Unraveling the Mechanics of Tough Hydrogels: A Micro-to-Macro Perspective

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Abstract

It is widely accepted that the emerging mechanical properties of polymer-based materials can be explained by analysing the macromolecular composition, structure and arrangement. Complex polymeric networks are indeed the result of a range of strong and weak interactions which dictate the energy landscape. Models which are able to predict the macrostructure response as an interplay of simpler microscopic phenomena are highly valuable in that they allow for engineering new materials with tailored behaviour.

In the present investigation, we turn our attention to tough hydrogels, which are gaining importance in tissue engineering and replacement. Inheriting from an approach which has revealed its capability in predicting the complex mechanical response of single copolymer networks (1,2), we expand it to double network hydrogels. The methodological framework for our novel constitutive model consists in the following main ingredients: a model for the response of a representative ductile chain deduced from well known statistical mechanics observations; a corresponding micro-to-macro bridge based on the so-called microsphere homogenization; a constitutive model for the soft I2-based behaviour of a second network which is brittle.

The obtained model is validated against the to-date most comprehensive experimental dataset encompassing many deformation states and cyclic loading history. Despite a narrow number of physics grounded parameters, the model shows its versatility in reproducing finite elasticity and internal hysteresis. Furthermore, the clarity of its derivation allows for an understanding of the contributions from both networks, which could be linked to the synthesis process. This understanding can enhance material design, particularly for biomedical applications.

References

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