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Effects of two scaling exponents on biaxial deformation and mass transport of swollen elastomers

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ABSTRACT

In this study, we investigate the effects of two scaling exponents on the biaxial deformation and mass transport of swollen elastomers. Two scaling exponents are included in an extended version of the Flory–Rehner model (Okumura et al., J. Mech. Phys. Solids, 2016); two scaling exponents are used to adjust the swelling effects on the Young's modulus and osmotic pressure of swollen elastomers, resulting in the ability to predict swelling-induced strain softening under uniaxial tensile loading. It is found that when biaxial tensile loading is given under stress control, strain softening is accelerated by increasing the biaxial stress ratio, while under strain control, the responses become more complicated, which can be interpreted by considering that Poisson's ratio at equilibrium free swelling can take negative values depending on the two scaling exponents. Further, the effect of the two scaling exponents on mass transport is discussed by analyzing the swelling kinetics of a gel layer constrained on a rigid substrate.

1. Introduction

The Flory–Rehner (F–R) model [1] is used to describe the mechanical and swelling behavior of elastomers [2,3]. The free energy function consists of the sum of two free energies associated with polymer stretching and the mixing of polymer and solvent molecules, which are derived from the Gaussian network theory (i.e., a Neo–Hookean solid model) and the Flory–Huggins solution theory, respectively. When the F–R model is assumed, the boundary value problem of swollen elastomers is equivalent to that of a hyperelastic solid [4,5], so that the F–R model is easily implemented into commercial finite element software using user-defined material subroutines. These subroutines allow researchers to perform finite element analysis of the swelling-induced surface instability of hydrogel films [6,7] and swelling-induced pattern transformation in porous gel films [4,8–11]. Further, solvent migration in the transient state can also be analyzed by introducing a diffusion model into the governing equations [12–18]. Thus, the F–R model provides a basis to interpret the mechanical behavior of swollen elastomers in both static and transient states, but it is not free from criticism.

Neo–Hookean solid models may be a poor choice to predict the stress-strain behavior of elastomers, especially at large strain and/or under biaxial deformation. When swollen elastomers are subjected to uniaxial loading, Neo–Hookean solid models predict that $E = E_{\mathrm{d}} J^{-1/3}$ and $\sigma_{\mathrm{r}} = E_{\mathrm{d}} / 3$, where E_{d} and E are the Young's moduli of the dry and swollen state, respectively, J is the volume swelling ratio, and σ_{r} is a transformed stress referred to as the swelling reduced stress [19,20]. However, experiments show that $E = E_{\mathrm{d}} J^{l}$ has various values of l depending on the elastomer, and σ_{r} is not constant but a function of J, and, especially under tension, σ_{r} is also affected by stretching in the loading direction [21–23]. To overcome these problems, a Neo–Hookean solid model in the F–R model can be superseded by advanced solid models, e.g., the Mooney–Rivlin, Arruda–Boyce, and Ogden models [2,3,13]. Flory and Erman [24] proposed the constrained chain model, in which a Neo–Hookean solid model was modified by adding terms to describe constrained chains. Drozdov and Christiansen [25,26] derived an equivalent expression of this model using stretch invariants and developed extended models by including phenomenological parameters to correctly predict the mechanical response of hydrogels under multi-axial deformation. Generally, advanced models involve complicated strain energy functions with a substantial number of phenomenological parameters. If the parameters are fitted to a particular set of mechanical responses, there may be no guarantee that other responses can also be correctly predicted using the same parameters.

As an extended version of the scaling approach [27,28], Okumura et al. [29] extended the F–R model using two scaling exponents, m

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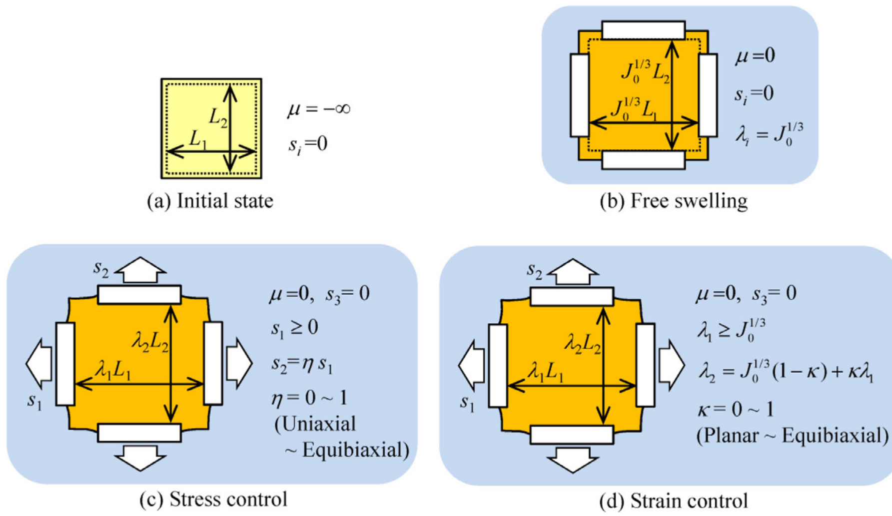

Fig. 1. Schematic illustration of biaxial tensile loading of an elastomer in a solvent; (a) initial state, (b) equilibrium free swelling, (c) stress control, and (d) strain control.

