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Functional nanogels applied to materials development

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Nanogels are commonly used in the controlled delivery of therapeutic agents, DNA/RNA and proteins as well as for imaging and sensing in other biomedical applications. Nanogels are rarely utilized to modify or create macroscopic polymeric structure, which is the focus of this report. Our unique version of reactive polymeric nanogel materials involves globular particles that have well-controlled swollen dimensions of approximately 10 nm. These particles are composed of many short yet highly interconnected primary chains that cumulatively provide particles with molecular weights of 10^4-10^6 Da.

The introduction of these high molecular weight, reactive prepolymeric nanogels has been evaluated here as a means to revise the structure and properties of polymer matrix networks derived from the monomer-dispersed and monomer-infiltrated nanogels. We also consider the use of nanogels dispersed in inert solvents, including completely water-compatible nanogels, to create macroscopic networks based exclusively on monomer-free covalently interconnected nanogel structures. Prior to the final macroscopic polymerization step, the discrete nanogels display an expected load-dependent viscosity profile; however, as the swollen particle size is decreased from about 50 nm to less than 5 nm, the size-dependent viscosity at any given nanogel loading level drops precipitously. This facilitates the formulation of highly nanogel loaded resins that still retain practically useful rheologic properties while also allowing dense nanogel-based structures to be achieved.

The composition and structure of nanogels can be varied widely with features such as particle size, branching density, hydro-philic/phobic balance, modulus/glass transition temperature, refractive index and appended functionality all readily controlled. The isolated particles easily redisperse into suitable monomers or inert solvents to give stable, transparent suspensions to at least 80 wt% loading levels. As the nanogel content in monomer is increased, substantive reductions are achieved in polymerization shrinkage and stress development profiles while the rate of polymerization of nanogel-infiltrated monomer can be enhanced, supressed or unchanged relative to bulk monomer polymerization. The nanogel-modified resins can be used to prepare polymer/polymer composites with dispersed, co-continuous or confluent nanogel distribution as well as to create composite materials that combine high inorganic filler contents along with high nanogel loadings.

The use of reactive nanogels offers a generic means to dramatically modify the physical, mechanical and chemical properties of existing polymeric materials. This approach also provides a route to high-performance, *in situ* curable polymers based on monomer-free formulations.