculated in the amine-ethylene glycol system.

The molecular packing parameter can be defined as:

\[ P = \frac{V_c}{l_c \lambda_0} \]

(2)

\[ V_c = V_{(\text{CH}_3)} + (n-1)V_{(\text{CH}_2)} \]

(3)

\[ V_{(\text{CH}_3)} = 0.0546 + 1.24 \times 10^{-4}(T - 298) \text{ nm}^3 \]

(4)

\[ V_{(\text{CH}_2)} = 0.0269 + 1.46 \times 10^{-5}(T - 298) \text{ nm}^3 \]

(5)

\[ l_c = 0.1265(n-1) + 0.2765 \text{ nm} \]

(6)


where \( V_c \) and \( l_c \) are the volume and length of hydrophobic tail and \( A_0 \) is the cross section area of the polar surfactant headgroup at the interface of the hydrophobic core-hydrophilic medium. It is well-known that molecular aggregates can undergo self-assemble from layered to vesicle structure when \( 1/2 < P \leq 1 \) in order to minimize the overall surface energy of the system. As a result the packing parameter of dodecylamine in ethylene glycol at 30 °C was determined using the plot of surface tension versus log dodecylamine concentration (c) obtained from the drop shape analysis, as shown in Figure 10. From the slope before critical micelle concentration, the interfacial \( A_0 \) is determined to be 0.794 nm² molecule⁻¹. It is noted that this \( A_0 \) is substantially higher than the cross section area of alkylamine from the literature (0.186 nm²). On the other hand, the cross section area of ethylene glycol should be incorporated, which is believed to be around 0.60 nm² per OH groups akin to 1,2-octanediol but ignoring the effect of carbon chain. A conclusion is drawn that the solvent molecule must have involved in the molecular self-assembly forming a structure similar to gemini surfactants where two hydroxyll groups of the ethylene glycol linked with the two dodecylamine molecules head to head to account for the drop shape data. This interpretation is consistent with the data from the SAXRD. Therefore, the packing parameter \( V_c \) should be calculated using twice the volume of hydrocarbon chain. According to eqs 2 to 6, \( P \) is determined to be 0.53. With longer chain length in alkylamine the packing parameter \( P \) is expected to increase, which implies that a giant vesicle is a favorable assembly structure.

Following this argument, it is noted that the lamella fiber structure of organogel at low temperature may not truly represent the most thermodynamically stable assembly as the edge segments are rather nonpolar in the presence of polar solvent molecules surrounding them (see Scheme 1). It is logical to assume that at low temperature these molecules in the form of lamella fibers do not possess sufficient kinetic energy to undergo phase transformation to the more stable vesicle form. Upon heating, a phase transformation from lamella fiber to vesicle (spherical) is favorably taken place in order to eliminate the nonpolar edges with apparent low activation energy (a narrow temperature range for the gel-sol conversion). Thus, the vesicle with all polar surface (low surface to volume ratio) minimizing contact between the nonpolar core with the external polar solvent molecules is perhaps a more stable assembly at elevated temperature. This may also account for the observed Ostwald ripening phenomenon of fusing smaller vesicles to larger ones in order to reduce the overall surface energy of the system. In molecular modeling point of view, the curling of the supramolecules could be achieved through bending of ethylene glycol molecule but the structural flexibility of the hydrocarbon tails and their packing could also contribute toward the formation of vesicle as bilayer or multilayered solute assembly. Upon the cooling the whole process is reversible. It is interesting to note that the introduced stress and rigidity of the molecules on the surface of the vesicle when cooled at low temperatures will revert part of vesicle structure to the lamella fibers and the structural mismatch will finally break up the vesicle. It is thus emphasized that the hydrophobic and hydrophilic interactions of gelator and solvent play a significant part in the final assemble morphologies and their thermoreversible interconversions.

**Conclusion**

In conclusion, we report a novel temperature and solvent-dependent morphological transformation of gel to sol vesicle based on hexadecylamine in ethylene glycol, the system has been widely used in nanoparticles synthesis. By combining *in situ* polarized optical microscopy with \(^1\)H NMR, FTIR, SEM, SAXRD, and drop shape analysis, the structures and morphologies unpinning the transformation process are for the first time revealed. In this work, it is also found that giant vesicles can be formed by alkylamine with chain length from 12 to 18 in ethylene glycol at elevated temperatures and that they can reversibly transform into organogels under cooling cycles. These findings may provide a new insight into the molecular self-assemble of amine gelator-polar solvent molecules. Furthermore, there has been recent interest to search a simple chemical system to mimic prebiotic assembly processes, in which amphiphiles such as amines play an important role. This present study may contribute a degree of understanding toward these interesting and fundamental biological self-assemble processes.

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