Nonlinear viscoelasticity and generalized failure criterion for polymer gels

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Polymer gels find ubiquitous applications in material science, from biological tissues to manufactured goods, among which food stuffs and medical products are the most widespread [1]. These materials commonly feature a porous microstructure filled with water, which results in solid-like viscoelastic mechanical properties. While soft polymer gels share common features with hard materials, including delayed failure [2, 3], crack propagation [4, 5] or work-hardening, their porous microstructure also confers upon them remarkable nonlinear viscoelastic properties. Indeed, such soft solids strongly stiffen upon increasing deformation, which stems from the inherent nonlinear elastic behavior of the polymer chains and the structure of the gel network [6, 7]. Polymer gels hence endure large strains to failure and dissipate substantial mechanical work. However, to date no quantitative link has been made between the nonlinear viscoelasticity of polymer gels and the failure that is subsequently observed as the strain-loading is increased beyond the initial stiffening regime.

Here we apply the concept of a strain damping function, traditionally used for polymeric liquids and rubber-like materials [8] to quantify the nonlinear viscoelastic response of a prototypical protein gel. The form of the damping function $h(\gamma)$ is determined through a series of stress relaxation tests performed on freshly prepared gels that allow us to probe large deformations while injecting very little energy into the gel. The damping function $h(\gamma)$ is used to construct a time-strain separable constitutive equation of K-BKZ (Kaye–Bernstein-Kearsley-Zapas) form [9, 10] that predicts the gel mechanical response to arbitrary loading histories:

$$\sigma(t) = \int_{-\infty}^{t} G(t - t')h(\gamma)\dot{\gamma}(t')\mathrm{d}t' \qquad (1)$$

This approach captures the strain-stiffening of the gel during start up of steady shear tests up to the appearance of a stress maximum that is accompanied by the onset of the first macroscopic crack. Moreover, in order to link the nonlinear viscoelastic response of the gel to its subsequent brittle-like rupture, we adopt the failure criterion introduced by J. Bailey, which successfully describes the rupture of much stiffer materials such as glasses [11] and elastomeric-like materials [12]. This criterion assumes that the failure process is Markovian, i.e. the failure of the material results from the accumulation of local failure events that are independent [13]. The combination of the stress response predicted by the K-BKZ constitutive formulation with the Bailey criterion allows us to predict the scaling of the critical stress and strain at failure with the applied shear rate.

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