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A phenomenological model for dynamic response of double-network hydrogel composite undergoing transient transition

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Abstract

We present a phenomenological model for dynamic deformation and mechanical response of double-network (with short-chained ionic network and long-chained covalent network) hydrogel composite based on theory of transient networks. Molecular structures and stress-strain relations of the hydrogel composite were investigated based on thermomechanical properties of the individual network. Constitutive relations were derived for its nonlinear viscoelastic responses and annihilation/reformation rates of active short chains were determined by means of Eyring formula. An extended Volokh model was proposed to separate effects of large strain hysteresis and anomalous viscoelastic relaxation on the hydrogel composite after strain reversal. Experimental results from rate-independent tests are well in agreement with that of the numerical simulations. This study

provides a fundamental simulation tool for modelling and predicting mechanics and mechanisms of viscoelastic response and mechanical responses in double-network hydrogel composite.

1. Introduction

Hydrogels are the embodiment of "soft matter" as they combine the chemomechanical complexity and rheological flexibility [1-3], of which enable them a large entropy change in response to the external stimuli, similar with dielectric elastomer [4,5] and shape memory polymer [6-12].

Hydrogels are a unique class of soft polymeric materials which exhibit nonlinear mechanical responses to external loading with superior stretchability [13]. They have been widely investigated for numerous engineering applications in biomedicine, sensors, actuators due to their excellent properties including large amount of water absorption, high elastic strain, low weight and biocompatibility [14]. Gong and co-workers proposed and synthesized double-network hydrogel composites with both high stiffness and strength compared to those of the conventional single-network hydrogel [15-18]. The double-network hydrogel composites are combined with an ionic network with short chains and a covalent network with long chains [19]. When they are stretched, the ionic network ruptures, thus resulting in a great deal of energy dissipated (a large stress-strain hysteresis) and hydrogels are therefore toughened [20,21]. Whereas during unloading after stretching, the elastic long chains make the hydrogels return back to its initial state owing to the configurational entropy, thus realizing the large stretchability of hydrogels, this process is vividly shown in figure 1(a). In the double-network, the ionic links are adaptive and the rates of annihilation and reformation follow the Eyring formula [22]. For a double network hydrogel composite, it is well-documented that configurational entropy can be neglected and the breakage of the ionic links is

sensitive to strain-rate during the loading process [23,24]. However, the mechanical response is not sensitive to the strain-rate because there is no breakage of the adaptive links during the unloading process. Furthermore, the ionic crosslinks without breakage are in process of the elastic stretching [25], this process is displayed in figure 1(b).

In this study, a phenomenological model was proposed to study the synergistic effect of ionic network and covalent network on the dynamic mechanical responses of the double-network hydrogel composites based on the theory of transient networks. The molecular structures and stress-strain relations of the double-network hydrogel composites were investigated based on the thermomechanical properties of the individual networks. The nonlinear viscoelastic and mechanical responses of the ionic network was initially modeled based on the theory of transient networks. An extended volokh model was then proposed to separate the effects of large strain hysteresis and anomalous viscoelastic relaxation on the covalent network. Finally, phenomenologically constitutive relations were established to explore the working mechanism and predict the mechanical behavior of the double-network hydrogel composites.

2. Modelling for ionic network

According to the theory of transient network, the viscoelastic ionic network can be treated as a transient network made from short chains and adaptive links [26]. All the short chains are repeatedly connected via the same adaptive links [27-29]. It is assumed that the short chains of ionic networks lead to annihilation even at a small tensile strain. Therefore, the mechanical response of the ionic network is viscoelastic.

We use the function $\Lambda(t,\vec{l})$ to represent the number of adaptive links with respect to the unit vector $\vec{l}$ (per unit mass) existed at time t. The quantity of breakage $\tilde{\Gamma}(t,\vec{l})$ within a constant interval

time equals to,

$$\tilde{\Gamma}(t,\vec{l}) = -\frac{\partial \Lambda}{\partial t}(t,\vec{l}) \tag{1}$$

