Mechanics of High Strain Rate Behavior of Elastomeric Random Copolymers

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Summary The mechanics of elastomeric random copolymers has been a focal point of interest due to the resilient, yet highly dissipative features of their mechanical response under extreme loading events such as impact, blast and ballistic penetration. In this research, the high strain rate behavior of an exemplar elastomeric polyurea is explored through experiments and computational modeling where a recently developed constitutive theory of polyurea is facilitated.

INTRODUCTION

Elastomeric random copolymers have a backbone structure comprised of "hard" and "soft" segments, where the hard/soft phases correspond to their thermodynamic glassy/rubbery state at ambient temperature. The mechanical performance of random copolymers is governed by the chemical composition as well as the morphology and provides hybrid properties of their constituents. The morphological structure of the hard and soft domains results in multiple molecular relaxations in association with distinct dissipation mechanisms such as rate-dependent viscoelasticity and viscoplasticity. In addition, *in situ* X-ray scattering data as a function of deformation revealed a microstructural evolution and breakdown of the structure, which governs a stretch-induced softening which, in turn, provides another major dissipation mechanism under large deformation. The large energy-dissipation via the microstructural breakdown together with the viscoelastic, viscoplastic behavior provide new avenues for improving resistance and energy-absorption against extreme environments such as impact, blast and ballistic penetration events. An exemplar polyurea exhibits a highly resilient yet dissipative behavior; its "rubbery" soft domain provides resilience and enhanced resistance to flow at high strain rates while its "glassy" hard domain provides enhanced stiffness as well as viscoplastic resistance. The mechanical principles crucial to realizing the versatility of elastomeric random copolymers are incompletely understood at present. Herein, the mechanics of the large strain behavior of polyurea over a wide range of strain rates is examined. Specifically, the high strain rate behaviors of polyurea are extensively addressed by introducing a large deformation viscoplasticity constitutive model for the hard/soft domains in association with their distinct relaxations, spanning a transition behavior in the vicinity of a moderate strain rate. Also, the highly resilient yet dissipative responses under extreme strain rates are elucidated through Taylor impact tests, where the study of such elastomeric materials has been largely unexplored, and the complementary finite element simulations where the constitutive model imparting the hard/soft domain contribution to macroscopic mechanical responses is implemented.

MECHANICAL BEHAVIOR OF POLYUREA

Dynamic mechanical analysis (DMA, Figure 1a) supports the phase-separated morphology of polyurea where the soft *aliphatic diamine* is in a rubbery state while the hard *diisocyanate* is in a glassy state at room temperature. The large deformation stress-strain behavior of polyurea is featured in relatively stiff response with the rate-dependent roll-over or yield to more compliant response; rate-sensitivity increase after a critical strain rate; nonlinear unloading accompanied with substantial hysteresis and resilience; a stretch-induced softening in subsequent reloading-unloading cycles via nonlinear elasticity; viscoelasticity; and viscoplasticity as depicted in Figure 1b and c.

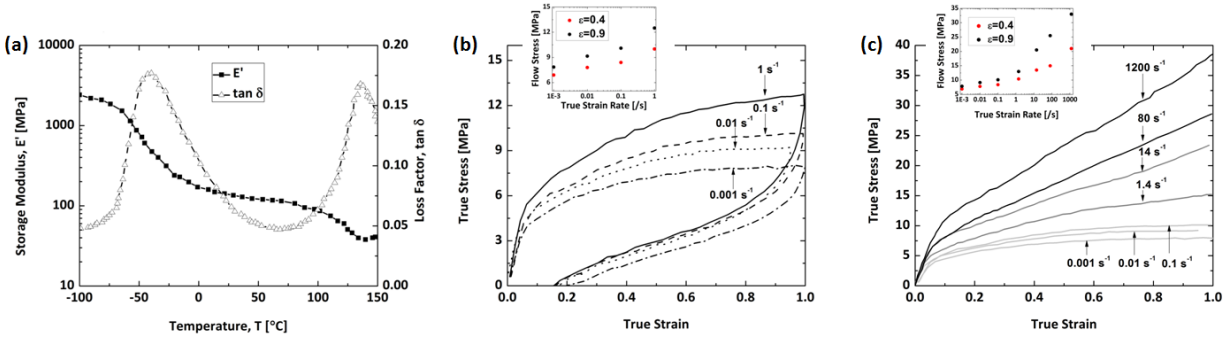

Figure 1 (a) dynamic mechanical analysis explaining co-continuous microstructures; two peaks in loss factor are associated with molecular relaxations in soft/hard domain [1]; (b) and (c) stress-strain behavior under low and high strain rate compression, respectively, with an inset of flow stress as a function of strain rate [2,3].

