Experimental report

05/08/2021

Proposal: 9-11-1964 **Council:** 10/2019

Title: Probing self-sorted networks usingdeuterated gelators

Research area: Materials

This proposal is a new proposal

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Samples: C30H28N2O5
C19H22N2O5
C30H2D26N2O5

Instrument	Requested days	Allocated days	From	To
D11	3	2	10/09/2020	12/09/2020

Abstract:

Low molecular weight gelators self-assemble in solution to give nanofibres. These entangle to form the gel networks. Normally, a single gelator is used, but there are real opportunities when multicomponent systems are used. In these cases, a mixture of gelators is used, all of which can individually assemble. However, in the multicomponent gel networks, either mixing occurs, so that each fibre contains more than one gelator, or self-sorting occurs, where each fibre contains only one gelator. Understanding the networks formed these cases is very difficult as they look the same by microscopy, and the scattering is an average of all of the fibres. One way around this is to use a mixture of a deuterated gelator and a non-deuterated gelator. In the gel phase, only the non-deuterated gelator should scatter and hence the network from this gelator only can be determined. This is the approach we will take here.

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Introduction Here we probed mixtures of different gelators. As one specific example, we used two gelators, one (2NapFF) that forms worm-like micelles at high pH and stable gels at low pH and one (2NapAA) that forms transient micelles at high pH, then gels at low pH, with crystallisation occurring from the gel phase over time. We examined mixtures of these two gelators at different ratios, then used a perdeuterated equivalent of the 2NapFF. This allowed us to compare mixtures at high and low pH where we could measure the scattering from both gelators or only from the non-deuterated 2NapAA. This is the first example of such methods being used to understand multicomponent low molecular weight gel systems.

Experimental The LMWG were prepared as described elsewhere.[1] Solutions at 10 mg/mL were prepared in D_2O at high pD by the addition of one molar equivalent of NaOD (0.1 M), followed by stirring until the LMWG had dissolved.
SANS experiments were performed on the D11 diffractometer, a neutron wavelength of $\lambda = 6$ Å was employed at three different detector distances, D = 1.5, 8 and 39 m. This set-up corresponds to a Q range from $1.0 \times 10\text{-}3$ to 0.31 $Å^{-1}$. All spectra were normalised and corrected using the scattering of the empty cell. Scattering data were corrected for electronic noise and incoherent background subtraction and normalised by the intensity scattered for a 1 mm H_2O sample corrected by the intensity scattered from the empty quartz cell. To induce gelation, the trigger (GdL[2]) was added, and then the solution immediately placed in the cell.

Results We were able to examine the mixture of the two gelators at a different relative concentrations. The data showed that the deuterated 2NapFF (in H_2O) scatters as for the non-deuterated 2NapFF in D_2O. In D_2O, the perdeuterated 2NapFF scatters very weakly as expected. Hence, deueteration does not affect the self-assembly. The scattering from the mixtures could be fitted to hollow cylinder models at high pH for 2NapFF and to cylinder models for the mictures. The data could be used to show that self-sorting occurs at high and low pH and have been used as part of a wider study into this mixed system which has now been published.[3]

References

1. L. Chen, S. Revel, K. Morris, L.C. Serpell and D.J. Adams, *Langmuir*, **2010**, *26*, 13466-13471.
2. D.J. Adams, W.F. Frith, M. Kirkland, L. Mullen and P. Sanderson, *Soft Matter*, **2009**, *5*, 1856-1862.
3. D. Giuri, L.J. Marshall, B. Dietrich, D. McDowall, L. Thomson, J.Y. Newton, C. Wilson, R. Schweins, and D.J. Adams, *Chem. Sci.*, **2021**, *12*, 9720 – 9725.