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Widely adaptable oil-in-water gel emulsions stabilized by an amphiphilic hydrogelator derived from dehydroabietic acid

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Abstract: A novel surfactant, 6-((1*R*,4a*S*)-7-isopropyl-1,4a-dimethyl-1,2,3,4,4a,9,10,10a-octahydro-phenanthrene-1-carboxamido)-*N*,*N*-dimethylhexan-1-amine oxide (abbreviated as R-6-AO) derived from dehydroabietic acid has been synthesized which behaves as a highly efficient low molecular weight hydrogelator with an extremely low critical gelation concentration (CGC) of 0.18 wt.% (4 mM). As a typical surfactant, R-6-AO not only stabilizes oil-in-water (O/W) emulsions at concentrations above its critical micelle concentration (cmc) of 0.6 mM, but also forms gel emulsions at concentrations beyond the CGC with the oil volume fraction freely adjustable between 2% and 95%. Cryo-TEM images reveal that R-6-AO molecules self-assemble into left-handed helical fibers with cross-sectional diameters of about 10 nm in pure water which can be turned to very stable hydrogels at concentrations above the CGC. Compared with traditional high internal phase emulsions, the gel emulsions stabilized by R-6-AO can be prepared with different oils such as n-dodecane, n-decane, n-octane, soybean oil, olive oil and tricaprylin, due to the tricyclic diterpene hydrophobic structure in their molecules which enable them to adopt a unique arrangement in the fibers. The intriguing properties of gel emulsions stabilized by R-6-AO are believed to be significant and helpful for the preparation of new soft materials and other applications.

Gel emulsions with properties of both emulsions and gels have received much attention in recent years. [1] Similar to traditional emulsions, gel emulsions are two phase systems with one phase dispersed as droplets in the other. On the other hand, they also show the behavior of a physical gel. The gel emulsions thus display unique rheological behavior and meso-scale structures so that they have found wide application in foods, cosmetics, drug delivery, tissue engineering and in the preparation of advanced porous materials. [2-8]

Traditionally, gel emulsions are referred to as high internal phase emulsions (HIPEs), in which the volume fraction of the dispersed phase can be higher than 74%, [9-12] a geometrical limit corresponding to the maximum packing of uniform spheres in a container [13] and beyond this limit droplets are normally

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deformed yielding a polyhedral morphology. HIPEs usually have strong elasticity arising from tight packing of the dispersed droplets, and have advantages of high stability, high ability to carry either oil or water and good resistance to external forces. However, the disadvantages of HIPEs are also obvious. For example, due to the geometrical requirement, the ratio of the two immiscible liquids in HIPEs can only be adjusted within a narrow range resulting in limited formulations, and the rather high amount of stabilizer [14-16] required in traditional HIPEs is unfavorable for some industrial processes.

It has been found that low molecular weight gelator (LMWG) molecules are able to form fibrillar networks through self-assembly in solutions and then immobilize organic solvents or water. [17] Therefore, gelling the continuous phase of a traditional emulsion using suitable LMWG molecules to form gel emulsions is an alternative method to overcome the defects of traditional HIPEs. [18-24] Using this method the gelation behavior of the gel emulsions does not rely on the stacking of the dispersed droplets, and the dispersed phase/continuous phase volume ratio can be varied continuously. However, successful implementation of such a strategy depends strongly on the molecular structure of the LMWG molecules involved, whose self-assembly is sensitive to their concentration, system composition, temperature and the immiscible liquids involved. The gel emulsions generated using different LMWG molecules therefore may exhibit quite different properties. For example, the water-in-oil gel emulsions prepared by Fang and coworkers [18, 19, 22] using a dicholesteryl derivative as gelator have limited stability, whereas the thermo-responsive structured oil-in-water gel emulsions prepared by Wan *et al.* [23, 24] using glycyrrhizic acid mono ammonium salt as gelator have a gelation temperature as high as 80 °C. It is therefore significant to develop highly efficient gelators, which can stabilize gel emulsions with freely adjustable droplet/continuous phase volume ratio by self-assembly at low concentration. It is thus expected to prepare such gelators by innovating the molecular structure of LMWGs, which may form mesoscopic fibrils through intermolecular interactions and which are more sensitive to environmental stimuli.