Here, the constant $\chi(\vec{l})$ represents the proportion of permanent links in the ionic network, i.e.:

$$\tilde{\Lambda}(\vec{l}) = \Lambda(0,\vec{l}) \tag{2}$$

where $\Lambda(0,\vec{l})$ is the number of links (peer unit mass) at $t=0$ with respect to the stretching direction $\vec{l}$. Therefore, the adaptive links exist at time t can be expressed by:

$$\Lambda(t,\vec{l}) = \chi(\vec{l})\,\tilde{\Lambda}(\vec{l}) + Z(t,\vec{l}) \tag{3}$$

where $Z(t,\vec{l})$ represents the content of the remaining adaptive links at t. The rate of breakage is a function of adaptive links, i.e.:

$$\Gamma(t,\vec{l}) = \frac{\tilde{\Gamma}(t,\vec{l})}{Z(t,\vec{l})} = b \tag{4}$$

where b is the breakage rate of the ionic network. Different from the covalent network, the breakage rate of the ionic network is much larger and more sensitive to the tensile rate [21,25]. Here, we can get the differential of $Z(t,\vec{l})$ by combining equations (1), (3) and (4),

$$\frac{\partial Z}{\partial t}(t,\vec{l}) = -b \cdot Z(t,\vec{l}) \tag{5}$$

At $t=0$, the initial condition is $Z(0,\vec{l}) = \tilde{\Lambda}(\vec{l})[(1-\chi(\vec{l})]$, therefore, we can obtain:

$$Z(t,\vec{l}) = \tilde{\Lambda}(\vec{l})[1-\chi(\vec{l})]\exp(-bt) \tag{6}$$

By substituting the equation (6) into the equation (3), we can obtain:

$$\Lambda(t,\vec{l}) = \tilde{\Lambda}(\vec{l})\{\chi(\vec{l}) + [1-\chi(\vec{l})]\exp(-bt)\} \tag{7}$$

Here, the function $w(t,\vec{l})$ is introduced to express the average potential energy of each link of the ionic network with respect to the vector $\vec{l}$, therefore, we can obtain the potential energy of the ionic network (per unit mass) as:

$$W(t) = \int_S \Lambda(t,\vec{l})w(t,\vec{l})f(0,\vec{l})dA(\vec{l}) \tag{8}$$

where $f(0,\vec{l})$ denotes the density of the ionic link, S is the boundary of a unit sphere with respect to the vector $\vec{l}$. As the ionic networks are considered to be isotropic [19], both the functions $\tilde{\Lambda}(\vec{l})$ and $\chi(\vec{l})$ are independent of the guiding vector, and the density of the adaptive link becomes a constant,

$$f(0,\vec{l}) = a \tag{9}$$

We use spherical angles φ and ϑ to locate the vectors in the Cartesian coordinate. $\vec{e}_i$ is the unit vector. Accordingly, we can obtain:

$$\vec{l} = \cos\vartheta \vec{e}_1 + \sin\vartheta(\cos\varphi \vec{e}_2 + \sin\varphi \vec{e}_3) \tag{10}$$

$$dA = \sin\vartheta d\vartheta d\varphi. \tag{11}$$

Here, we can further write the total equations by substituting the equations (7), (9) and (11) into (8),

$$W(t) = a\int_0^\pi \sin\vartheta d\vartheta \int_0^{2\pi} [\chi + (1-\chi)\exp(-bt)]w(t,\vec{l})d\varphi \tag{12}$$

The nominal strain of an adaptive link in the form of Cartesian coordinate system frame has the following form [27]:

$$\varepsilon_* = \vec{l}\cdot\hat{\varepsilon}(t)\cdot\vec{l} = \varepsilon_1\cos^2\vartheta + \sin^2\vartheta(\varepsilon_2\cos^2\varphi + \varepsilon_3\sin^2\varphi) \tag{13}$$

where $\hat{\varepsilon}(t)$ denotes the strain tensor, which represents the transition from the initial configuration to the current configuration at t. ε_i represents the eigenvalues of the tensor $\hat{\varepsilon}$.

In the following, we will consider the mechanical energy of a nonlinear elastic spring for the constitutive stress-strain relation of the ionic network in hydrogel, which can be written as:

$$w(t,\vec{l}) = \tilde{w}(\varepsilon_*(t,\vec{l})) = \tilde{w}(\vec{l}\cdot\hat{\varepsilon}\cdot\vec{l}) \tag{14}$$

Because the strain of ionic network is small, we can assume that the potential energy of the ionic network obeys the power-law stress-strain principle [26]:

$$\tilde{w}(\varepsilon_*) = \frac{K}{1+k}|\varepsilon_*|^{1+k} \tag{15}$$

where K and k both denote adjustable parameters.

