

Slippery When Wet: Gradient Surface Functionalization of Hydrogels for Lubricity Control

Brandon A. Krick, Angela A. Pitenis, Alison C. Dunn and W. Gregory Sawyer.

Department of Mechanical and Aerospace Engineering, University of Florida.

Statement of Purpose: The body is approximately 60% water; as we develop new biomaterials, we must match local tissue properties as well as design for compatibility with physiological functions of biological tissues (e.g. ion and oxygen transport, cell compatibility and optimal mechanical properties). Hydrogel polymer networks can be developed to contain from less than 20% to more than 90% water for a wide variety of hydration states and mechanical properties. Hydrogels can be tuned to behave as solids, liquids or anywhere in between and can be used in joint implants, heart valves, drug delivery and other biomedical applications. The water content of a hydrogel directly impacts its mechanical properties due to the poroelastic nature of hydrogel polymer networks. Lubricity, which has been shown to directly correlate to comfort in contact lenses¹, can be dramatically altered by tuning the surface water content of hydrogel polymer networks. By functionalizing the surface of hydrogels, we can tune the friction coefficient and other interfacial properties of the hydrogel. These techniques are used to create gradient functionalized surfaces approaching complexities in real biological systems and tissues.

Methods: Polyhydroxyethylmethacrylate (pHEMA) hydrogels were molded into flat rectangular prisms with dimensions of 75 x 25 x 4 mm; gels were crosslinked with N,N'-Methylenebisacrylamide (MBAm) 0.025%, activated in ~ 30 minutes by ammonium persulfate (APS) and tetramethylethylenediamine (TEMED). One 75 x 25 mm face was treated by wetting it with a 50% TEMED solution. Once treated, gradient brushes of polyacrylamide were grown from the surface of the pHEMA using a computer controlled servo motor stage to perform controlled dip of the hydrogel into an acrylamide solution. The TEMED at the surface of the pHEMA hydrogel activates the polymerization of the acrylamide, which attach to the surface of the pHEMA hydrogel. By controlling the speed of the dip, a gradient polyacrylamide brush is applied to the surface of the pHEMA.

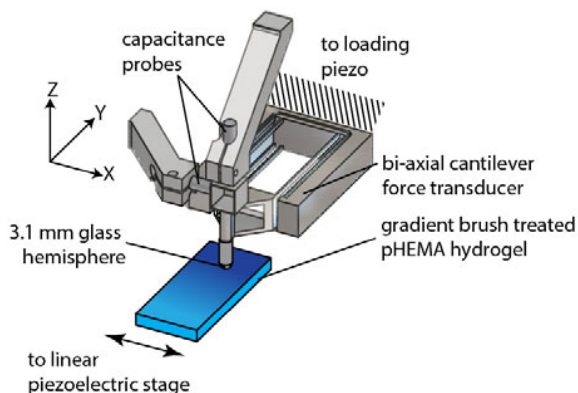


Figure 1. Microtribometer for friction force measurement on functionalized hydrogel surface.

Friction coefficient was measured using a custom microtribometer (Fig. 1) to evaluate lubricity of the material as a function of surface position (i.e. brush length). Externally applied normal loads ranging from 100 to 10,000 μN and a sliding velocity of 200 $\mu\text{m/s}$. Friction coefficients are calculated as the ratio of the measured tangential force (friction force) and the externally applied normal force. Friction coefficients and standard deviations are reported for each experiment.

Results: Polyacrylamide brushes dramatically affect the lubricity of the hydrogel surface. Friction coefficient was measured as a function of position down the coating dip-axis to confirm the presence of a gradient surface layer (Fig. 2). It was shown that friction coefficient decreased with increasing acrylamide exposure during the dip process. Friction coefficient varied from as low .047 under 10,000 μN at the thick brush end to as high as .096 at 2,500 μN at a position with minimal acrylamide exposure/growth time. Furthermore, the friction coefficient decreased with increasing normal force for all sample locations.

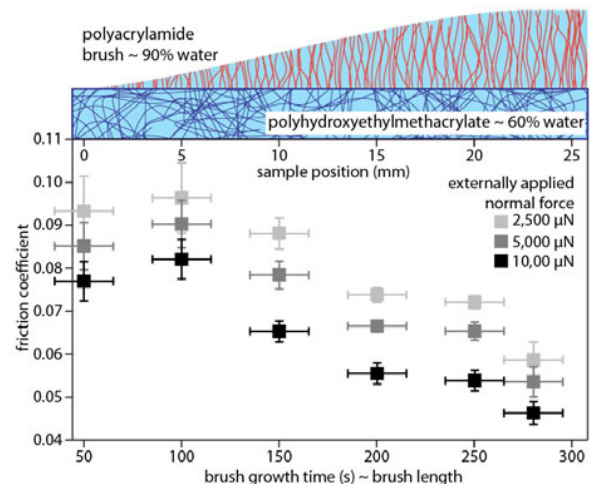


Figure 2. Friction coefficient along the gradient polyacrylamide brush sample. Brush growth time was varied linearly with position on the hydrogel.

Conclusions: Polyacrylamide surface functionalization can dramatically reduce the friction coefficient of pHEMA hydrogels. It is hypothesized that the polyacrylamides affinity for water allows for fluid support of the normal load at the interface, dramatically reducing the interaction between the polymer and the glass probe. By using a high precision dip coating method, gradient surface functionalization is achieved to create complex surface properties.

References: (1) Brennan NA. AAO Annual Meeting Abstr. 2009