Herein we report that a new structured amine oxide surfactant, 6-((1*R*,4a*S*)-7-isopropyl-1,4a-dimethyl-1,2,3,4,4a,9,10,10a-octahydro-phenanthrene-1-carboxamido)-*N*,*N*-dimethyl−hexan-1-amine oxide, abbreviated as R-6-AO, has been synthesized using dehydroabietic acid derived from rosin as a starting material (Figures S1-S3). It behaves as a highly efficient LMWG with an ultralow critical gelation concentration (CGC) of 0.18 wt.% (4 mM) in pure water and a critical gelation temperature (T_{gel}) of 35 °C (Figure S4). Using R-6-AO as gelator at rather low concentration (5 mM), stable O/W gel emulsions of various long chain oils can be prepared, in which R-6-AO molecules containing a tricyclic diterpene structure gelate the aqueous phase *via* unique self-assembly and the oil volume fraction can be freely adjusted between 2% and 95%. This strategy greatly enriches gel emulsion formulations and their applications such as in the preparation of new soft materials.

R-6-AO has a molecular structure and a surface tension (γ)-concentration (C) curve shown in Figures 1(A) and (B) which gives a critical micelle concentration (cmc) of 0.6 mM, γ_{cmc} of 35.1 mN/m and a minimum molecular area (a_{min}) of 0.57 nm^2 at the air-water interface. This a_{min} is only slightly larger than that of the traditional dodecyl dimethylamine oxide surfactant (0.47 nm^2) [25] which adopts a conformation with its long alkyl chain perpendicular to the air-water interface, indicating that R-6-AO adopts a similar conformation at liquid interfaces. The synthesis procedures, molecular characterization as well as various performance assessments of the product are detailed in the Supporting Information (Figures S1-S5). Amazingly, an aqueous solution of R-6-AO can be gelated to a clear and transparent hydrogel at concentrations as low as 0.18 wt.% or 4.0 mM, when the solution was heated to 50 °C and then cooled to room temperature (20-25 °C), as shown in Figure 1(C). Hydrogels cannot be formed without such a heating-cooling process. R-6-AO therefore is a super LMWG since each R-6-AO molecule can immobilize 13,888 water molecules on average. The dynamic rheological measurements of the hydrogel samples formed by R-6-AO at different concentrations (Figure S5) show that within the range of oscillation frequencies investigated the values of the storage modulus (G') of all gels are always higher than those of the loss modulus (G''), indicating typical gel-like behavior. It was also observed that at a fixed frequency, the values of G' and G'' increase upon increasing the surfactant concentration, indicating formation of a denser meso-scale structure at higher concentration.
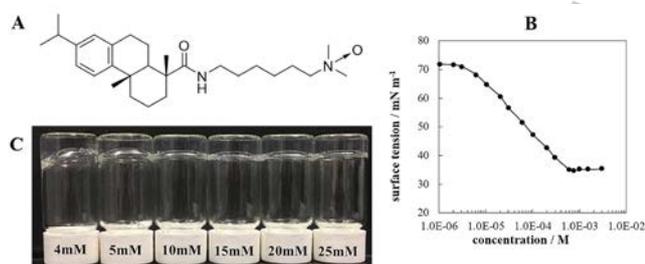

Figure 1. (A) Molecular structure, (B) surface tension of aqueous solution as a function of concentration at 25 °C and (C) appearance of gelled aqueous solutions of different concentration at 25 °C of the synthesized R-6-AO surfactant.

