Theory of nonlinear creep under large deformation in polymer glasses

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Goal

- Creep is defined by the time dependence of strain at constant stress

- At low stress, the typical ideal creep curve grows monotonically and gradually, bending down approaching the steady flow state (constant strain rate) after an instantaneous jump

- But, at large stress and/or deformation, a dramatic upturn occurs, and followed by a bending-over at very large strain
  - What’s the origin of the upturn and bending-over?
  - What causes the dynamics change during these processes?
  - How to model the creep response?

Microscopic theory of glassy dynamics

- Dynamical “free energy” for single-segment movement:

\[
\dot{\gamma} = -\frac{\partial}{\partial r} F_{eff}(r(t)) + \eta(t)
\]

- Strain hardening at large deformation

State variable under Gaussian thread model

\[\dot{S}_0 = S(q = 0)\]

Mechanical and Dynamical properties:

Elastic modulus: \(E'\)

Alpha relaxation time:

\[\tau_a(T) = \tau_0(T)e^{\alpha_\tau/S_k T}\]

Model of aging and rejuvenation

\[
\frac{dS}{dt} = S_0 - S_0 \tau_a(S_0 - S_0) - \frac{\tau}{E(S_0, \tau)\tau_e(S_0, \tau)}\]

Constitutive equation of nonlinear mechanical response

- Generalized Maxwell Model:

\[\dot{\gamma} = \frac{d\tau}{dt} E'(\tau) + \frac{\tau}{E'(\tau)\tau_a(\tau)}\]

- Nonexponential decay:

\[
\tau(t) = \int_0^t d\tau'E'(\tau(t))e^{-\int_0^{\tau(t)} E'(\tau)^{\beta_k} \dot{\gamma}(\tau')d\tau'}
\]

\(\beta_k\) : related to heterogeneity

Results: Creep at large stress

- Strain hardening at large deformation

State variable decreases for stretched chain

\[\lambda = e^{\alpha_\tau/S_k T}\]

Conclusion

- The upturn behavior is caused by rejuvenation process.
- The bending-over happening at very large deformation is due to strain hardening.
- Strain response and relaxation time are well predicted except for the recovery process because of the lack of relaxation mechanism of stretched chain in the theory.