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Research article

Multi-Scale Computation-Based Design of Nano-Segregated Polyurea for Maximum Shockwave-Mitigation Performance

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Abstract: A multi-length-scale computational analysis is used to carry out the design of polyurea for maximum shockwave-mitigation performance. The computational analysis involves a combined all-atom/coarse-grained molecular-level investigation of shockwave-propagation within polyurea and a finite-element analysis of direct quantification of the shockwave-mitigation capacity of this material as a function of its chemistry (or, more specifically, of its soft-segment molecular weight). The results obtained suggest that the approach employed can correctly identify the optimal chemistry of polyurea and, thus, be of great benefit in the efforts to develop new highly-efficient blastwave-protective materials, in a cost-effective manner.

Keywords: Polyurea; Materials by design; Shockwave mitigation

1. Introduction

Polyurea is an elastomeric co-polymer that is formed by a rapid co-polymerization reaction between two groups: (a) an isocyanate (an organic compound containing isocyanate -N=C=O group); and (b) an amine (organic compound containing amine $-NH_2$ group). A schematic of a polyurea repeat unit is shown in Figure 1. As seen in this figure, polyurea chains contain two distinct types of segments: (a) hard segments (HS), which are formed by highly polar (i.e., containing centers/poles of negative and positive charge) urea linkages (-NH-CO-NH-) and adjoining diphenyl methane ($C_6H_5-CH_2-C_6H_5$) functional groups; and (b) soft segments (SS), which consist of a series of aliphatic functional groups. For clarity, the urea linkages in the hard segments are depicted using enlarged spheres.



Figure 1. Schematic of polyurea repeat unit consisting of hard segment (HS) and soft segment (SS). For clarity, elements are color-coded and labeled, and urea linkages are indicated using enlarged spheres.

In bulk polyurea, strong hydrogen bonding between urea linkages of the adjacent chains (or between neighboring portions of the same chain) promotes microphase-segregation (self-assembly) of hard segments into the so-called nanometer-sized, ribbon-shaped hard (i.e., high glass-transition temperature, often crystallized) domains. The remaining hard and the soft segments of the polyurea chains are fairly well-mixed and form the so-called soft (i.e., low glass-transition temperature, amorphous) matrix. It should be noted that strong hydrogen bonding between hard segments, within the hard domains, provides non-covalent inter-chain cross-linking. This is the reason that polyurea is often referred to as a thermoplastically cross-linked elastomer. In addition, depending on the amount of higher-functionality isocyanate present in the precursor, polyurea may contain different extents of covalent inter-chain cross-linking. Based on the preceding descriptions of the polyurea molecular-and domain-level microstructures, this material can be considered as an elastomeric-matrix-based nanocomposite, in which nanometer-sized hard domains act both as stiff/strong reinforcements and as inter-chain linkages.

Recent work by Runt et al. [1] clearly established that microstructure and properties of polyurea are highly sensitive to the chemical composition and functionality of the precursors, synthesis route (e.g., solution-synthesis vs. bulk synthesis) and synthesis conditions (e.g., temperature, humidity, etc.). In particular, the microstructure and properties of polyurea were found to be highly sensitive to the soft-segment molecular weight (i.e. the number of tetra-methylene-oxide, C_4H_8O , groups). Consequently, and considering the fact that polyurea is used in various shockwave-mitigation applications, the present work is focused on the effect of soft-segment molecular weight on the blast-protection potential of polyurea.

In recent years, polyurea external coatings and/or internal linings have been applied to: (a) structures to protect them against blasts produced by detonation of bombs, ordnance and improvised explosive devices (IEDs) [2]; (b) metallic armor in order to improve its ballistic-penetration resistance [3] and (c) personnel-protective gear (e.g., helmet) in order to minimize the danger of traumatic brain injury (TBI) through mitigation of the blast-induced shockwaves [4].

The main objective of the present work is to carry out a multi-length-scale computational analysis in order to identify optimal soft-segment molecular weight of polyurea for maximum shockwave-mitigation performance. Recent computational work [5] revealed that the dominant shockwave-mitigation mechanism in the case of blast-induced shockwaves is associated with the phenomenon of a trailing release-wave catching up with, and attenuating the leading shockwave. Consequently, a metric quantifying the capacity of polyurea (as a function of its soft-segment molecular weight) to mitigate blast-induced shockwaves via this mechanism will be introduced and assessed for different polyurea grades and different shockwave-strength conditions.