The microstructure of the gelled aqueous solution of R-6-AO at 5 mM was then investigated with cryo-TEM, as shown in Figure 2(A), which clearly shows that the self-assembly aggregates of R-6-AO are fibers, mostly with lengths > 2 μm. In addition, there are no fractured fibers, indicating their excellent mechanical strength. This may account for the fact that R-6-AO can gel water at extremely low concentrations.

In many reported supramolecular hydrogel systems, the LMWGs tend to form thick bundles. For example, a bola-amphiphile with two carboxylic acid groups as end groups and a diketopyrrolopyrrole (DPP) chromophore can self-assemble into fiber bundles of 30-100 nm width, [26] and a cyclohexane-based surfactant was reported to self-assemble into fiber bundles of 50-80 nm width. [27] However, as revealed in Figure 2(A), the

cross-sectional diameter of the fibril formed by R-6-AO is as low as 10 nm, indicating that R-6-AO may follow a self-assembly mechanism different from other LMWGs reported.
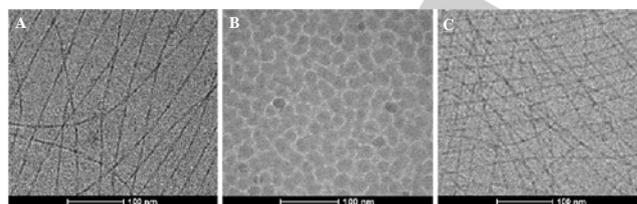
Figure 2. Cryo-TEM images of a gelled R-6-AO aqueous solution at (A) 5 mM and (B) after addition of ethanol (0.5 mL ethanol into 2 mL R-6-AO solution of 5 mM); (C) cryo-TEM image of the continuous phase of an O/W gel emulsion (50% oil) stabilized by 20 mM R-6-AO after storing for one year at 20 °C.

As a typical amphiphilic surfactant (Figure 1(A)), R-6-AO is likely to form aggregates of core-shell structure with the hydrophobic group inside and the hydrophilic amine oxide group on the surface of the fibers. The arrangement of R-6-AO molecules within the fibers is similar to that in worm-like micelles formed by certain surfactants. It is worth noting that in Figure 2(A) alternate sections of different darkness can be observed along a fiber, probably indicating a helical structure. Since the fibers are too fine to be recognized in detail, they were further investigated with circular dichroism (CD), a powerful means to characterize the chirality of systems.
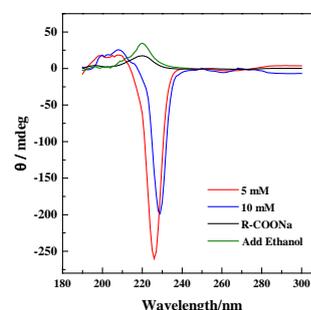
Figure 3. CD spectra of R-6-AO and R-COONa in different environments.

To rule out the possible effect of molecular chirality originating from R-6-AO, the CD spectrum of sodium dehydroabietic acid (R-COONa) containing only the dehydroabietic acid unit was also investigated, which gives a positive absorption peak at 225 nm attributed to the molecular chirality of the dehydroabietic acid unit, as shown in Figure 3. On the other hand, all the R-6-AO samples give totally different strong CD signals at concentrations above the CGC of 4 mM. For example, the 5 mM sample gives a positive absorption peak at 203 nm and a large negative absorption peak at 226 nm, proving the formation of left-handed chiral aggregation structures. As the concentration increases, the CD adsorption peaks gradually move towards longer wavelength. However, addition of 0.5 mL of alcohol into a 2 mL sample of 5 mM R-6-

AO results in dissociation of the fibrils, as shown in Figure 2(B), leaving only the CD signals corresponding to molecular chirality.

As a surfactant, R-6-AO can effectively reduce the surface tension of water (Figure 1(B)). The surface tension beyond the cmc was measured just before the solution was gelled after increasing the temperature in order to render R-6-AO soluble. The size of the micelles in an aqueous solution of 5 mM at 50 °C was determined to be about 4 nm by dynamic light scattering, as shown in Figure S6. Upon reducing the temperature to 25 °C, fibrils are formed gradually *via* the self-assembly of R-6-AO.