So substituting the equations (13) and (15) into (14), we can obtain the average potential energy of an adaptive link in the ionic network,

$$\vec{w}(t,\vec{l}) = \tilde{w}(\vec{l} \cdot \hat{\varepsilon} \cdot \vec{l}) = \frac{K}{1+k}[\varepsilon_1(t)\cos^2 \vartheta + \sin^2 \vartheta(\varepsilon_2(t)\cos^2 \varphi + \varepsilon_3(t)\sin^2 \varphi)]^{1+k} \tag{16}$$

Here, we have calculated the average potential energy $\overset{\text{ш}}{w}(t,\vec{l})$, so the modified mechanical energy of the viscoelastic ionic network can be rewritten as:

$$W(t) = a\tilde{\lambda}\int_0^\pi \sin \vartheta d\vartheta \int_0^{2\pi} [\chi + (1-\chi)\exp(-bt)] \cdot \tilde{w}(\vec{l} \cdot \hat{\varepsilon}(t) \cdot \vec{l})d\varphi \tag{17}$$

Differentiating equation (17) with respect to time t, we obtain the following equation:

$$\frac{dW}{dt}(t) = a\tilde{\lambda}\int_0^\pi \sin \vartheta d\vartheta \int_0^{2\pi} \{[\chi + (1-\chi)\exp(-bt)] \cdot \frac{\partial \tilde{w}}{\partial \hat{\varepsilon}(t)}(\vec{l} \cdot \hat{\varepsilon}(t) \cdot \vec{l})d\varphi\} : \frac{d\hat{\varepsilon}}{dt}(t)$$
$$-ab\tilde{\lambda}(1-\chi)\int_0^\pi \sin \vartheta d\vartheta \int_0^{2\pi} \exp(-bt) \cdot \tilde{w}(\vec{l} \cdot \hat{\varepsilon}(t) \cdot \vec{l})d\varphi \tag{18}$$

Considering the thermodynamic potential of the ionic network without the effect of thermal expansion, we can use the following equation [26],

$$\frac{dW}{dt} = \frac{1}{\rho}\hat{s} : \frac{d\hat{e}}{dt} - \frac{dQ}{dt} \tag{19}$$

where Q denotes the specific dissipation of energy in the ionic network per unit mass and $\hat{s}$ denotes the deviatoric part of $\hat{\sigma}$. The purpose of this article now is to derive constitutive equation for an incompressible medium, this condition implies that the first invariant of the strain tensor $\hat{\varepsilon}$ vanishes, so $\hat{\varepsilon}(t) = \hat{e}(t)$.

By combining equations (18) and (19), the deviatoric part of $\hat{\sigma}$ has the form:

$$\hat{s}(t) = a\rho\tilde{\lambda}\int_0^\pi \sin \vartheta d\vartheta \int_0^{2\pi} [\chi + (1-\chi)\exp(-bt)] \cdot \frac{\partial \tilde{w}}{\partial \hat{\varepsilon}(t)}(\vec{l} \cdot \hat{\varepsilon}(t) \cdot \vec{l})]d\varphi \tag{20}$$

and,

$$\frac{dQ(t)}{dt} = ab\,\tilde{\lambda}(1-\chi)\int_0^\pi \sin\vartheta d\vartheta \int_0^{2\pi} \exp(-bt) \cdot \tilde{w}(\vec{l} \cdot \hat{\varepsilon}(t) \cdot \vec{l})d\varphi \tag{21}$$

3. Modelling for covalent network

Volokh model is also introduced here to characterize the mechanical response of covalent network [30-33], which can be described by [30]:

$$\sigma = 2J^{-1}\mathbf{F}\frac{\partial\psi}{\partial C}\mathbf{F}^T \tag{22}$$

Where ψ is the strain energy, tensor σ denotes the Cauchy stress tensor and $\mathbf{F}$ denotes the deformation gradient of the tensor. The strain energy ψ is a function of W (the strain energy of intact material), and their relationship can be written as follows [31]:

$$\psi(W) = \Phi - \Phi\exp(-W/\Phi) \tag{23}$$

where function Φ denotes the maximum strain energy of the covalent network without failure in a infinite volume.

