A Physically-Based, Quasilinear Viscoelasticity Model for the Dynamic Response of Polyurea

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Abstract

Polyurea, a promising material for damage mitigation in impact scenarios, has been investigated through plane-wave, pressure-shear plate impact (PSPI) experiments to obtain its mechanical response at high pressures and high strain rates. Based on these experimental results, a physically-based, quasi-linear, viscoelasticity model is introduced to capture the observed nonlinear pressure-volume behavior, the strong dependence of shearing resistance on pressure, and the strong relaxation of deviatoric stresses. This model has been implemented in finite element software ABAQUS to simulate the response of polyurea P1000 under the impact conditions of a variety of PSPI experiments. Simulation results agree reasonably well with those of the experiments.

Keywords: Polyurea, High Pressure, High Strain Rate, Pressure-shear, Quasilinear Viscoelasticity

1. Introduction

Polyurea is an elastomer that has been used increasingly in defense applications involving explosive and impact loading. In spite of having significantly lower wave speeds and acoustic impedances than structural metals, it appears to have attractive features for mitigating damage caused by high-rate loading. Many experiments have been done to measure the dynamic behavior of polyurea at medium strain rates (10^2 - 10^3 s⁻¹) using a Split-Hopkinson bar [1-4] and to obtain the Hugoniot under plate impact loading [5]. Our recent PSPI experiments have provided considerable data on the mechanical response of polyurea at high strain rates (10^5 - 10^6 s⁻¹) and pressures up to 18 GPa [6-11]. These experiments have shown that polyurea has remarkable shearing resistance at high pressures and high strain rates.

Previous models for describing the inelastic, rate dependent response of polyurea appear to be inadequate, especially at higher strain rates and higher pressures. Moreover, it is evident from experiments that Lagrangian shear wave speeds depend significantly on the volumetric strain of the region into which the shear wave is propagating. Therefore, an acceptable model needs to include a dependence of shear wave speed on pressure.

The present study introduces a quasi-linear viscoelasticity model to capture: the observed nonlinear pressure-volume behavior; the strong dependence of shearing resistance on pressure; and the strong relaxation of deviatoric stresses. This constitutive model has been implemented in ABAQUS/Explicit using a VUMAT user material subroutine. Simulation results are compared with experimental results on the mechanical response of polyurea P1000 for a variety of high-strain-rate, high-pressure, loading conditions [6-11] to gain better understanding of the viability of the proposed model.

2. Constitutive Model

The overall approach is to model the response of polyurea as consisting of an instantaneous elastic response followed by relaxation with a continuous distribution of effective relaxation times. The analysis begins by considering the instantaneous elastic response to be that of a nonlinear, isotropic solid undergoing finite deformations. Subsequently, relaxation is considered within the framework of quasi-linear viscoelasticity.

2.1 Instantaneous Elastic Response

Unlike the traditional additive decomposition of strain energy (e.g. the generalized Mooney-Rivlin model[12, 13]), a multiplicative decomposition is used to include the pressure dependence of the shear wave speed:

$$\bar{W} = f(J) \hat{W}(\bar{I}_1, \bar{I}_2) \quad (1)$$

where $f(J)$ is a factor that depends on specific volume and is taken to have the ‘‘Lennard-Jones-like’’ form[14]

$$f(J) = (J^{-M} - 2J^{-N}) + 2 \quad (2)$$

where J is the ratio of the current volume to the initial volume. Material constants $N = M / 2$, and the function $f(J)$ meet the requirement $f(1) = 1$.

$\hat{W}(\bar{I}_1, \bar{I}_2)$ is the distortional strain energy and is taken to have the Mooney-Rivlin form:

$$\hat{W}(\bar{I}_1, \bar{I}_2) = C_{00} + C_{01}(\bar{I}_2 - 3) + C_{10}(\bar{I}_1 - 3) \quad (3)$$

where $\bar{I}_1$ and $\bar{I}_2$ are the normalized first and second principal invariants of the left Cauchy Green deformation tensor B . For pressure-shear loading the deformation gradient tensor F and the corresponding tensor B are:

$$F = \begin{pmatrix} \lambda & 0 & 0 \\ -\kappa & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}; \quad B \equiv FF^T = \begin{pmatrix} \lambda^2 & -\kappa\lambda & 0 \\ -\kappa\lambda & \kappa^2 + 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (4)$$