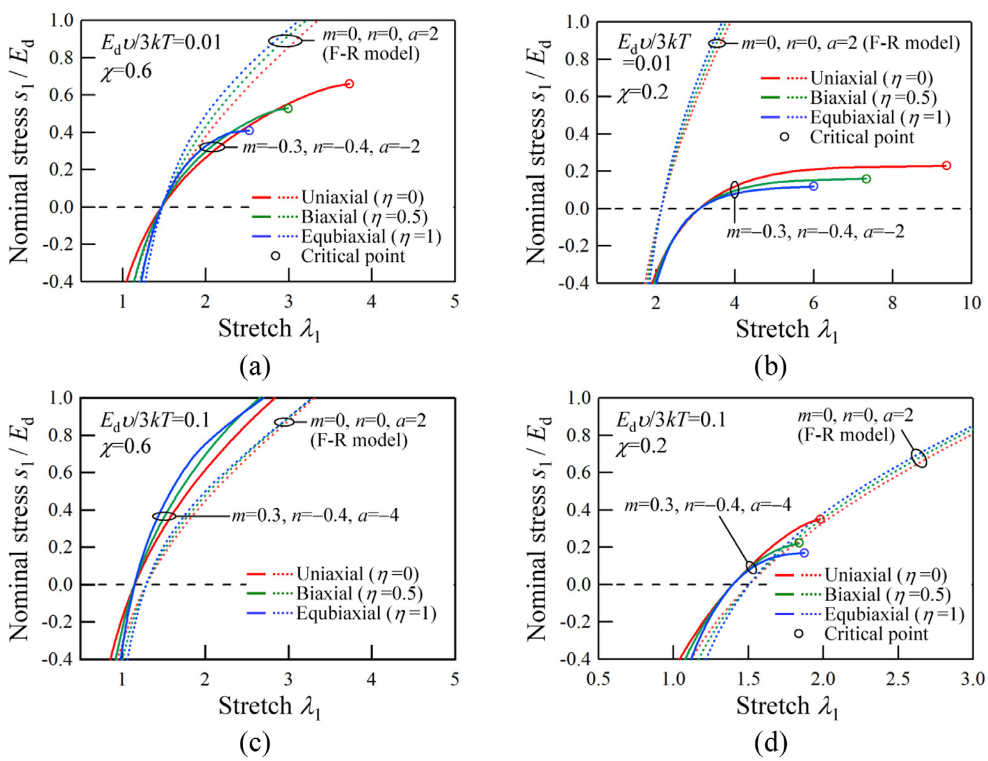

Fig. 2. Stress-stretch responses at equilibrium swelling under uniaxial loading ($s_2 = 0$), biaxial loading ($s_2 = 0.5\, s_1$) and equibiaxial loading ($s_2 = s_1$); (a) $E_d\upsilon/(3kT) = 0.01$ and $\chi = 0.6$, (b) $E_d\upsilon/(3kT) = 0.01$ and $\chi = 0.2$, (c) $E_d\upsilon/(3kT) = 0.1$ and $\chi = 0.6$, and (d) $E_d\upsilon/(3kT) = 0.1$ and $\chi = 0.2$.

and n. The two scaling exponents, which are introduced into strain energy functions separated into deviatoric and volumetric components, are used to independently adjust volumetric and deviatoric elastic contributions, respectively. If $m = n = 0$, the extended model reduces to the original F–R model. In contrast, when the two exponents are adjusted based on experimental data, m spans a wide range of values depending on the elastomer, while n is a negative value that is almost independent of the elastomer. Consequently, the extended model successfully reproduces the effects of swelling on the Young's modulus and osmotic pressure of swollen elastomers, respectively. Further, under uniaxial tensile loading at equilibrium swelling, the extended model is able to predict strain softening, which is thus related with strain localization followed by swelling-induced rupture. This prediction is in good agreement with the tendency observed by experiments with natural rubbers [30]; swelling-induced rupture can occur when a small extension is applied in good solvents. This means that the two scaling exponents enable the extended model to obtain a special ability to predict swelling-induced rupture, resulting in elucidating the mechanism causing swelling-induced rupture of swollen elastomers.

Recently, Okumura and Mizutani [31] analyzed swelling-induced strain softening under equibiaxial and planar extensions using the two scaling exponents. They showed that under equibiaxial extension, the tensile stress in a lateral direction enables swelling-induced strain softening to occur in relatively poor solvents and accelerates the onset point. Under planar extension, a compressive stress in the constrained direction can occur in good solvents, preventing the elastomer from causing swelling-induced strain softening. The effects of two scaling exponents are fairly complicated under biaxial deformation. In addition, when a