The Mooney-Rivin mode is further applied to model the elastic covalent network in a uniaxial tensile loading [25]. For the isotropic network, I_1, I_2 and I_3 are set as the invariants of the right Cauchy-Green tensor,

$$I_1 = tr\mathbf{c} = c_{ii} \tag{24}$$

Accordingly, we can write the strain energy of the intact material as,

$$W = W(I_1) = \frac{\alpha}{2}(I_1 - 3), \tag{25}$$

where α is a constant. Substituting equations (23) and (25) into (22), we can get the final constitutive equation of the covalent network as follows [30],

$$S = \alpha(\lambda^2 - \lambda^{-1})\exp\frac{-\alpha(\lambda^2 + 2\lambda^{-1}-3)}{2\Phi} \tag{26}$$

Figure 2 shows the stress-strain response of a single-network under a uniaxial loading predicted by the theoretical model according to equation (26), by assuming $\alpha = 0.67$ and $\Phi = 18.82$. A comparison is made between the experimental data obtained from reference [25] and the fitting plots using equation (26). It is found that the model well predicts the stress-strain relation of the single-network hydrogel. The theoretical results are very close to the experimental results [25].

4. Simulation and prediction of stress-strain hysteresis of double-network hydrogel composite

If the hydrogel is assumed incompressible, the strain tensor $\hat{\varepsilon}$ will change according to its deviatoric part $\hat{e}$. Therefore, we can write the strain tensor $\hat{\varepsilon}$ and the deviatoric part of the tensor $\hat{e}$ as follows [27], and $s_0(t)$ represents the deviatoric part of $\hat{s}$ which has the same form:

$$\hat{\varepsilon}(t) = \varepsilon_0(t)\vec{e}_1\vec{e}_1 - \frac{1}{2}\varepsilon_0(t)(\vec{e}_2\vec{e}_2 + \vec{e}_3\vec{e}_3) \tag{27}$$

$$\hat{s}(t) = s_0(t)\vec{e}_1\vec{e}_1 - \frac{1}{2}s_0(t)(\vec{e}_2\vec{e}_2 + \vec{e}_3\vec{e}_3) \tag{28}$$

Substituting equation (27) into (16), we can obtain:

$$\tilde{w}(\vec{l}\cdot\hat{\varepsilon}(t)\cdot\vec{l}) = \frac{K}{1+k}\left|\varepsilon(t)(\cos^2\vartheta - \frac{1}{2}\sin^2\vartheta)\right|^{1+k} \tag{29}$$

and the corresponding partial derivative equation of $\tilde{w}(\vec{l}\cdot\hat{\varepsilon}(t)\cdot\vec{l})$ is,

$$\frac{\partial\tilde{w}}{\partial\varepsilon(t)}(\vec{l}\cdot\hat{\varepsilon}(t)\cdot\vec{l}) = K\varepsilon(t)^k\left|\cos^2\vartheta - \frac{1}{2}\sin^2\vartheta\right|^{1+k} \tag{30}$$

Combing equations (20), (28) and (30), we can obtain the function of $s_0(t)$ as follows:

$$s_0(t) = 2\pi a\rho\tilde{\Lambda}K\int_0^\pi \sin\vartheta[\chi + (1-\chi)\exp(-bt)]\varepsilon(t)^k\left|\cos^2\vartheta - \frac{1}{2}\sin^2\vartheta\right|^{1+k}d\varphi. \tag{31}$$

In a uniaxial loading, the stress functions are presented as follows,

$$\sigma = \text{-p} + s_0, \quad \sigma_2 = \sigma_3 = -p - \frac{1}{2}s_0 = 0 \tag{32}$$

where σ represents the external stress in the loading direction, σ_2 and σ_3 represent the two external stresses which are perpendicular to the σ, and p is the atmospheric pressure.

Therefore, we can obtain the following equation for the stress:

$$\sigma(t) = \frac{3}{2}s_0(t) = 3\pi a\rho\,\tilde{\Lambda}\,K\int_0^\pi \sin\vartheta[\chi + (1-\chi)\exp(-bt)]\varepsilon(t)^k \left|\cos^2\vartheta - \frac{1}{2}\sin^2\vartheta\right|^{1+k} d\varphi \qquad (33)$$