where λ is the stretch, in the direction of the normal to the wave front, and κ is the shear. The invariants $\bar{I}_1, \bar{I}_2$:

$$\begin{aligned} \bar{I}_1 &\equiv J^{-2/3} I_1 = \lambda^{4/3} + \kappa^2 \lambda^{-2/3} + 2\lambda^{-2/3} \\ \bar{I}_2 &\equiv J^{-4/3} I_2 = 2\lambda^{2/3} + \kappa^2 \lambda^{-4/3} + \lambda^{-4/3} \end{aligned} \quad (5)$$

are so-defined that their value is 3 for a purely dilatational deformation.

The symmetric Piola-Kirchoff stress tensor $\bar{\tau}$ can be expressed in terms of derivatives of the strain energy as,

$$\bar{\tau}_{ij} = \frac{\partial \bar{W}}{\partial \bar{I}_1} \frac{\partial \bar{I}_1}{\partial E_{ij}} + \frac{\partial \bar{W}}{\partial \bar{I}_2} \frac{\partial \bar{I}_2}{\partial E_{ij}} + \frac{\partial \bar{W}}{\partial J} \frac{\partial J}{\partial E_{ij}}. \quad (6)$$

The Cauchy stress tensor T for pressure-shear loading is obtained from $\bar{\tau}$ by

$$T = \frac{1}{(\det F)} F \bar{\tau} F^T. \quad (7)$$

2.2 Quasilinear Viscoelasticity

To account for the relaxation of stresses after polyurea's relatively stiff initial response to a rapid change in strain, consider a simple relaxation model — examined by Pipkin and Rogers[15], and having similarities with a model that has been used successfully by Fung [16] to account for stress relaxation in soft biological tissues. Following Fung[16], a reduced relaxation function $R(t)$ is introduced that relates the relaxed stress $\bar{\sigma}$ to the instantaneous elastic stress σ^e by

$$\bar{\sigma}(t) = \sigma^e(t) + \int_0^t \sigma^e(t-t') \frac{dR(t')}{dt'} dt' \quad (8)$$

where $(0, t)$ is the time interval over which the loading has occurred. The model is described here for the case of uniaxial stress σ , but will be implemented subsequently for the components of the deviatoric stress tensor.

The reduced relaxation function $R(t)$ is motivated by the relaxation function for a standard linear solid:

$$E(t) = E_\infty + (E_0 - E_\infty) e^{-t/t_R} \quad (9)$$

where E_0 , E_∞ are, respectively the instantaneous (“glassy”) elastic modulus and the late-time (“rubbery”) elastic modulus. The time t_R is the effective relaxation time. The reduced relaxation function $R(t)$ for the standard linear solid is given by

$$R(t) \equiv \frac{E(t)}{E(0)} = \frac{E_\infty}{E_0} + \left(1 - \frac{E_\infty}{E_0}\right) e^{-t/t_R} \quad (10)$$

or

$$R(t) = \varsigma + (1 - \varsigma) e^{-t/t_R} \quad (11)$$

where $\varsigma \equiv E_\infty / E_0 < 1$ is the ratio of rubbery and glassy moduli. Motivated by the complexity of the micro-structure of polyurea, its response will be described by a model having a continuous distribution of relaxation times. Such a model, that reduces to (11) for the case of a single relaxation time, can be described by

$$R(t) = \int_0^\infty \left\{ \zeta + (1-\zeta)e^{-t/t'} \right\} S(t') dt'; \quad \int_0^\infty S(t') dt' = 1 \quad (12)$$

where $S(t')dt'$ is the fraction of states with relaxation times in the interval $(t', t' + dt')$. For $S(t') = \delta(t' - t_R)$, where $\delta(u)$ is the delta function, Eqn. (12) reduces to Eqn. (11) for all relaxation times t_R . From Eqn. (12) the reduced relaxation function $R(t)$ has the properties $R(0) = 1$ and $R(\infty) = \zeta$.

