Modelling the Chemomechanics of Hydrogels Gelation: Reactive-Diffusive Processes and Swelling-Shrinking Mechanisms

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ABSTRACT

Hydrogels are extensively used in additive manufacturing for biomedical applications. Because of their intelligent behaviour, biocompatibility and unique mechanical properties, they are ideal materials for producing scaffolds in tissue engineering applications or smart devices for drug delivery. Hydrogels can be stabilized by chemical gelation: an ionic stimulus (e.g., Calcium ions) induces the formation of cross-links in the polymer network as a consequence of diffusive-reactive mechanisms [1]. Gelation will produce inelastic deformations, in turn responsible for residual stresses, and will affect the mechanical properties, shape fidelity and structural resolution of the printed construct. This will affect, for instance, cell behaviour inside bioprinted scaffolds or active behaviors of smart devices.

This work presents a computational model for the chemomechanics of the gelation of alginate hydrogels. The system is described by introducing: elastic, shrinking and swelling deformation gradients; concentrations of fluid, free-alginate, gel and Calcium; the cross-linking degree. Swelling is associated with the movement of the fluid within the polymer network. Cross-linking is quantified from the chemical reaction of Calcium ions binding to alginate and it activates a re-organization of the polymer connectivity which is modeled as a source of shrinking deformation. A chemo-mechanical free energy couples the different mechanisms in a variational formulation [2, 3]. Constitutive laws are defined from thermodynamical principles. Balance of linear and angular momentum, as well as mass balance for the different molecular species, represent the solving equations of the coupled system. The effect of cross-linking on hydrogel permeability is also taken into account.

The developed computational framework allows to quantitatively predict stress distribution and polymer volume fraction inside the hydrogel as a function of internal chemical reactions and fluid movements. Numerical approaches for multiphysics mechanisms have been developed and implemented in a finite element framework, considering a monolithic coupling between chemical transport and mechanics. Benchmark tests are presented in order to verify the computational framework. The set-up of some available experimental tests is also virtually reproduced for the validation of obtained results. In particular, the importance of accounting for the coupling between the counteracting effects associated with fluid movements (inducing swelling) and cross-linking reactions (inducing shrinking) is highlighted.

REFERENCES

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