It is assumed that deformation of the ionic networks has a linear relationship with the deformation of the hydrogels,

$$\lambda - 1 = \beta\varepsilon \quad t = (\lambda - 1)/\dot{\lambda} \qquad (34)$$

where λ denotes the axial stretch (dimensionless strain) of the hydrogels , ε denotes the small strain of the ionic network, $\dot{\lambda}$ represents the stretch rate and β is a constant. A constant of z is introduced here to simplify the equation (33) as follows:

$$z = 3\pi a\rho\,\tilde{\Lambda}\,K\int_0^\pi \sin\vartheta\left|\cos^2\vartheta - \frac{1}{2}\sin^2\vartheta\right|^{1+k} d\varphi / \beta^k \qquad (35)$$

Substituting equations (34) and (35) into (33), in dimensionless form of strain, the constitutive relation of the stress-stretch of double-network hydrogel composite can be rewritten as:

$$S = z\{\chi + (1-\chi)\exp[-b(\lambda-1)/\dot{\lambda}]\}(\lambda-1)^k \qquad (36)$$

where the stress S is the new expression of σ when the strain is in dimensionless form.

To analyze the mechanical response of the ionic networks, the parameters of $\chi = 0$, $z = 29.69$ and $k = 1.33$ were determined from the experimental results [25]. The breakage rates of short ionic chains as a function of strain rate are summarized in Table 1. A comparison between the experiment results [25] and simulation results (derived from equation (36)) is presented in Figure 3(a). It is found that the mechanical response of the double-network is sensitive to the strain rate, and a higher stress is needed to enable the double-network to reach a constant value of stretching extension ratio

with an increased strain rate. A standard deviation of the experimental results from the simulation results is plotted in Figure 3(b). It is found that the deviation of the simulation and experimental results is in a range from -10% to 15%. This indicates that the model is reasonably good to describe the experiment data, and well predicts the constitutive relation and mechanical response of the double-network hydrogel composite.

The effects of strain rate on the constitutive stress-strain relation and mechanical response were further studied. As shown in Figure 4, a higher stress is needed to stretch the network with a higher values of stretching extension ratios (where strain rate is increased from 1/min, 5/min, 10/min to 100/min). These simulation results confirm that the constitutive relation of the stress-strain of the double network hydrogel is sensitive to the strain rate.

The proposed model is further used to analyze the mechanical response of the double-network in the uniaxial unloading process. In the double-network, the ionic links are adaptive and the rate of reformation obeys the Eyring formula [27]. The reformation process obeys the similar rule of the annihilation. Therefore, Equation (36) could be rewritten as,

$$S = S_0 - m\{\chi + (1-\chi)\exp[-c \cdot (\lambda - 1)]\}(\lambda - 1)^k \tag{37}$$

where S_0 denotes the initial stress in the unloading process and the final stress in the loading process, m is a given constant to present the number of ionic links and determined by the strain rate, and c is also a given constant and is related with the stain parameter.

According to the experiment results, it is obtained that $\chi = 0$, $c = 0.19$ and $k = 0.60$ [25]. At a strain rate of 0.4/min, it is found that $s_0 = 35.4$ and $m = 33.1$. At a strain rate of 2.2/min, it is found that $s_0 = 40.4$ and $m = 37.48$. At a strain rate of 8.8/min, it is found that $s_0 = 43.8$ and $m = 42.1$. A comparison between the numerical and experimental results of the stress as a function of the

stretching ratio for the double-network hydrogel composite is plotted in Figure 5(a), in which the data were obtained using the equation (37), with given strain rates of 0.4/min, 2.2/min and 8.8/min, respectively. It is found that the stress gradually decreases with an increase in the strain, mainly due to the annihilation of the ionic network. The working mechanism behind is that the annihilation reduces the contribution of the ionic network to resist the external stress. The simulation results are well agreement with the experimental ones [25]. A deviation of simulation and experimental results is presented in Figure 5(b), which reveals that the stress deviation is limited to 0.15 kPa with a strain increased from 1 to 4.25, and the strain rate is increased from 0.4/min to 8.8/min.

Effect of number of ionic links (m) on the constitutive stress-strain relation is presented in Figure 6, with given numbers of ionic links of m =10, 20, 30, 37.48, 40, 50 and 60, respectively. The initial stress is given as s_0 =40.4. Simulation results reveal that a larger stress is needed to achieve a constant value of strain with an increase in the number of ionic links. It is implied that the contribution of the ionic network to resist to the external stress becomes more significant with a larger number of the ionic links. The simulation results are in agreement with the experimental ones.

