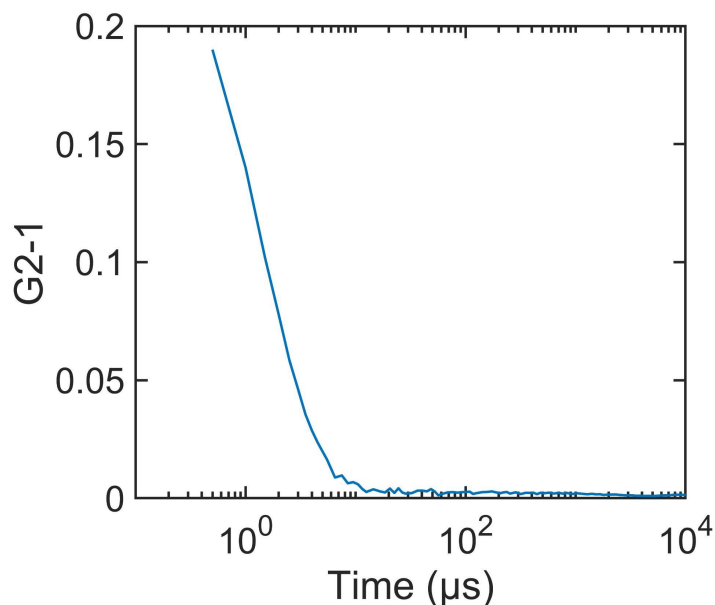


Supplementary Information

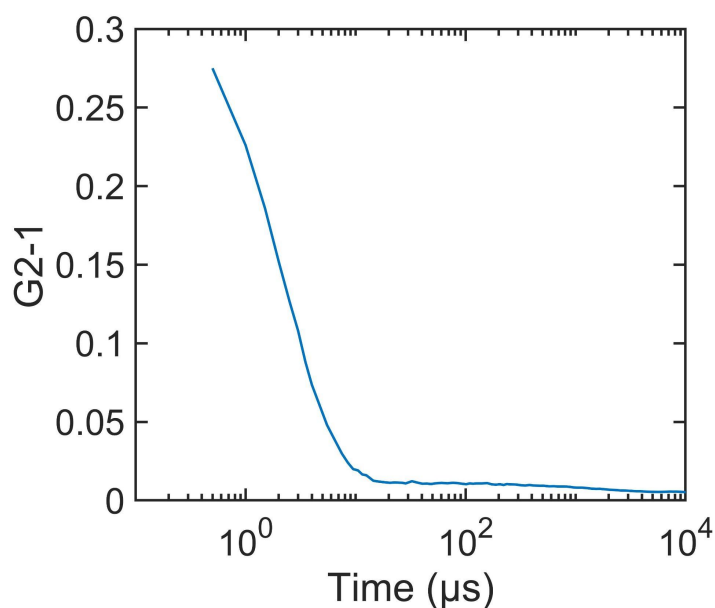
DLS measurements: G2 versus lag time

Appended below are the G2 versus lag time plots corresponding to the DLS measurements presented in Fig. 4b of the main text. The composition of the mixture in the tube is printed above each plot.

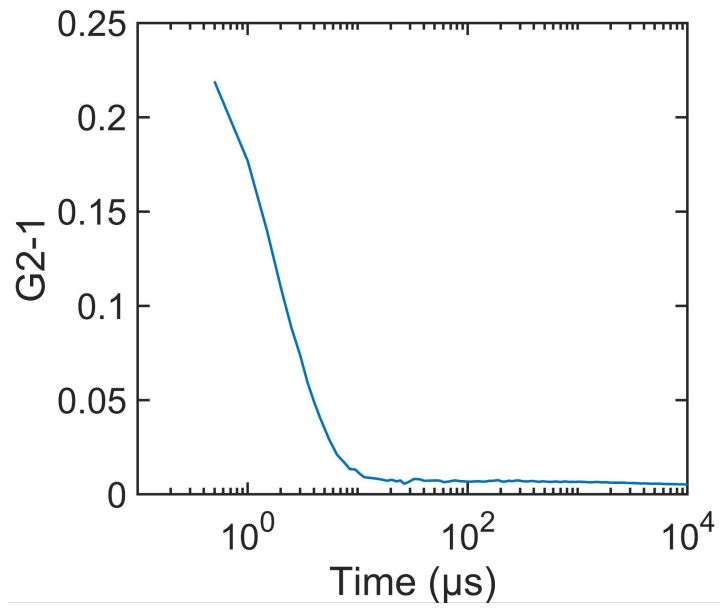
0.5 g PMMA, 10 g water, 2 g AAC:



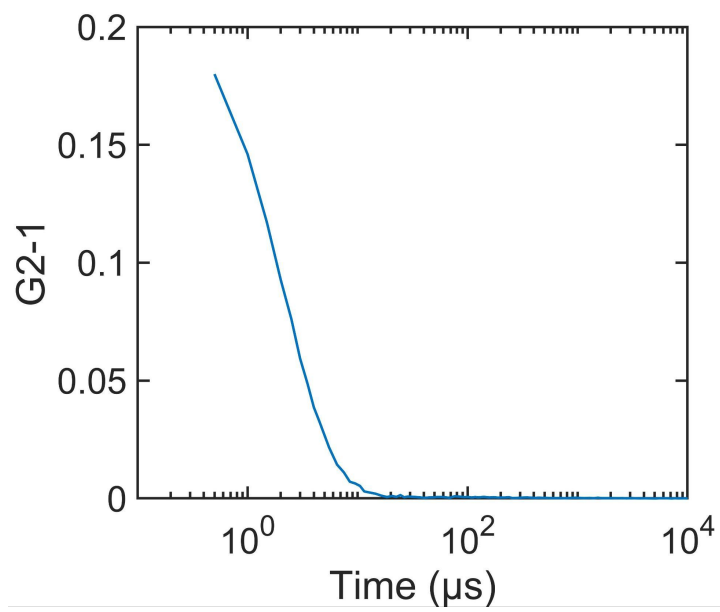
0.5 g PMMA, 10 g water, 4 g AAC:



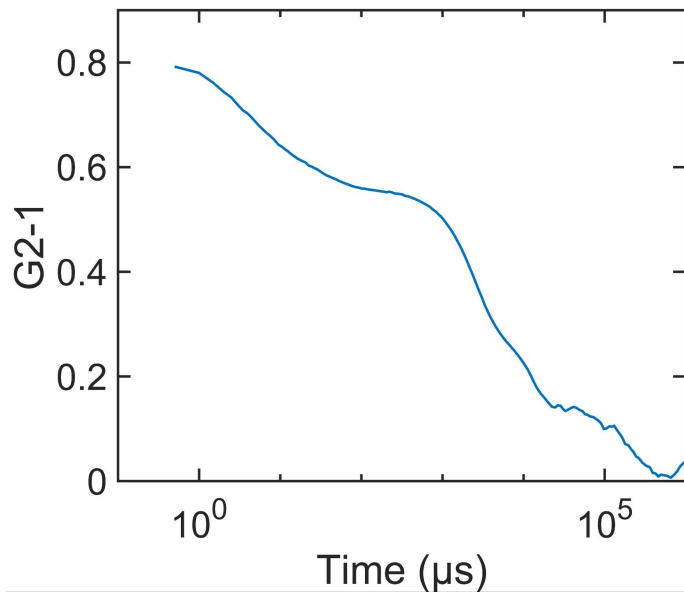
0.5 g PMMA, 10 g water, 6 g AAC:



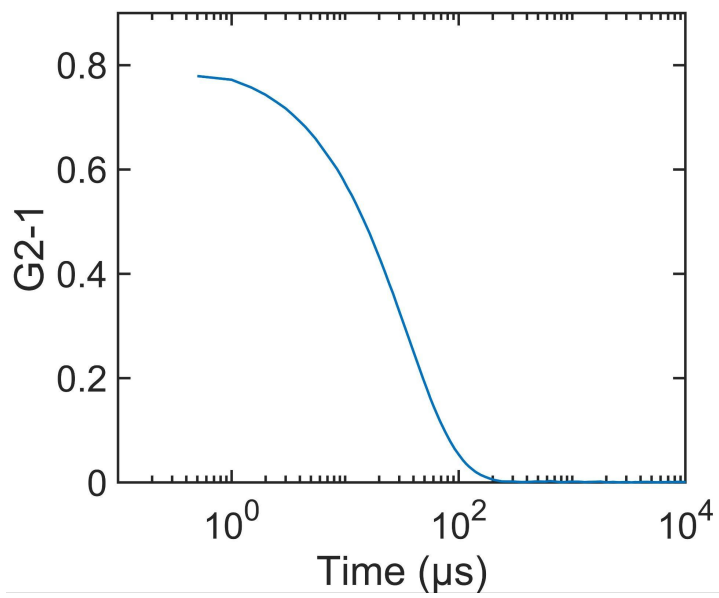
0.5 g PMMA, 10 g water, 8 g AAC:



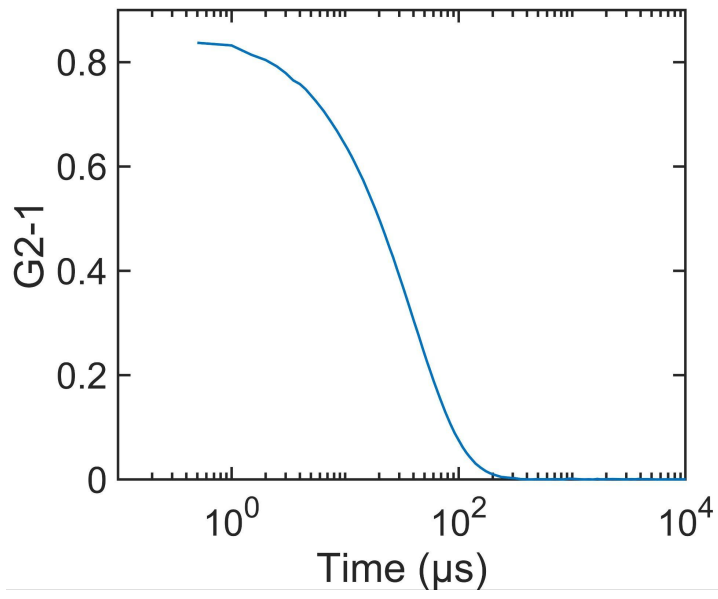
0.5 g PMMA, 10 g water, 10 g AAc:



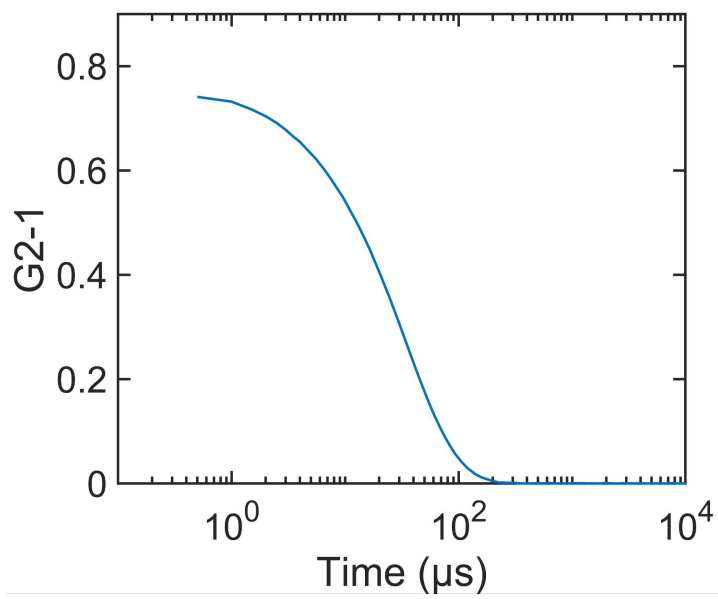
0.5 g PMMA, 10 g water, 12 g AAc:



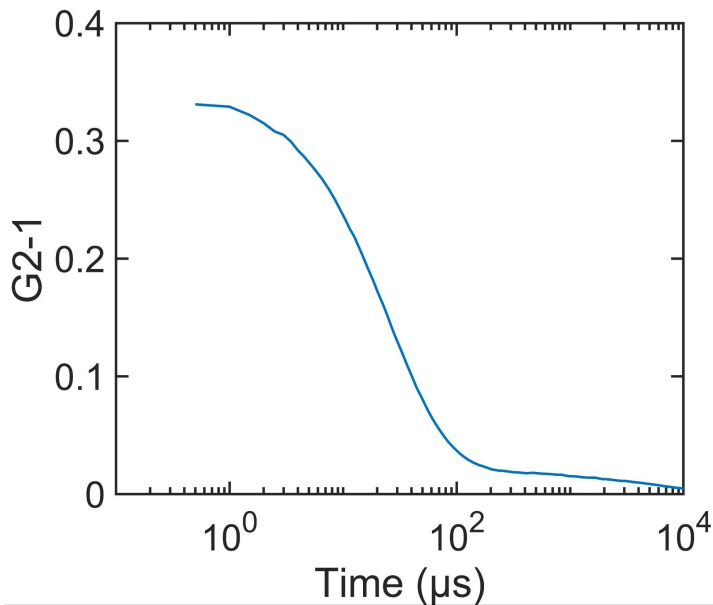
0.5 g PMMA, 10 g water, 14 g AAc:



0.5 g PMMA, 10 g water, 16 g AAc:



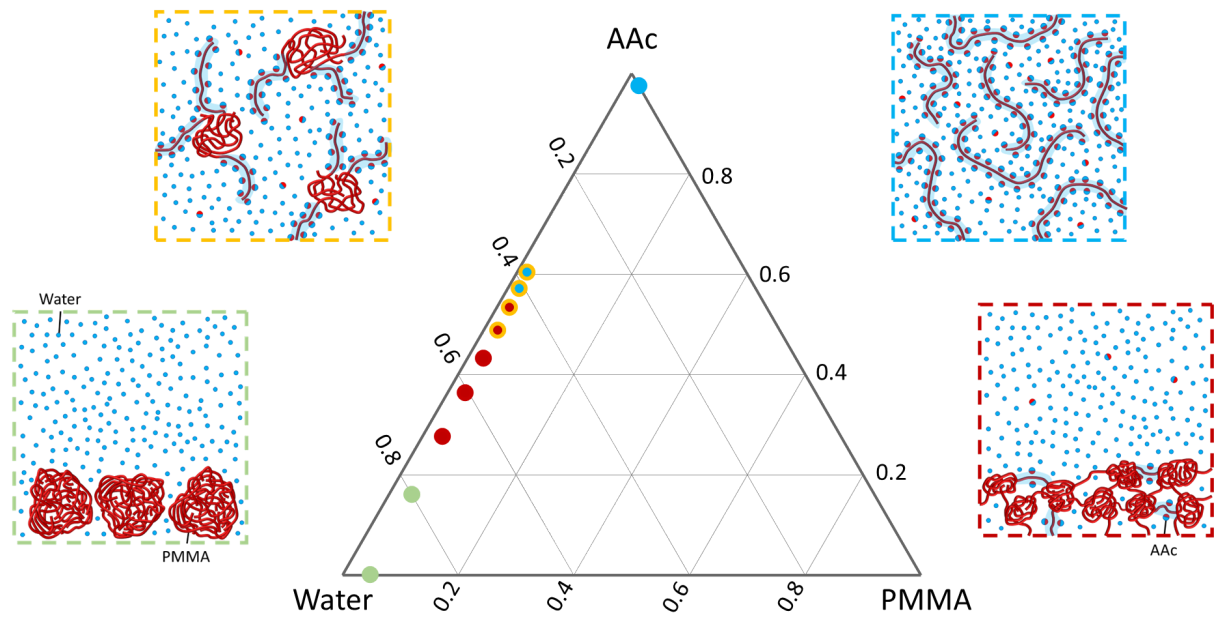
0.5 g PMMA, 0 g water, 20 g AAc:



Morphologies in the composition triangle

In Fig. 2 of the main text we present four different morphologies. In Figs. 3 and 4, and the corresponding text, we discuss the experimental manifestation of these morphologies. Following this, upon discussing the phase behaviour in Fig. 5, we focus solely on if the sample forms one or two layers. This is because we envision applications in which the AAc monomers are polymerized to manufacture materials of unusual mechanical properties. In a recent publication we demonstrated our procedure, for which phase separation is a prerequisite [1].

To nevertheless provide a connection between the two, below we show the data points corresponding to the measurements of Figs. 3 and 4 in the composition triangle. Green dots correspond to mixtures in which PMMA forms glassy particles (Fig. 2a) and red dots correspond to mixtures in which PMMA forms a gel (Fig. 2b). Blue dots correspond to mixtures that form a polymer solution (Fig. 2d). In addition to this, the PMMA in a mixture can form micelles (Fig. 2c), which may coexist either with the gel phase or the polymer solution. We indicate mixtures in which micelles form with data points that have a yellow border. The phase of glassy particles and the gel phase result in a tube in which two layers form (red dots in Fig. 5). The polymer solution phase results in a tube in which a single transparent liquid forms (blue dots in Fig. 5). Micelles may, in general, exist in both cases.



[1] G. Zhang, J. Kim, S. Hassan and Z. Suo, *PNAS*, 2022, **119**, e2203962119.