Examination of the public-domain literature revealed no study of the polyurea soft-segment molecular weight on its shock-mitigation capability. Presently, as part of our ongoing research effort, a comprehensive experimental investigation of the role of polyurea chemistry on its shock-mitigation performance is being investigated [6]. The experimental approach utilized employs a high-speed camera to image a polyurea sample struck by a projectile from a gas gun. To help resolve the structure of the resulting shockwave-front, the sample is illuminated using an arc-lamp, the light from which passes through a condenser and beam mask before reaching the sample. Details of this investigation will be reported in a future correspondence.

2. Materials and Method

The material investigated in the present work is polyurea, in which the number of tetra-methylene-oxide (C_4H_8O) groups was varied between 3 and 14. In other words, the molecular weight of the soft-segment was varied between 216 and 1008 amu. As far as the computational methods employed are concerned, they involved: (a) all-atom non-equilibrium molecular dynamics [7]; (b) coarse-grained non-equilibrium molecular dynamics [8]; and (c) transient nonlinear dynamics, fluid/structure interaction finite-element analysis [4]. Since details of these computational methods can be found in the cited references, these methods will be only briefly reviewed here.

2.1. All-atom molecular-level computational analysis

Within this approach, a material is considered as an assembly of interacting (and bonded) discrete particles (i.e., atoms, ions, etc.), and potential-energy minimization-based (molecular statics) and Newton's second-law-based (molecular dynamics) algorithms are utilized to examine and quantify the behavior and properties of the material under investigation.

In general, complete definition of an all-atom molecular-level computational model and analysis requires specification of the following:

(a) Geometrical (e.g., atomic positions, computational cell size, etc.) and chemical (e.g., atomic species, bond order, etc.) details of the computational model. An example of the computational model is represented by its unit cell for nano-segregated polyurea P650 (i.e., in polyurea with nine tetra-methylene-oxide units in its soft-segment, resulting in a molecular weight of approximately 72 9 = 648 a.m.u.) in Figure 2(a). Periodic boundary conditions are applied across the faces of the unit cell in order to model polyurea bulk behavior. To highlight nano-phase segregation in Figure 2(a), blue-colored nitrogen atoms are assigned a larger diameter;

(b) A set of fully parameterized force-fields (i.e., a set of mathematical expressions that quantify the potential energy contribution of various bonding and non-bonding interactions between the

constituents of the molecular-scale model). The present work utilizes the so-called "COMPASS" (Condensed-phase Optimized Molecular Potentials for Atomistic Simulation Studies) force-field [9]. This highly accurate force-field has an ab-initio character since most of its parameters were established by matching the predictions made by ab-initio quantum mechanics calculations with their experimental counterparts;



Figure 2. Examples of the: (a) all-atom; and (b) coarse-grained molecular-level computational cells for nano-segregated polyurea used in the present work.

(c) Details regarding the type, number, and usage sequence of the molecular-level computational algorithms/methods to be used in the simulation. To obtain the initial equilibrium state of polyurea, a schedule of combined prolonged equilibrium molecular-dynamics and molecular-statics simulations is employed. To generate and drive planar longitudinal shockwaves within the unit cell, the cell was first replicated fourfold in the direction of the shockwave propagation, and a procedure consisting of sequential axial compressions of the newly-formed unit cell employed. As seen in Figures 3(a)–(d), this procedure results in the formation of two converging, planar longitudinal shockwaves. It should be noted that, for improved clarity, covalent bonds are not displayed in this figure and all the atoms are represented with the same color. Approximate locations of the two shockwave fronts are indicated using arrowheads in Figures 3(a)–(d). It is seen that the material swept by the shockwaves acquires a significantly higher mass/number density. To generate two trailing release-waves, the unit-cell axial-contraction procedure is terminated after the leading shockwaves have each traveled, say one-sixth of the unit-cell axial length; and



Figure 3. Temporal evolution of the molecular-level material microstructure accompanying generation and propagation of a pair of converging planar shockwaves in polyurea.

