Simulations of controlled degradation in hydrogels – a Dissipative Particle Dynamics approach

*Vaibhav Palkar, Chandan K. Choudhury, Olga Kuksenok

Graduate Student, Department of Materials Science and Engineering, Clemson University

Postdoctoral Researcher, Department of Materials Science and Engineering, Clemson University

Associate Professor, Department of Materials Science and Engineering, Clemson University

Keywords: transient crosslink, dissipative particle dynamics, adaptive hydrogels

Recent advances in chemistry have enabled the synthesis of polymers with labile crosslinks that can degrade in response to a stimulus [1]. This degradation can be performed using stimuli such as light, heat and ultrasound [1-3]. Such degrading crosslinks enable the development of active stimuli responsive materials. As an example, photo cleavable crosslinks have enabled the development of a platform for generating user directed neural networks [4]. Simulation of hydrogels with degrading crosslinks would provide a means to elucidate the physics behind this interesting phenomenon and enable computer aided design of such materials. We have developed a Dissipative Particle Dynamics (DPD) simulation framework aimed at reproducing transient crosslinking. DPD allows us to simulate larger size hydrogel samples compared to classical molecular dynamics. The crosslink breaking rate in our simulations can be controlled using a single parameter and thus, our approach enables the simulation of controlled bond breaking in a crosslinked polymer system. We demonstrate the controlled degradation of crosslinks and enhancement in spreading of these gels on an oil-water interface.

References:

1. Wojtecki, Rudy J., Michael A. Meador, and Stuart J. Rowan. "Using the dynamic bond to access macroscopically responsive structurally dynamic polymers." Nature materials 10.1 (2011): 14.
2. Fuhrmann, Anne, et al. "Conditional repair by locally switching the thermal healing capability of dynamic covalent polymers with light." Nature communications 7 (2016): 13623.
3. Paulusse, Jos MJ, D. J. M. Van Beek, and Rint P. Sijbesma. "Reversible switching of the sol− gel transition with ultrasound in rhodium (I) and iridium (I) coordination networks." Journal of the American Chemical Society 129.8 (2007): 2392-2397.
4. McKinnon, Daniel D., et al. "Design and characterization of a synthetically accessible, photodegradable hydrogel for user-directed formation of neural networks." Biomacromolecules 15.7 (2014): 2808-2816.