# Time-Strain Separability and Inseparability in Multiaxial Stress Relaxation of Polymer Gels with Permanent and Transient Crosslinks

Takuro Kimura[1], Takuma Aoyama[1], Masaki Nakahata,[2] Yoshinori Takashima,[2]
Motomu Tanaka,[3] Akira Harada,[2] and Kenji Urayama[4]

[1]Kyoto Institute of Technology, Kyoto, Japan
[2]Osaka University, Osaka, Japan
[3] Heidelberg University, Heidelberg, Germany
[2]Kyoto University, Kyoto, Japan

Introduction of transient physical cross-links into polymer gels with permanent cross-links has attracted much attention as an effective and useful method to enhance the viscoelasticity and mechanical toughness. Indeed, such polymer gels have significant viscoelasticity: they exhibit large degrees of stress relaxation in response to a step strain, a definite relaxation peak in linear dynamic viscoelasticity, and a considerable hysteresis in the stress-strain relationships under loading-unloading cycles. Separability of the time and strain effects on stress is a key to construct the constitutive models of viscoelastic materials, but the time-strain separability is not a rule derived from fundamental laws, it must be assessed experimentally.

We investigate the nonlinear stress relaxation for the two types of polymer gels with permanent and transient cross-links using biaxial stretching with various combinations of two orthogonal strains as an imposed strain. Such general biaxial strain covers a wide range of physically accessible deformations, while conventional uniaxial stretching is only a particular deformation among them. Thus the biaxial stress relaxation data provide a definite basis to access the time-strain separability. We have validated unambiguously the time-strain separability for the dual cross-link poly(vinyl alcohol) (PVA) hydrogels[1)] with covalent bonds and the transient bonds via borate ions.[2)] In contrast, we observe that the time and strain effects are not separable in the supramolecular gels[3)] with permanent rotaxane cross-links utilizing the host-guest interactions as the transient cross-links.[4)] The relaxation is accelerated in a short time region by an increase in the magnitude of strain, whereas it is retarded in a longer time region by an increase in the anisotropy of the imposed biaxial strain.

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