20th European Mechanics of Materials Conference - EMMC20
Florence, Italy | May 27th-29th, 2026
www.emmc20.unifi.it

Dehydrated hydrogels: when volume becomes shape-blind

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Paper ID: 143
Symposium S1: Mechanical behavior of polymers and biopolymers: theory, simulations and testing

Abstract

Hydrogels are soft, water-rich polymer networks with broad applications in medicine, bioengineering, and food science. Their mechanical behavior is generally taken as the combination of polymer elasticity, interstitial fluid pressure, and a chemically driven mixing pressure that captures the polymer-liquid affinity. Traditionally, their characterization and modelling have largely focused on fully hydrated states, treating dehydration as a secondary consequence of environmental exposure with no profound effects on the overall mechanics. As a result, the interplay between the main participating physical phenomena upon dehydration remains insufficiently characterized, which contrasts with the practical relevance of partially-to-highly dehydrated states. Basic physical reasoning already hints a mechanistic dominance in drying hydrogels, since the strong liquid-polymer affinity increasingly resists further fluid loss, while the high compliance cannot heavily penalize large shape distortions. This disparity thus leaves a basic question unresolved: do dehydrated hydrogels behave substantially different than fully hydrated ones? To address this issue, we combine experimental and theoretical analyses to determine how and when the volumetric response of dehydrating hydrogels becomes largely insensitive to shape deformations. Our findings show that this regime arises from a previously overlooked asymmetry: resistances to volumetric and shape-preserving (deviatoric) deformation scale very differently with drying. The bulk modulus, dominated by polymer-water affinity, increases sharply with water loss, whereas the shear modulus, governed by polymer elasticity, remains comparatively

small. We find that this imbalance appears even at mild dehydration and is largely insensitive to mechanical loading, indicating the broad relevance of the dominance regime. This separation of scales weakens the coupling between volumetric and deviatoric responses and enables significant simplification of the governing mechanics: the originally multiscale, strongly coupled problem can be recast as two sequential, weakly coupled subproblems. Conceptually, this demonstrates that polymer-water affinity governs hydrogel shrinkage, with the resulting shape being mostly set by a subsequent volume-constrained elastic deformation.

Keywords:
Hydrogel Dehydration Mixing pressure Shrinkage