Report for newly appointed faculty startup

1. Name of project leader： Tetsuya Yamamoto
2. Project title: Mechanical properties of self-growing double network gels
3. Report

Gong and coworkers (ICReDD, Hokkaido University) have developed a double-network hydrogel that increases strength and mass under repetitive mechanical loading (*Matsuda et al., Science, 2019*). The stretching of the hydrogel breaks bonds of extended polymers and thus creates radicals that drive the radical polymerization to produce new network strands. The objective of the proposed research is to make a molecular theory to predict the mechanical properties of self-growing gels as a function of the conditions to heal the crack, reinforce, and strengthen the network.

As a first step towards this goal, we started from developing a scaling theory of the swelling and deswelling of single network gels in an extension of the non-affine tube model (*Panyukov and Rubinstein, 1997*) and the fractal loopy globule models (*Ge et al., 2016*). Strands of networks behave as liquid in small length scales because they are insensitive to the connectivity to the rest of the network and they behave as solid in large length scales because their fluctuations are suppressed by the potentials due to the crosslinks and entanglements. The length scale at the crossover between the two length regimes determines the mechanical properties of polymer gels. The topological potentials (that account for entanglements) decrease upon network swelling due to the separation of the neighboring strands, whereas the crosslinking potentials (that account for crosslinking) are constant. Our theory predicts that entangled networks (where their elasticity is dominated by the topological potentials) in the preparation condition become unentangled networks (where their elasticity is dominated by the crosslinking potentials) upon swelling. Upon network deswelling, network strands produce `entanglements', while the topology of the network is fixed. This situation is analogous to concentrated solutions of ring polymers. We used this analogy to predict that network strands form fractal loopy globules. This theory is a necessary step to develop a molecular theory of the mechanical properties of self-growling double-network gels.

In FY2019, the start-up grant was used to the travel expense of T.Y. to Duke University, NC to closely collaborate with Prof. Michael Rubinstein.

4. Research achievement