In order to obtain a physically-based distribution $S(t')$ that characterizes the weight of relaxation times in the interval $(t', t' + dt')$, consider a distribution motivated by statistical mechanics in which the Boltzmann distribution

$$p(U)dU = C_b e^{-\beta U} dU \quad (13)$$

is used to characterize the fraction of states of a system that lie in the energy interval $(U, U + dU)$. In (13), $\beta = 1/KT$ where K is Boltzmann's constant, T is absolute temperature, and U is the energy per atom. The coefficient C_b is a constant obtained from the normalization requirement:

$$\int_0^\infty p(U)dU = 1 \Rightarrow C_b = \frac{1}{\int_0^\infty e^{-\beta U} dU} = \beta. \quad (14)$$

To relate a relaxation time to a corresponding energy, consider the Arrhenius relation for the frequency of successful jumps across an energy barrier (e.g. the net frequency of breaking and reformation of molecular links [17]):

$$\nu(U) = \nu_0 e^{-\beta(U_b - U)} \quad (15)$$

where U_b is the height of the energy barrier and ν_0 is the attempt frequency. The relaxation time $t'(U)$ is inversely proportional to the success frequency $\nu(U)$. Then, from (15),

$$t' = \frac{1}{\nu(U)} = \frac{1}{\nu_0} e^{\beta(U_b - U)}. \quad (16)$$

From (16), states with small relaxation times (but greater than $1/\nu_0$) correspond to states with large energies $U(t')$ —approaching U_b as t' approaches $1/\nu_0$. Differentiation of (16) gives

$$1 = \frac{-\beta}{\nu_0} e^{\beta(U_b - U)} \frac{dU}{dt'} \Rightarrow \frac{dU}{dt'} = -\frac{\nu_0}{\beta} e^{-\beta(U_b - U)}. \quad (17)$$

Equating the fraction of states in the energy interval $(U, U + dU)$ to the fraction of states in the relaxation-time interval $(t', t' + dt')$ one obtains

$$S(t')dt' = -p(U)dU \quad (18)$$

where, in view of (17), the negative sign has been introduced so that the distribution functions $S(t')$ and $p(U)$ are both positive. Substitution of (13) and (17) into (18) gives

$$S(t') = \nu_0 e^{-\beta U_b}. \quad (19)$$

Because the right side of (19) is independent of t' , all relaxation times are weighted equally. Nevertheless, in developing a model to describe measured response over a limited range of loading durations, one needs to recognize that there are relaxation times that are so short that they are not measurable with available equipment and there are relaxation times that are so long that they exceed the total time of the measurements. For these reasons the distribution(19) will be cut off at upper and lower limits to obtain the distribution:

$$S(t') = \begin{cases} S_0 & \text{for } t_1 \leq t' \leq t_2 \\ 0 & \text{for } t' < t_1, t' > t_2 \end{cases} \quad (20)$$

where $[t_1, t_2]$ spans the range of relaxation times that are measurable in experiments for which simulations are to be performed. From the normalization condition (12), the constant S_0 is

$$S_0 \equiv \nu_0 e^{-\beta U_b} = \frac{1}{t_2 - t_1}. \quad (21)$$

Then, the reduced relaxation function becomes

$$R(t) = \varsigma + \frac{(1-\varsigma)}{(t_2 - t_1)} \int_{t_1}^{t_2} e^{-t/t'} dt' = \varsigma + \frac{(1-\varsigma)}{(t_2 - t_1)} \int_{1/t_1}^{1/t_2} e^{-tu} \left(\frac{-1}{u^2} \right) du \quad (22)$$

and its derivative, required in (8), is

$$\dot{R}(t) = \frac{(1-\varsigma)}{(t_2 - t_1)} \int_{1/t_1}^{1/t_2} e^{-tu} \frac{du}{u}, \quad (23)$$

which can be integrated to give

$$\dot{R}(t) = \frac{(1-\varsigma)}{(t_2 - t_1)} \left\{ \ln \left(\frac{t_1}{t_2} \right) + \sum_{n=1}^{\infty} \frac{(-t/t_2)^n - (-t/t_1)^n}{n \cdot n!} \right\}. \quad (24)$$

To gain understanding of the mechanical response characterized by (23) it is instructive to derive expressions for the associated complex moduli. To do so, consider the Laplace transform of $R(t)$ given by

$$\tilde{R}(s) = \frac{s(t_2 - t_1) + (1-\varsigma) \log[(1+st_1)/(1+st_2)]}{(t_2 - t_1)s^2}. \quad (25)$$

