Ideal Reversible Hydrogels: Theory and Applications

German A. Parada
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Hydrogels are crosslinked polymer networks with high water content that can be designed to have similar properties as native tissue. Their tunability and unique properties make them ideal biomaterial candidates, and as such, they have been explored for a wide range of tissue engineering and biomedical applications. The widespread adoption of hydrogel materials outside research lab settings, however, has been hampered by multiple technical and non-technical limitations. In this work we have addressed two of the current technical limitations identified: Poor mechanical robustness and weak integration with non-hydrogel surfaces, and the lack of quantitative predictions of the hydrogel properties based on the hydrogel's composition and structure.

In this thesis we introduce a set of tough hydrogel materials based on an interpenetrating network architecture using a combination of synthetic and bio-derived polymers. We also introduce several strategies used to robustly adhere these materials to inorganic and elastomeric substrates such as glass, metals, ceramics, silicones, and natural rubbers. The bonding strategies are used to coat thin hydrogel layers on flat surfaces and selected medical devices (urinary catheters, IV tubing, and cardiovascular catheters). Subsequently, we characterize the mechanical (i.e. tensile, friction), biocompatibility, antifouling, functional and blood compatibility properties of various hydrogel-coated surfaces, as compared to those of pristine surfaces, for medical device applications.

Addressing the second limitation, we have developed an Ideal Reversible Polymer Network (IRPN) system featuring 4-arm end-functionalized macromers with reversible crosslinks to correlate the polymer structure to its mechanical properties. The IRPNs show a single relaxation timescale due to the minimization of defects present on their structure, a feature that enables the prediction of the IRPNs' viscoelastic properties under small- and medium-strain shear deformation using Maxwell-based frameworks. These predictions have been validated using a PEG-based hydrogel containing boronic acid-diol dynamic covalent bonding as the reversible crosslinking strategy. The rheology data matches well the model predictions for most experimental conditions tested up to a frequency-dependent critical strain. This boundary is then estimated using scaling arguments. We hope the work presented in this thesis enables the design and formulation of hydrogel-based materials and devices that can be used to reduce clinical complications and address healthcare-related challenges.

Thesis Supervisor: Xuanhe Zhao
Title: Noyce Career Development Associate Professor, Mechanical Engineering