Relationship between viscoelastic properties and molecular behavior of physical gels with ideal network cross-linked by duplex DNA

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Dynamically crosslinked gels exhibit a time-dependent mechanical responses, which is solid for a short time but fluid for a long time. These unique properties are attributed to the repeated breaking and reforming of the crosslinkers that bridge the polymer chains in the gels. DNA duplexes are promising dynamic crosslinkers because its thermodynamic and kinetics properties of DNA duplexes can be continuously adjusted by changing the sequences and the length. Ideally, dynamic crosslinked gels with arbitrary viscoelastic properties can be fabricated by designing the DNA sequence. Here, we demonstrated a homogeneous DNA gel with highly predictable mechanical behaviors.

We used a star polymer strategy to improve the homogeneity of the gel network[1,2] and designed a pair of DNA sequences showing a two-state transition as the dynamic crosslinkers. The UV spectroscopy analysis of the DNA gels revealed the good correspondence between the thermodynamic free energy of the DNA crosslinkers in the gel and the simulated values via software package of DNA. Viscoelastic properties tests and dissociation kinetics measurements showed that the macroscopic stress relaxation time of the DNA gels is approximately equal to the lifetime of the duplexes over four orders of magnitude from 0.1-2,000 sec. Furthermore, a series of durability tests found that the DNA gels has self-healing ability for temperature and mechanical stimuli. These results exhibit the great potential of star-polymer-DNA precursors for building gels with tunable and predictable viscoelastic properties, suitable for various applications.

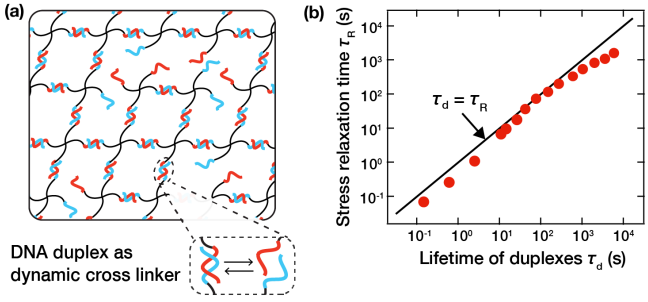

Figure 1(a) An illustration of star-polymer-DNA network. (b) Correlation plot of lifetime of duplexes and stress relaxation time.

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