Annihilation and reformation of the short chains (or ionic links) have been discussed above. However it is still necessary to characterize and predict the overall response of the double-network hydrogel composite under a cycle of mechanical loading. At a given strain rates of 0.4/min and 35.3/min, annihilation and reformation models are combined to characterize the constitutive stress-strain relation, and the results are shown in Figure 7. Clearly the simulation results are in a well agreement with the experimental ones, and the models are reasonably good to predict the mechanical response of the double-network hydrogel composite. Furthermore, it is also applicable to combine the two models to simulate and predict the mechanical response at various strain rates.

5. Conclusions

We proposed an explicit model to study the mechanics and mechanical response of the double-network hydrogel composites based on the transient theory. Initially, the annihilation and reformation of ionic network were both modelled on the origin of Eyring rule for the hydrogel with reversible large deformation. Two constitutive stress-strain relations were found to be suitable to describe and predict the mechanical responses in double-network hydrogel composite in terms of the mechanical properties of the ionic network (with short chains) and covalent network (with long chains). Furthermore, a combined model was proposed to characterize the overall mechanical response and constitutive stress-strain relation in hydrogels induced by both annihilation and reformation of ionic network. The simulation by the proposed model was compared with the experimental results reported in the literature. The simulation results were well agreement with and close to the experimental results. This study is therefore expected to provide an effective way to estimate and model the mechanical behavior of the hydrogels.

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Table caption

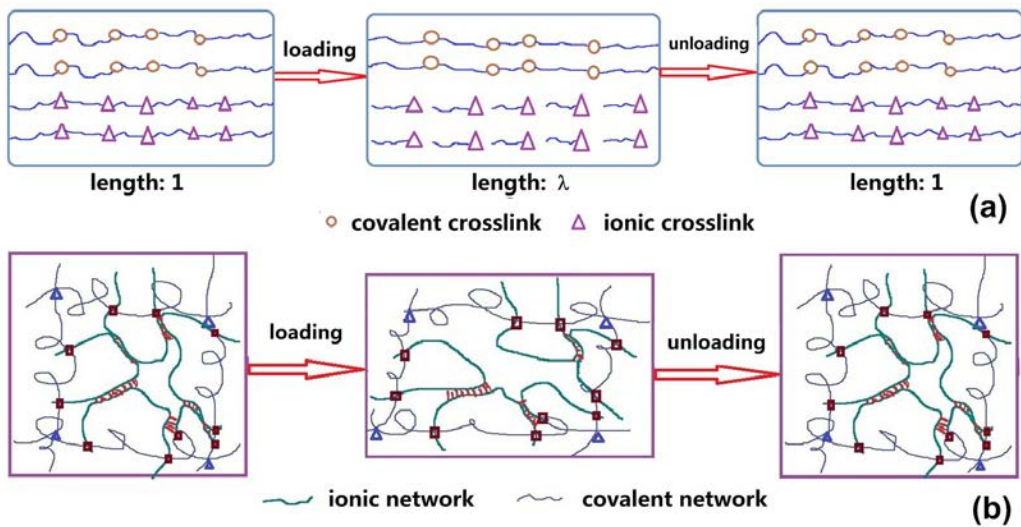**Table 1**. The breakage rate at a variety of tensile strain rate (1/min)

Figure 1. (a) Dissipation much energy by the breakage of the ionic crosslinks and the configurational entropy of the covalent network makes the hydrogel return to the initial state. (b) The change of the ionic network during the loading and unloading process: some of the ionic crosslinks break, the other is in elastic process. Meanwhile, the elastic long chains have a big deformation during the loading process and return to its initial state during the unloading process.

Figure 2. Numerical simulation for the stress as a function of strain under a uniaxial loading and at the stretch rate of 8.8/min in the single-network hydrogel.

Figure 3. (a) A comparison between the simulation and experimental results of the stress-strain of the double-network as a function of extension ratio under a constant strain rate of 0.4/min, 2.2/min, 8.8/min and 35.3/min, respectively. (b) The deviation of simulation and experimental results at a variety of strain rate.

Figure 4. Numerical simulation for the relation between the stress of the double-network as a function of extension ratio under a constant strain rate of 1/min (where b=0.41), 5/min (where b=1.85), 10/min (where b=3.4) and 100/min (where b=28), respectively.

Figure 5. (a) Comparison of the experiment data with the model curve at different rates. The triangle points are the experiment data and the three lines are the model curves. This figure also shows little sensitive to the rate and a large hysteresis during the unloading process. (b) The deviation degree of every data at different tensile rate is shown in this figure. We can see the deviation degree of the data is in the range from -0.15 to 0.15 kPa. This model commendably accord with the experiment data.