R-6-AO displays obvious emulsifying ability at concentrations as low as 1.0 mM. To probe the emulsification properties of R-6-AO as well as the ability to form gel emulsions, *n*-dodecane-in-water emulsions with different oil:water volume ratios were investigated at a constant aqueous surfactant concentration of 20 mM. The oil-water mixtures were homogenized at 11,000 rpm for 2 min at 50 °C to form fluid emulsions, which were kept at 50 °C for 0.5 h to allow creaming to occur, followed by cooling to 20 °C to form gel emulsions as shown in Figure 4(A); both the emulsion and the resolved aqueous phase are gelled in each sample. All the samples are stable O/W gel emulsions which remain unchanged at 20 °C for one year as shown in Figure 4(B). The gel emulsions with similar stability can also be prepared by hand shaking a biphasic system for a few seconds, but they possess larger droplet sizes (Figure S7) than those prepared by homogenization. The excellent long term stability of the emulsions is attributed to the gelled continuous phase, which restricts movement of the dispersed droplets and thus prevents their flocculation and coalescence. Besides, the oil volume fraction in the emulsions can be freely selected between 2% and 95% when preparing gel emulsions, which is difficult to realize in traditional HIPE systems. Interestingly, when the freshly emulsified samples (at 50 °C) are rapidly cooled to 20 °C or below, homogeneous gel emulsions were obtained even in the presence of excess water, as shown in Figure 4(C), which is different from many HIPEs where the excess water will separate after creaming. This makes it possible to randomly adjust the volume fraction of the droplets in the gel emulsions and the average inter-droplet distance, both of which are beneficial for preparing mesoscopic materials of variable structure.

The shape and size of the droplets in the gel emulsions were observed by confocal laser scanning microscopy (CLSM), as shown in Figure 5. It is seen that at low oil volume fraction the oil droplets are spherical and are dispersed sparsely in the continuous phase, and both the droplet concentration and size increase with increasing oil volume fraction. At an extremely high oil volume fraction (95%), droplets become polyhedral, similar to that observed in other HIPEs. These emulsions also show good thermal reversibility when heated to 45 °C and cooled to 20°C, with the droplet size remaining almost the same after several cycles (Figure S8 and S9).

The appearance of gel emulsions containing 50% oil stabilized by R-6-AO at different concentrations is shown in Figure 6(A). It is observed that at concentrations beyond the CGC (4 mM) the fluid emulsions become gelled emulsions which reveals that R-6-AO can stabilize gel emulsions at concentrations as low as 0.2 wt.% (5 mM), much lower than that in other HIPEs (normally beyond 5 wt.%). [14-16] The droplet

size can be further reduced by increasing the R-6-AO concentration and approaches a limit of around 7-8 μm at 50 mM, as shown in Figure S10. As expected both the emulsification and emulsion stability can be improved by increasing the R-6-AO concentration.