Following Ferry[18], one obtains the complex reduced relaxation “modulus” from

$$R^*(\omega) \equiv i\omega \tilde{R}(i\omega) = 1 - \frac{(1-\varsigma)}{\omega(t_2 - t_1)} \left[\tan^{-1}(\omega t_2) - \tan^{-1}(\omega t_1) \right] + i \frac{(1-\varsigma)}{2\omega(t_2 - t_1)} \ln \left[\frac{1 + \omega^2 t_2^2}{1 + \omega^2 t_1^2} \right]. \quad (26)$$

The storage modulus $G'(\omega)$ and loss modulus $G''(\omega)$ can be expressed as

$$G'(\omega) \equiv \text{Re}[R^*(\omega)] = 1 - \frac{(1-\zeta)}{\omega(t_2 - t_1)} \left[\tan^{-1}(\omega t_2) - \tan^{-1}(\omega t_1) \right], \quad (27)$$

$$G''(\omega) \equiv \text{Im}[R^*(\omega)] = \frac{(1-\zeta)}{2\omega(t_2 - t_1)} \ln \left[\frac{1 + \omega^2 t_2^2}{1 + \omega^2 t_1^2} \right], \quad (28)$$

and the loss tangent becomes

$$\tan[\delta(\omega)] \equiv \frac{G''(\omega)}{G'(\omega)} = \frac{\frac{(1-\zeta)}{2\omega(t_2 - t_1)} \ln \left[\frac{1 + \omega^2 t_2^2}{1 + \omega^2 t_1^2} \right]}{1 - \frac{(1-\zeta)}{\omega(t_2 - t_1)} \left[\tan^{-1}(\omega t_2) - \tan^{-1}(\omega t_1) \right]}. \quad (29)$$

3. Numerical Simulations

To conduct the simulations the model has been implemented in ABAQUS/Explicit using a VUMAT user material subroutine. Values of parameters used for the simulations are listed in table 1.

Table 1: Parameter values used in the model of polyurea

ρ_0 (kg / m ³)	ν	c_1 (km / s)	α	M	$\rho_0 c_1^2$ (GPa)	C_{00} (GPa)	C_{01} (GPa)	C_{10} (GPa)	ζ	t_1 (ns)	t_2 (ns)
1070	0.42	1.725	0.5	4.4	3.184	0.2684	0.1098	0.1098	0.0132	1	1500

The instantaneous elastic response is determined by the four parameters (c_1, ν, α, M) . The value, c_1 , for the elastic longitudinal wave speed at zero pressure (i.e. at $J=1$) is taken from the unloading wave speed in release wave experiments[6]. The value of M is determined primarily by the need to match Lagrangian longitudinal wave speeds at high pressures [6]. The value of Poisson's ratio ν is a measure of the relative stiffness in shear to that of dilatation. The value $\alpha = 0.5$ corresponds to assuming $C_{01} = C_{10}$ in the absence of data to weight one of these shear stiffness coefficients heavier than the other. Choosing a value $\alpha = 0.5$ in the relationships $C_{01} = (1 - \alpha)\mu / 2$, $C_{10} = \alpha\mu / 2$ —where μ is the elastic shear modulus — may seem arbitrary, especially in view of the value $\alpha = 0$ obtained for the Neo-Hookean model derived from the theory of rubber elasticity. The applicability of a rubber elasticity model for a block co-polymer in its glassy regime is unknown. Therefore, the choice $\alpha = 0.5$ is used to minimize uncertainty in the absence of an established theory for the impact loading regime of the experiments. This choice turns out not to be very important as simulations with α varying from 0 to 1 led to simulated velocity-time profiles varying by only a few percent.

The linear elastic shear modulus at zero pressure is $\mu = 2(C_{01} + C_{10}) = 0.4392 \text{ GPa}$. Relaxation parameters ζ, t_1, t_2 are determined by a modest, best-fit comparison of

transverse velocity-time profiles. Parameter values for relaxation of pressure are not included because previous study[6] shows the effects of pressure relaxation are small.

