Genipin-Crosslinked Gelatin Bioplastics for Edible Origami Actuators

Spencer Matonis, Bozhong Zhuang[1], Siya Scindia[1,2], Christopher J Bettinger[1,2]
[1]Department of Materials Science and Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA.
[2]Department of Biomedical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA.

Statement of Purpose: The gastrointestinal (GI) tract provides a convenient, non-invasive access point for medical therapeutics and diagnostics. Ingestible devices have traditionally been limited to transient operation and rigid, inorganic structures, largely due to a lack of robust biodegradable materials. As shown in this work, genipin-crosslinked gelatin (Gelapin) biopolymers offer a high degree of mechanical tunability and can be fabricated into a diverse range of functional geometries. Leveraging auxetic origami design and laser processing techniques, we demonstrate the potential for edible, self-deployable Gelapin actuators. Such structures can be fabricated into gastrorentitive interfaces capable of packing up to ~10cm^2 of surface area into a standard "000" pill for future bioelectronic and drug-delivery platforms.

Methods: All raw materials were acquired from Millipore-Sigma (Milwaukee, WI, USA) and used as-received unless otherwise noted. For this study, genipin dissolved in ethanol and water was used to crosslink 300-Bloom, Type A gelatin derived from porcine skin. Approximate crosslinking percentage was determined by the occupation of free amine sites using a ninhydrin assay. Varying amounts of glycerol plasticizer were incorporated into solution and the resulting biogels were dehydrated for 24 hours at 70°C. Auxetic origami designs were etched using a Rabbit CO_2 laser (Middletown, Ohio) to create hill and valley crease lines. Using a 3D-printed polymer mold, the structures were pressed to 10% of their final actuation. Mechanical and viscoelastic properties of biogel and bioplastic samples were evaluated using a tabletop RSA-G2 Solids Analyzer and Discovery HR 20 Rheometer by TA Instruments (New Castle, DE) along with a Instron Universal Testing System (Norwood, MA).

Results: In order to survey the mechanical tunability of gelapin as a function of crosslinking density, a variety of mechanical tests were performed. Figure 1, shows the complex modulus of gelapin samples over a 36hr time sweep operating in the linear viscoelastic region (1Hz) as samples cured *in-situ*. After 36 hours, the un-crosslinked sample exhibited a complex modulus of 1.95kPa, while the fully crosslinked sample measured 11.09kPa, an increase of 469%.

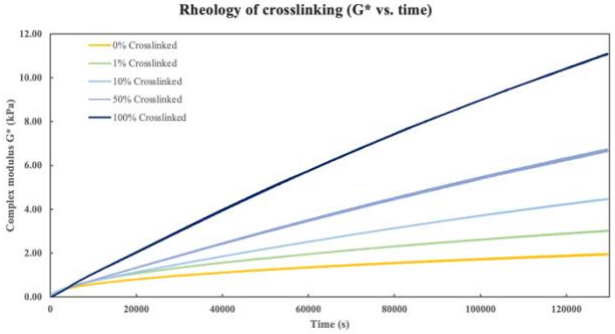

Figure 1: In-situ rheological testing of Gelapin biogels during crosslinking.

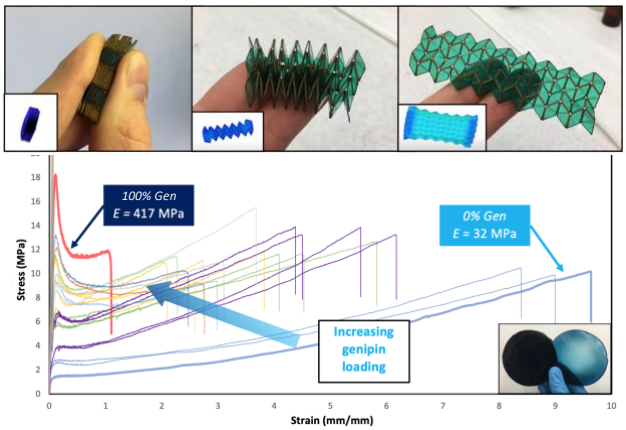

Figure 2: A laser-etched plastic model of a miura-ori origami array alongside FEA analysis (top). Stress-strain plots from uniaxial testing of Gelapin bioplastic samples (inset compares two crosslinked samples).

A similar degree of mechanical tunability was demonstrated via uniaxial tensile tests of Gelapin bioplastic dog bones. In Figure 2, we see an increase of over 1200% in Young's modulus as crosslinking percentage is increased from 0% to 100%. These samples were prepared using a constant glycerol loading (35% mass relative to gelatin) and laser-cut to Type V dog-bones for testing.

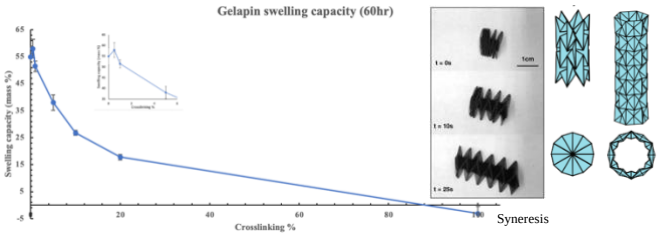

Figure 3: Gelapin swelling capacity as a function of crosslinking (left). Laser-etched gelapin origami self-unfolding over a 25 second period alongside an expanded model of tubular auxetic origami (right).

Conclusion: Here we have shown thorough mapping of the Gelapin system's mechanical, rheological and swelling properties (Figure 3). Additionally, Gelapin bioplastics are demonstrated to be compatible with a highly scalable CO_2 etching process for the production of detailed, expandable origami structures. In future research, we aim to leverage this material knowledge in order to construct and evaluate a semi-empirical model for the bimodal deployment of gelapin origami actuators. This model will be applied to optimize geometric and material design parameters of an auxetic origami tube capable of dilating to a diameter of 25mm, while exerting sufficient radial pressure to be retained in the small intestine for prolonged periods of time (>8hrs).