Chemo-mechanics of biodegradable polymer networks

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Biodegradable hydrogels are hydrophilic polymer networks designed to break down after having fulfilled their intended function. The combined hydrophilicity and biodegradability make them very promising in biomedical applications such as drug delivery and tissue engineering. In many biodegradable hydrogels, the dominant degradation pathway is hydrolysis, which involves the scission of polymer chains by reaction of susceptible backbone bonds with water, leading to a reduction in mechanical properties. Conversely, stresses also impact the rate of hydrolysis. For example, stress-assisted hydrolysis can advance cracks in rubbery networks under small loads, which has been attributed to forces transmitted along the polymer chains, helping to reduce the energy barrier for hydrolysis [1].

In this work, we develop discrete and continuum modelling approaches to describe hydrolytic degradation coupled to mechanics in rubbery networks. At the mesoscale, we use random discrete networks [2,3] to describe the partitioning of forces along polymer chains in Representative Volume Elements of the network structure. Polymer chains are represented as entropic springs, and the equilibrium configuration of the network is determined from energy minimisation. The discrete mechanical model is coupled to a Kinetic Monte Carlo procedure to simulate stochastic force-accelerated chain scission by hydrolysis. The model is used to investigate degradation-induced swelling under free and constrained conditions, as well as hydrolytic crack growth. At continuum scale, we develop a micromechanics-based constitutive theory for coupled elasticity, swelling and degradation within a consistent thermodynamic framework [4]. A general implementation of the theory in FEM is proposed through Abaqus sub-routines. Predictions of the continuum and discrete models are compared.

References

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