Figure 4. Digital photos of *n*-dodecane-in-water gel emulsions (inverted) prepared at a constant R-6-AO concentration of 20 mM but with different oil volume fractions as shown. After homogenization at 50 °C samples were kept at 50 °C for 0.5 h, and then (A), (B) cooled naturally to 20 °C or (C) cooled quickly from 50 °C to 20 °C in 15 min followed by storage at room temperature. Photos were taken (A) 7 days and (B, C) one year after storage.
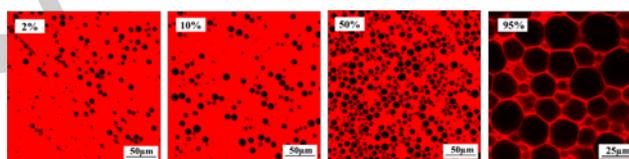
Figure 5. Confocal fluorescence images of *n*-dodecane-in-water gel emulsions with different oil volume fractions (shown) stabilized by 20 mM R-6-AO, taken 7 days after storage at 20 °C. The aqueous phase was labelled with rhodamine B excitable by a laser at 540 nm.
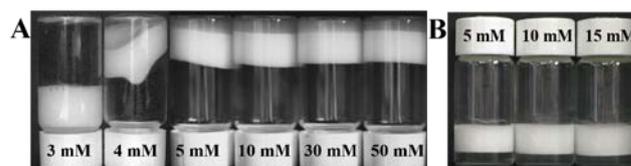
Figure 6. Digital photos of *n*-dodecane-in-water (50% oil) gel emulsions stabilized by R-6-AO at different concentrations as shown, taken (A) 7 days after storage at 20 °C and (B) 30 days after storage at 45 °C. The water resolved in (A) is gelled but not in (B).

The microstructure of the continuous phase in the gel emulsions (50% oil) stabilized by 20 mM R-6-AO was also characterized by cryo-TEM, as shown in Figure 2(C). Unfortunately, the image only shows the fibers and not the oil droplets, which may have been damaged during sample preparation (wiping excess sample using filter paper) or evaporated under vacuum. Anyway, the image confirms that the fibers are still formed in the presence of oil, which is essential for the formation of the gel emulsions here. The chiral fibers with length of over 2 μm can be clearly seen, and the morphology of

the fibers is the same as that formed in pure water (Figure 2(A)). Since fibers can also stabilize emulsions, [27-30] it is important to establish whether R-6-AO molecules or self-assembled fibers are responsible for emulsion stabilization. In order to answer this, gel emulsions at 5, 10 and 15 mM R-6-AO were prepared and held at 45 °C (over T_{gel}) at and beyond which fibrils will not be formed. As shown in Figure 6(B), emulsions with a fluid aqueous phase still remain stable even after 30 days, and the droplet size decreases with increasing R-6-AO concentration (Figure S11) indicating that it is molecules of R-6-AO that adsorb at the oil-water interface to stabilize the emulsions. In another experiment, a 10 mM R-6-AO hydrogel was mixed with dodecane at an oil:water ratio of 1:1 for homogenization at 25 °C, where the originally formed intact fibers were broken into pieces and cannot be recovered. However, the prepared emulsion still shows favorable stability. When further heated to and kept at 45 °C to eliminate all fiber pieces, the emulsion also remains stable after 30 days. Since R-6-AO monomers with low concentrations coexist with fibers as proved by the regular surface tension curves (Figure 1(B)) and the above experiment, it can be concluded that at room temperature it is still R-6-AO molecules that stabilize emulsion droplets, where the hydrated headgroup layer covering droplet interfaces endows steric stabilization to approaching droplets. These results indicate that in the gel emulsions reported here R-6-AO molecules not only gel the continuous phase by forming fibers, but also adsorb at the oil-water interface which is quite different from other gel emulsions stabilized solely by fibrils. [28-31] For example, the gel emulsions stabilized by fibers formed by dipeptide derivatives demulsified when kept at 60 °C for 3 h [28].

In addition, the dynamic rheological measurements of *n*-dodecane-in-water gel emulsions (50% oil) stabilized by R-6-AO at different concentrations are shown in Figure S12. It is found that the storage modulus G' is significantly higher than the loss modulus G'' at any R-6-AO concentration, demonstrating elastic gel behavior. This is similar to the gels formed by R-6-AO in the absence of oil. The gel emulsion droplets are therefore ductile when they are forced to flow as proved by stirring an emulsion at 2000 rpm for 1 h, where most of the fibrous structures are broken leading to fluid emulsions without the release of oil, as shown in Figures S13-S15.