(d) Lastly, a different post-processing/data-reduction procedure generally needs to be employed in order to characterize and quantify material response and properties of interest. For example, in the present work, shockwave (as well as release-wave) speeds had to be determined. Towards that end, the extended computational cell is partitioned (in the axial direction) into (typically) 40 equal-size Lagrangian bins (the term "Lagrangian" is used here in order to denote that the bins are defined in the material reference/initial configuration). Then, all the atoms associated with each bin are identified. Lastly, average material/atomic properties for each cell (e.g., material-particle velocities) are computed at each simulation time. An example of the typical results obtained (for the right-propagating shockwave only, and before the right-propagating trailing release-wave is generated) through the use of the procedure described above is displayed in Figure 4. It should be noted that the results displayed in Figure 4 pertain to P650, and to the imposed unit-cell axial-contraction rate (i.e., imposed particle velocity, quantifying the shockwave strength) of 600 m/s. The results presented in Figure 4 (and the corresponding results obtained at additional imposed particle longitudinal velocities, and the additional polyurea chemistries) can be used to determine a functional relationship between the (Lagrangian) shockwave speed, Us (set equal to a ratio of the steady shockwave-front advancement and the associated elapsed time) and the imposed particle velocity, up.



Figure 4. Temporal evolution of the particle velocity associated with a right-propagating planar shockwave in polyurea under an imposed (maximum) particle velocity of 600 m/s. Curve labels pertain to the respective post-shock initiation times.

2.2. Coarse-grained molecular-level computational analysis

Within this framework, the material under investigation is still treated as an assembly of discrete, interacting particles (referred to as "beads," hereafter). Each bead represents a collection of atoms and their bonds, and accounts for the collective degrees of freedom of the constituent atoms.

Adjacent beads residing within the same chain are bonded using "connectors" to form bead-based chains. Beads of the neighboring segments of different, or the same chain, are also allowed to interact in a van der Waals-like and/or Coulomb-like non-bond fashion. Due to the larger size of the beads relative to the atoms used within the all-atom computational framework, larger computational domains could be analyzed using the same number of particles, whose treatment could be handled using the available computational and storage resources. In addition, since the high-frequency vibrations of the constituent atoms are integrated out during the coarse-graining procedure, the system dynamics is controlled not by the stiff/hard, atom/atom interactions but rather by the compliant/soft, bead/bead interactions. Consequently, significantly longer time-steps (and, thus, simulation times) can be employed. It should be noted that the all-atom simulations were limited to the analysis of strong (i.e., small wavefront-width) shockwaves. In the case of coarse-grained simulations, the larger computational domains enable analysis of medium-strength and weak shockwaves (associated with a relatively large wave-front width). The ability to analyze intermediate-strength/weak shocks and to use longer simulation times was the main reason for adding the coarse-grained computational framework to its all-atom counterpart.

In general, a complete formulation of a coarse-grained molecular simulation problem involves the specification of the same four items as in the all-atom case:

(a) Geometrical and chemical details of the computational model. An example of the computational model, as represented by its unit cell (with imposed periodic boundary conditions) for nano-segregated P650, is depicted in Figure 2(b). It is seen that two bead types (i.e., larger blue-colored hard H-beads and smaller white-colored soft S-beads) are used to coarse-grain polyurea. It should be noted that the edge-lengths of the unit cell displayed in Figure 2(b) are more than an order of magnitude larger than their counterparts shown in Figure 2(a);

(b) The coarse-grained force-field used has the following features: (i) it includes only two (functionally distinct) contributions to the system potential energy a bond-stretch term and a non-bond van der Waals-type term; (ii) the bond-stretch term is represented using a harmonic-type potential-energy function, and includes three (H–H, H–S and S–S) types of connectors; (iii) the non-bond van der Waals-type bead/bead interactions are accounted for through the use of the 12–6 Lennard-Jones types of functions, and also includes three (H–H, H–S and S–S) types of bead-pair interactions. Details regarding the forcefield functions and their parameterization can be found in [5]; (c) The same (molecular-statics/molecular-dynamics) computational methods as in the all-atom case are employed to determine the initial equilibrium structure of nano-segregated polyurea. Furthermore, the same unit-cell replication procedure and the subsequent sequential axial contraction were employed to generate two converging (medium-strength/weak) planar longitudinal shockwaves; and (d) The same procedure based on the use of Lagrangian bins was employed to reveal the formation and propagation of steady shockwaves, and to establish the functional relationship between the shockwave speed and the particle velocity in the medium-strength/weak shockwave regime.

