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## Advances in Thermoplastic Elastomer Gels: Lightweight High-Strain Materials

As examples of multifunctional macromolecules that can spontaneously microphase-separate to form a variety of **nanstructures**, block copolymers continue to receive tremendous attention from the scientific and technological communities. Of particular interest here are thermoplastic elastomers: triblock and higher-order multiblock copolymers that consist of rigid endblocks and one or more flexible midblocks. Selective swelling of these examples of synthetic rubber with a low-volatility oil yields physically-crosslinked molecular networks referred to as thermoplastic elastomer gels (TPEGs). Unlike their chemically-crosslinked analogues, these materials can be recycled and reused by solvent dissolution or thermal processing. This interesting **class of materials** also possesses composition-tunable moduli that can extend to levels below the plateau modulus, as well as strain levels that exceed those, of conventional elastomers. Because these materials contain as much as 90 wt% liquid, they afford a lightweight alternative to dense polymers. In this presentation, we first discuss some of the fundamental **mesophase-level** characteristics of TPEGs from experimental and simulation results. Of particular interest here is the network topology, expressed in terms of the fraction of midblock bridges. For this purpose, both on-lattice Monte Carlo simulations and off-lattice Dissipative Particle Dynamics simulations are employed, and the most probable molecular conformations are identified by defining the Midblock Conformation Index. This analysis will be followed by examples of contemporary technologies, such as dielectric elastomers, that can benefit from the development of such materials. While most TPEGs rely on copolymers possessing rigid (glassy) polystyrene endblocks and flexible polydiene or polyolefin blocks swollen with an aliphatic oil to form a spherical micellar morphology, another possibility consists of copolymers with crystallisable polyethylene endblocks and a styrene/olefin midblock. We use transmission electron microscopy and small-angle X-ray scattering (SAXS) to examine the competition between **crystallization** and self-assembly as these copolymers form discrete semi-crystalline aggregates in solution as the temperature is reduced. The morphologies and properties of the resultant films are then monitored as oil is introduced into the copolymers to generate TPEGs, and their properties are compared to copolymers composed of high-density polyethylene hard blocks and poly(ethylene-co- -octene) soft blocks, which can attain strain levels beyond 4000%.

### Biography:

Dr. Richard J. Spontak is a Distinguished Professor of Chemical & Biomolecular Engineering, Professor of Materials Science & Engineering and Alumni Distinguished Graduate and Undergraduate Professor at NC State University. He received his B.S. degree from Penn State University in 1983 and was awarded his Ph.D. from UC Berkeley in 1988. He then pursued post-doctoral research in at the University of Cambridge (U.K.) and the Institute for Energy Technology (Norway) before joining the Procter & Gamble Company in 1990. In 1992, he accepted a faculty position at NC State University, where he supervises the Macromolecular Materials & Morphology Group.



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