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The Structure and Gelation Mechanism of Tunable Guanosine-Based Supramolecular Hydrogels Z. LI, L. BUERKLE, CWRU, M. ORSENO, K.A. STRELETZKY, CSU, S. SEIFERT, ANL, A.M. JAMIESON, S.J. ROWAN, CWRU — SLS experiments on 50/50 mixtures of Guanosine (**G**) and 2',3',5'-Tri-O-Acetylguanosine (**TAcG**) in aqueous 0.354 M KCl show a sudden increase in molecular weight between c of 0.1 and 0.2 wt%, indicating the critical concentration (C_c) for self-association of **G/TAcG** quartets into columnar assemblies. Guinier plot of SLS data above C_c indicates a radius of gyration that decreases with c and unphysically small values of molecular weight, suggesting that SLS is probing internal structure of microgel particles. Polarized and depolarized DLS indicate translational and rotational diffusion of a bimodal distribution of particles. The fast DLS modes appear to originate from fibrillar agglomerates of **G/TAcG** columnar quartet assemblies, and the slow modes from microgel domains. SAXS probes structure of individual columnar **G/TAcG** quartet assemblies, and indicates that, above C_c it is described by c -independent rigid cylinder, whose dimensions are consistent with earlier SANS results. Collectively, the experiments suggest that sol and microgel phases coexist in solution below macroscopic gel point, and that sol phase contains individual columnar stacks of **G/TAcG** quartets and fibrillar aggregates formed via lateral aggregation of these columnar assemblies. With increasing c , DLS indicates progressive increase in the volume fraction of microgel domains, which ultimately leads to macroscopic gelation.

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