2.3. Finite-element analysis

As will be discussed later, application of the two aforementioned computational analyses will result in the prediction of the optimal polyurea soft-segment molecular weight at different shockwave strengths. To validate these predictions more directly, a simple finite-element analysis of a problem involving impact of an airborne shockwave with a polyurea protective structure, generation and propagation of shockwaves and release-waves within polyurea, and the interaction of these waves with the polyurea/scalp interface is conducted. Since details of this analysis can be found in Ref. [10], they will only be briefly overviewed here.

The geometrical/meshed model used in this portion of the work is of an elongated rectangular parallelepiped shape, consisting of three distinct material domains stacked (going from left to right) in the axial direction as follows: (a) an air-filled Eulerian domain; (b) a polyurea-filled Lagrangian domain; and (c) a scalp-filled Lagrangian domain. This type of geometrical model is often referred to as the "core sample" and it represents a portion of the air/helmet/head finite-element model often used in the finite-element analysis of blast-induced TBI [11]. To mimic blast-loading conditions, lateral displacements/velocities are set to zero. A blastwave of a given strength is generated at the leftmost face of the computational domain by prescribing the appropriate time-dependent pressure and axial-velocity boundary conditions. The incident right-propagating blastwave ultimately reaches the air/polyurea interface, at which point a right-propagating shockwave is introduced in polyurea (as well as a reflected left-propagating blastwave in air). After the shockwave reaches the polyurea/scalp interface, a right-propagating shockwave is generated within the scalp domain (while a left-propagating release-wave is generated within the polyurea). It is the ratio of the incident-blastwave peak pressure to the scalp-borne transmitted-shockwave peak axial stress that is used as a measure of the shockwave-attenuation capacity of the polyurea intermediary.

To solve the governing (mass, linear momentum and energy conservation) equations along with the material constitutive equations and the equations defining the initial, boundary, contact and kinematic-constraint conditions, a second-order accurate, transient non-linear dynamics, fluid-structure interaction finite-element analysis is employed. Within this approach, the potential problems associated with large motions and deformations of the fluid (air, in the present case), are avoided by treating the fluid-filled region as an Eulerian (control-volume) region. On the other hand, the solid structure (the polyurea/scalp assembly, in the present case), which experiences considerably less motion and deformation, is analyzed using a Lagrangian scheme. Details regarding the material models and various contact algorithms used can be found in Ref. [11].

3. Results

An example of the coarse-grained computational results revealing the operation of the mechanism for the "shockwave-capture-and-neutralize" effect by the trailing release-wave in P650 is depicted in Figure 5. It is seen that the trailing release-wave propagates faster than the leading shockwave, and that, by 20.61 ps post-shockwave-initiation time, the release-wave has effectively captured the shock-wave and has begun to neutralize it. It should be noted that the corresponding all-atom computational results are obtained (but not shown here) for higher shockwave strengths.

The effect of polyurea soft-segment molecular weight and the shockwave strength (as quantified by the imposed particle axial-velocity) on the ratio of the release-wave speed to the shockwave speed, is depicted in Figure 6. The results shown in this figure are obtained from the particle-velocity vs. axial-position vs. post-shockwave-initiation-time results (like the ones displayed in Figure 5). These results were generated using all-atom (for higher-strength shockwaves) and coarse-grained (for intermediate-strength and weak shockwaves) computational analyses. It should be noted that the release-wave and shockwave speeds can be determined experimentally using the acoustic pulse-echo-overlap method [12] and the flyer-plate impact test [13], respectively. Unfortunately, no

experimental data for the release-wave and the shockwave speeds as a function of the polyurea soft-segment molecular weight are presently available in the open literature.



Figure 5. An example of the capture and neutralization of a slower-propagating leading shockwave by a faster-propagating trailing release-wave in nano-segregated polyurea P650.