In stress-strain behaviors under high strain rate compression displayed in Figure 1c, dramatic enhancement in both initial stiffness and flow stress was observed. An inset of Figure 1c on the flow stress at strains of 0.4 and 0.9 as a function of strain rates clearly indicates there is a transitional regime in the rate-sensitivity in the vicinity of 1 – 10 s^{-1}. As described

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in DMA curves in Figure 1a, there are two distinct relaxations for the hard and soft domain in polyurea, which, at high strain rates, leads to an additional resistance from the soft domain which is not fully relaxed at these short timescales.

A LARGE DEFORMATION CONSTITUTIVE MODEL

The macromolecular network of a polymer responds to mechanical deformation through intermolecular and network resistance of the molecular chains. As described in a schematic in Figure 2a, the random copolymer is comprised of interpenetrating hard and soft domains in co-continuous structures via macromolecular networks. Thus, the model requires two sets of the intermolecular and network resistance for both hard and soft domains. A rate-dependent elastic, viscoplastic constitutive model is constructed to predict the macroscopic responses over a wide range of strain rates, imparting the Neo-Hookean elasticity with thermally-activated viscoplasticity and the Arruda-Boyce rubber elasticity with nonlinear reptation model for the intermolecular and network resistance, respectively, for hard and soft domains. As described in Figure 2b, the simulation results agree excellently with experimental data, spanning the transition in the stress response around the moderate strain rate that supports multiple distinct relaxation processes in polyurea.

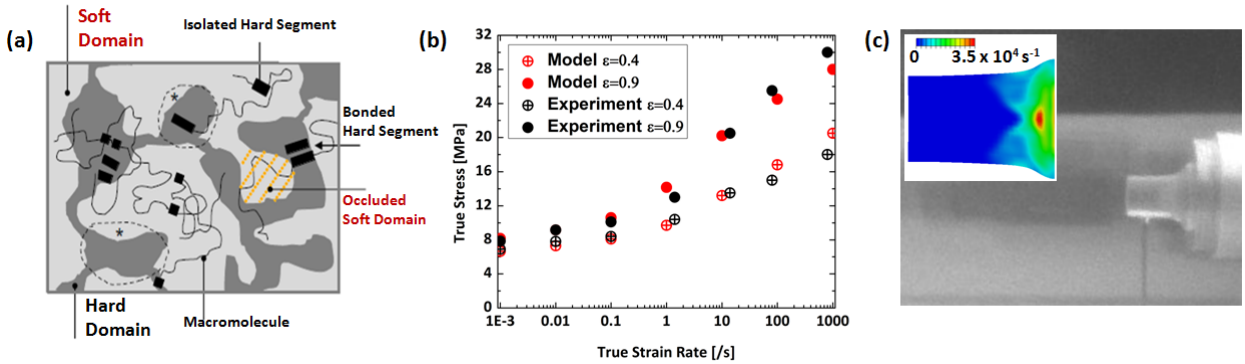

Figure 2 (a) schematic for polyurea irregular microstructure representing a segmented block copolymer with segregated interpenetrating hard/soft domains; (b) flow stress at different true strains of 0.4 and 0.9 as a function of strain rates; (c) polyurea rod subjected to Taylor impact with a piston velocity 245 m/s with an inset of a contour of plastic strain rate $\dot{\gamma}_p$ in simulation

TAYLOR IMPACT TEST: EXPERIMENTS and FINITE ELEMENT SIMULATIONS

Ultra-high strain rates (> 1000 s^{-1}) localized during realistic ballistic or blast loading events can be reached via Taylor impact experiments (Figure 2c). Taylor impact tests on polyurea rods are conducted in experiments and finite element simulations to examine the mechanics of the elastomeric random copolymer under extreme strain rates and dynamic inhomogeneous deformation. The highly resilient yet dissipative behaviors during extreme deformation events as incurred during Taylor impact tests are understood, via the finite element analysis (inset of Figure 2c) utilizing the present large deformation viscoplasticity constitutive theory. The dynamic deformation profiles with residual and recovered shapes in the simulations are extensively compared to those in the experiments. Furthermore, the energy storage and dissipation in the random copolymeric materials are addressed by analyzing the elastic, plastic wave front propagations, revealing a critical role of the rubbery soft domain in both elastic and plastic responses during the extreme loading events.

CONCLUSIONS

Elastomeric random copolymers have been found to be versatile materials in a wide range of applications from self-healing microstructures to ballistic protective coatings via their highly resilient yet dissipative features. In this research, a general framework for understanding the mechanics of an exemplar polyurea under high strain rate events is proposed ranging from the large deformation constitutive model to the inhomogeneous finite element simulations in association with the high strain rate experiments. The ability to accurately model the high rate mechanical responses will enable predictive design of the random copolymeric systems operating under extreme mechanical environments.

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