It is worth noting that although a large number of hydrogels have been prepared using LMWG molecules, O/W gel emulsions using LMWG as stabilizer are still rare. The main reason is that the fibril structures in the aqueous phase are formed by the self-assembly of LMWG molecules *via* intermolecular interactions such as van der Waals attraction and hydrogen bonding. However, the mesoscopic structures of the self-assembly will become extremely fragile and may easily dissociate into small molecules once in contact with an organic solvent so as to lose the ability to trap water. However, the R-6-AO reported here is highly resistant to long chain organic compounds such that O/W gel emulsions can be formed using a variety of oils such as *n*-octane, *n*-decane, soybean oil, olive oil and tricaprylin in addition to *n*-dodecane, as shown in Table S1. This may be attributed to the tricyclic diterpene hydrophobic structure in R-6-AO molecules, which has a topology different from alkyl tails so that R-6-AO molecules can adopt a unique arrangement in the formed fibers preventing insertion of long

alkyl chains in organic compounds and maintaining integrity of the fibers.

In summary, a new low molecular weight hydrogelator (R-6-AO) has been synthesized from dehydroabietic acid derived from rosin, which can gel pure water at concentrations as low as 0.18 wt.% or 4 mM, and stabilize *n*-dodecane-in-water gel emulsions with the oil volume fraction freely adjustable between 2% and 95% at concentrations beyond 5 mM. Since the fibril structures in these systems are formed through molecular interactions, they are especially sensitive to the external stimuli. These characters are superior to typical covalently bonded biopolymers as water thickening agents such as collagen, locust bean gum, carrageenan, gelatin, fish gelatin, gum arabic and xanthan gum, [32-36] which require much high concentrations to form gel emulsions. On the one hand, R-6-AO molecules adsorb at the oil-water interface to stabilize emulsions *via* steric stabilization like ordinary nonionic surfactants, and the self-assembly of R-6-AO in water into fibers is responsible for gelation of the aqueous phase which restricts the movement of droplets and thus endows the gel emulsions with long term stability. Different from other LMWG molecules, R-6-AO molecules containing the tricyclic diterpene hydrophobic structure and can adopt a unique arrangement in the fibers preventing insertion of long alkyl chains in oils and maintaining integrity of the fibers. This made it possible to stabilize gel emulsions with a variety of oils other than *n*-dodecane, such as *n*-octane, *n*-decane, soybean oil, olive oil and tricaprylin. It is believed that the excellent properties of the gel emulsions stabilized by R-6-AO derived from rosin make them potentially applicable in many fields such as the preparation of new porous materials, transportation of oil products as well as in the cosmetic and pharmaceutical sectors.

Acknowledgements

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COMMUNICATION

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Layout 1:

COMMUNICATION

A novel amine oxide surfactant was synthesized using dehydroabietic acid derived from rosin, which behaves as a highly efficient low molecular weight hydrogelator by self-assembly into left-handed helical fibers in aqueous solution with an extremely low critical gelation concentration (CGC) (0.18 wt.% or 4 mM). O/W gel emulsions of various oils with the oil volume fraction freely adjustable between 2% and 95% are prepared at surfactant concentrations beyond the CGC.
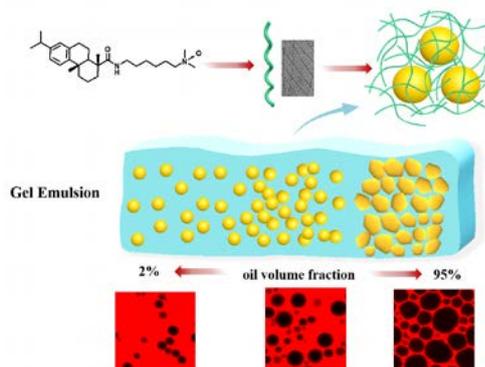
T. T. Yan, B. L. Song*, X.M. Pei, Z. G. Cui*, B. P. Binks*, and H.Q. Yang

Page No. – Page No.

Widely adaptable oil-in-water gel emulsions stabilized by an amphiphilic hydrogelator derived from dehydroabietic acid

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