It should be noted that the larger is the ratio of release-wave speed to shockwave speed, the more efficient is the capture-and-neutralize shock-mitigation mechanism. Examination of the results displayed in Figure 6 reveals that, at each shockwave-strength level, there is an optimum soft-segment molecular weight, the weight which yields the largest value of the speed ratio.



Figure 6. Effects of polyurea soft-segment molecular weight and the shockwave-strength (as quantified by the upstream imposed particle axial-velocity) on the ratio of release-wave speed to shockwave speed.

The effect of polyurea soft-segment molecular weight and the shockwave strength (as quantified by the imposed particle axial-velocity) on the ratio of the incident-blastwave peak pressure to the scalp-borne transmitted-shockwave peak axial stress, is depicted in Figure 7. It should be noted that these results were obtained using the aforementioned transient non-linear dynamics, fluid-structure interaction finite-element analysis, and the recently reported polyurea-chemistry-dependent material model [14–18]. Also, it should be recalled that larger is the ratio of incident-blastwave peak pressure to the scalp-borne transmitted-shockwave peak axial stress, the higher is the shock-mitigation capacity of polyurea. Examination of the results displayed in Figure 7 reveals that, at each shockwave-strength level, there is an optimum soft-segment molecular weight, the weight which yields the largest value of the pressure-to-stress ratio and, thus, the largest shock-mitigation capacity of polyurea.



Figure 7. Effects of polyurea soft-segment molecular weight and the shockwave-strength (as quantified by the upstream imposed particle axial-velocity) ratio of incident-blastwave peak pressure the scalp-borne on the to transmitted-shockwave peak axial stress.

Results presented in Figures 6 and 7 are next used to generate the results displayed in Figure 8. In each case, at a given level of the shockwave strength, the soft-segment molecular weight associated with the maximum value of the contour quantity is identified and plotted in Figure 8. The two resulting functional relationships are labeled as "Molecular-level" (in the case of the results depicted in Figure 6) and "Finite-element" (in the case of the results shown in Figure 7). A comparison of the two sets of results shows that both the all-atom/coarse-grained molecular-level calculation-based results and their finite-element counterparts are in reasonably good agreement. This finding suggests that the employed multi-length-scale computational approach for the design of polyurea for maximum shockwave-mitigation performance yields reliable results and that it could be used to complement and, perhaps, partially substitute the traditional purely-empirical

material-development efforts. Furthermore, the present computational approach enabled a direct comparison of the computational efficiencies of the all-atom, coarse-grained and finite-element methods. It was found that the computational speed roughly increases by an order of magnitude going from the all-atom to the coarse-grained and then to the finite-element methods.

The results displayed in Figure 8 suggest that the optimal polyurea soft-segment molecular weight is approximately in the 490–535 a.m.u. range. Since polyurea containing seven tetra-methylene-oxide units and having a molecular weight of 504 a.m.u. falls in this range, it could be considered as having the optimal chemistry with respect to its shock-mitigation capability.



Figure 8. Identification of the optimal polyurea soft-segment molecular weight with respect to its maximum shockwave-mitigation capacity at different shockwave-strengths (as quantified by the upstream imposed particle velocity).

4. Conclusion

Based on the results obtained in the present work, the following summary remarks and main conclusions can be drawn:

- 1. Combined all-atom/coarse-grained molecular-level computations are used to identify optimum polyurea chemistry for maximum shockwave-mitigation performance, over a range of shockwave strengths.
- 2. Predicted optimal polyurea chemistries are more directly validated by carrying out a finite-element analysis of a problem involving interaction of a blastwave with a polyurea protective structure placed in front of the protected scalp-like domain.
- 3. Following the recent reports, within the approach employed, the dominant shockwave-mitigation mechanism is taken to be based on the capture-and-neutralize effect of the release-wave.
- 4. To quantify the shockwave-mitigation capacity of polyurea for this type of shockwave-mitigation mechanism, the appropriate shockwave-mitigation metrics are introduced.

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5. The results obtained indicate that the optimal soft-segment molecular weight of polyurea is approximately in a 490–535 a.m.u. range, suggesting that polyurea containing seven tetra-methylene-oxide units in its soft segment possesses nearly optimal chemistry.

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Conflict of Interest

The authors declare that there are no conflicts of interest related to this study.

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