The storage modulus $G'(\omega)$, loss modulus $G''(\omega)$, and loss tangent $\tan \delta$ are shown in Figure 1 for the relaxation parameters of Table 1. Storage moduli increase from their “rubbery” values at low frequencies to their “glassy” values at high frequencies. The loss tangent $\tan \delta$ has a peak in the rubbery regime. The onset of the rubber-to-glass transition is characterized by a pronounced reduction in the loss tangent beginning at $2.11 \times 10^4 \text{ Hz}$. The loss tangent then decreases until it becomes essentially constant in the glassy regime where the amorphous chain conformations of the polymer can be regarded as essentially frozen into a network. The range of frequencies over which the rubber-to-glass transition occurs is approximately the interval $(1/t_2, 1/t_1)$ where t_1, t_2 are the cut-off times for the distribution of relaxation times $S(t')$ of (20). Relaxation is largest at frequencies near $1/t_2$. Correspondingly, simulations of relaxation are more strongly affected by the value of t_2 than that of t_1 .

Figure 1: Storage modulus G' , loss modulus G'' , and loss tangent $\tan \delta$ for the normalized modulus $R^*(\omega/2\pi)$ for the relaxation parameter values of Table 1.

A schematic of the sandwiched pressure-shear plate impact experiment is shown in Figure 2. The polyurea sample is cast between two hard plates. Depending on the pressure regime being targeted, the bounding plates are made from either tool-steel or tungsten carbide. The target assembly is impacted by a flyer plate, made from the same material as the bounding plates, skewed to the direction of travel by the angle θ . The flyer, front, and rear plates are lapped to a flatness of $0.6 \mu\text{m}$ over a 50 mm diameter. The rear surface of the rear plate is polished to a mirror finish. A thin film of photoresist

is spun onto its rear surface and used to produce a diffraction grating, with a frequency of 600 lines/mm . Diffraction of light from this grating is used to monitor the normal and transverse motion of the rear surface. Details of the techniques involved in conducting the pressure-shear plate impact experiment are given by Clifton and Klopp[19, 20].

Figure 2: A schematic of sandwiched pressure-shear plate impact experiment(TDI and NDI are, respectively, acronyms for Transverse Displacement Interferometer and Normal Displacement Interferometer ; VISAR is the acronym for Velocity Interferometer for Any Reflector)

PSPI experiments examined in our study are listed in Table 2, which include constant-pressure shots (#1303, #1201, #404), a pressure-change shot (#704) and symmetric PSPI shots (#1301, #1302). Details of the PSPI experiments are given in[6].One row of plane-strain (2D) elements is used in the calculations. Simulations are performed with a Lagrangian mesh size $\Delta X = 1.72 \times 10^{-6} m$ and a time step $\Delta t = 10^{-10} s$. Impact configurations and boundary conditions are chosen to correspond to the experimental conditions.

Table 2: Pressure-shear impact experiments simulated

Shot No.	h (mm)	h_{front} (mm)	h_{rear} (mm)	h_{flyer} (mm)	Φ (mm)	Hard plates	v_0 (m/s)	Inclination Angle ($^\circ$)	Tilt Angle (mrad)
1303	0.1	2.938	5.915	9.957	50	Pure WC	357.1	16	0.5
1201	0.097	3.582	5.578	7.411	50	Pure WC	183.5	18	0.296
404	0.11	2.896	7.041	6.991	60	Steel	112.6	18	0.3
704	0.14	2.325	4.427	10.996	57.15	D2	109	18	1.15
Shot No.	$H_{\text{p\_anvil}}$ (mm)	h_{anvil} (mm)	$H_{\text{p\_flyer}}$ (mm)	h_{flyer} (mm)	Φ (mm)	Hard plates	v_0 (m/s)	Inclination Angle ($^\circ$)	Tilt Angle (mrad)
1301	0.069	4.051	0.074	4.043	50	WC504	188	18	1
1302	0.098	4.032	0.086	8.075	50	WC504	178	18	0.3

4. Results and Discussion

Simulated velocity-time profiles for so-called “constant-pressure” (i.e. experiments where the pressure in the sample rings up to its final level before the shear wave arrives) PSPI experiments (shots #1303, #1201, #404) of the type shown in Figure 2 are compared with experimental results in Figure 3. Oscillations in the experimental transverse velocity-time profiles are indicative of data reduction difficulties for some of the shots because of low signal levels. Velocity-time profiles for the normal component of rear surface velocity agree well for all relaxation models and even without relaxation (i.e. instantaneous elastic response only). Thus, relaxation of shear stresses has relatively little effect on measured normal velocities. However, velocity-time profiles for the transverse component of rear surface velocity are affected strongly by the relaxation of shear stresses— as could be expected because measured transverse velocities reflect the shearing resistance of the material. Without relaxation the simulated shear stresses and transverse particle velocities are much greater than those observed in the experiments. With relaxation of shear stresses the simulated shear stresses agree reasonably well with those measured in the experiments.