Figure 6. The influence of the parameters m: (a) when the value of the parameter m is less than the true value, the stress changes less value with the strain. (b) when the value of the parameter m is

more larger than the true value, the stress changes more value with the strain.

Figure 7. A comparison between the simulation and experiment results [12] of the stress-strain relation for the double network hydrogel at the strain rate of 0.4/min and 35.3/min.

Table 1. The breakage rate at a variety of tensile strain rate (1/min)

$\dot{\lambda}$ /min	35.3	8.8	2.2	0.4
b	11.299	3.140	0.871	0.177

Figure 1. (a) Dissipation much energy by the breakage of the ionic crosslinks and the configurational entropy of the covalent network makes the hydrogel return to the initial state. (b) The change of the ionic network during the loading and unloading process: some of the ionic crosslinks break, the other is in elastic process. Meanwhile, the elastic long chains have a big deformation during the loading process and return to its initial state during the unloading process.

Figure 2. Numerical simulation for the stress as a function of strain under a uniaxial loading and at the stretch rate of 8.8/min in the single-network hydrogel.
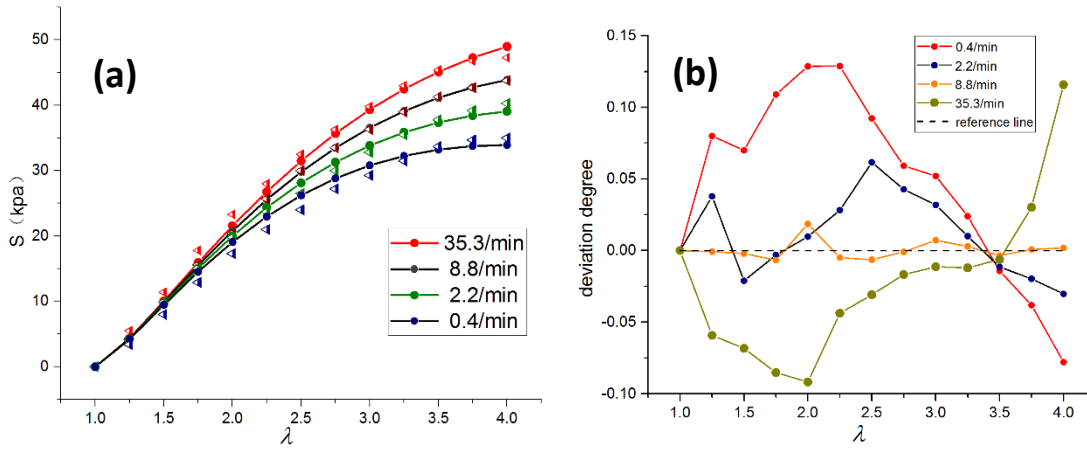
Figure 3. (a) A comparison between the simulation and experimental results of the stress-strain of the double-network as a function of extension ratio under a constant strain rate of 0.4/min, 2.2/min, 8.8/min and 35.3/min, respectively. (b) The deviation of simulation and experimental results at a variety of strain rate.
