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## **Electronic Supplementary Information**

## Thixotropic stiff hydrogels from a new class of oleoyl-D-glucamine-based low-molecular-weight gelators

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Fig. S1 Evaluation of hydrogelation in aqueous solutions: (a) OG-G, (b) OG2-G, (c) OG3-G and (d) OA-G.



Fig. S2 Evaluation of hydrogelation in PBS.



Fig. S3 DSC curves of hydrogels (deionised water, heating and cooling rate: 2 °C/min).

Table S1 Transition temperatures of hydrogels obtained from DSC measurements (deionised water,heating and cooling rate: 2 °C/min).

Sample	T <sub>gel→sol</sub> /°C	T <sub>sol→gel</sub> /°C
	(∆H/mJ mg <sup>-1</sup> )	(∆H/mJ mg <sup>-1</sup> )
OG-G 3.0 wt% hydrogel	71 (3.3)	50 (3.2)
OG2-G 2.0 wt% hydrogel	85 (3.0)	78 (3.2)
OG2-G 3.0 wt% hydrogel	85 (3.6)	78 (3.7)
OG3-G 2.0 wt% hydrogel	104 (1.9)	103 (1.9)
OG3-G 3.0 wt% hydrogel	104 (3.4)	102 (3.3)
<b>OA-G</b> 3.0 wt% hydrogel	83 (3.6)	57, 66* (3.5)

\* Bimodal peak (peak temperatures).

Similar transition temperatures were obtained for **OG2-G** and **OG3-G** hydrogels for 2.0 and 3.0 wt% (generally,  $\Delta$ H increases with sample weight). This means that **OG2-G** and **OG3-G** hydrogels for 2.0 and 3.0 wt% may have a similar gel network. Thus, we believe that the comparison of experimental results at the CGCs was appropriate.



Fig. S4 Periodical step-shear test results for OG-G (3.0 wt%), OG2-G (2.0 wt%), OG3-G (2.0 wt%) and OA-G (3.0 wt%) hydrogels (full version).



Recovery

Fig. S5 Thixotropic behaviour of hydrogel formed with OG3-G (3.0 wt% in PBS).



**Fig. S6** IR spectra of the hydrogelators in different states in the region of carbonyl stretching: (a) **OG-G**; (b) **OG2-G**; (c) **OG3-G**; (d) **OA-G**.

To clarify the driving force for fibre formation of hydrogelators, the infrared (IR) spectra of hydrogelator systems in different states, in the region of the carbonyl stretching, were measured (Fig. S10). One of the main absorption peaks of the carbonyl group in the higher wavenumber region (near 1650 cm<sup>-1</sup>) increases and that of a lower wavenumber region (near 1550 cm<sup>-1</sup>) decreases when the state changes from solution to gel and from gel to xerogel. The absorption peak of higher wavenumber (over 1650 cm<sup>-1</sup>) could be attributed to the carbonyl stretching of non-interactive, free molecules, and that of lower wavenumber (1650 cm<sup>-1</sup> and 1550 cm<sup>-1</sup>) could be attributed to the carbonyl stretching of those with intermolecular interactions due to hydrogen bonding. This trend in the IR carbonyl stretching frequencies is observed in all hydrogelator systems. From these results, the driving force for the tendency for self-assembly likely depends on hydrogen bonding of hydrogelators.

## References

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