Figure 3: Comparison of simulated and experimental velocity-time profiles for constant-pressure PSPI experiments. Upper (lower) curves for each shot number correspond to normal (transverse) velocities. Pressures for the three experiments are: 18 *GPa* (#1303), 9 *GPa* (#1201) and 2.5 *GPa* (#404).

Comparisons of simulated and experimental velocity-time profiles are shown in Figure 4 for a so-called “pressure-change” experiment (shot #704) in which the pressure is changed during the shearing by the arrival of an unloading wave reflected from the rear face of the flyer. The rise times of the simulated profiles for normal velocity are less than those of the experimental profile. This discrepancy has been suggested as being due to initial porosity

of the sample obtained from a different source [6]. When relaxation of the shear stresses is included, the simulated transverse velocity follows quite well the decrease in shearing resistance with reduction in pressure that is observed in the experiments.

Figure4: Comparison of simulated and experimental velocity-time profiles for a pressure-change experiment (Shot 704) and for a low-pressure experiment (Shot 710).

Simulated and measured velocity-time profiles for twosymmetric pressure-shear plate impact (SPSPI) experiments are shown in Figure 5. In SPSPI experiments, approximately one-half of the thin sample is attached to the front face of the flyer plate and the remaining half is attached to the front face of the target (anvil) plate. The principal difference between these two experiments is the difference in total sample thicknesses for the two shots: 0.143 mm for #1301 and 0.184 mm for #1302. Because of the greater thickness of the sample thickness for Shot 1302, the risetimes for its velocity-time profiles are significantly longer than those for Shot 1301. Agreement between simulated and measured velocity-time profiles for the normal component of rear surface velocity is reasonably good — best when shear stress is relaxed in the simulations. The simulated transverse velocity with the relaxation of shear stresses rises more slowly than observed in the experiments, which may be an indication that the model does not adequately reflect the early increase in stiffness as the pressure is ramping up.

Figure5: Comparison of simulated and experimental velocity-time profiles for symmetric PSPI experiments (#1301, #1302) for peak pressures of approximately 9 GPa

5. Conclusion

A physically-based quasilinear viscoelasticity model for polyurea has been introduced and examined through comparisons with results of PSPI and SPSP experiments. The model was developed to capture the observed nonlinear pressure-volume behavior, the strong dependence of shearing resistance on pressure, and the strong relaxation of deviatoric stresses. Simulated velocity-time profiles show qualitative agreement with measured profiles over the full range of impact velocities and target configurations. Quantitative agreement is also quite good for the modest number of parameters that appear in the model.

Whether or not this model represents an improvement over other models, such as the model introduced in [6] for which the distribution of relaxation times lacks a physics-based motivation, remains to be determined. Simulations using each of these models show comparable agreement with experimental velocity-time profiles. The model reported here is attractive in that the coefficient S_0 has an interpretation in terms of the physical parameter $\nu_0 e^{-\beta U_b}$. However, concern about broader applications of the model arise when one observes that the relaxation time t_2 that has the greatest effect on the simulated velocity-time profiles is also approximately the duration of the experiments. Thus, one should be cautious in interpreting the time $t_2 = 1.5 \mu s$ as a dominant relaxation time for the high strain rate response of polyurea P1000. Sorting out these matters

appears to require additional experiments over a wider range of loading durations. Molecular dynamics calculations may also be helpful in this regard as these calculations may provide a link between the mechanistic interpretation of S_0 in (21) and its interpretation, also in (21), as a parameter related to cut-off limits for the distribution of relaxation times. Collaborative work with Stefanie Heyden and Michael Ortiz is currently underway to examine the possibility of such a connection [21].

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