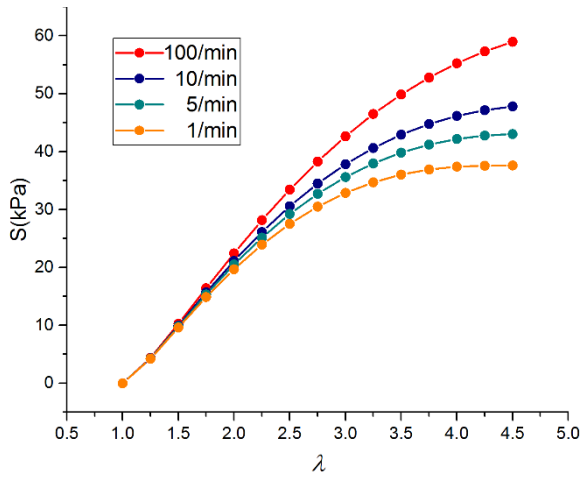
Figure 4. Numerical simulation for the relation between the stress of the double-network as a function of extension ratio under a constant strain rate of 1/min (where b=0.41), 5/min (where b=1.85), 10/min (where b=3.4) and 100/min (where b=28), respectively.
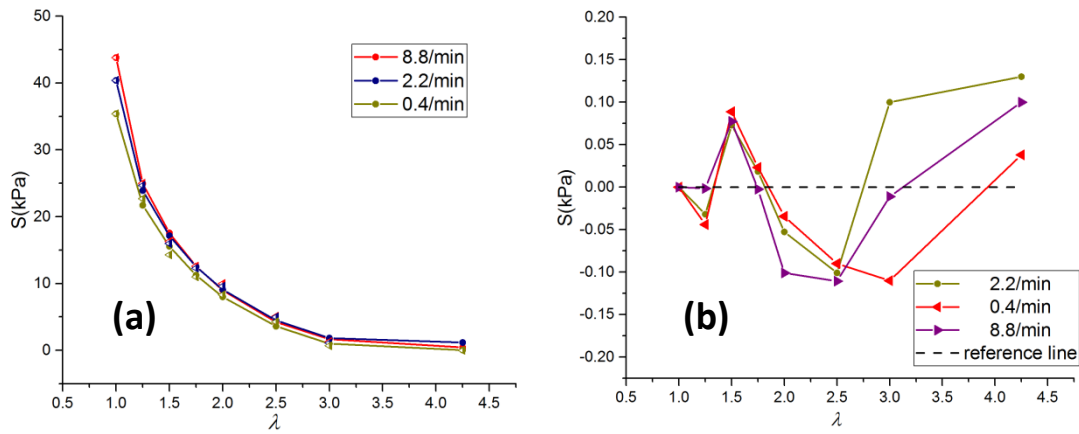
Figure 5. (a) Comparison of the experiment data with the model curve at different rates. The triangle points are the experiment data and the three lines are the model curves. This figure also shows little sensitive to the rate and a large hysteresis during the unloading process. (b) The deviation degree of every data at different tensile rate is shown in this figure. We can see the deviation degree of the data is in the range from -0.15 to 0.15 kPa. This model commendably accord with the experiment data.
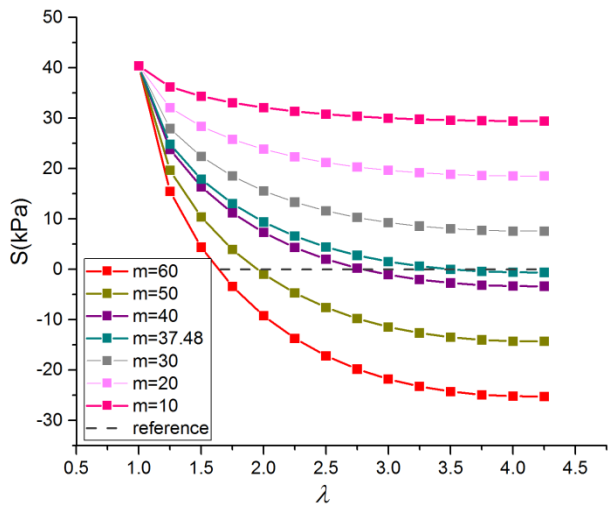
Figure 6. The influence of the parameters m: (a) when the value of the parameter m is less than the true value, the stress changes less value with the strain. (b) when the value of the parameter m is more larger than the true value, the stress changes more value with the strain.
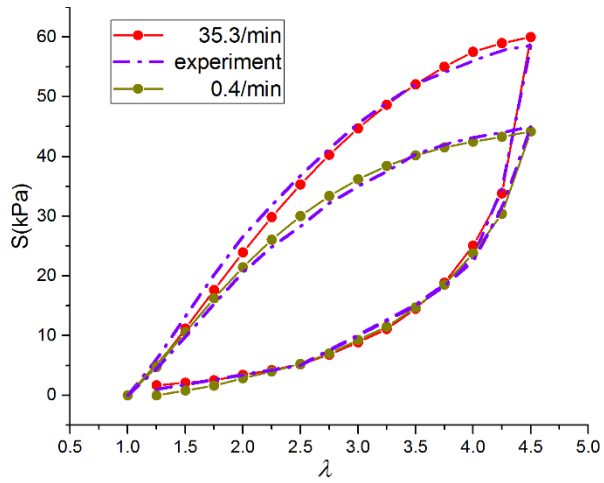
Figure 7. A comparison between the simulation and experiment results [12] of the stress-strain relation for the double network hydrogel at the strain rate of 0.4/min and 35.3/min.