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Assembly and Gelation of Four-Armed DNA Dendrimers¹

FRANCIS STARR, Wesleyan University

The disordered-arrested state of matter is ubiquitous in existing materials like glasses, and is prevalent in many new materials, due to the uncommon mechanical, thermal and electrical properties. The “bottom-up” construction of new materials is one of the central aims of nanotechnology. DNA is potentially an optimal choice for the construction of three-dimensional supramolecular assemblies since it can self-assemble into long and fairly rigid helices, based simply on sequence complementarity. We numerically study a model designed to mimic the behavior of recently synthesized single-stranded DNA dendrimers. Complementarity of the base sequences of different strands results in the formation of strong cooperative intermolecular links with a valency controlled by the number of strands. We simulate the bulk behavior of a system containing many 4-armed DNA dendrimers and find that in an extremely narrow temperature range the system forms a large-scale, low-density network via a thermo-reversible gel transition. The sharpness of the crossover the gel state can be controlled by the length of the DNA strands, since longer strands will form cooperative bonds over a narrower temperature range. As a result, the percolation temperature of the network formed by the dendrimers can be made arbitrarily close to the gel transition by tuning the length of DNA strands. This is in contrast with recent model systems designed to understand thermo-reversible gelation. Given that gelation and percolation coincide in irreversible chemical gels, this system provides an excellent model material to bridge the understanding between reversible and chemical gels.

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